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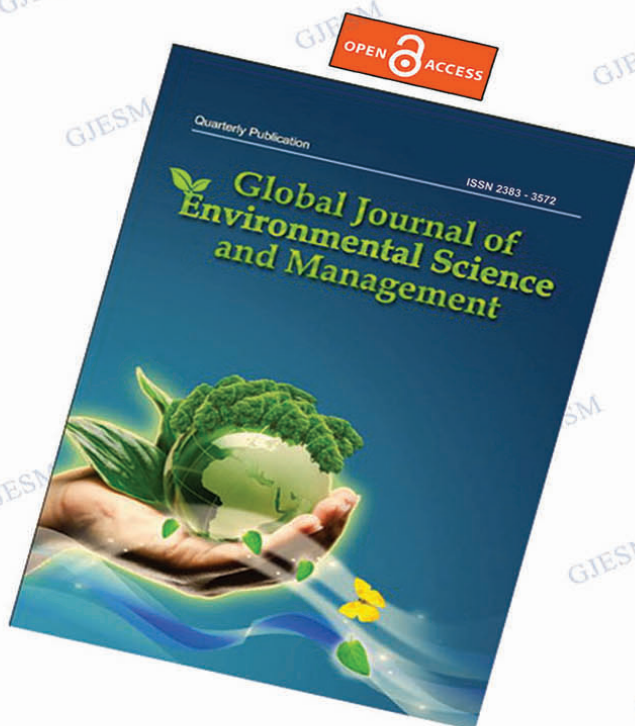
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ORIGINAL RESEARCH PAPER

Application of amorphous zirconium (hydr)oxide/MgFe layered double hydroxides composite in fixed-bed column for phosphate removal from water

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ABSTRACT

BACKGROUND AND OBJECTIVES: Fixed-bed column has been considered an industrially feasible technique for phosphate removal from water. Besides the adsorption capacity, the effectiveness of an adsorbent is also determined by its reusability efficiency. In this study, phosphate removal by a synthesized amorphous zirconium (hydr)oxide/MgFe layered double hydroxides composite in a fixed-bed column system was examined.

METHODS: The effects of flow rate, bed height, phosphate concentration, solution pH, and adsorbent particle size on the phosphate adsorption ability were examined through a series of continuous adsorption experiments. The appropriate breakthrough curve models, phosphate adsorption from real anaerobic sludge and synthetic seawater, column regeneration and reusability, and adsorption mechanism were also investigated for practical application feasibility.

FINDINGS: The results showed that the increased bed height and phosphate concentration, and reduced flow rate, pH, and adsorbent particle size were found to increase the column adsorption capacity. The optimum adsorption capacity of 25.15 mg-P/g was obtained at pH 4. The coexistence of seawater ions had a positive effect on the phosphate adsorption capacity of the composite. Nearly complete phosphate desorption, with a desorption efficiency of 91.7%, could be effectively achieved by 0.1 N NaOH for an hour. Moreover, the initial adsorption capacity was maintained at approximately 83% even after eight adsorption-desorption cycles, indicating that the composite is economically feasible. The high phosphate adsorption capacity of the composite involves three main adsorption mechanisms, which are electrostatic attraction, inner-sphere complexation, and anion exchange, where the amorphous zirconium hydr(oxide) on the surface of the layered double hydroxides likely increased the number of active binding sites and surface area for adsorption.

CONCLUSION: The amorphous zirconium (hydr)oxide/MgFe layered double hydroxides composite, with its high adsorption capacity and superior reusability, has the potential to be utilized as an adsorbent for phosphorus removal in practical wastewater treatment. This study provides insights into the design of amorphous zirconium (hydr)oxide/MgFe layered double hydroxides composite for phosphorus removal and recovery in a practical system.

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INTRODUCTION

Phosphates are vital for the sustainability of organisms and several industrial activities. However, their widespread use, along with the increase in population, has induced superfluous levels of phosphates in the aquatic environment, leading to water eutrophication (Dash *et al.*, 2020). Thus, it is essential to minimize or eliminate phosphorus contaminations from wastewater prior to its discharge into the receiving water body to maintain water quality and improve human health. Additionally, the global increase in food demand and limited carrying capacity of land has increased the dependence on phosphorus, which is used to increase agricultural productivity to fulfill human and livestock needs (March *et al.*, 2016). As phosphorus is a non-renewable natural resource and cannot be substituted by other resources (Wang *et al.*, 2021), the recovery of phosphates from wastewater is important for the sustainability of global phosphorus reserves.

In the last decade, numerous phosphate removal techniques have been designed, ranging from adsorption and chemical precipitation to the application of polyphosphate-accumulating organisms (PAOs) (Ramasahayam *et al.*, 2014). Among these techniques, adsorption is constantly gaining popularity owing to its simplicity and the possibility of phosphorus recovery from wastewater (Zou *et al.*, 2020). However, the cost-effectiveness depends on the reusability of the adsorbent (Kumar *et al.*, 2019). Consequently, adsorbents with high chemical stability are being preferred for phosphorus adsorption because of their stability during the desorption process under various stripping agent chemistry conditions, which usually use a solution with extreme acidity or alkalinity.

Nuryadin *et al.* (2021) synthesized a composite of amorphous zirconium (hydr)oxide/MgFe layered double hydroxides (am-Zr/MgFe-LDH) using a novel two-stage synthesis. A series of batch experiments showed that the uncalcined am-Zr/MgFe-LDH composite with a Zr:Fe ratio of 1:5 showed a high adsorption performance towards phosphates and retained good removal efficiency over eight adsorption-desorption cycles. The high adsorption capacity is related to the high surface area and large number of hydroxyl groups on the composite as a consequence of small crystal size and low

agglomeration of amorphous zirconium (hydr)oxide (am-Zr) in the composite. However, batch settings are not suitable for large-scale applications and continuously generated wastewater (Taka *et al.*, 2020). Therefore, it is important to understand the application of the composite in continuous fixed-bed column adsorption to investigate its performance in practical applications. Fixed-bed column adsorption is very popular in wastewater treatment because of its continuous, high yield, simple, and economical operation and the ability to be scaled-up from the laboratory-scale (Jiang *et al.*, 2018; Kumar *et al.*, 2011). Sun *et al.* (2014) found that MgFe-LDH in a fixed-bed column had a high phosphate adsorption capacity and acceptable desorption efficiency. However, no studies have been conducted on the phosphate removal performance of the am-Zr/MgFe-LDH composite in a fixed-bed column.

In this study, the effects of design parameters essential for practical applications, such as flow rate, bed height, influent phosphate concentration, pH, and adsorbent particle size on the am-Zr/MgFe-LDH composite performance for phosphate removal were examined. Furthermore, the three most widely used models, Thomas, Yoon-Nelson, and modified dose-response (MDR) models were applied to interpret the phosphate adsorption breakthrough data. The applicability of the composite for real anaerobic sludge and phosphate-enriched seawater and its reusability were also evaluated. In addition, an adsorption mechanism of phosphate on the composite was proposed for designing a practical system for the removal and recovery of phosphorus. The objective of the current study is to investigate the phosphate adsorption performance and reusability of a fixed-bed column filled with the am-Zr/MgFe-LDH composite. This study has been carried out in Environmental Planning Laboratory, Graduate School of Sciences and Technology for Innovation, Yamaguchi University, Ube, Japan during 2018-2020.

MATERIALS AND METHODS

Adsorbent and adsorbate preparation

Analytical grade $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (99.0% pure), $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ (98.0% pure), $\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$ (99.0% pure), Na_2CO_3 (99.8% pure), NaOH (97.0% pure) were purchased from Wako Pure Chemical Industries, Ltd. for preparing the am-Zr/MgFe-LDH composite, and used directly in this study without further purification.

A composite with a Zr:Fe molar ratio of 1.5:1 was synthesized using an established method (Nuryadin *et al.*, 2021). Briefly, a solution containing Mg^{2+} and Fe^{3+} ions with a molar ratio of 3:1 was poured into 200 mL of deionized (DI) water. The solution pH was maintained at ~ 10 using a solution of 1 M NaOH and 1 M Na_2CO_3 with a 3:1 volume ratio under vigorous stirring for 30 min. The separated and neutralized gel obtained was then mixed with $\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$ (Zr to Fe molar ratio of 1.5:1) in 500 mL of DI water. Simultaneously, a 25% NaOH solution was used to precipitate the am-Zr under constant stirring until a pH of ~ 10 was obtained. After aging at 353 K for 24 h, the composite was filtered, washed with DI water for neutralization, and then dried in an oven at 353 K for 24 h. The dried composite was ground in a mortar and sieved to the desired particle size.

K_2HPO_4 (99.0% pure) was obtained from Wako Pure Chemical Industries, Ltd., Japan. A phosphate stock solution (500 mg-P/L) was prepared by dissolving an appropriate amount of K_2HPO_4 in 1 L of DI water. The stock solution was diluted to the desired working solution using DI water. The pH of the working solution was adjusted using 0.1 M HCl and NaOH solutions and monitored using a pH meter (Horiba, LAQUA SS131 D-71).

Adsorbent characterization

The X-ray diffraction (XRD) pattern of the adsorbent, with particle size of $d \leq 0.1$ mm, was recorded on a Rigaku Ultima IV Protectus diffractometer using Cu-K α radiation at 40 kV. Fourier transform infrared (FTIR) spectra of the composites before and after adsorption were recorded on a Jasco FT/IR-4600 spectrophotometer. The X-ray photoelectron spectroscopy (XPS) spectra were obtained using a Thermo Scientific K-alpha X-ray photoelectron spectrometer and fitted with XPSPEAK41 and Origin 9.4 software. The high-resolution scans of XPS focused on Mg, Fe, Zr, C, and O. The pH at the point of zero charge (pH_{pzc}) of the composite was investigated using potentiometric mass titration (PMT), as described by Bourikas *et al.* (2003). Three different doses of the composite (5, 15, and 25 g/L) were immersed in Erlenmeyer flasks containing 100 mL of 0.03 M KNO_3 and shaken under an N_2 atmosphere at 200 rpm for 24 h. In each flask, 0.5 mL of 1 M KOH was added to deprotonate sites in the composite surface, and the shaken samples were

titrated using a 0.1 M HNO_3 solution. The titration curve was constructed by plotting the pH value as a function of HNO_3 volume.

Adsorption experiment

Continuous fixed-bed column adsorption experiments were conducted at room temperature (~ 298 K) using a non-jacketed liquid chromatography column (Sigma-Aldrich, Co.) with an inner diameter of 1.0 cm and a length of 10 cm. Fig. 1 shows a schematic diagram of the fixed-bed column system used in this study. A 1 cm thick glass wool was placed on the top and bottom of the sample to prevent the adsorbent from being washed out by the liquid flow. A particular weight (0.7, 1.0, and 1.4 g) of the composite with a maximum particle size of 106 μm was packed into the column to attain the desired bed height (h) (1.0, 1.5, and 2.0 cm). The artificial influent phosphate solution with various concentrations (C_0) (5 mg-P/L, 12 mg-P/L, and 20 mg-P/L) at pH 7 was pumped in an up-flow configuration through the packed composite inside the column at varying flow rates (Q) (1.5, 2.5, and 3.5 mL/min) using a peristaltic pump (EYELA MP-1000). The effects of phosphate solution pH (4, 7, and 10) and composite particle size (d) (≤ 0.1 , 0.1–0.5, and 0.5–1.0 mm) on phosphate adsorption were examined. To obtain the breakthrough curves, the effluent solutions were automatically collected at specified intervals using a fraction collector (Advantec SF-3120) for measuring phosphate concentrations. In this study, the breakthrough time was assigned when the ratio of effluent to influent phosphate concentration (C/C_0) was 10%. An exhaustion time is the time when the influent solution passes through the adsorbent without any significant decrease in concentration (Marzbali and Esmaili, 2017). A preliminary experiment showed that the breakthrough curves became relatively flat after C/C_0 reached approximately 85% and the phosphate adsorption became insignificant. Therefore, the exhaustion time in this study was determined to be the time when C/C_0 reached 85%.

Breakthrough curves of phosphate adsorption were constructed by plotting the ratio of effluent phosphate concentration at an interval time to influent phosphate concentration (C/C_0) as a function of time (t). The breakthrough curves demonstrate the mass transfer characteristics of phosphate onto the

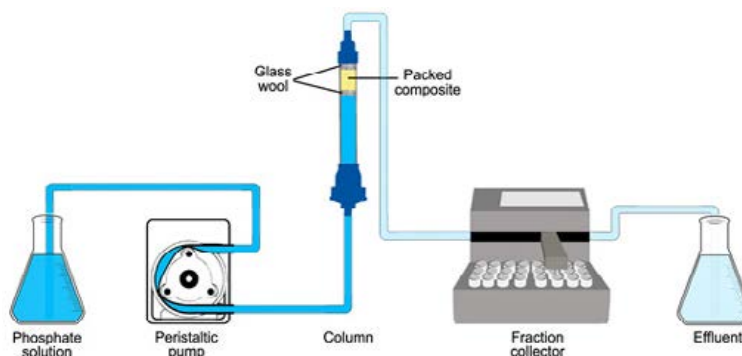


Fig. 1: Schematic diagram of lab-scale fixed-bed column adsorption system

packed composite. The total amount of adsorbed phosphate, $q_{adsorbed}$ (mg), during adsorption and the equilibrium adsorption capacity, q_e (mg/g), were calculated as follows (Eqs. 1 and 2, respectively) (Gouran-Orimi *et al.*, 2018).

$$q_{adsorbed} = \frac{QC_0}{1000} \int_{t=0}^{t=t_e} \left(1 - \frac{C}{C_0}\right) dt \quad (1)$$

$$q_e = \frac{q_{adsorbed}}{m} \quad (2)$$

Where, Q is the flow rate (mL/min), C_0 and C are the influent and effluent phosphate concentrations (mg/L), respectively, t_e is the time required to reach exhaustion time (min), and m is the weight of the composite packed inside the column (g).

Regeneration and reusability experiment

Before the regeneration experiment, the exhausted bed in the column was washed with DI water at a flow rate of 2.5 mL/min for 1 h to remove residual phosphorus. Subsequently, 0.1 M NaOH was pumped in the opposite direction to desorb the adsorbed phosphate. In this study, regeneration was done until the effluent concentration reached approximately 0.5 mg-P/L and it was assumed that the column had been regenerated. In the first cycle of desorption, the desorption effluent solutions were collected at the pre-determined intervals to obtain the desorption curve. The amount of phosphorus eluted from the composite (EAP) (mg/g) was calculated using the following equations (Eqs. 3 and

4) (Nguyen *et al.*, 2015):

$$EAP = \frac{1}{m} q_{desorbed} \quad (3)$$

$$q_{desorbed} = \sum_{q=1}^{n2} CV_q \quad (4)$$

Where, m is the weight of adsorbent in the column (g), $q_{desorbed}$ is the total amount of desorbed phosphate (mg), V_q is the effluent volume of the q -th fraction (L), and $n2$ is the number of the last fraction in the desorption operation. Desorption and regeneration experiments were conducted for eight cycles of the adsorption-desorption process.

RESULTS AND DISCUSSION

Fixed-bed column adsorption studies

The effect of phosphate solution flow rate on the phosphate adsorption on the composite in the column was investigated at three different flow rates (1.5, 2.5, and 3.5 mL/min), where the bed height (1.5 cm) and the influent phosphate concentration (12 mg-P/L) were kept constant. Fig. 2a shows the effect of the flow rate on the breakthrough curve of phosphate adsorption on the composite. The increase in flow rate from 1.5 to 3.5 mL/min resulted in reduction in the breakthrough time from 14.9 to 5.5 h and in the exhaustion time from 24.0 to 8.9 h. The increase in the flow rate led to an increase in the volume speed, leading to a reduction in the time required to reach breakthrough and exhaustion states. The adsorption capacity, calculated using Eq. 2, decreased from 20.40 to 18.23 mg-P/g as the flow rate increased from 1.5 to 3.5 mL/min (Table 1). At a high flow rate, the phosphate anions have a short

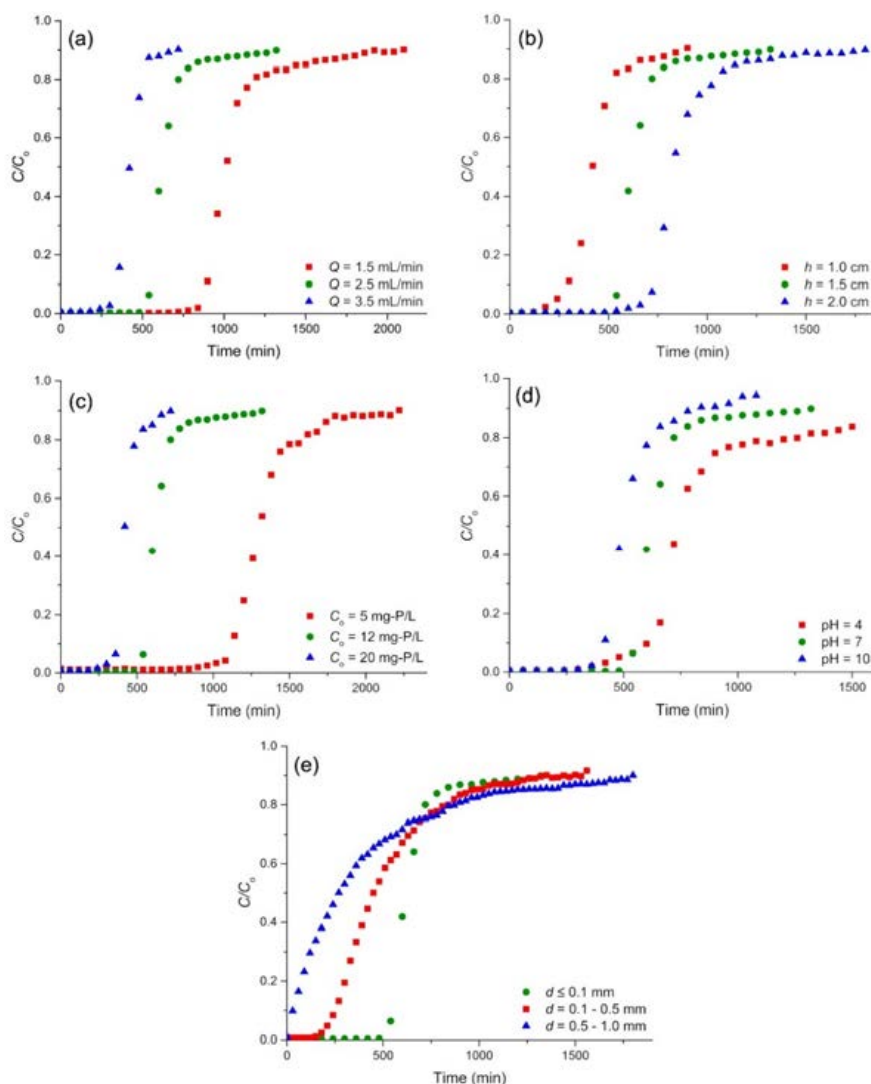


Fig. 2: The Effect of (a) flow rate ($h = 1.5$ cm, $C_0 = 12$ mg-P/L, $\text{pH} = 7$, and $d \leq 0.1$ mm), (b) bed height ($Q = 2.5$ mL/min, $C_0 = 12$ mg-P/L, $\text{pH} = 7$, and $d \leq 0.1$ mm), (c) influent phosphate concentration ($h = 1.5$ cm, $Q = 2.5$ mL/min, $\text{pH} = 7$, and $d \leq 0.1$ mm), (d) solution pH ($h = 1.5$ cm, $Q = 2.5$ mL/min, $C_0 = 12$ mg-P/L, and $d \leq 0.1$ mm), and (e) composite particle size ($m = 1.01$ g/cm³, $Q = 2.5$ mL/min, $C_0 = 12$ mg-P/L, and $\text{pH} = 7$), on the fixed-bed column studies

residence time in the bed, where composite particles in the fixed-bed column have a short contact time with the phosphate anions as the solution flows through the bed (Gouran-Orimi *et al.*, 2018). Consequently, the possibility of phosphate anions passing through the column before being completely adsorbed increased because of the insufficient contact time for adsorption equilibrium. In addition, the steepness of the breakthrough curves increased with the increase in flow rate. The different slope of the breakthrough

curves was due to the retention process. A high flow rate led to an increase in mass transfer rate, increasing the amount of phosphate captured onto the adsorbent surface in the beginning of operation. This caused an early saturation with a steeper slope given in the breakthrough curves.

Breakthrough curves resulting from fixed-bed column experiments at different bed heights (1.0, 1.5, and 2.0 cm) with a constant flow rate (2.5 mL/min) and influent phosphate concentration (12 mg-

Table 1: Fixed-bed column parameters for phosphate adsorption on am-Zr/MgFe-LDH composite

No..	Q (mL/min)	h (cm)	C_o (mg-P/L)	m (g)	pH	d (mm)	t_b (h)	τ (h)	t_e (h)	q_e (mg-P/g)
1	1.5	1.5	12	1.01	7	≤ 0.1	14.9	17.33	24.0	20.40
2	2.5	1.5	12	1.01	7	≤ 0.1	9.1	10.65	13.5	19.04
3	3.5	1.5	12	1.01	7	≤ 0.1	5.5	7.19	8.9	18.23
4	2.5	1.0	12	0.68	7	≤ 0.1	4.8	7.24	10.7	18.75
	2.5	1.5	12	1.01	7	≤ 0.1	9.1	10.65	13.5	19.04
5	2.5	2.0	12	1.36	7	≤ 0.1	12.1	14.48	19.0	19.56
6	2.5	1.5	5	1.01	7	≤ 0.1	18.7	22.28	28.7	17.08
	2.5	1.5	12	1.01	7	≤ 0.1	9.1	10.65	13.5	19.04
7	2.5	1.5	20	1.01	7	≤ 0.1	4.4	6.04	7.8	19.13
8	2.5	1.5	12	1.01	4	≤ 0.1	10.0	12.82	25.0	25.15
	2.5	1.5	12	1.01	7	≤ 0.1	9.1	10.65	13.5	19.04
9	2.5	1.5	12	1.01	10	≤ 0.1	6.9	8.63	11.5	15.73
	2.5	1.5	12	1.01	7	≤ 0.1	9.1	10.65	13.5	19.04
10	2.5	1.8	12	1.01	7	0.1–0.5	4.2	7.51	16.0	15.80
11	2.5	2.0	12	1.01	7	0.5–1	0.5	4.49	19.0	12.45

P/L) are depicted in Fig. 2b. The figure demonstrates that the breakthrough and exhaustion times were extended when the bed height increased. Table 1 shows that the adsorption capacity increased from 18.75 to 19.56 mg-P/g as the bed height increased from 1.0 to 2.0 cm. This was predictable as there were more active binding sites at a higher bed height, and these increased the adsorption areas (Mohan and Dutta, 2020). Furthermore, a higher bed height provides a longer packed composite through which the phosphate anions pass, which allows a longer residence time of phosphate, enabling it to be deeply adsorbed within the composite. The bed height of 2.0 cm had a flatter breakthrough curve than the lower bed height, which might be due to a broad mass transfer zone provided by a high bed height.

Fig. 2c shows the adsorption curves of various influent phosphate concentrations (5, 12, and 20 mg-P/L) at a constant bed height (1.5 cm) and flow rate (2.5 mL/min). An increase in influent phosphate concentration from 5 to 20 mg-P/L decreased the breakthrough time from 18.7 to 4.4 h and exhaustion time from 28.7 to 7.8 h. The concentration gradient between the solution and adsorbent surface increased at high phosphate concentrations, which increased the driving force for phosphate adsorption (Lee et al., 2019). Under these conditions, the limited active binding sites in the column were inhibited

rapidly, and saturated conditions were achieved in a short period. This led to a steeper breakthrough curve at higher influent phosphate concentrations. This steeper curve at the high load of phosphate anions signifies that the am-Zr/MgFe-LDH is likely favorable for phosphate adsorption. The adsorption capacity increased from 17.08 to 19.13 mg-P/g when the influent phosphate concentration was raised from 5 to 20 mg-P/L. A higher influent concentration of phosphate can deliver more phosphate anions into the outer and inner surface of the composite to attach with the adsorption sites, which is possibly the reason for an increase in adsorption capacity with influent concentration increasing. In addition, a high concentration gradient at high influent phosphate concentration can overcome the mass transfer resistance in the adsorbent surface, leading to an increase in the adsorption capacity.

Another important parameter that affects the dynamic phosphate adsorption process in a fixed-bed column is the pH solution. Fig. 2d shows the effect of phosphate solution pH (4, 7, and 10) on phosphate adsorption breakthrough curves, while maintaining a constant bed height (1.5 cm), flow rate (2.5 mL/min), and influent phosphate concentration (12 mg-P/L). It can be seen clearly that the breakthrough time was reached in a longer time at a lower pH. The breakthrough time increased from 6.9 to 10.0

h with decreasing pH from 10 to 4. As shown in Table 1, a higher adsorption capacity (25.15 mg-P/g) was obtained at a lower pH (acidic solution). The adsorption capacity decreased significantly when the influent pH increased from 4 to 10, signifying that pH played a key role which affected the physical-chemical interaction between the phosphate and composite surface. The adsorbent surface was positively charged at low pH, attracting the negatively charged phosphate anions. Conversely, the number of hydroxyl ions (OH⁻) increased at higher pH, which competed with phosphate anions to occupy more active binding sites on the adsorbent surface and led to the reduction of phosphate adsorption onto the composite surface (He *et al.*, 2016). The lower pH showed a broader tailing edge in the breakthrough curve shape, reflecting that the phosphate adsorption onto composite bed in lower pH was extended with a slow adsorption.

The effect of composite particle size on the breakthrough curve of phosphate adsorption was investigated by isolating the composite into three groups of particle sizes using sieves with apertures of different sizes (≤ 0.1 mm, 0.1–0.5 mm, and 0.5–1.0 mm). The bed height (1.5 cm), flow rate (2.5 mL/min), and influent phosphate concentration (12 mg-P/L) were kept constant. Fig. 2e shows the breakthrough curves of phosphate adsorption resulting from different composite particle sizes. The bed performance of the smallest particle size (≤ 0.1 mm) was better than that of the larger particle sizes, particularly for the initial part of the curve. The reduction in particle size led to a longer breakthrough time but a shorter exhaustion time. As the particle size of the composite increased, the surface area per unit volume decreased. Consequently, the number

of active binding sites on the surface of composite with large particle size, that directly interact with the phosphate ions in the solution, smaller than the composite with a lower particle size resulted in an earlier breakthrough time corresponding to the completion of surface adsorption. However, after the breakthrough point was achieved in the large particle composite, the pore adsorption continued at a lower rate, which resulted in a delayed exhaustion time. Additionally, for smaller particle sizes with a higher surface area (Oguz, 2017), phosphate adsorption mostly occurred on the surface of the composite with a large number of exposed active binding sites. A decrease in particle size appears to have increased the sharpness of the breakthrough curves and delayed the breakthrough time.

Table 2 summarizes the phosphate adsorption capacity of am-Zr/MgFe-LDH composite in this study and calcined MgFe-LDH as well as other Zr-based adsorbents in fixed-bed column adsorption. It was demonstrated that am-Zr/MgFe-LDH composite was found to have higher adsorption capacity than calcined MgFe-LDH and positively comparable to most of the Zr-based adsorbents reported in the literature. This result confirmed that the combination of am-Zr and MgFe-LDH resulted in favorable modification in the adsorbent surface for effective phosphate adsorption. The high phosphate adsorption capacity of am-Zr/MgFe-LDH composite can be associated with the development of a high amount of hydroxyl groups in the adsorbent surface by am-Zr, where the hydroxyl groups act as the effective binding sites for phosphate anions.

Breakthrough curve modeling

Generally, the modeling of breakthrough curves

Table 2: Comparison of the phosphate adsorption capacity of am-Zr/MgFe-LDH composite with various adsorbents in fixed-bed column

Adsorbent	Experimental condition					q_e (mg-P/g)	Reference
	Q (mL/min)	h (cm)	C_o (mg-P/L)	pH	d (mm)		
Zr-FPS	1.77	1.6	0.411	2.01	-	1.73	Awual <i>et al.</i> , 2011
Zirconium ferrite	0.083	-	20	7	0.7	2.6	Biswas <i>et al.</i> , 2008
A-Zr-NP	6	4.8	20	5.4	-	5.59	Husein <i>et al.</i> , 2017
Zr(IV)-loaded SOW gel	0.083	-	20	7	0.075–0.15	10	Biswas <i>et al.</i> , 2008
Zr(IV)-loaded okara	12	9	5.5	7.6	0.3–0.15	14.97	Nguyen <i>et al.</i> , 2015
Zr(IV)-loaded okara	12	23	5.6	3	1–0.6	16.43	Nguyen <i>et al.</i> , 2015
Calcined MgFe-LDH	0.4	12	20	-	0.075–0.028	21.09	Sun <i>et al.</i> , 2014
CS-Zr-PEPA	6	12	2.4	6.5	-	32.50	Chen <i>et al.</i> , 2020
Am-Zr/MgFe-LDH	2.5	1.5	12	4	≤ 0.1	25.15	Present study

resulting from fixed-bed column adsorption is essential for designing a column adsorption system for practical applications. To describe the behavior of dynamic phosphate adsorption onto the am-Zr/MgFe-LDH composite in a fixed-bed column system, three nonlinear mathematical model equations, Yoon-Nelson (Eq. 5) (Yoon and Nelson, 1984), Thomas (Eq. 6) (Thomas, 1944), and MDR models (Eq. 7) (Yan *et al.*, 2001) were used.

$$\ln\left(\frac{C}{C_0 - C}\right) = k_{YN}t - \tau k_{YN} \quad (5)$$

$$\ln\left(\frac{C_0}{C} - 1\right) = \frac{k_{Th}q_{Th}m}{Q} - k_{Th}C_0t \quad (6)$$

$$\ln\left(\frac{C}{C_0 - C}\right) = a \ln\left(\frac{C_0Qt}{1000}\right) - a \ln q_{mdr}m \quad (7)$$

Where, k_{Th} and k_{YN} are the Thomas kinetic coefficient (L/mg min) and the Yoon-Nelson kinetic coefficient (1/min), respectively, q_{Th} and q_{mdr} are the predicted adsorption capacities (mg/g), τ is the time required for 50% phosphate adsorption breakthrough (min), and a is an MDR model parameter.

The main parameters of each mathematical model obtained by non-linear fitting of the breakthrough data under different experimental conditions are presented in Table 3. A simple Yoon-Nelson model was applied to examine the phosphate adsorption behavior of the packed composite. This model assumes that the decrease in the adsorption probability of

each molecule is proportional to the adsorption probability of the molecule and the probability of molecule breakthrough on the adsorbent (Sotelo *et al.*, 2012). Although this model is based on the adsorption probability, the R^2 (0.864–0.984) values showed a good fit with the experimental data. Additionally, the predicted values of τ were similar to those obtained from experiments (Table 1), except for d values of 0.1–0.5 and 0.5–1.0 nm, where the breakthrough curves deviated from ideal adsorption system with an S-shaped profile characteristic. The τ values decreased with increasing flow rate, influent phosphate concentration, pH, and adsorbent particle size, and by decreasing the bed height. Conversely, the k_{YN} values indicated opposite trends in τ values. For instance, increasing the flow rate or decreasing the bed height resulted in an increase in phosphate ions passing the packed adsorbent for a given period of time. Consequently, k_{YN} increased and τ decreased.

The Thomas and MDR models were also applied to the experimental phosphate adsorption curves to depict the entire breakthrough curve. The Thomas model is based on the assumption that adsorption is only controlled by the surface reaction between the adsorbate and adsorbent surface, where the effects of intra-particle diffusion and external film resistances are neglected (Chitto and Sutherland, 2020). The MDR model was developed based on mathematical issues to reduce the error resulting from the Thomas model (Sana and Jalila, 2017). The values of predicted q_{Th} and q_{mdr} were close to the experimental q values (percentage errors < 3%), except for d values of 0.5–1.0 nm. Additionally, their trends in response to

Table 3: Yoon-Nelson, Thomas, and MDR model parameters for phosphate adsorption by am-Zr/MgFe-LDH composite in fixed-bed column at various experimental conditions

Experimental conditions No.	Yoon-Nelson			Thomas			MDR		
	k_{YN} ($\times 10^{-2}$)	τ	R^2	k_{Th} ($\times 10^{-3}$)	q_{Th}	R^2	q_{mdr}	a	R^2
1	0.96	17.33	0.958	0.76	19.91	0.958	19.94	8.157	0.966
2	1.59	10.65	0.962	1.29	19.02	0.962	19.01	8.583	0.969
3	1.92	7.19	0.984	1.55	18.44	0.984	18.31	7.796	0.990
4	1.82	7.11	0.961	1.01	19.10	0.972	18.79	4.845	0.985
5	0.95	14.48	0.960	0.76	19.49	0.960	19.45	6.987	0.970
6	0.70	22.28	0.974	1.33	17.16	0.974	17.10	8.693	0.981
7	2.19	6.21	0.968	0.81	19.05	0.964	19.05	6.980	0.975
8	0.81	12.83	0.916	0.48	24.67	0.926	24.66	4.418	0.953
9	1.89	8.40	0.974	1.16	15.84	0.978	15.73	6.840	0.986
10	0.52	8.66	0.943	0.42	15.61	0.943	15.33	2.516	0.985
11	0.25	5.78	0.864	0.21	10.21	0.864	10.80	1.370	0.931

various experimental conditions were similar to those of the experimental q . However, the Thomas model was not completely fit with the experimental curves as the phosphate adsorption on the am-Zr/MgFe-LDH composite involved intra-particle diffusion as the rate-limiting step. The slow intra-particle diffusion of phosphate on the composite pores was signified by the considerably slow adsorption from the C/Co ratio of 0.85 to reach 1 (Chu, 2004). As can be seen from Table 3, the MDR model showed larger R^2 values than those of the Yoon-Nelson and Thomas models for all experimental conditions. This indicates that the phosphate adsorption behavior of the packed am-Zr/MgFe-LDH composite in a fixed-bed column system was better described by the MDR model in each experimental condition. As the MDR model based on the Thomas model, the results suggest that adsorption follows the Langmuir isotherm, which suggests that the driving forces obey second-order reversible reaction kinetics with no axial dispersion in the column (Sana and Jalila, 2017).

Phosphate adsorption from real municipal anaerobic sludge filtrate and synthetic seawater

The anaerobic sludge used in this study was collected from an eastern municipal wastewater treatment plant in Ube, Japan. Before use in the column adsorption test, the sludge was allowed to stand for 24 h for deposition, and the supernatant was filtered using a 0.45 μm filter membrane. The effect of seawater ions on phosphate adsorption in seawater was also investigated using synthetic phosphate-containing seawater (P seawater). As 99% of the seawater electrolyte concentration consists of NaCl, Na_2SO_4 , KCl, MgCl_2 , and CaCl_2 , synthetic seawater was prepared using these salts, corresponding to a salinity of 15 PSU (Zhang *et al.*, 2020). The 15 PSU

salinity was chosen to represent the salinity of the mesohaline estuary (salinity ranges from 5 to 18 PSU), an area that has great potential to be eutrophic and needs attention (Brush, 2009). For comparison, phosphate adsorption was also conducted in synthetic phosphate-containing water (P water). The details of the sludge, synthetic P seawater, and P water are listed in Table 4. In this experiment, an adsorbent particle size range of 0.1–0.5 mm was used to prevent column clogging by extremely small particles present in the sludge filtrate. Each water sample was pumped through the 1.0 g of adsorbent column at flow rate of 2.5 mL/min.

Fig. 3 shows a comparison of the breakthrough curves for phosphate adsorption onto the composite-packed column using real anaerobic sludge filtrate, synthetic P water, and seawater. The earlier breakthrough of phosphate adsorption in anaerobic sludge might be due to the high concentration of carbonate (CO_3^{2-}), which is signified by high alkalinity (Table 4). Nuryadin *et al.* (2021) found that CO_3^{2-} (or bicarbonate) was highly competitive with phosphate for the active binding sites on the adsorption surface. Furthermore, the presence of extremely fine suspended solids that passed through the filter during filtration in the filtrate could interfere with the transfer of phosphate from the bulk liquid to the am-Zr/MgFe-LDH composite surface (Sun *et al.*, 2014). The adsorption capacity of the sludge filtrate (8.99 mg-P/L) was lower than that in synthetic P water (13.12 mg-P/L). However, phosphate adsorption was not negatively affected by the seawater ions, but was enhanced after the breakthrough point passed. The adsorption capacity in synthetic P seawater was 19.99 mg-P/L. The positively charged seawater ions (such as Na^+ , Mg^{2+} , Ca^{2+} , and K^+) in the solution were easily attracted by an adsorbent surface that became more

Table 4: The details of anaerobic sludge filtrate, synthetic P water, and synthetic P seawater used in fixed-bed column phosphate adsorption

Anaerobic sludge filtrate		Synthetic P seawater		Synthetic P water	
Parameter	Value	Parameter	Value	Parameter	Value
pH	8.51	pH	8.10	pH	8.51
Phosphate ($\text{PO}_4\text{-P}$)	14.24 mg/L	Phosphate ($\text{PO}_4\text{-P}$)	14.58 mg/L	Phosphate ($\text{PO}_4\text{-P}$)	14.23 mg/L
TSS	168.67 mg/L	NaCl	10519.20 mg/L		
COD	400.00 mg/L	MgCl_2	2094.64 mg/L		
Chloride (Cl^-)	574.82 mg/L	Na_2SO_4	1704.48 mg/L		
Nitrate ($\text{NO}_3\text{-N}$)	14.58 mg/L	CaCl_2	488.31 mg/L		
Alkalinity (CaCO_3)	109.70 mg/L	KCl	298.21 mg/L		

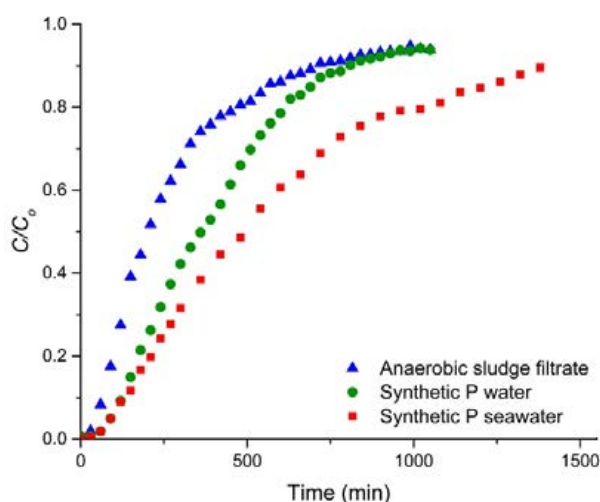


Fig. 3: Breakthrough curves for phosphate adsorption from anaerobic sludge filtrate, synthetic P water, and synthetic P seawater ($m = 1.01$ g, $Q = 2.5$ mL/min, $d = 0.1$ - 0.5 mm)

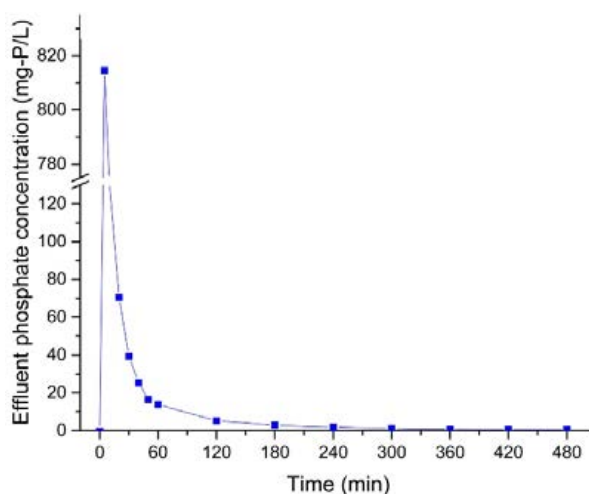


Fig. 4: Continuous phosphate desorption curve on am-Zr/MgFe-LDH composite column by 0.1 N NaOH ($h = 1.5$ cm and $Q = 2.5$ mL/min)

negatively charged after initial phosphate adsorption. This increased positive charge on the adsorbent surface and promoted subsequent phosphate adsorption. According to the results, the composite adsorption capacity reduced due to the highly carbonated wastewater in the continuous adsorption system. However, it exhibited more favorable phosphate adsorption in synthetic seawater.

Column regeneration and reusability

The effectiveness of the adsorbent for practical

applications depends on the adsorption capacity and efficiency of adsorbent regeneration and reusability. However, the reuse of an adsorbent requires a desorption agent to desorb phosphate from the adsorbent for regeneration. To ensure that the adsorbent was saturated with phosphate before the regeneration study, a phosphate solution ($C_0 = 12$ mg-P/L, pH = 7) was pumped ($Q = 2.5$ mL/min) to the column ($h = 1.5$ cm, $d \leq 0.1$ mm) for 24 h. The exhausted composite was regenerated with 0.1 N NaOH at a flow rate of 2.5 mL/min. The effluent

phosphate concentrations in a continuous desorption curve of phosphate at particular time points were determined and are shown in Fig. 4.

The desorption curve was an asymmetrically shaped curve with significantly high desorption at the beginning, followed by a flattened decline. The regeneration study was conducted for 8 h when the effluent concentration was 0.51 mg-P/L. Fig. 4 shows that the maximum phosphate concentration was obtained within 5–10 min, indicating that the majority of the phosphate amount desorbed during this period. The flattened curve was observed after a one-hour desorption, indicating that almost complete desorption was achieved after 1 h with a desorption efficiency ($100\% \times q_{\text{desorbed}}/q_{\text{adsorbed}}$) of 91.7% and an effluent concentration of 13.82 mg-P/L. However, a total regeneration of 98.8% was achieved after an eight-hour operation. The regenerated column was reused for subsequent adsorption to assess the reusability of the column, and its breakthrough curves and regeneration capacity after eight adsorption-desorption cycles are shown in Fig. 5. A significant decrease in the column adsorption capacity (~8.0%) was observed in the second cycle, as depicted by the earlier breakthrough point after the first cycle (Fig. 5a). These breakthrough points appeared to shift slowly to an earlier time, signifying a slow decrease in the adsorption capacity after the second cycle (Fig. 5b). The gradual decrease in adsorption performance is attributed to the loss of active binding sites from

the previous adsorption process, particularly in the first cycle, by strongly attached phosphate, which is difficult to desorb. The decrease in adsorption capacity from the first to the eighth cycle was approximately 17%, indicating that the composite may be suitable for practical applications.

Adsorption mechanism

The possible mechanism of phosphate adsorption by the composite was further investigated using FTIR, XRD, point of zero charge (PZC), and XPS analyses. The functional groups of the composite before and after adsorption were identified using FTIR spectroscopy, and the spectra are shown in Fig. 6a. The intense and broadband centered at 3430 cm^{-1} and the band at 1640 cm^{-1} could be attributed to strong OH stretching and structural OH bending vibrations due to the presence of structural hydroxyl groups and physically adsorbed water molecules in the samples, respectively (Su *et al.*, 2013; Tang *et al.*, 2018). The bands of the Zr-OH bending vibration were observed at approximately 1353 cm^{-1} and 1565 cm^{-1} , and the band of CO_3^{2-} antisymmetric stretching in the interlayer was detected at 1358 cm^{-1} (Dou *et al.*, 2012; Magri *et al.*, 2019; Zhang *et al.*, 2013). These bands were weakened after phosphate adsorption, indicating that phosphate anions were successfully adsorbed by replacing hydroxyl bonds and by exchanging some carbonate anions in the LDH interlayer. In addition, phosphate adsorption

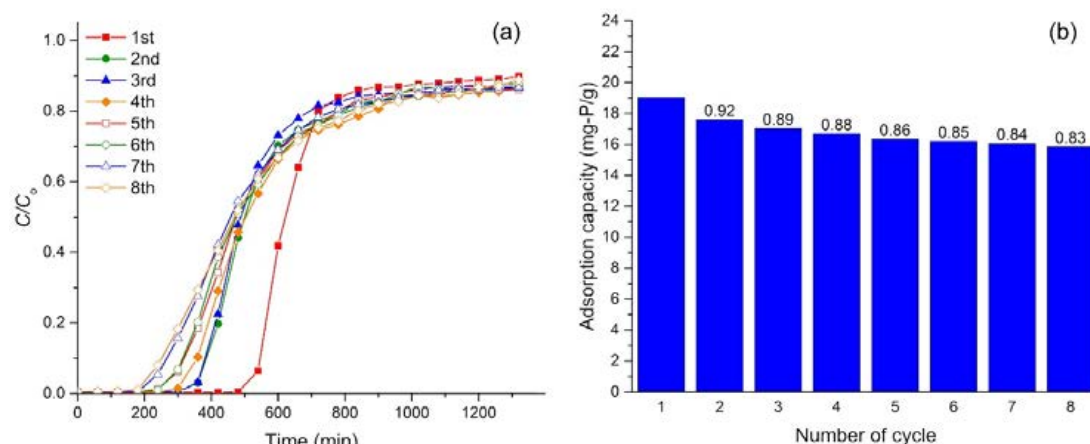


Fig. 5: Column reusability study: (a) phosphate adsorption breakthrough curve on am-Zr/MgFe-LDH composite for each consecutive eight adsorption-desorption cycles at a flow rate 2.5 mL/min, and (b) column adsorption capacity calculated for each adsorption cycle (the numbers above the bar are adsorption retain ratio of phosphate adsorption capacity)

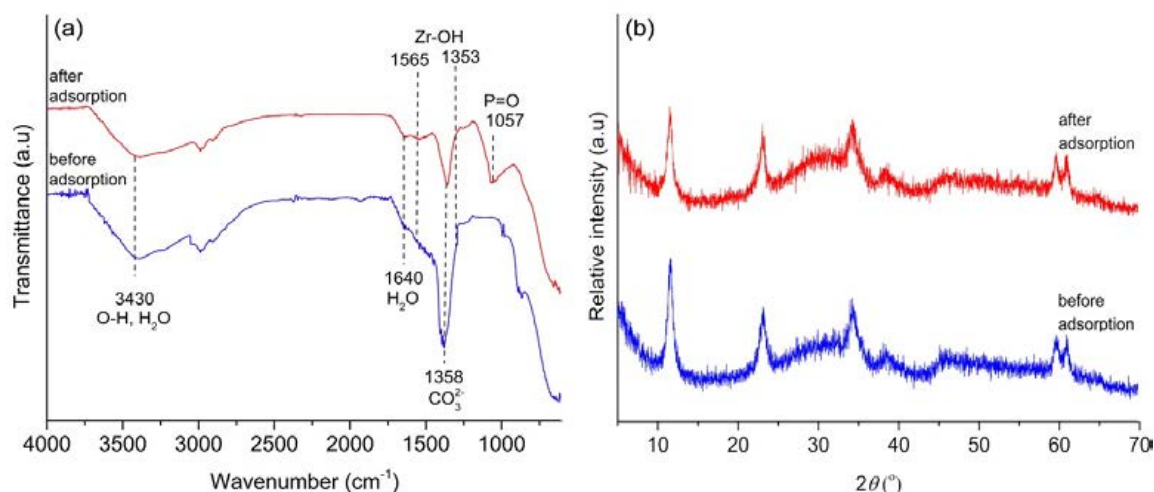


Fig. 6: (a) FTIR spectra and (b) XRD patterns of am-Zr/MgFe-LDH composite before and after adsorption

in the composite was confirmed by the appearance of a new peak near 1057 cm^{-1} after the adsorption process, which could be attributed to the P=O bond (Zha *et al.*, 2018).

Fig. 6b shows a comparison between the XRD patterns of the composite, before and after phosphate adsorption. The patterns exhibited the presence of an LDH structure with a typical halo pattern of amorphous zirconium hydroxide. There was no significant change in the structure and no additional characteristic peak was observed in the patterns after the adsorption process. However, the structure showed a lower crystallinity. The decrease in crystallinity confirmed the occurrence of phosphate adsorption, which caused a disordered structure in the composite surface.

The phosphate adsorption mechanism was further evaluated by deconvolution of the XPS spectra. As depicted in Fig. 7a and Fig. 7b, the peaks of the Mg 1s and Fe 2p spectra shifted to higher binding energies after phosphate adsorption by about 0.2 eV for Mg 1s, 0.4 eV for Fe 2p₁, and 0.2 eV for Fe 2p₃. This enhancement of binding energy values was caused by electron withdrawal from the negatively charged O atoms of the phosphate species to the valence band of Mg s₁ and Fe 2p and the formation of Mg-O-P and Fe-O-P through surface complexation (Hong *et al.*, 2019; Liu *et al.*, 2019). This indicated that the surface of LDH in the composite was also involved in the phosphate adsorption process. As presented

in Fig. 7c, the two peaks centered at 181.85 eV and 184.20 eV were assigned to Zr-O-Zr bonds of Zr 3d_{5/2} and Zr 3d_{3/2} in the composite, respectively. The main peaks of Zr 3d were shifted to a higher binding energy of approximately 0.2 eV after phosphate adsorption, indicating that electron transfer occurred in the valence band and Zr-O-P inner-sphere complexation was formed by substituting the hydroxyl groups of am-Zr (Zhang *et al.*, 2019). This shift also can be attributed to the overlap from two new Zr-P peaks in higher binding energies (182.35 eV and 184.79 eV) which formed through ligand exchange (Zong *et al.*, 2016). These indicated that phosphate was adsorbed chemically on the composite surface. As shown in Fig. 7d, the C 1s spectra of the composite can be divided into four different peaks: C-C (284.5 eV), C-O (285.2 eV), C=O (288.8 eV), and metal carbonate (M-CO₃) (289.6 eV) (Lai *et al.*, 2020). After phosphate adsorption, the relative area of the metal carbonate decreased from 14.7% to 5.2%. This signified that the carbonate in the LDH interlayer took part in the phosphate adsorption process via ion and ligand exchange.

Nuryadin *et al.* (2021) reported that the adsorption was high at low pH (particularly at pH 2 and 3) and decreased at higher pH (notably at pH 10). To explain this condition, the pH_{pzc} of the composite was investigated by PMT, and the plotted experimental curves are shown in Fig. 8. The curves show an intersection point at a pH of approximately 9.7, which

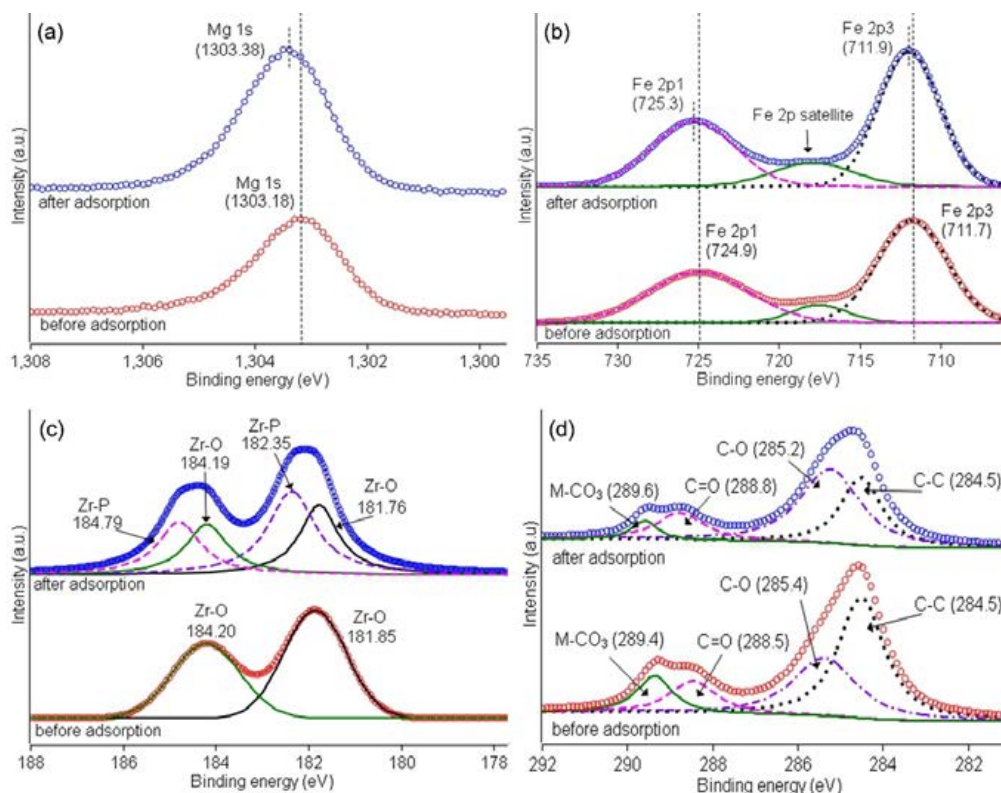


Fig. 7: XPS analysis of am-Zr/MgFe-LDH composite before and after phosphate adsorption: (a) Mg 1s, (b) Fe 2p, (c) Zr 3d, and (d) C 1s

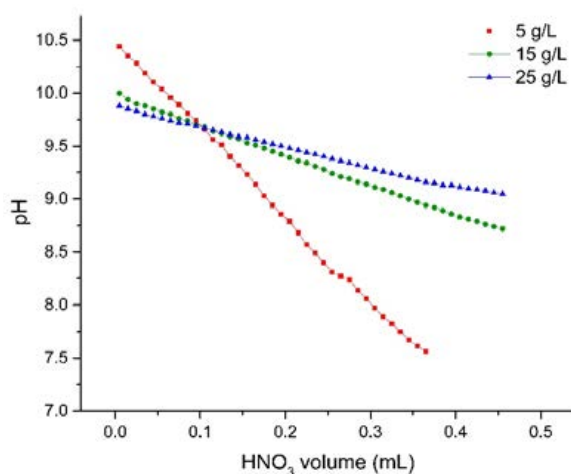


Fig. 8: Potentiometric mass titration curves of am-Zr/MgFe-LDH composite samples using 0.1 M HNO_3

was identified as the pH_{pzc} of the composite. This indicates that the significant decrease in adsorption efficiency at pH 10 was triggered by the negatively

charged composite surface as the solution pH was higher than pH_{pzc} . This result indicates that the electrostatic bond also contributed to the phosphate

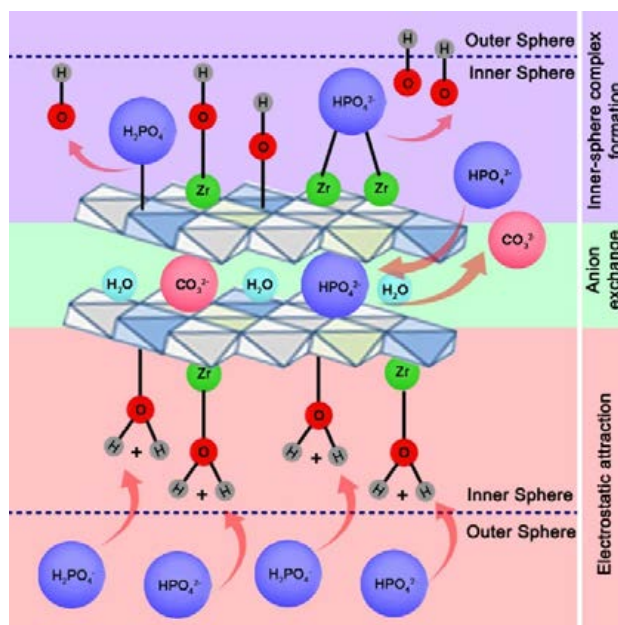


Fig. 9: Proposed mechanism of phosphate adsorption on am-Zr/MgFe-LDH composite

adsorption mechanism between the composite surface and phosphate anions, particularly at pH values lower than 9.7.

Based on the analysis of adsorption in varying pH solutions, FTIR spectra, XRD patterns, and XPS spectra, the potential adsorption mechanisms involved in the composite during the adsorption process were found to mainly consist of electrostatic attraction between the positively charged adsorbent surface and phosphate anions (particularly at low pH), inner-sphere complex formation between metal hydroxides and phosphate anions on the adsorbent surface, and exchange of carbonate anions in the LDH interlayer. The proposed phosphate-adsorption mechanism is illustrated in Fig. 9. The presence of am-Zr on the surface of the LDH likely increased the adsorption capacity of the composite. The zirconium species, which is in amorphous form, have a massive number of hydroxyl groups as the active binding sites for the phosphate anions. Moreover, the crystallization of am-Zr on the LDH matrix was presumably suppressed during synthesis, leading to the formation of nanosized am-Zr (Nuryadin *et al.*, 2021). The nanosized materials are preferable for adsorbent because the crystal has a large surface area for adsorption sites. Therefore, we suggest that

the high phosphate adsorption of the composite is mainly associated with the presence of highly hydrated am-Zr collaborated with the ability of MgFe-LDH for adsorbing phosphate by anion exchange and surface adsorption.

CONCLUSION

This study involves the application of a synthesized am-Zr/MgFe-LDH composite as a phosphate sorbent in a continuous fixed-bed column system as well as the investigation of the feasibility of its reusability using NaOH solution in regeneration process. The results of the column experiments showed that the phosphate adsorption capacity of the composite was affected by the flow rate, bed height, influent phosphate concentration, influent pH, and composite particle size. The increase in the flow rate, pH, and adsorbent particle size, decreased the phosphate adsorption capacity of the composite. Contrarily, the phosphate adsorption capacity of the composite increased with increasing bed height and influent phosphate concentration in the fixed-bed column system. The highest adsorption capacity of 25.15 mg-P/L was obtained when the influent pH was 4, indicating that the adsorption of phosphate onto the composite surface is favorable in acidic. From the

non-linear fitting of the breakthrough data, the MDR model was found to be the best model for predicting the experimental phosphate adsorption behavior of a packed composite in a fixed-bed column system, as compared to the Yoon-Nelson and Thomas models. The phosphate adsorption capacity of the composite decreased when it was used in wastewater with a high (bi)carbonate concentration. However, it exhibited a more effective phosphate adsorption in synthetic seawater. The column regeneration studies revealed that 91.7 % of adsorbed phosphorus was desorbed during the one-hour desorption, and 83% of the fresh composite adsorption ability could be retained even after eight adsorption-desorption cycles. The pH_{pzc} , FTIR, XRD, and XPS analyses indicated that electrostatic attraction, inner-sphere complexation, and anion exchange played an important role in phosphate adsorption. The am-Zr/MgFe-DH composite has the potential to be utilized as an adsorbent for phosphorus removal from aqueous solutions as it has high phosphate adsorption capacity and reusability. Although, this study provides helpful information for the design of a practical system for phosphorus removal and recovery, some considerations should be taken to improve the performance of the composite adsorption system in future work. First, the application of economical pretreatment to reduce (bi)carbonate from wastewater before process, such as chemical precipitation, since (bi)carbonate is the most competitive anion for phosphate adsorption onto the composite. Second, the use of adsorption byproduct to create an economic adsorption system, such as the processing of desorbed phosphorus in the regeneration process to a valuable product like hydroxyapatite, or the application of saturated composite as the slow release phosphate fertilizer.

AUTHOR CONTRIBUTIONS

A. Nuryadin performed the literature review, conception and design, acquisition of data, analysis and interpretation of data, drafting of the manuscript, and statistical analysis. T. Imai performed conception and design, critical revision of the manuscript for important intellectual content, supervision, and support for administrative, technical, and material.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

a	A modified dose-response model parameter
<i>A-Zr-NP</i>	Alginate-/Zirconium-Grafted Newspaper Pellets
<i>am-Zr</i>	amorphous zirconium (hydr)oxide
<i>am-Zr/MgFe-LDH</i>	amorphous zirconium (hydr)oxide/MgFe layered double hydroxides
<i>C</i>	Carbon
<i>C</i>	Effluent concentration
C_o	Influent concentration
Ca^{2+}	Calcium ion
$CaCl_2$	Calcium chloride
$CaCO_3$	Calcium carbonate
<i>Cl⁻</i>	Chloride ion
<i>cm</i>	Centimeter
CO_3^{2-}	Carbonate ion
<i>COD</i>	Chemical oxygen demand
<i>CS-Zr-PEPA</i>	Polyethylene polyamine grafted chitosan-zirconium(IV) composite beads
<i>Cu-Kα</i>	Copper K-alpha
<i>d</i>	Particle size
<i>DI</i>	Deionized
<i>EAP</i>	Eluted amount of phosphorus
<i>eV</i>	Electron volt
<i>Fe</i>	Iron
$FeCl_3 \cdot 6H_2O$	Iron(III) chloride hexahydrate
<i>FTIR</i>	Fourier transform infrared
<i>g</i>	Gram

h	Hour	OH	Hydroxyl
\bar{h}	Bed height	OH^-	Hydroxyl ion
HCl	Hydrochloric acid	P	Phosphorus
HNO_3	Nitric acid	$P \text{ seawater}$	Synthetic phosphate-containing seawater
K	Kelvin	$P \text{ water}$	Synthetic phosphate-containing water
K^+	Potassium ion	$PAOs$	polyphosphate-accumulating organisms
K_2HPO_4	Dipotassium hydrogen phosphate	pH_{pzc}	pH at the point of zero charge
KCl	Potassium chloride	PMT	Potentiometric mass titration
KNO_3	Potassium nitrate	PO_4-P	Phosphate as phosphorus
KOH	Potassium hydroxide	PSU	Practical salinity unit
k_{Th}	Thomas kinetic coefficient	PZC	Point of zero charge
kV	Kilovolt	Q	Flow rate
K_{YN}	Yoon-Nelson kinetic coefficient	$q_{adsorbed}$	Total amount of adsorbed phosphate
L	Liter	$q_{desorbed}$	Total amount of desorbed phosphate
LDH	Layered double hydroxides	q_e	Equilibrium adsorption capacity
M	Molar	Q_{mdr}	Predicted adsorption capacity of modified dose-response model
m	Mass		Predicted adsorption capacity of Thomas model
MDR	Modified dose-response	R^2	Coefficient of determination
Mg	Magnesium	rpm	Rotation per minute
mg	milligram	t	Time
$mg-P$	Milligram as phosphorus	t_b	Breakthrough time
Mg^{2+}	Magnesium ion	t_e	Time required to reach exhaustion time
$MgCl_2$	Magnesium chloride	TSS	Total suspended solid
$MgCl_2 \cdot 6H_2O$	Magnesium chloride hexahydrate	V_q	Effluent volume of the q -th fraction
$MgFe-LDH$	Magnesium iron layered double hydroxides	XPS	X-ray photoelectron spectroscopy
min	Minute	XRD	X-ray diffraction
mL	Milliliter	Zr	Zirconium
mm	Millimeter	$Zr(IV)\text{-loaded okara}$	Soybean residue (okara) loaded with zirconium(IV)
N	Normal	$Zr(IV)\text{-loaded SOW gel}$	Saponified orange waste gel loaded with zirconium(IV)
N_2	Nitrogen		Fiber containing phosphonate and sulfonate (FPS) loaded with zirconium(IV)
n_2	Number of the last fraction in the desorption operation	$ZrOCl_2 \cdot 8H_2O$	Zirconyl chloride octahydrate
Na^+	Sodium ion	μm	Micrometer
Na_2CO_3	Sodium carbonate	τ	Time required for 50% adsorption breakthrough
Na_2SO_4	Sodium sulfate	$\%$	Percent
$NaCl$	Sodium chloride		
$NaOH$	Sodium Hydroxide		
nm	Nanometer		
$NO_3^- - N$	Nitrate as nitrogen		
O	Oxygen		

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ORIGINAL RESEARCH PAPER

Modelling the impact of environmental responsibility on the development of enterprises

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ABSTRACT

BACKGROUND AND OBJECTIVES: The requirements of stakeholders for the qualitative transformation of the approaches of industrial enterprises – environmental users to introduction of economic activities cause an increase in the role of environmental responsibility as their strategic priority for development. The purpose of the study is to improve the practical tools for calculating and developing strategies for the development of environmental responsibility of industrial enterprises.

METHODS: In the study, tools for assessing the level of environmental initiative of an industrial enterprise as an integrated indicator of environmental initiative are suggested. It is expressed as the arithmetic mean of ten factors of environmentally proactive behavior of industrial enterprises. To achieve the objectives of the study, methods of criterion-qualitative assessment are suggested, such as expert assessment and survey. The assessment system includes ten factors rated on a 10-point scale and weighted by significance. The integrated indicator of the environmental initiative acquires values within, is a stimulant of environmental responsibility.

FINDINGS: One justified a scientific and methodological approach to assessing the level of environmental responsibility of the industrial enterprise, including calculation of the corresponding taxonomic indicator with consideration of the quantitative factors of its environmental responsibility and the qualitative and quantitative factors of environmental initiative. This enables the possibility to assess the relevant state of environmental responsibility and determine the volumes for further correction of the ecological strategy for the development of the industrial enterprise.

CONCLUSION: As a conclusion, practical recommendations are made for the implementation of organizational and economic support of environmental responsibility in the development strategy of an industrial enterprise taking into account the impact of its environmental initiative and environmental duty. The level of environmental responsibility of industrial enterprises taken into account when developing recommendations on adjustment of their environmental strategy of development is determined, which as a whole allowed to form organizational and economic support for environmental responsibility of the enterprises under study. Thus, an enterprise, which only ensures compliance with environmental emission standards (compensation strategy or sufficiency strategy) cannot be characterized by a high level of environmental responsibility. To ensure a high level of environmental responsibility, an industrial enterprise must rely on a proactive strategy or a strategy for sustainable use of nature.

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INTRODUCTION

In the context of increasing environmental problems in the 21st century, enterprises, which use natural resources, face the task to strike a balance between economic development and minimizing the negative environmental impacts of economic activity. The requirements of stakeholders, concerning the qualitative transformation of approaches, taken by industrial enterprises, which use natural resources, to their economic activity, give rise to the role of environmental responsibility, as their strategic development priority (Zare et al., 2016). The issue regarding the observance of environmental responsibility principles becomes increasingly relevant at the level of interaction between the community and enterprises in the areas, where the latter are located (Hadj, 2020). Since industrial enterprises particularly place the greatest stress on the environment, it is important to provide an organizational-economic mechanism for ensuring their environmental responsibility in due course (Panyam et al., 2018). The need for ecologically friendly and equitable economic development remains the most difficult issue in the context of growing environmental-economic problem (Hovardas, 2016). The pursuit of the sustainable development of the ecological-economic system, providing a wide range of technological capabilities and high productiveness of business activities requires today's organizations to be more involved in managing these processes (Chen and Hamilton, 2020). The projection of the concept of sustainable development on the enterprise level causes the escalation of ecological and socio-economic conflict, which, on the one hand, is aimed at profit maximization, and on the other hand, is stemming from the requirements of the social sector and ecological control authorities to increase the social and ecological standards of economic activity (Fujii and Managi, 2016). However, the ecological efficiency is determined not only by the industrial base. It can be achieved by changing goals and targets, which are being pursued by industries, operational management and the methods of its implementation (Hong et al., 2016). It is, therefore, necessary to refuse outright the purely consumerist approach and to follow a policy that takes account of ecological and social factors (Singh et al., 2016). Consequently, the top management of industrial companies should be aware of the fact that positive or negative assessment

of the ecological activity of a company on the part of stakeholders and, first of all, the local community significantly affects the future success of products on the market, and business competitiveness as well (Malá et al., 2017).

Thus, in the context of increasing environmental problems in the 21st century, the enterprises, which use natural resources around the world, face an important task to strike a balance between the development of their economic activities and minimizing environmental problems, which arise due to these activities (Yue et al., 2020). The concept of socio-environmental responsibility of business was originally interpreted as the enhanced principle "polluter pays", well-known in the economics of nature use, which evolved into the requirement of environmental entrepreneurship initiative (Jovovic et al., 2017). At the same time, experts introduced a broad interpretation of the principle of sustainability within the "Sustainable Development Triangle" (Ji and Long, 2016). According to this approach, modern-day business activities are challenged to combine three interrelated goals in practice, namely, economic efficiency, environmental responsibility and social activity (Sáez-Martínez et al., 2016). All highly-developed countries nowadays recognize the need for theoretical underpinning and taking practical steps to shape enterprise environmental policy, which would envisage the care for nature conservation, environmental quality, the wise use of existing and potential natural resources, maintaining the ecological balance of nature and ensuring the living conditions of human (Elsawah et al., 2017). In accordance with these statements, the logical response of companies to the demands of public opinion would be the observance of environmental responsibility principles at all stages of industrial activity (Zhu and Zhao, 2018). At the same time, a range of issues still remains insufficiently developed in scientific literature. Among them, in particular, are the issues related to the meaning of the concept of industrial enterprise's environmental responsibility, assessing its level, determining the nature of the impact, exerted by environmental initiative and commitment, on the level of environmental responsibility. Consequently, one should continue to study the issue related to the organizational-economic mechanism for ensuring environmental responsibility of industrial enterprises according to

the requirements of stakeholders, as well as the issue of choosing an appropriate development strategy, considering ecological factors. The relevance of the research problem, its practical importance and the deficient theoretical elaboration of the mentioned aspects led to the choice of the topic, objective and tasks of the research paper. The purpose of this study is to improve the practical tools for calculating and developing strategies for the development of environmental responsibility of industrial enterprises. This study were set to achieve the theoretical foundations and the current condition of the system of ecological responsibility of the industrial enterprises; to develop the scientific and methodological approach to assessing the level of ecological responsibility of the industrial enterprises with the consideration of the level of their ecological initiative and ecological responsibility; to calculate the integrated indicator of environmental responsibility for Volkswagen AG; to justify the scientific basis for choosing an environmental strategy for the development of the industrial enterprises in terms of considering the impact of the existing level of environmental responsibility of the enterprise in these strategies. This study was conducted for Volkswagen AG (Germany, Poland) in the period of 2012-2018.

MATERIALS AND METHODS

The local community can act both as an object of environmental responsibility, and as a fully-fledged entity in the process of its formation at an industrial enterprise (Domenech and Bahn-Walkowiak, 2019). The assessment system should provide a general idea about the level of industrial enterprise's environmental responsibility, confirmed by outcomes, social (non-financial) reports, statistical data, as well as to form the information base for further analysis, detecting potential risks and the decision-making process (Borland et al., 2016). To assess the environmental initiative of an enterprise, one considers it appropriate to use the questionnaire method, as the concept of enterprise's environmental initiative is distinctly subjective. A questionnaire refers to the qualitative methods of information assessment and has a range of advantages, such as the ability to cover large target groups, a high level of result formalization, the minimal impact of the researcher on the interviewee, responsiveness, and

disadvantages, associated with the need for data verification, consistency check, etc. That is why a questionnaire should be necessarily combined with the evaluation of statistical error or the consistency of experts' opinions (Jiang et al., 2019). It is time to review the key advantages and disadvantages of expert methods. A direct estimation method can be applied, when the subjects of examination, which determine the final results, are directly comparable since these subjects are of similar nature and therefore have a common benchmark standard (Wang et al., 2016a). The method of direct estimation makes it possible to take into account the degree of predominance of a particular indicator in relation to others (Graafland, 2018). But when estimating the composite index of effectiveness, the wrong estimate of the indicator with lower value, if its position relative to other indicators has been determined correctly, can lead to the serious distortion of the final result (Wang et al., 2016b).

The ranking method and its types can be applied in the following cases:

- to arrange any kinds of objects or phenomena in time or space (Wilmer et al., 2018);
- to arrange objects according to any indicator, which does not need to be accurately measured (Cao et al., 2017);
- if a particular indicator should be measured, but can not be measured at the moment for practical or theoretical reasons (Anthony et al., 2016).

Environmental initiative of industrial enterprises is a category that is difficult to quantify. Given this statement, it is advisable to use high-quality, heuristic research methods to assess it. Thus, the integrated indicator of environmental initiative is determined on the basis of the method of expert survey conducted among employees of the enterprise under study and environmentalists. During this study, a survey to determine the scores of the level of environmental initiative of enterprises in each assessment category was conducted among 50 employees of the industrial enterprise under study, including middle managers and technical staff. Experts were asked to assign ranks (from 10 - the most significant, to 1 - the least significant) to each of the assessment categories. This choice of the target audience of the survey was due to the need to ensure the impartiality of expert opinions. Thus, none of the experts was interested in certain results of the survey, their opinion was completely

independent. When assessing the objects of a survey, experts often disagree on the problem to be solved. In this regard, there is a need to quantify the level of agreement of experts. Obtaining a quantitative measure of agreement allows a more reasonable interpretation of the reasons for differences of opinion. To this end, the level of agreement between the opinions of experts was determined through the use of the concordance coefficient. The concordance coefficient determines the degree of agreement of expert assessments, varies in the range of $0 < K < 1$, with 0 meaning complete disagreement, and 1 meaning complete agreement. The results of the survey are considered statistically significant at $K > 0.55$. The calculated concordance coefficient for the enterprise under study is higher than its control value of $K = 0.77$, thus, it can indicate a high level of agreement of experts. Therefore, the results of the conducted study are credible and suitable for further use. Given that the environmental responsibility of industrial enterprises is considered as a system of their environmental duty and environmental initiative, the indicators of the level of environmental responsibility are suggested to be divided into two blocks — the block of environmental duty and the block of environmental initiative. At the same time, by the nature of the impact on the level of environmental responsibility, all indicators are divided into stimulants, the increase of which contributes to the increase of the level of environmental responsibility, and disincentives, the increase of which causes a decrease of the level of environmental responsibility.

During the second stage, the standardization of indicators is carried out using Eqs. 1, 2 and 3.

$$r_k = \frac{y_k - \bar{y}_k}{V_k} \quad (1)$$

Where,

$$\bar{y}_k = \frac{1}{z} \sum_{i=1}^z y_k \quad (2)$$

$$V_k = \left[\frac{1}{z} \sum_{i=1}^z (y_k - \bar{y}_k)^2 \right]^{\frac{1}{2}} \quad (3)$$

Where, $k=1,2,...,n$; y_k - the value of the index k for the unit i ; \bar{y}_k - arithmetic mean value of the index k ; V_k - standard deviation of the index k ; r_k - the

standardized value of the index k for the unit i .

The highest values of incentives and the lowest values of disincentives form the coordinates of desired development benchmark using Eqs. 4, 5 and 6.

$$r_{0_i} = \max r_{ik}, \text{ if index } k \text{ is an incentive;} \quad (4)$$

$$r_{0_i} = \min r_{ik}, \text{ if index } k \text{ is a disincentive.} \quad (5)$$

The development benchmark will therefore have coordinates:

$$D_0 = (r_{0_1}, r_{0_2}, \dots, r_{0_n}) \quad (6)$$

To obtain the taxonomic index of development the following equations are used. According to interpretation, the closer its value is to 1, the higher level of development it shows as Eqs.7, 8, 9 and 10.

$$p_i = 1 - \frac{b_{i0}}{b_0} \quad (7)$$

$$\text{Where, } b_{i0} = \left[\sum_{v=1}^n (r_{iv} - r_{0v})^2 \right]^{\frac{1}{2}}, n=1,2,\dots,t \quad (8)$$

$$b_0 = \bar{b}_0 + 2V_0 \quad (9)$$

$$\bar{b}_0 = \frac{1}{t} \sum_{i=1}^t b_{i0} \quad (10)$$

Where, b_0 - the distance between separate points-units and the point D_0 , which presents the development benchmark, V_0 - standard deviation of the index b_{i0} .

For Volkswagen Aktiengesellschaft, the overall taxonomic environmental responsibility index will change by 1.79%, along with the change of index from the environmental commitment block by 1% and the constant value of the index from the environmental initiative block. If the environmental initiative index changes by 1% while the environmental commitment index remains sustainable, the overall taxonomic environmental responsibility index will change by 1.27%. In the light of the above, for the researched Volkswagen Aktiengesellschaft, both environmental commitment and environmental initiative blocks have relatively equal impact on the resultant environmental responsibility index, with a slight prevalence of environmental commitment block. The composite taxonomic environmental responsibility

index, constructed from the environmental commitment and environmental initiative blocks, synthetically characterizes the changes in property values of the researched groups. One of the important advantages of this indicator is the characterization of direction and the scope of changes in the processes described by a set of initial data. The taxonomic method enables one to determine the level of environmental responsibility of industrial enterprises by assessing the distance from the coordinates of the benchmark-point to the standardized values of indicators. The fictitious benchmark has been defined by taking into account the differentiation of incentive and disincentive features of impact on the object under study. Through series of manipulations, the calculated distance has been transformed into the index with range (0; 1). Thus, the level of ecological responsibility of the enterprises is proposed to be assessed using the taxonomy method, which is used to reduce the factor space, manifesting itself in the aggregation of the information space, in the result of which one observes the formation of a general index. The methodology for the assessment of industrial enterprise's environmental responsibility, presented in this research, makes it possible to specify the areas of ecological strategy, which need improvement, as well as to define the level of its sustainability. It also provides a basis for the comparison of the actual level of environmental responsibility among different companies, which can be performed in further researches.

RESULTS AND DISCUSSION

It is believed that in order to build an effective system of nature management, it is necessary to form new strategic areas of intersubjective relationships in the natural resource block of the economy, which will allow to create a certain margin of safety of the economy from negative external or internal influences. It is believed that this important strategic area is a multilevel approach to management decision-making, which takes into account the external and internal environment of the main industrial enterprises polluting the region, the creation of intersectoral partnership in the field of environmental protection. In modern economic conditions, the basis is created for the implementation of environmental management actions through the business environment, which must take into account

the emerging contradictions of stakeholders. It is there that the most complete interaction of regional and local (enterprise level) levels of transition to sustainable development takes place. It is the environmental responsibility of industrial enterprises that is the key to taking into account the environmental interests of their stakeholders. The transition of economic systems at the macro, meso and micro levels to the path of sustainable development is due to the importance of preserving the environment not only for the existing generation, but also for future generations. Analysis of environmental costs as a component of assessing the level of environmental initiative of industrial enterprises allows to assess the functioning of the environmental management system, to monitor environmental protection costs, to analyze the relationship of environmental characteristics with financial and economic indicators of company activities. At each stage of the process of implementation of the environmental initiative of an industrial enterprise, the costs for the generation of adequate reporting information, effective management decisions and further optimization of environmental costs should be monitored. It should be stressed that the process of optimizing environmental costs should, in addition to the objectives of the enterprise, take into account the requirements of the local community and other stakeholders. Optimization should not cause deterioration of the environment due to reduced costs for the restoration of the damaged state of the environment. Environmental responsibility of industrial enterprises is one of the fundamental factors for shifts towards sustainable development at both micro and macro levels. Environmental responsibility of an industrial enterprise is defined as a set of mechanisms, tools, measures for the implementation of its environmental initiative and environmental duty. Environmental duty is defined as a set of environmental obligations to the local community and administrative bodies. The environmental initiative of an enterprise is considered as a set of measures of the environmental program of the enterprise, which go beyond normatively established requirements and meet requirements of the local community concerning environmental protection. This creates an appropriate categorical framework in the field of research of environmental responsibility of industrial enterprises for theoretical cognition of its forms

and relationships with the internal and external environments of industrial enterprises. In accordance with the objective of the environmental initiative, there arises a problem related to the assessment of its level, since it is a category that cannot be fully quantified. Assessing the environmental initiative of industrial enterprises should, first of all, heighten the interest of the main stakeholders, represented by local authorities and the community, since the level of the environmental initiative expresses enterprise's interest in participating in social dialogue regarding the ecologization of industrial activity and regional development strategy. The assessment of the environmental initiative level should be carried out using both qualitative methods, such as expert surveys and questionnaires, and quantitative methods. It should be noted that particular attention

should be put to the analysis of environmental costs and investment in nature protection measures, since these categories specifically reveal the efficiency of the environmental management system of industrial enterprises. Thus, one offers to carry out the assessment of the level of the ecological initiative of the industrial enterprises upon the following scheme in Fig. 1.

During the first stage, one forms a statistical database based on the listed sources of information. During the second and third stages, one carries out the processing of data, which have been obtained using the questionnaire and expert survey, and further calculates the integral environmental initiative index based on data processing. The fourth stage covers the analysis of environmental costs and capital investment in nature protection activities. At

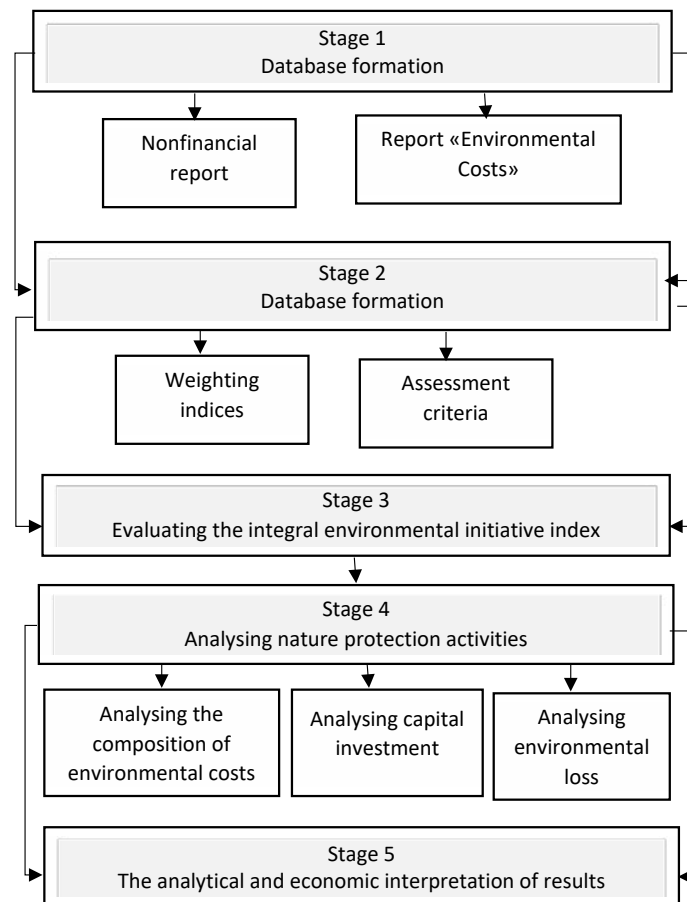


Fig. 1: Stages of assessing the environmental initiative of industrial enterprises

the last stage, one suggests the economic-analytical interpretation of attained results, more specifically, the analysis of growth rate and dynamic pattern of indices, obtained during the researched period. It is time to consider the process of calculating the integral environmental initiative index. The fundamental phase of this process is to form a system of evaluation categories, which would reveal the most substantial and significant aspects of the environmental initiative activities of industrial enterprises and meet the main international ecological standards. As to the system of evaluation categories for assessing the environmental initiative of industrial enterprises, special attention should be paid to the existence and scope of the compensatory policy. Since ecologically responsible development strategy always includes environmental restoration from damages caused by industrial activity, one considers it appropriate to add a corresponding category to the range of enterprise initiatives. It is called-activity, concerning the compensations for environmental damage to the community. Thus, one proposed a system of evaluation categories for determining the level of the ecological initiative of the industrial enterprises

(Table 1).

It includes 10 entries and can be implemented within the process of expert survey and the analysis of enterprise non-financial reports. The given system enables one to take into account both the external indicators of environmental activities and the subjective evaluations of experts. This makes it possible to conduct the most optimized assessment of the enterprise environmental initiative level and specify the priority areas for activities, which are of particular interest to the main stakeholders of industrial enterprises led by the local community. Consequently, the implementation of enterprise's own environmental initiative requires a specific set of methods depending on the peculiarities of industrial activity. The role of the environmental initiative of industrial enterprises in the implementation of their environmental responsibility is to restore the damaged environment and provide compensations for environmental damage to the community. It is implemented through a series of measures taken beyond the regulatory requirements. The issue of environmental responsibility assessment is one of the most essential problems of managing the socially

Table 1: The system for assessing the environmental initiative of industrial enterprises

Evaluation category	Key assessment criteria
Activity, concerning the compensations for environmental damage to the community	Compensatory ecological payments to employees, availability of sanitary protection zone of the enterprise, environmental restoration
Undertaking the environmental activities and measures	Own initiatives to improve environmental education of the community, environmental enhancement due to the public activities, held during the entire analyzed period
Atmospheric air protection	Availability of air quality monitoring system, amount of capital investment in this field, the quality of air cleaning equipment
Optimization of waste management areas	Waste disposal policy, the level of waste utilization, amount of capital investment in this field
Protection of water bodies, which are used by the enterprise	Activities aimed at restoration of the damaged coastline, availability of wastewater quality monitoring system, amount of capital investment in this field
Willingness to cooperate with the community	Conducting public hearings and consultations, ensuring feedback, availability of community engagement programs
Condition of the surrounding area	Pollution condition of the surrounding area, restoration and landscaping activities, sanitary protection zones
Non-financial reporting, full disclosure of the environmental information	The frequency of non-financial reports publication, their compliance with international standards, the quality and transparency of the reported information
Effectiveness of environmental management system	Availability of ISO Series Certificates, amount of capital investment in this field
Conducting an internal environmental audit	The frequency of internal ecological audits, corrective actions and removal of disadvantages, which were revealed during the audit

responsible practices of industrial enterprises. This question remains relevant both for all stakeholders and for industrial enterprises themselves. The gradually increasing amount of industrial companies, which claim themselves to be highly supportive of environmental responsibility principles, cause the need for verifying if the information about the level of their environmental responsibility is factually accurate. Consequently, it is a crucial task nowadays to develop a methodology for the assessment of environmental responsibility, which would be accessible and easily understandable for all interested parties. The enhanced and comprehensive assessment of the environmental responsibility of the industry, corporation, or a particular enterprise cannot be carried out independently by the concerned parties alone, for the reason of resource sufficiency, mainly information and time. Thus, one of the major tasks of this research was to develop the methodology for assessing the environmental responsibility of industrial enterprises, which would be relatively easy to use and contain accessible information database. Considering the details given above, one suggests the following scheme to be used for assessing the environmental responsibility of an industrial enterprise. During the first stage, one carries out the qualitative and quantitative analysis of indicators, calculates the numerical score of the enterprise environmental initiative level and the coefficients-indices of environmental commitment and environmental initiative for the researched enterprises, defines their incentive or disincentive impact on the resultant index. The standardization of the calculated coefficients, caused by the different dimensions of initial data, is performed at the second stage. One determines the benchmark indices, calculates the partial taxonomic indices for the environmental responsibility and environmental initiative levels. During the third stage, one conducts the comprehensive evaluation of the obtained indices, determines the impact of each partial index on the overall level of environmental responsibility of the researched industrial enterprises, identifies the resources for further development and improvement. The generalized algorithmic scheme, which displays the procedure for the assessment of environmental responsibility (ER), is shown in [Fig. 2](#). The suggested method for the assessment of environmental responsibility is based on the determination of

so-called taxonomic distance, that is, the distance between points in multivariable space, the dimension of which is determined by the number of features describing the object under study. The undoubted advantage of a taxonomic method is the procedure of the so-called standardization of indicators, as a result of which the object properties, represented by different qualitative and quantitative indicators, are transformed into a single standardized measurement system. The algorithm for determining the taxonomic index of development includes several stages. At the first stage, one forms an observation matrix, which contains the most complete property characteristic of the researched set. The metrics, included in the matrix, are diverse since these metrics describe different object properties and are measured in different units.

After the standardization procedure, all selected indices should be divided into incentives and disincentives based on their impact on the level of environmental responsibility. Indices, high values of which are desirable in terms of the chosen aspect of the research, are considered to be incentives. In this research, one has examined 10 indices from two blocks, environmental commitment and environmental initiative, among which all indicators of the environmental commitment block are characterized by an incentive effect, proceeding from their mathematical expressions. Among the indices of the environmental initiative block, some are characterized by disincentive features. This includes, in particular, the share of stranded environmental costs (environmental fines, penalties, other sanctions) and the share of running NE (nature environment) protection costs, as its growth indicates the obsolescence of cleaning equipment and low efficiency. Other indices of the environmental initiative block are characterized by an incentive impact on the resultant index, proceeding from their mathematical expressions. Following the strategy of cleaner production, which is expressed in waste management, reducing emissions into the atmosphere and water is considered the main area of implementation of the environmental duty of industrial enterprises. It should be stressed that in recent years a high interest in the implementation of measures to improve energy efficiency, due to high prices for fuel and energy resources has become a characteristic feature for industrial enterprises.

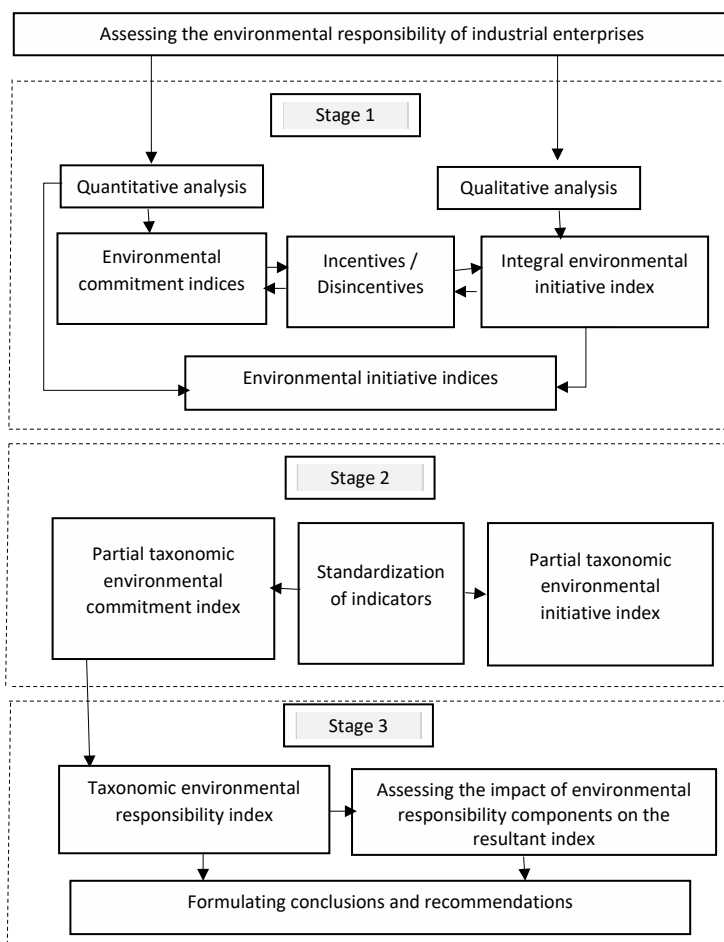


Fig. 2: Stages of assessing the environmental responsibility of industrial enterprises

This conditioned the inclusion of the coefficients of waste intensity for emissions into the environment by categories of “atmospheric air”, “water”, and “wastes”, the coefficient of waste utilization, energy intensity of production (Table 2) in the system of indicators of the bloc of environmental duty.

Nevertheless, the indicators of efficiency of a system of environmental management should be included in the indicators of the block of environmental initiative as presence of environmental management at an enterprise allows to speak about the sufficient level of environmental responsibility. Thus, these indicators include indicators of the structure of environmental costs, namely capital investments, current environmental protection costs, the coefficient of environmental damage as an indicator

of compliance with environmental legislation, which is also a result of the implementation of an environmental management system (Table 3).

Summing up the above, it should be noted that all the criteria for the level of environmental responsibility of industrial enterprises are suggested to be divided into two groups — those, which are related to environmental obligations, in other words to environmental responsibility, and those, which are related to the own environmental initiative of a company and defined as voluntary. The environmental duties of an industrial enterprise include responsibility for environmental damages caused by production activities, compliance with environmental regulations and limits on emissions to NE. The group of criteria for own environmental initiative should include

two subgroups of indicators — activities concerning protection and restoration of the damaged nature environment, the existence of environment quality monitoring system, environmental management system, and the effectiveness of communication with

stakeholders. The results of the calculation of the integrated indicator of the environmental initiative for 2012-2018 are given in Table 4.

However, in addition to the directly integrated indicator of environmental initiative, considerable

Table 2: Indicators for assessing the level of environmental responsibility of industrial enterprises — block of environmental commitment

Indicator name (y_{ik}), nature of impact on the level of environmental responsibility (stimulator/disincentive)	Calculation procedure	Notations
Coefficient of waste intensity by category of "air emissions", stimulator, (y_1)	$y_1 = 1 - \frac{W_A}{P}$ $y_1 \rightarrow 1$	W_A — total air emissions, thousands of tons; P — volume of manufactured products, thousands of tons
Coefficient of waste intensity by category of "discharges into water bodies", stimulator, (y_2)	$y_2 = 1 - \frac{W_o}{P}$ $y_2 \rightarrow 1$	W_o — total discharges into water bodies, thousands of tons
Coefficient of waste intensity by category of "waste", stimulator, (y_3)	$y_3 = 1 - \frac{W}{P}$ $y_3 \rightarrow 1$	W — total wastes, thousands of tons
Coefficient of waste utilization, stimulator, (y_4)	$y_4 = \frac{W_w}{W}$ $y_4 \rightarrow 1$	W_w — volume of waste used, thousands of tons
Coefficient of energy intensity of production, stimulator, (y_5)	$y_5 = 1 - \frac{E}{C}$ $y_5 \rightarrow 1$	E — energy costs for the manufacture of products, thousands of euros; C — net cost of manufactured products, thousands of euros

Table 3: Indicators for assessing the level of environmental responsibility of industrial enterprises — block of environmental initiative

Indicator name (y_{ik}), nature of impact on the level of environmental responsibility (stimulator/ destimulator)	Calculation procedure	Notations
Share of capital investments in environmental measures in the total costs of NE protection, stimulator, (y_6)	These indicators will enable management to assess the operation of the environmental management system, the level of implementation of objectives of enterprise "greening", to monitor the costs of NE protection, and to analyze the relationship of environmental characteristics with financial and economic indicators of an enterprise.	
Share of current costs for NE protection in the total costs of NE protection, destimulator, (y_7)		
Share of unrecovered environmental costs paid from profits, destimulator, (y_8)		
Coefficient of environmental loss capacity of production, stimulator, (y_9)	$y_9 = 1 - \frac{T+S}{C}$ $y_9 \rightarrow 1$	T — amount of accrued environmental tax, thousands of euros; S — sanctions for violation of environmental legislation, thousands of euros.
Integrated indicator of environmental initiative, stimulator, (y_{10})	It reflects the activities of an enterprise for the implementation of self-initiated measures to protect the environment. It includes expert assessments of measures to compensate the community for losses from NE pollution, optimization of waste management, the state of the surrounding area in the sanitary zone of an enterprise, etc.	

attention should also be paid to the environmental costs of industrial enterprises (Table 5), priority areas of environmental financing.

At the fourth stage, one constructs the benchmark vector of development with coordinates r_0 . It also involves determination of the taxonomic environmental responsibility index for Volkswagen Aktiengesellschaft for the period 2012-2018. First of all, one should form an observation matrix (Table 6).

To convert the observation matrix into a dimensionless form, it is necessary to standardize its elements using the equations (1-3) to obtain a new matrix (Table 7). In accordance with the results of grouping indices into incentives and disincentives with the help of the equations (4-6), one forms a reference vector with the corresponding coordinates: $D_0 = (1.179; 1.392; 1.501; 1.556; 1.274; 1.689; -0.897;$

$-1.328; 1.603; 1.018)$.

Based on the calculated values, one determined the distance between the elements of a standardized matrix and the elements of the reference vector using Formula 8. Further calculations of intermediary indices and the taxonomic index, which reflects the dynamics of environmental responsibility in 2012-2018, were conducted using the Excel software package and are shown in Table 8.

As a result of the made analysis of the assessment of the levels of significance of the suggested categories, the following conclusions can be drawn. Experts have identified the following most important assessment categories, which have the greatest impact on the level of environmental initiative of an enterprise: activities to compensate the community for environmental damage, protection of air and

Table 4: Data for the analysis of the block of ecological initiative of the enterprise under study for 2012–2018

Environmental costs, thousands of euros	2012	2013	2014	2015	2016	2017	2018
Environmental payments	69,300.0	82,100.0	96,100.0	148,300.0	145,850.0	193,715.7	216,961.6
including fines, sanctions	19,000.0	22,620.0	32,650.0	45,300.0	73,000.0	0.0	0.0
Capital investment in environmental measures	235,469.0	307,600.0	289,102.0	693,883.0	1,034,304.0	430,945.6	486,968.5
Current costs of NE protection	26,984.5	21,612.1	16,707.1	12,114.0	18,384.1	15,331.8	16,865.0
Total costs of NE protection	331,753.5	411,312.1	401,909.1	854,297.0	1,198,538.1	639,993.1	720,795.1
Costs by category, thousands of euros	2011	2012	2013	2014	2015	2016	2017
Atmospheric air protection	142,000.0	237,200.0	325,600.0	718,400.0	730,250.0	209,957.6	228,853.8
Return water treatment	55,000.0	36,500.0	29,800.0	24,100.0	30,080.0	8,793.0	9,408.5
Waste management	78,000.0	106,600.0	133,800.0	161,000.0	169,000.0	22,636.1	269,935.0
Environmental management	7,600.0	10,800.0	7,200.0	5,900.0	5,500.0	372.5	383.7

Table 5: Coefficients characterizing the level of environmental initiative of the enterprise under study for 2012–2018

Indicators	2012	2013	2014	2015	2016	2017	2018
Share of capital investments in environmental measures in the total costs of NE protection	0.710	0.748	0.719	0.812	0.863	0.673	0.676
Share of current costs of NE protection in the total costs of NE protection	0.081	0.053	0.042	0.014	0.015	0.024	0.023
Share of unrecovered environmental costs	0.274	0.276	0.340	0.305	0.501	0.000	0.000
Coefficient of environmental loss capacity of production	0.996	0.997	0.997	0.996	0.997	0.996	0.996
Integrated indicator of environmental initiative	0.361	0.305	0.401	0.508	0.489	0.546	0.549

Table 6: Observation matrix for the assessment of environmental responsibility

Indicators		2012	2013	2014	2015	2016	2017	2018
Environmental commitment block	Atmospheric emission capacity index (Y_1)	0.949	0.955	0.956	0.963	0.963	0.968	0.967
	Waste water discharge capacity index (Y_2)	0.590	0.537	0.598	0.586	0.426	0.505	0.509
	Waste capacity index (Y_3)	0.071	0.053	0.046	0.050	0.014	0.143	0.174
	Waste utilization index (Y_4)	0.586	0.528	0.552	0.552	0.615	0.749	0.728
	Product's energy-output ratio index (Y_5)	0.885	0.876	0.880	0.887	0.897	0.883	0.900
Environmental initiative block	Share of capital investment in nature protection measures (Y_6)	0.730	0.748	0.719	0.812	0.863	0.673	0.676
	Share of running NE protection costs (Y_7)	0.061	0.053	0.042	0.014	0.015	0.024	0.023
	Share of stranded environmental costs (Y_8)	0.374	0.276	0.340	0.305	0.501	0.201	0.302
	Product's ecological loss capacity index (Y_9)	0.994	0.997	0.997	0.996	0.997	0.996	0.996
	Integral environmental initiative index (Y_{10})	0.461	0.305	0.401	0.508	0.489	0.546	0.549

Table 7: Standardized matrix for the assessment of environmental responsibility

Indicators		2012	2013	2014	2015	2016	2017	2018
Environmental commitment block	(Y_1) incentive	-1.574	-0.775	-0.597	0.373	0.475	1.079	1.018
	(Y_2) incentive	-0.474	0.027	1.140	1.292	-1.657	-0.206	-0.122
	(Y_3) incentive	0.163	-0.490	-0.619	-0.538	-1.175	1.057	1.601
	(Y_4) incentive	-1.140	-0.726	-0.488	-0.485	0.133	1.458	1.247
	(Y_5) incentive	0.730	-1.336	-0.901	-0.140	0.948	-0.575	1.274
Environmental initiative block	(Y_6) incentive	-0.468	0.068	-0.334	0.974	1.689	-0.981	-0.949
	(Y_7) disincentive	1.958	0.677	0.227	-0.897	-0.849	-0.496	-0.519
	(Y_8) disincentive	0.165	0.183	0.535	0.347	1.416	-1.328	-1.328
	(Y_9) incentive	-0.076	1.603	0.347	-1.734	0.243	-0.478	0.096
	(Y_{10}) incentive	-0.941	-1.525	-0.524	0.591	0.393	0.987	1.018

water bodies used by enterprises, the effectiveness of environmental management and environmental audit. It is a matter of deep concern that the survey showed the lowest level of satisfaction with these indicators. Therefore, the data obtained during the study can be used to assess the level of environmental initiative of

industrial enterprises in terms of the implemented measures and their level of significance directly for a particular enterprise and local community. The integrated indicator of the environmental initiative is within (0; 1) and is interpreted as follows: the higher the indicator, the higher the level of environmental

initiative of the industrial enterprise under study. Thus, using the suggested methodology, one obtained the partial taxonomic indices of environmental commitment and environmental initiative for the researched enterprises, as well as the overall taxonomic environmental responsibility index. Since the taxonomic indicators of the level of ecological responsibility of the studied industrial enterprises have multi-directional dynamics, one considers it appropriate to use the method of economic and mathematical modeling for the explanation of the influence of the blocks of environmental responsibility and environmental initiative on the resulting indicator (Gast *et al.*, 2017). One of the possible ways to evaluate the quality of property package for result analysis is to apply generalized Harrington desirability function (Bombiak and Marciniuk-Kluska, 2018; Cai *et al.*, 2018). The interpretation of obtained results using this approach is one of the easiest ways to transform the natural values of individual responses into the dimensionless scale of desirability and priority (Xu *et al.*, 2020). To construct a generalized function means to generate the obtained values of property indices (which have different units of measurement, including qualitative, esthetic, psychological and personal features) into a dimensionless desirability scale. The function of the desirability scale is to set up a correspondence between the obtained values of property indices and expert's evaluations of desirability for a particular index. A standard scale contains five valuation levels in the total range from 0 to 1, which reflect different desirability rates. The value 0.37 is a critical point, where unsatisfactory indices turn into satisfactory ones. The distinct advantage of this scale in the interpretation of the numerical values of environmental responsibility is its versatility, the ability to evaluate calculated data in a quality manner for separate companies and the entire industry. The desirability scale has a range from 0 to 1. Under this approach to valuation, it is convenient to use the additional scores 0.2 and 0.8 to make the desirability function more "sensitive" to the changes of informative index x within the "satisfactory" area and less sensitive out of it. Then, one should analyze the values and dynamics of calculated environmental responsibility indices. Table 4 shows that the environmental responsibility of Volkswagen Aktiengesellschaft in the period 2012-2018 was at the sufficient level. Since the taxonomic indicators

of the level of ecological responsibility of the studied industrial enterprises have multi-directional dynamics, one considers it appropriate to use the method of economic and mathematical modeling for the explanation of the influence of the blocks of environmental responsibility and environmental initiative on the resulting indicator. As noted in (Sharma and Gupta, 2020), multiplicative models, and Cobb–Douglas production function, in particular, reflect the development of socio-economic systems in the most objective way, avoiding the influence of subjective factor. It helps to detect isolated factors influencing the resultant index, which is the objective of this study. For the given two-factor regression, the partial elasticity coefficient indicates, how much the taxonomic environmental initiative index will change in percentage terms, if one of the factors changes by one percent while the other factor keeps the constant values. It helps to identify leaders and outsiders in the field of corporate environmental responsibility and provides a basis for the construction of development benchmark vectors, taking into account ecological factors. To implement environmental responsibility policy at an industrial enterprise one should coordinate objectives, tasks, areas of activity for the whole range of its subsystems. The deliberate integration of environmental responsibility principles into the enterprise development strategy requires designing a coherent, systematic plan for achieving this aim. The procedure of implementing environmental responsibility at an industrial enterprise should therefore take place according to a certain scheme. To implement environmental responsibility at an industrial enterprise, the following steps must be taken: using the developed mechanism for its implementation, it is necessary to analyze the internal and external factors, which affect company's capability to undertake particular environmental activities. The next step is to examine the ability of an industrial enterprise to impose the key principles of environmental responsibility on economic activity. Provided that an enterprise is capable of environmental responsibility implementation, the next step would cover the search for methods and tools, the formation of an appropriate organizational-economic mechanism for ensuring environmental responsibility of industrial enterprises. The whole range of selected tools should be based on the well-defined information base of research. Thus, one

can proceed to the next step of the scheme, that is, the creation of the information base. The final step provides control over the effectiveness of previous steps.

The key outcomes, which can be achieved by implementing the system of environmental responsibility, include the following:

- improvement of the natural environment;
- reduced number of fines and penalties for the violations of established limits on discharges into NE;
- improved communication with representatives of the external environment of an enterprise;
- positive environmentally-conscious reputation of a company;
- optimization of investment policy in the field of NE protection;
- higher environmental compatibility of enterprise's production system.

Particular attention should be put to the implementation of the developed environmental responsibility system into the general development strategy. Talking about a large industrial company, when designing its strategy, one often defines the key directions for the development, upon which separate mini-strategies are similarly being designed and further integrated into the general one, therefore implementing the integrated approach to solving this task. One of the key factors, which influence the choice of a development strategy for any industrial company, is competitive growth. For industrial enterprises, the ecological compatibility of production systems and demonstration of environmental responsibility serve as one of such factors. However, the major problem for the existing types of development strategies is still the lack of a long-term vision of the environmental component. It is obvious that designing an organization's development strategy from the perspective of environmental responsibility concept is that integrant, which is essential for the fulfilment of core business tasks.

As it is commonly known, the concept of an environmentally responsible industrial enterprise involves the existence of a separate environmental strategy, which is part of its overall development strategy. A decision to develop such a strategy is usually enhanced by the following facts:

- consumer requirements for the open information about the ecological safety of production are

increasingly spreading out across the world;

- banks and insurance organizations take into account company's business reputation, which includes its ecological status when making decisions on granting of credits or choosing an insurance policy;
- ecological control, provided by the government, is getting increasingly tightened;
- strengthening the international standards of environmental management, etc.

Thus, the organizational-economic system for ensuring the environmental responsibility of an industrial enterprise should become an integral part of its development strategy. When designing the development strategy of an industrial enterprise, which is aimed at adherence to the concept of environmental responsibility, it is essential to take into account not only external factors, which reflect the state control, consumer opinion, ecological status of competitors, but also observe the internal principles of ecological safety. Only in this case, an organization will be able to achieve balanced environmental management and economic growth at the same time (Fig. 3).

Thus, designing an organization's strategy following the environmental responsibility principles is that integral part of its general economic policy, without which it is impossible to meet the targets of economic development for a separate enterprise and a country as a whole. When choosing a strategy for the development of industrial enterprises, one should take into account regional ecological development strategies. In fact, the harmonious combination of ecologization tasks at the local and regional levels makes it possible to achieve sustainable socio-economic development. Thus, the adjustment of an enterprise's development strategy should be made in two directions – in accordance with the level of industrial enterprise's environmental responsibility and taking into account the ecological living conditions of inhabitants in a particular region. Inhabitants, who live in districts with different living conditions, will have consequently different requirements for the development of their settlement areas and, accordingly, for the main industrial enterprises-pollutants of the environment. Thus, the inhabitants of areas with worsened and strained living conditions will demand ecologization strategy and environmental improvement, while

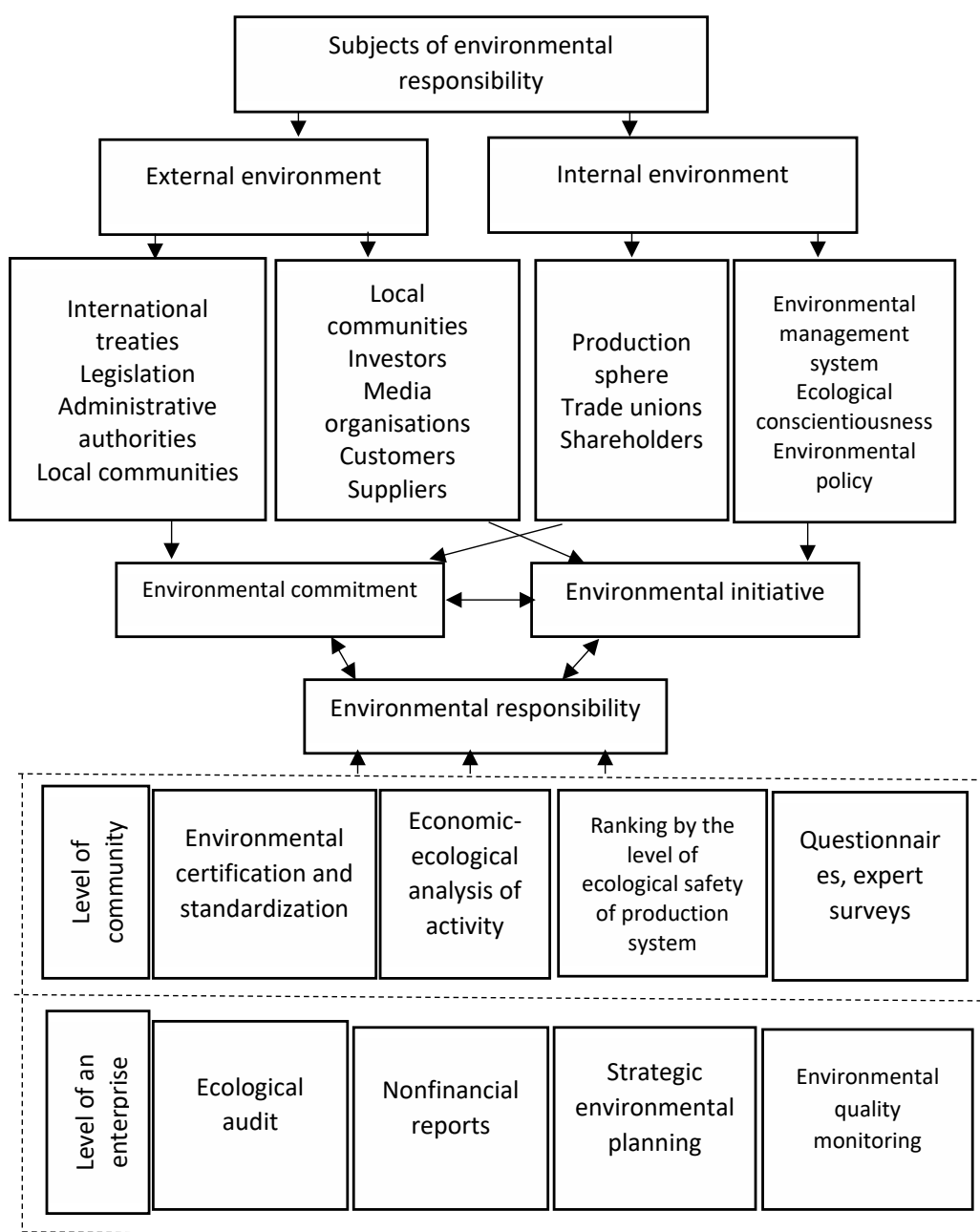


Fig. 3: Organizational-economic support of environmental responsibility of industrial enterprises

the local communities in areas with satisfactory living conditions will pay greater attention to environmental initiatives, the implementation of new environmentally friendly ways to manufacture products at industrial enterprises. In the light of

the above, it becomes evident that the type of the development strategy of an enterprise is related to its environmental responsibility level and the ecological living conditions of inhabitants since the community acts as a full participant in the process of designing

Table 8: Taxonomic indices of environmental responsibility, environmental commitment and environmental initiative for the period 2012-2018

Indicators	2012	2013	2014	2015	2016	2017	2018
Overall taxonomic environmental responsibility index	0.683	0.673	0.678	0.684	0.691	0.701	0.712
Partial taxonomic environmental commitment index	0.618	0.591	0.598	0.585	0.538	0.614	0.679
Partial taxonomic environmental initiative index	0.668	0.690	0.684	0.745	0.653	0.632	0.683

Table 9: Ecological strategy for the development of industrial enterprises*

level of environmental responsibility	Areas of enterprise activity	Type of an ecological strategy	Type of territory depending on ecological living conditions
Unsatisfactory (0-0.37]	Purification of emissions, discharges, development of a strategy concerning waste management at the end of the manufacturing-technological cycle. Undertaking pollution abatement measures. Compliance with the requirements for disclosure of external environmental reporting.	Protective strategy Regulatory environmental management	3.4
Satisfactory (0.37-0.63]	Reduction of emissions to the regulatory level. Internal ecological audit (of different types and purposes). Long-term environmental measures, including technological changes. Responsibility of department managers to uphold ecological parameters of the production system.	Compensation strategy (sufficiency strategy) Effective environmental management	2.3
Sufficient (above 0.63)	Preventive reduction of environmental emissions. Adherence to the eco-standards of the products in the international markets. Including the concept of sustainable development in the company's reporting. External and internal ecological audit of the enterprise.	Proactive strategy (balanced nature management strategy) Management of the environmentally responsible organization	1; 2; 3

*All regions of the country were divided into 4 groups according to the level of favourable living conditions: 1- satisfactory conditions; 2- worsened conditions; 3 – strained conditions; 4- ecological disaster zone.

and implementing the environmentally-friendly strategy for the development of industrial enterprises. Thus, the choice of a development strategy for an industrial enterprise should be adjusted, having into consideration its environmental responsibility level and the ecological conditions of the region. An enterprise, which only adheres to the regulatory requirements for ecological quality (sufficiency strategy), can not have a high level of environmental responsibility. Moreover, against the background of increasing ecological decay, the reduction of emissions to the efficacy level (compromise strategy) is only a mean step of environmental responsibility. To this end,

the environmental situation in the regions of Poland and Germany, where the enterprises of Volkswagen Aktiengesellschaft are located, was analyzed in terms of territorial concentration of production, economic development of land, population density, environmental pollution, the level of adverse natural and anthropogenic processes. All regions of the countries under study were divided into 4 types according to the level of favorable living conditions: 1 - satisfactory conditions; 2 - deteriorating conditions; 3 - stressful conditions; 4 - environmental disaster zone. A highly responsible industrial company in respect of the environment should be guided by the balanced

nature management strategy, which responds to the trends in the field of strategic management and sustainable development (Table 9).

In this way, the choice of ecological strategies for the development of industrial companies can have various scenarios. However, companies, possessing outmoded equipment and poor state of technology in use, are not able to perform the prompt implementation of the proactive strategy. For that reason, one highly recommends the researched industrial companies, ranking as temporarily satisfactory, to align with the compensation strategy. In addition, one considers it appropriate to formulate for each of the researched companies a set of recommendations for increasing their environmental responsibility level. The key advantages of each type of suggested development strategies in relation to the external and internal environment of the company are as; 1) sufficiency strategy – the reduction of costs, increase in transparency (public control); 2) compromise strategy – the social comprehensiveness, establishing a social dialogue with stakeholders; 3) balanced nature management strategy - uniqueness (the specific nature of organization), the efficient cooperation between the business community, government and public organizations. In general, to optimize the environmental losses of the enterprise, the following measures are recommended: accelerated depreciation of fixed assets, which have environmental significance (treatment facilities, environmental quality monitoring systems), as well as obsolete production equipment; material and intangible incentives for staff in the area of development of engineering solutions and innovation proposals for the greening of production activities and environmental management system, energy efficiency and optimization of waste management; compliance with ISO and EMAS standards in the area of environmental protection, control over the effectiveness of the system of environmental responsibility in enterprises using a number of scientifically sound indicators suggested in the study. It should be emphasized that in addition to optimizing the environmental strategy of an enterprise, these measures will create a positive image of a company, increase the competitiveness of products through “green” advantages, which is especially important in European markets.

CONCLUSION

Based on the conducted study, the following conclusions can be made.

1) The scientific-methodical approach to assessing the level of environmental responsibility of an industrial enterprise is substantiated, which provides for the calculation of the relevant taxonomic indicator taking into account the quantitative factors of its environmental commitment (taxonomic indicator of environmental commitment) and qualitative and quantitative factors of environmental initiative (taxonomic indicator of environmental initiative), as well as their stimulating or destimulating effect on the level of environmental responsibility of an industrial enterprise. 2) The thus obtained numerical values of the taxonomic indicator of the level of environmental responsibility of an industrial enterprise allow to assess its current state and determine the scope and nature of the impact of mandatory and initiative components on the overall level of environmental responsibility for further correction. 3) Based on a critical analysis of the content of development strategies of industrial enterprises in the environmental sphere, there was identified the need to take into account in these strategies the current level of their environmental responsibility differentiated by the desirability scale, and environmental living conditions in the cities where these enterprises are located. This makes it possible to take into account both economic and environmental-social factors when selecting an environmental strategy for the development of industrial enterprises. 4) The level of environmental responsibility of industrial enterprises taken into account when developing recommendations on adjustment of their environmental strategy of development is determined, which as a whole allowed to form organizational and economic support for environmental responsibility of the enterprises under study.

AUTHOR CONTRIBUTIONS

M. Pinskaya performed an experimental design and analyzed the data. O. Meleshchenko defined the concept and methodology of the research. O. Kovalchuk ranked the data into tables and figures. O. Karpenko performed the literature survey. E. Kompanets customized the manuscript to meet the requirements of the journal.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

b_0	Distance between separate point-units and the point D_0 , which presents the development benchmark
C	Net cost of manufactured products, thousands of euros
D_0	Development benchmark
E	Energy costs for the manufacture of products, thousands of euros
<i>EMAS</i>	Eco-Management and Audit Scheme
<i>Eqs.</i>	Formula
<i>ER</i>	Corporate Environmental Responsibility
<i>etc</i>	And so on (et cetera)
f	Environmental responsibility dependence function
<i>Fig.</i>	Figure
Fp	Fisher's test
<i>HR</i>	Human resources
<i>ISO</i>	International Organization for Standardization
K	Concordance coefficient
k	Number of indicators
max	Maximum value
min	Minimum value
n	Interval value
<i>NE</i>	Nature environment
P	Volume of manufactured products, thousands of tons
R^2	Determination coefficient
r_{ik}	Standardized value of the index k for the unit i

S	Sanctions for violation of environmental legislation, thousands of euros
	The amount of accrued environmental tax, thousands of euros
t	Time lag
V_0	Standard deviation of the index b_{i0}
V_k	The standard deviation of the index k
W_A	Total emissions into the atmosphere, thousands of tons
W_0	Total discharges into water bodies, thousands of tons
W	Total wastes, thousands of tons
W_w	Volume of waste used, thousands of tons
y_1	Atmospheric emission capacity index
y_2	Wastewater discharge capacity index
y_3	Waste capacity index
y_4	Waste utilization index
y_5	Product's energy-output ratio index
y_6	Share of capital investment in nature protection measures
y_7	Share of running NE protection costs
	Share of stranded environmental costs
y_9	Product's ecological loss capacity index
y_0	Integral environmental initiative index
y_k	Value of the index k for the unit i
\bar{y}_k	The arithmetic mean value of the index k
z	Integer

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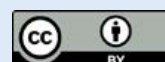
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ORIGINAL RESEARCH PAPER

Estimation and mapping of the contribution of nitric acid to atmospheric corrosion of zinc

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ABSTRACT

BACKGROUND AND OBJECTIVES: Atmospheric zinc corrosion in the Mexico City Metropolitan area has long been attributed mainly to the effect of pollutants such as sulfur dioxide. There are changes in the urban atmosphere's chemical composition due to the implementation of air quality policies focused on reducing the emission of sulfur dioxide and other pollutants. This study's objectives were to estimate and map the contribution of nitric acid on zinc's atmospheric corrosion process.

METHODS: The impact of nitric acid on zinc is feasible to estimate using a function for a multi-pollutant situation. This function contemplates the sum of two contributions: one of nitric acid and another that includes sulfuric acid and climatic parameters. The multi-pollutant function is suitable to apply in areas without the strong influence of chlorides and tropical and subtropical climates, comparable to the Mexico City Metropolitan area.

FINDINGS: The results showed that spatial and temporal estimation of corrosion rates in grams per square meter of zinc was made for 2015-2019, using data modeling in a geographic information system. The maps of corrosion rates allowed us to visualize that, in general, the "southwest" zone has the most significant effects and that the lowest corrosion rates were presented in 2019 as an outcome of the implementation of air quality programs. Furthermore, a contribution of nitric acid up to 32% to the zinc corrosion rate was estimated.

CONCLUSION: The construction of corrosion rate maps provides a spatial and temporal estimate that allows visualizing areas where zinc materials are at risk corrosion due to the dispersion of atmospheric pollutants and climatic parameters. Likewise, it can represent a decision-making tool for the implementation of atmospheric corrosion studies of materials.

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INTRODUCTION

The atmospheric corrosion process of hot-dip zinc-coated steel occurs in environments influenced by industrial activities with emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_x), and hydrogen sulfide (H_2S). In marine environments, the reaction occurs with chlorides, together with carbon dioxide (CO_2) and water (H_2O) (Friel, 1986). The hydrogen potential (pH) of the aqueous surface film depends on the atmospheric concentration of SO_2 and NO_x , essential for the corrosion process because it controls the dissolution of the passive oxyhydroxide surface (Graedel, 1989; Graedel and Frankenthal, 1990). Corrosive atmospheric trace substances, such as SO_2 , nitrogen dioxide (NO_2), and hydrogen chloride (HCl), accelerate metallic corrosion by acidifying the surface electrolyte and accelerate metal corrosion (Johansson, 1990). Knotková and Bartoň (1992) analyzed the contribution of acid deposition as a heterogeneous dynamic system of atmospheric corrosion metals. Kucera and Fitz (1995) considered that an acid effect in the atmosphere due to a multi-pollutant situation was the cause of the damage to materials. The deterioration of various materials, to some extent, is caused by acid gases such as SO_2 , nitric acid (HNO_3), and HCl (Ferm et al., 2005). Current studies indicate that the corrosion effect of HNO_3 exceeds SO_2 , with a factor between 2 and 20 depending on the exposed material (Tidblad et al., 2009). Atmospheric corrosion metals has long been attributed to SO_2 and chlorides (Cl^-), which has been demonstrated in laboratory weathering exposures and proven in field exposures. So, the promulgation of environmental protection laws in industrialized countries, in addition to the efforts inside the limits of the Convention of the United Nations Economic Commissions for Europe (UN ECE) on Long-range Transboundary Air Pollution (CLRTAP), led to a notable decrease in SO_2 levels. However, the concentration levels of other air pollutants, such as NO_x and ozone (O_3), remained or even increased slightly. Therefore, the effect of SO_2 needs to be considered in a multi-pollutant situation with other polluting gases such as NO_2 and O_3 , including particles (Oesch and Faller, 1997; Tidblad et al., 2002a). The levels of environmental concentration for HNO_3 are lower concerning SO_2 ; however, depending on the aggressiveness, HNO_3 can be compared to SO_2 in a multi-pollutant situation (Tidblad et al., 2009).

Atmospheric sulfur levels have been better controlled, and there has been a considerable reduction in SO_2 emissions, while the concentration of NO_x and HNO_3 has increased due to emissions from automobile traffic (Graedel and Frankenthal, 1990; Pantani et al., 1998). HNO_3 has higher concentrations in urban areas than in rural areas, but they are lower than NO_2 in urban areas (Ferm et al., 2005). The current study's main objective is to estimate nitric acid's spatial and temporal contribution to zinc's atmospheric corrosion in the Mexico City Metropolitan Area (MCMA) for 2015-2019. Maps of corrosion rates in grams per square meter (g/m^2) were constructed to achieve this objective. Also, the maps provide a spatial estimate of the corrosion risk of zinc materials when exposed to a polluted atmosphere. This study was carried out in the Mexico City Metropolitan Area in 2020.

The behavior of HNO_3 in the atmosphere of the Mexico City Metropolitan area

The Mexico City Metropolitan Area (MCMA) is located between north $19^\circ 03'$ to $19^\circ 54'$ latitude and $98^\circ 38'$ to $99^\circ 31'$ west longitude. It is part of an endorheic basin, with an average altitude of 2,240 meters above sea level. It is in the "East" area of the region known as the Transverse Neovolcanic System. The latitude of the MCMA can accelerate the photochemical reactions of pollutants in the atmosphere due to intense solar radiation. Likewise, there are anticyclonic systems in the area that keep the sky clear and increase the photochemical capacity of the atmosphere (SMA, 2008). MCMA is one of the largest megacities worldwide, as it has a population of more than 21 million people (UN, 2016). MCMA lies in central Mexico, at a tropical latitude where the climate is semi-dry temperate tropical in the northeast region; in the center, the temperate subhumid predominates, and in the upper, semi-cold subhumid type regions. Generally, the main entry of wind is located in the north, although there may be wind flow from south to north (Jáuregui, 2000; INEGI, 2007). The rainy season with high relative humidity occurs from May to October (Jáuregui and Romales, 1996), causing the levels of some pollutants to drop due to atmospheric instability. The upper levels of precipitation are registered in the mountainous areas and the lowest mainly in the northeast (INEGI, 2007). A study of the chemical composition of rain samples in Mexico City through multiple regression

analysis for the period 1994-2000 showed that the contribution of Cl^- (chloride-based sea salt) with sodium ion (Na^+) and potassium ion (K^+) was 2.2 percentage (%) at the prediction of the hydrogen ion (H^+) concentration. It allows us to assume that the chlorides found in rainwater come from sources other than sea salt. Likewise, Mexico City is placed at more than 300 kilometres (km) from the nearest shore, surrounded by high mountains. The Sierra Madre Oriental, with mountains above 4000 meters above sea level, is located near the Gulf of Mexico coast, where heavy rains are produced that intervene in further eliminating sea salt aerosols (Báez *et al.*, 2006). A study of the humid atmospheric deposition in the 2013-2016 period in the MCMA indicated that the calcium Ca^{2+} and Cl^- ions represented total annual deposits of 8% to 9% and 3% to 8% (Avila, 2018). The presence of chlorides could be attributed to emissions of approximately 30,000 small, medium, and heavy industries located in the northern part of the MCMA and the residential use of hypochlorite. There are hypochlorite production, ferrous and non-ferrous foundries, glass producers, motor vehicle manufacturers (a foundry), automobile assembly, plant factories, lime, brick, ceramics, cement, and tires (García *et al.*, 2009). The altitude of the MCMA leads to a lower oxygen content of the air, approximately 23% lower than at sea level. It causes the combustion processes to operate poorly and diffuse a greater quantity of pollutants into the atmosphere, together with the surrounding mountain chain, which favors its stagnation (Molina and Molina, 2002). In 2016, Mexico City registered 2.3 million vehicles; the most abundant are those for private use, such as cars, Sport Utility Vehicle (SUVs), and motorcycles, which account for 83% of the total fleet. Total emissions of nitrogen oxides from this vehicle fleet represented a total of 60,907 Megagrams per year [Mg/y], with private cars and trucks being the main generators with an amount of 52,437 [Mg/y], which represents 86% of the total (SEDEMA, 2018a). The concentrations of SO_2 and NO_2 in the ambient air remained during 2017 below the limit concentrations required by the Official Mexican Environmental Health Standards. The Tula-Tepeji corridor remains the most important source of sulfur dioxide for Mexico City. An analysis of the trend indicated that since 2000, SO_2 concentrations maintained a downward trend, while nitrogen oxides did not show a significant trend. NO_2 contributes

about 60% of the total concentration of nitrogen oxides. Also, acid rain samples were recorded in the rainy season at places south and east of Mexico City (SEDEMA, 2018b). Studies on the monitoring of the atmospheric concentration of HNO_3 in the MCMA and its spatial distribution are scarce. HNO_3 shows a behavior similar to ozone, reaching a maximum level during the day and a low level at night. Also, it tends to increase its concentration from north-northeast to south-southwest (Moya *et al.*, 2004; Zheng *et al.*, 2008; Wood *et al.*, 2009; Cuevas, 2014). A map of the annual average concentration of HNO_3 micrograms per cubic meter ($\mu\text{g}/\text{m}^3$) for the year 2007 shows that the highest concentration levels are located in the northwest area of the MCMA (Castillo-Miranda *et al.*, 2017). The climatic conditions in the MCMA area are as follows: In 2015, average temperature 16.4 °C. April hottest 18.7 °C. Relative humidity (RH) average 62%, atypical rain during march and April (35 and 17 mm). Average precipitation 672 millimeters (mm). Barometric pressure average 584 millimeters of mercury (mmHg) (581-588 interval). Average wind speed 2.0 meters per second (m/s), highest values during October 2.3 m/s (SEDEMA, 2016). In 2016, average temperature 16.2 °C, May hottest 19.5 °C. RH 57% average. Average precipitation 965 mm. Barometric pressure average 585 mmHg (575 - 591 interval). Average wind speed 2.1 m/s (SEDEMA, 2017). In 2017, average temperature 16.8 °C, May hottest 19.7 °C. RH 54% average. Average precipitation 965 mm. Barometric pressure average 585 mmHg (580 - 591 interval). Average wind speed 2.1 m/s (SEDEMA, 2018c). In 2018, average temperature 16.7 °C, May hottest 19.4 °C. RH 57.1% average. Average precipitation 786.2 mm. Barometric pressure average 584.9 mmHg (583 - 589 interval). Average wind speed 2.0 m/s (SEDEMA, 2020b). In 2019, average temperature 17.4 °C. RH 53.2%, Average precipitation 565.3 mm. Average wind speed 2.1 m/s (CONAGUA, 2019). For pollutant SO_2 has a reduction between 2015 to 2019 from near 70 parts per billion (ppb) go 47 ppb. In case of the NO_2 from near 130 ppb to 110 ppb (SEDEMA, 2020c).

Effects of nitric acid on zinc corrosion

In the modern urban atmosphere, the concentration levels of SO_2 tend to decrease, increasing the relative importance of other trace gases such as NO_2 . The importance of NO_2 lies in being the

precursor of HNO_3 that participates in the materials' degradation processes (Graedel and Schwartz, 1977; Svensson and Johansson, 1993). HNO_3 is a molecule that quickly adsorbs to surfaces, specifically if there is water on the surface. Due to its high solubility in water, HNO_3 is rapidly deposited on surfaces and in water droplets (Seinfeld and Pandis, 2016; Finlayson-Pitts and Pitts Jr., 2000). It experiences dry and wet deposition rapidly, with deposition rates in the range of 1-5 cm/s (Finlayson-Pitts and Pitts Jr., 2000). The atmospheric removal processes for nitric acid gas are by wet and dry deposition. The estimated half-life for dry and wet nitric acid deposition is 1.5 to 2 days and 2 to 3 days, respectively. There is also an efficient removal of nitric acid during precipitation events (Hamilton and Crabbe, 2009). Laboratory studies (Edney and Stiles, 1986; Svensson and Johansson, 1993) have been conducted on the effect of SO_2 , NO_x , and oxidants on the early stages of the galvanized steel atmospheric corrosion. This corrosion is due to the dry deposition of SO_2 and HNO_3 , and HNO_3 is likewise a source of wet NO_3^- (Edney et al., 1986). They also found that the deposition rate was 0.02 centimeter per second (cm/s) for NO_2 , and HNO_3 was 2.0 (m/s). The only relevant role of atmospheric nitrogen is likely to serve as an acidifying species in the precipitation and deposited aerosol particles, as shown in Fig. 1.

Nitric acid is the main gaseous component that dissolves in the aqueous surface layer, and nitrates

come from both particles and the quantity of precipitation. Trace amounts of nitrates in the zinc corrosion layers were discovered, and it was possibly zinc nitrate ($\text{Zn}(\text{NO}_3)_2$) (Graedel, 1989). Friel (1986) obtained nitrate ions in low concentrations from the zinc corrosion layers. Oesch and Faller (1997) reported the formation of basic zinc nitrate $\text{Zn}_5(\text{NO}_3)_2(\text{OH})_8 \cdot 2\text{H}_2\text{O}$, which is formed when zinc is exposed to parts per million (ppm) levels of NO_2 . HNO_3 has a very high dry deposition rate (V_d) that is generally independent of relative humidity and is rapidly adsorbed on most surfaces, making it relatively more harmful for dry and hot climates (Kucera, 2003; Tidblad et al., 2012). Deposited nitrate particles are associated with coarse particles. These coarse particles are commonly alkaline and can also contain sodium chloride. This composition makes them a sink for nitric acid. The reaction can be carried out in the atmosphere or deposited particles (Tidblad et al., 2012). Laboratory experiments and data from the literature show that the relatively high (V_d) of HNO_3 has mainly been attributed to the extreme adherence and reactivity of HNO_3 (Samie, 2007b). Also, the solubility of HNO_3 in water, according to Henry's law coefficients (2.1×10^5 mole per liter atmosphere (mol/L atm), is higher than SO_2 and NO_2 , which have 1.24 and 0.01 mol/L atm, respectively (Seinfeld and Pandis, 2016; Oesch, 1996). It allows a high solubility of HNO_3 in the electrolytic layer and increases the corrosion rate (Samie et al., 2007b). HNO_3 is soluble in water,

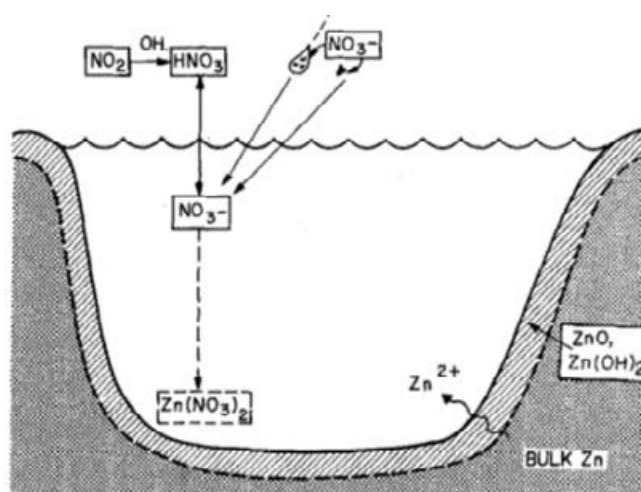


Fig. 1: A schematic representation of the processes involved in the formation of components containing nitrogen during the atmospheric zinc corrosion (Graedel, 1989)

and the concentration in urban environments is relatively low (1-4) $\mu\text{g}/\text{m}^3$ compared to SO_2 (1-20) $\mu\text{g}/\text{m}^3$. Nitrate salts (NO_3^-) are formed when metals react with HNO_3 ; however, nitrate salts are soluble in water, and this may be one reason for the rare occurrence of nitrates in layers of corrosion products (Samie *et al.*, 2007a). Pure zinc plates were exposed in the field for a year, within the Model for Multi-pollutant Impact and Assessment of Threshold Levels for Cultural Heritage (MULTI-ASSESS) and Regional Air Pollution in Developing Countries (RAPIDC) programs in Europe, Asia, and Africa. The Fourier transform spectroscopy (FTIR) technique demonstrated the presence of nitrates as corrosion products. Likewise, nitrates with high solubility are found in greater quantity on the plate's backside since they are less affected by precipitates. Furthermore, essential nitrates ($\text{Zn}_5(\text{NO}_3)_2(\text{OH})_8 \cdot 2\text{H}_2\text{O}$) are generally more soluble in acidic solutions than in water. Therefore, the pH decreases in the absorption layer when acidic contaminants dissolve. Consequently, the essential nitrates are dissolved and removed from the surface by the presence of rain. Accordingly, the zinc front side corrodes more because it has a less protective layer of basic nitrate (Samie *et al.*, 2007a).

Damage functions

The development of various exposure studies has shown that atmospheric corrosion is a phenomenon that varies considerably in each locality (Guttman and Sereda, 1968). The development and use of a damage function applying the fundamental principles of thermodynamics and kinetics of atmospheric corrosion allow predicting galvanized steel structures corrosion rates due to dry and wet deposition (Spence and Haynie, 1990). The dose-response functions should reflect the physicochemical nature of atmospheric corrosion and each parameter's contribution to the global effect (Leuenberger-Minger *et al.*, 2002). Since atmospheric corrosion affects all structures exposed to potentially corrosive environments, many countries have established programs dedicated to understanding and monitoring (Roberge *et al.*, 2002). In some cases, atmospheric exposure programs are conducted to assess the corrosivity of specific atmospheres. This type of information can help choose coatings or corrosion protection systems at particular sites (Dean Jr., 2005). A set of exhibition programs such as International

testing program [] (ISOCORRAG), Ibero-American Map of Atmospheric Corrosiveness (MICAT), International Co-operative Programme on Effects on Materials, including Historical and Cultural Monuments (ICP materials), Model for Multi-pollutant Impact, and MULTI-ASSESS and RAPIDC have been developed in the last decades in different parts of the world. The International Testing Program (ISOCORRAG) began in 1987-1989 with International Organization for Standardization/Technical Committees 156 (ISO/TC 156) test methods and procedures to provide environmental and corrosion data. It includes 53 test sites in 14 countries located in Europe, Argentina, Canada, Japan, New Zealand, and the United States (Knotková, 1993). However, the ISOCORRAG and MICAT programs lack data on gases other than SO_2 and wet deposition (Tidblad *et al.*, 2009). However, atmospheric corrosivity can be estimated with consistent environmental information to establish damage functions, based on a statistical analysis of the databases' information (Morcillo *et al.*, 2002). The ICP Materials program had its first stage (1987-1995), where dose-response functions were developed with long-term data on corrosion and contamination to investigate the effect of acid deposition on materials. It includes non-marine sites, lacks data on the dry deposition of chlorides and HNO_3 and particles. In this period, SO_2 concentrations were still relatively high but decreasing. It led to the need for a new multi-pollutant exposure program (1997-2001). This program aimed to determine both the effect of SO_2 and other important air pollutants such as HNO_3 and particles. In the 2002-2003 period, the MULTI-ASSESS project included HNO_3 and particles as mandatory parameters (Tidblad *et al.*, 2012). Also, RAPIDC project included measurements of HNO_3 and particulate matter. Also, the program was set up with 18 sites in the South and Southeast of Asia and South Africa, which have tropical and subtropical climates. Both the MULTI-ASSESS Program and RAPIDC are the most suitable for investigating the effects of multiple pollutants in non-marine environments (Tidblad *et al.*, 2009). After one year (2002-2003), zinc RAPIDC exposure's corrosion is lower than expected compared to values calculated (Tidblad *et al.*, 2007) using the best available dose-response functions. However, due to the relatively high measured SO_2 levels, it was correct to use functions for the dominant SO_2 situation developed within the ICP Materials program (Tidblad

et al., 2001). The International Organization for Standardization (ISO) ISO 9223 standard is intended to evaluate the corrosivity category for metals. This category explicitly describes the corrosiveness of outer atmospheres through the development of dose-response functions. Considering the above, the dose-response function for zinc is supported on data after one year of exposure. It can, therefore, only be used for classification objects (Zn, N=116, R²=0.78), using Eq. 1 (Tidblad et al., 2002b).

$$\begin{aligned} C_{Zn} &= 0.0053 \text{ SO}_2^{0.43} \text{ TOW}^{0.53} \exp \{f_{Zn}\} + 0.00071 \\ &\text{Cl}^{0.68} \text{ TOW}^{0.30} \exp \{0.11T\} \quad (1) \\ f_{Zn}(T) &= 0 \text{ when } T \leq 10^\circ\text{C}, \text{ otherwise } -0.032(T-10) \end{aligned}$$

Where;

C_{Zn} = Corrosion attack after 1 year of exposure in μm of zinc; T =Temperature in $^\circ\text{C}$; TOW =Time of wetness in hour per year (h/y); SO_2 = SO_2 deposition in milligram per square meter day ($\text{mg}/\text{m}^2\text{day}$); Cl = Cl deposition in $\text{mg}/\text{m}^2\text{day}$.

Corrosivity mapping

Mapping the regional distribution of corrosivity is a method that provides general information on corrosion, rational use, and selection of material protection measures. The mapping must be based on the knowledge of the deterioration and kinetic processes based on external factors, and it generalizes the information supported on local environmental conditions. Likewise, it requires available information on the levels of atmospheric environmental components and converts it into values on the effects of corrosion. The above is achieved using damage equations (dose-response functions) obtained from empirical procedures applied to data processing using GIS technologies (Knotková and Kreislova, 2007). The classification of the different atmospheric basins based on corrosivity serves to select optimal metallic materials and coatings for adequate protection against corrosion in a country (Genescá and Rodríguez, 1992). The atmospheric corrosion maps for metals in Mexico were designed according to the ISO 9223 classification system. They required annual mean corrosion values in test stations, meteorological data, and atmospheric pollution such as SO_2 and chloride ions (Cl^-) (Mariaca et al., 1999). Corrosivity maps were constructed for zinc, copper, and aluminum in the MCMA, by characterizing the

atmospheric aggressiveness on these materials based on the ISO 9223 standard. Furthermore, atmospheric aggressiveness does not necessarily correspond to the most polluted areas since it depends on the relative humidity and wetting time (Muñoz and Uruchurtu, 2002).

MATERIALS AND METHODS

Selection of the dose-response function

The dose-response functions are empirical expressions and then simplifications that indicate the relationship between the rate of corrosion or deterioration and the levels of pollutants combined with climatic parameters (Kucera, 2004). At first, the dose-response functions were more focused on SO_2 , as the main stimulator of corrosion. These functions were denoted as dose-response functions for the dominant SO_2 situation. However, as there have been changes in the atmosphere's chemical composition, there are higher levels of concentration of nitrogen, ozone, and particles than of SO_2 . The above made it possible to propose "multi-pollutant" type functions. Multi-pollutant functions are preferable at high levels of particulate and nitrogen pollutants in the air. The above occurs in urban atmospheres dominated by traffic (Kucera, 2005). Other important factors in applying the multi-pollutant dose-response function are chloride concentration [Cl^-] and Temperature (T). This function is applicable in geographic areas without the influence of chlorides, have been developed and suggested to be implemented in countries with tropical and subtropical climates, characteristic of the MCMA. Likewise, in these climates, the corrosion rate of zinc in field tests consisting of a rack for placing zinc samples and holding passive samplers for gases is lower compared to the corrosion rates obtained with the best available dose-response functions (Tidblad et al., 2007). Table 1 shows the annual average concentration levels of SO_2 , NO_2 , and Cl^- and the yearly average temperature obtained from the monitoring stations located in the MCMA in the period 2015-2019 (SEDEMA, 2020a). The concentration levels of SO_2 , in general, are low and are more noticeable for the year 2019. The concentration levels of NO_2 are higher than SO_2 . The Cl^- concentration is less than 5 milligrams per liter (mg/L), and the temperature is greater than 10°C , which suggests the use of a multi-pollutant dose-response function that includes the effect of HNO_3 .

Table 1: Measurements of [SO₂], [NO₂], [Cl⁻] and T annual average in the period 2015-2019

Parameter	Year				
	2015	2016	2017	2018	2019
SO ₂ (µg/m ³)	8.45	8.48	9.63	8.54	6.87
NO ₂ (µg/m ³)	36.46	35.28	36.61	36.30	31.58
Cl ⁻ (mg/L)	0.45	0.29	0.25	0.24	0.21
T (°C)	16.51	16.42	16.44	16.26	17.17

Dose-response functions are importantly applied to map areas of increased risk corrosion and to calculate corrosion costs. Furthermore, it is possible to estimate the loss of mass of zinc for a multi-pollutant situation (Kucera *et al.*, 2007). The statistical processing of data from materials exposed in the field generated a dose-response function for the loss of zinc mass after one year of exposure, using Eq. 2 (Kucera, 2005).

$$ML = 3.53 + 0.471[SO_2]^{0.22} e^{0.018RH+f(T)} + 0.041Rain [H^+] + 1.37[HNO_3] \quad (2)$$

$$f(T) = 0.062(T-10) \text{ when } T \leq 10^\circ C,$$

$$f(T) = -0.021(T-10) \text{ otherwise}$$

Where ML is the corrosion rate in g/m², [SO₂] and [HNO₃] is the concentration in micrograms per cubic meter (µg/m³), RH is the relative humidity in %, Rain is the amount of precipitation in millimeters (mm), [H⁺] is the H⁺ concentration of rainfall in mg/L, and T is the temperature in °C. Eq. 2 shows an evident effect of HNO₃ on zinc corrosion and represents the sum of two contributions, one from HNO₃ and the other from corrosion-enhancing factors including SO₂ (Samie *et al.*, 2007a). They also found a relatively good correlation between the extrapolated zinc mass loss, based on laboratory exposures, and the dose-response function based on a statistical analysis of field data. The multi-pollutant function includes nitric acid that any MCMA monitoring station does not measure. Eq. 3 developed within the MULTI-ASSESS project can be used to calculate the annual concentration of HNO₃ when there are no values (Kucera, 2005):

$$[HNO_3] = 516 \cdot e^{-3400/(T+273)} ([NO_2] \cdot [O_3] \cdot RH)^{0.5} \quad (3)$$

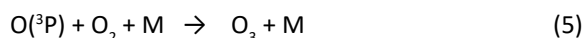
Where [HNO₃], [NO₂] and [O₃] is the concentration in µg/m³; RH is the relative humidity in %, and T is the temperature in degree Celsius (°C). The oxidation process from NO₂ to HNO₃ described in equations (4-

21) contemplates the chemical species of Eq. 3.

The NO₂ molecule absorbs sunlight at wavelengths <430 nm and can break one of the NO bonds and generate the reactive ground-state oxygen atom, the triplet-P oxygen atom, O (³P), and a NO molecule, using Eq. 4 (Seinfeld, 1980).



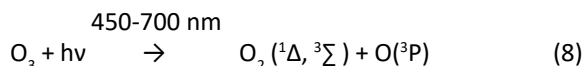
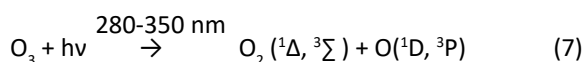
The exceptionally reactive triplet-p oxygen atom formed regularly collides with oxygen molecules to form ozone, using Eq. 5.



Under typical polluted atmosphere conditions, the ozone molecule will often react with NO to regenerate NO₂, using Eq. 6.



Ozone can also photolyze, using Eqs. 7 and 8.



The singlet-D oxygen atom is considerably more reactive than the ground-state triplet-P oxygen atom, using Eq. 9. Apart from deactivation.

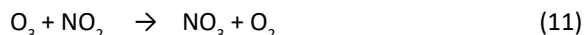


It reacts with water to form the hydroxyl radical, using Eq. 10.



Ozone can react with nitrogen dioxide to produce

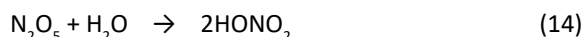
symmetrical nitrogen trioxide, using Eq. 11.



The NO_3 species forms dinitrogen pentaoxide, by reaction with nitrogen dioxide, using Eq. 12.



Dinitrogen pentaoxide can rearrange to form NO_3 and NO_2 or maybe react with water to form nitric acid, using Eqs. 13 and 14.



The subsequent reaction can take place between oxygen atoms and NO_2 , using Eq. 15.



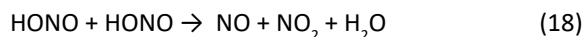
In addition, NO and NO_3 can react to regenerate NO_2 , using Eq. 16.



Nitrous acid is generated by using Eq. 17.



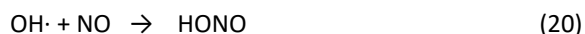
And can react bimolecular to regenerate the original reactants, using Eq. 18.



Photolysis of nitrous acid in the wavelength range 280-400 nm guide to producing hydroxyl radical and NO , using Eq. 19.



The hydroxyl radical may react with NO and NO_2 to yield nitrous and nitric acid using Eqs. 20 and 21, respectively.



Applied method

Mapping of the corrosion rates was carried out through the following procedure: Integration of the available data from measurements of concentrations of sulfur dioxide (SO_2), estimated nitric acid (HNO_3), relative humidity (RH), rainfall, or amount of precipitation (PP), temperature (T) and the acidity of the rain $[\text{H}^+]$. The data and results were applied in suitable mathematical functions to construct, by interpolation, maps of their spatial distribution. Subsequently, with these values, the multi-pollutant function corresponding to zinc is applied to obtain the corrosion layers. A description of these steps is presented in the following paragraphs.

Damage function parameter database

The database was integrated with the multi-pollutant function parameters to calculate the corrosion rate in the period 2015-2019. These parameters are obtained from the different monitoring stations as described down below. For this study, SO_2 data generated by the Red Automática de Monitoreo Atmosférico (Automatic Atmospheric Monitoring Network, RAMA) for the period (2015-2019) are used. The estimation of HNO_3 required annual average values per hour of the parameters: Nitrogen dioxide (NO_2) and ozone (O_3) from the (RAMA); in addition to temperature (T) and relative humidity (RH) from the databases of the Red de Meteorología y Radiación Solar (Atmospheric Monitoring Network and the Meteorology and Solar Radiation Network, REDMET). The values of the acidity of the precipitation $[\text{H}^+]$ and the amount of rainfall or rain (PP) included in the multi-pollutant function were obtained from data reported by the Red de Depósito Atmosférico (Atmospheric Deposit Network, REDDA) for the period (2015-2019). The values of temperature (T) and relative humidity (RH) included in the dose-response function were obtained from the database of the Red de Meteorología y Radiación Solar (Meteorology and Radiation Network, REDMET) for the period (2015-2019) (SEDEMA, 2020a).

Creating layers using geographic information systems Selecting the interpolation method

Geographic information systems (GIS) are an indispensable technology for capturing, storing, analyzing, modeling and displaying spatially

referenced data (Moreno, 2006a). Conesa (1996) indicated that GIS could process large databases with information about the land surface globally and globally. One of a GIS's purposes is to provide a spatial framework to support decisions about the efficient use of the earth's resources and manage the anthropogenic environment (Zeiler, 1999). The kriging method combined with deterministic modeling improves the spatial analysis of acid deposition (Venkatram, 1988). Modeling temperature and relative humidity were interpolated using the kriging method, and the standard deviation (SD) was used to measure the spatial uncertainty (Phillips and Marks, 1996). The inverse distance weighting (IDW) and kriging spatial interpolation techniques are regularly used to estimate pollutant concentration levels in areas with limited sample points. The value of each parameter is determined by cross-validation until the least mean square error (RMSE) is obtained (Rojas-Avellaneda, 2007; Diem and Comrie, 2002). Pollutant concentration maps using the kriging method are used in the European Monitoring and Evaluation Materials Program (EMEP) (Denby *et al.*, 2005). Modeling the temporal behavior of SO₂ using the ordinary kriging method provides an estimate of the air quality in Europe. It is suggested to use a semivariogram with a spherical model to obtain the SO₂ layer (Denby *et al.*, 2010). In this study, spatial representation was developed using Aeronautical Reconnaissance Coverage Geographic Information System (ArcGIS) software (ArcGIS, 2014). ArcGIS uses the extension called "geostatistical analysis". It employs advanced tools to perform exploratory analysis of spatial data and a wizard with which statistical surfaces are constructed. The layers' construction was decided to build vector layers with both IDW and ordinary kriging for each of the parameters. The selection of the interpolation method for each parameter was based on obtaining the lowest value of the Root Mean Square (RMS) of the cross-validation. The coefficient of determination R² of the relationship observed value vs. predicted value resulting from the cross-validation is obtained. The ordinary kriging interpolation method was used to obtain the layers of the following parameters, with their corresponding annual coefficient of determination R²: [SO₂]: 2015 = 0.60; 2016 = 0.52; 2017 = 0.76; 2018 = 0.71 and 2019 = 0.67. [HNO₃]: 2015 = 0.55; 2016 = 0.76; 2017 = 0.73; 2018 = 0.60 and 2019 = 0.60. HR: 2015 = 0.02;

2016 = 0.17 and 2018 = 0.15. T: 2015 = 0.44; 2016 = 0.60; 2017 = 0.41; PP: 2016 = 0.67; 2017 = 0.87; 2018 = 0.70 and 2019 = 0.72. [H⁺]: 2017 = 0.01; 2018 = 0.20 and 2019 = 0.21. The IDW interpolation method was employed to obtain the following parameters' layers, with their corresponding annual coefficient of determination R²: HR: 2017 = 0.03 and 2019 = 0.01. T: 2018 = 0.40 and 2019 = 0.32. PP: 2015 = 0.26. [H⁺] 2015 = 0.09 and 2016 = 0.52.

RESULTS AND DISCUSSION

Layer maps of the parameters of the dose-response function and the corrosion rate

The representation of the maps of the dose-response function parameters focuses mainly on SO₂ as one of the main factors in the corrosion process and HNO₃ regarding corrosion in the urban atmosphere. The sulfur dioxide [SO₂] layers for the period 2015-2019 are shown in Fig. 2. The figure indicates spatially that the highest [SO₂] levels are located to the "northwest" and the lowest to the "east, northeast" and "southeast" and a lesser extent south of the MCMA. From a temporal perspective, the highest levels of [SO₂] are present in 2017 and 2018, while the lowest levels occur in the year 2019, followed by 2015 and 2016. The [HNO₃] layers for the period 2015-2019 are represented in Fig. 3. The figure shows spatially that the highest [HNO₃] levels are located in the "central" and "southwest" areas and the lowest in the "east" and "northeast" areas of the MCMA. From a temporal viewpoint, the highest [HNO₃] levels are in 2015 and 2016, while the lowest levels are in 2016, followed by 2018. The importance of the annual change in the levels of HNO₃ concentration observed in the maps allows visualizing the urban atmosphere's complexity in relation to its acidity.

The corrosion layers for zinc used in the evaluation were constructed using the "raster calculator" tool included in the ArcGIS 10.2.2 spatial analysis extension. It performs analysis operations with mathematical functions. The process consists of applying the multi-pollutant function, introducing the raster layers of the corresponding parameters. These layers represent the terrestrial space's property through a set of square cells of the same size, called pixels. The raster model is appropriate to represent continuous variables in space, such as meteorological variables and air pollution (Moreno, 2006b). The corrosion rate maps construction results are shown in Figs. 4 and 5.

The layers of the corrosion rates employing the multi-pollutant function of equation 2 for 2015-2019 are shown in Fig. 4. The images show a corrosion rate of 5.81-8.19 of mass loss (g/m²). Spatially, the highest corrosion rates are located in the “southwest” of the

MCMA. On the other hand, the lowest corrosion rates are seen in the “northeast-southeast” of the MCMA. The highest corrosion rates present in 2015 and 2016, while the lowest corrosion rate occurs in the year 2019. Likewise, the corrosion rates’ layers using the

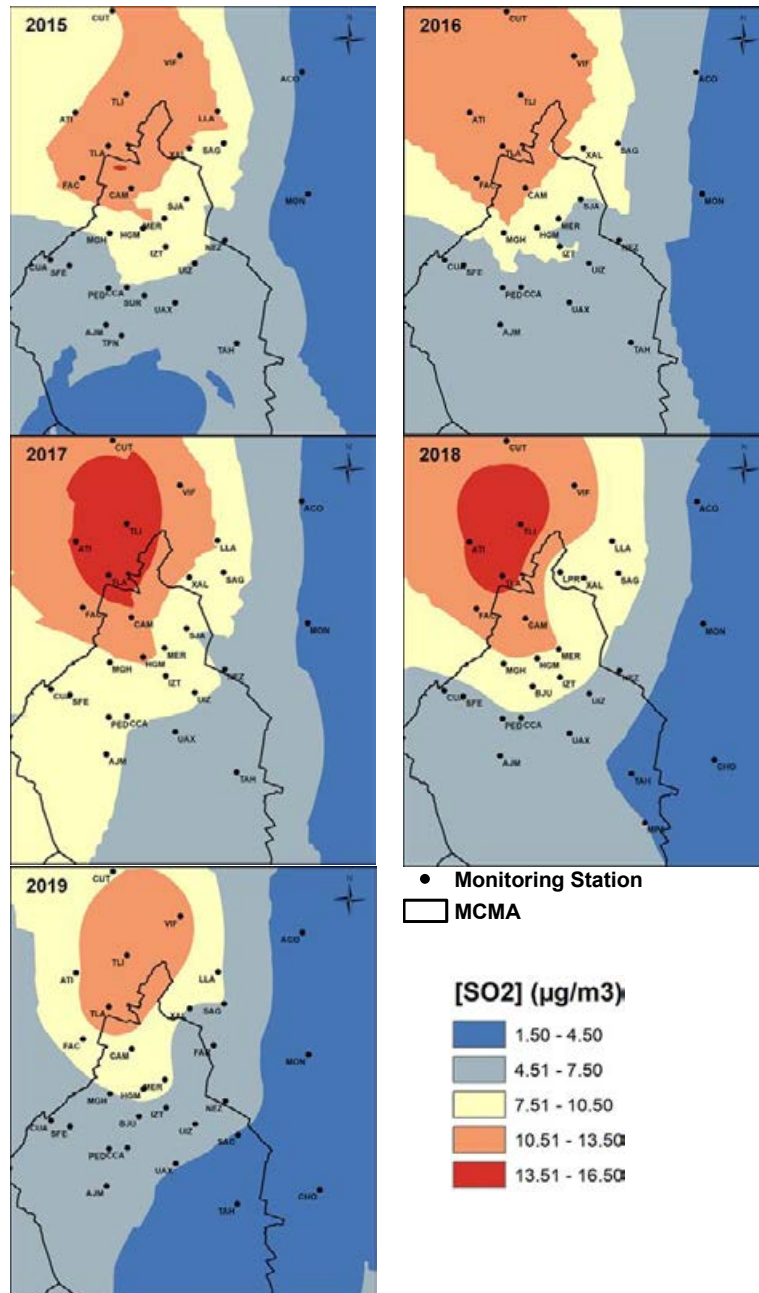


Fig. 2: Maps of concentration layers of the annual average of sulfur dioxide [SO₂] (µg/m³) for the period 2015-2019 in the MCMA

multi-pollutant function of equation 2, without the component that includes HNO_3 for 2015-2019, are shown in Fig. 5. The images establish a corrosion rate interval of [4.86-6.64] of mass loss (g/m^2). Spatially, the corrosion rates represent the same trend as in Fig.

4. The highest rates are located in the “southwest” of the MCMA, although the area covered is smaller. Meanwhile, the lowest corrosion rates are seen in the “northeast-southeast” of the MCMA, covering larger areas. In the same way, temporarily, the highest

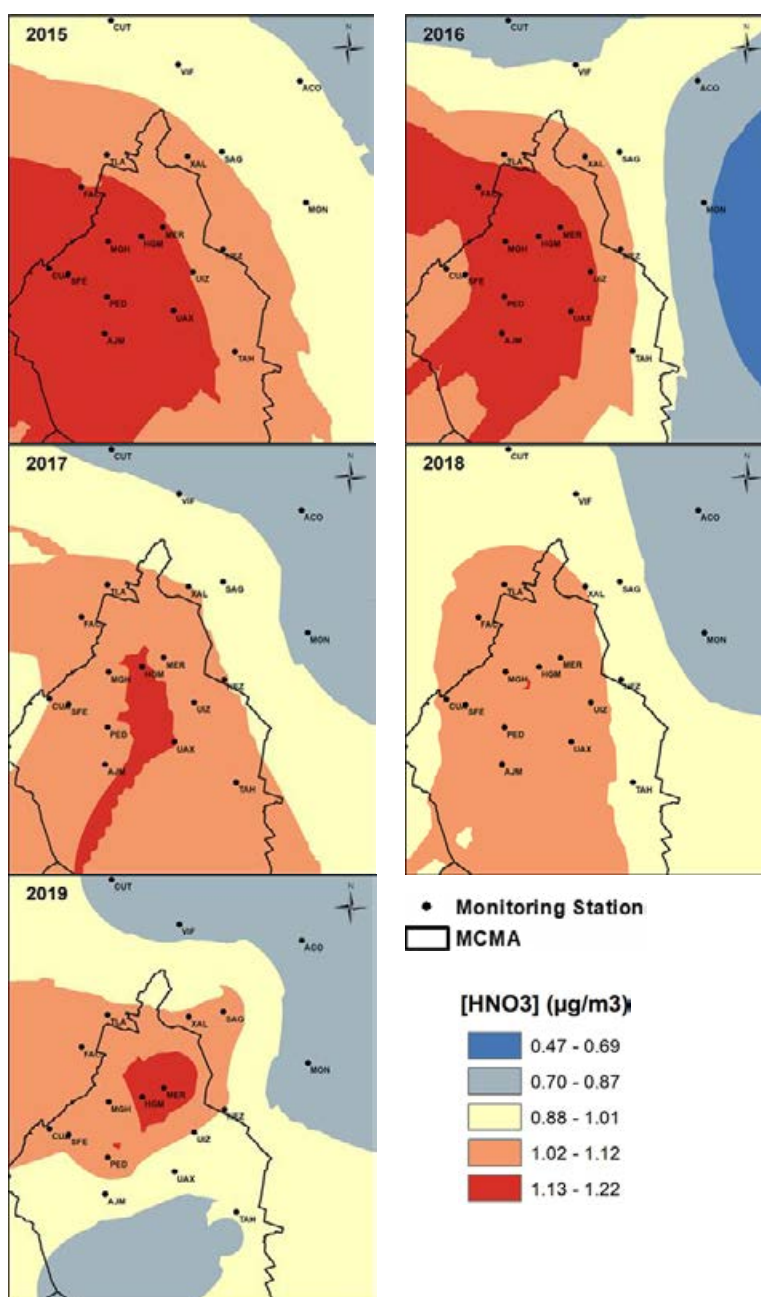


Fig. 3: Maps of concentration layers of the annual average of [HNO_3] ($\mu\text{g}/\text{m}^3$) for the period 2015-2019 in the MCMA

corrosion rates present in 2015 and 2016, while the lowest corrosion rate occurred in 2019. The highest corrosion rates are presented in the years 2015 and 2016. It is probably due to the high levels of $[\text{HNO}_3]$ observed in Fig. 3. In contrast, the lowest corrosion

rate occurred in 2019. The above is probably due to both sulfur dioxide and nitrogen dioxide, which present the lowest concentration levels in the 2015-2019 period. However, the fact that the corrosion rate for 2019 is the lowest in magnitude compared

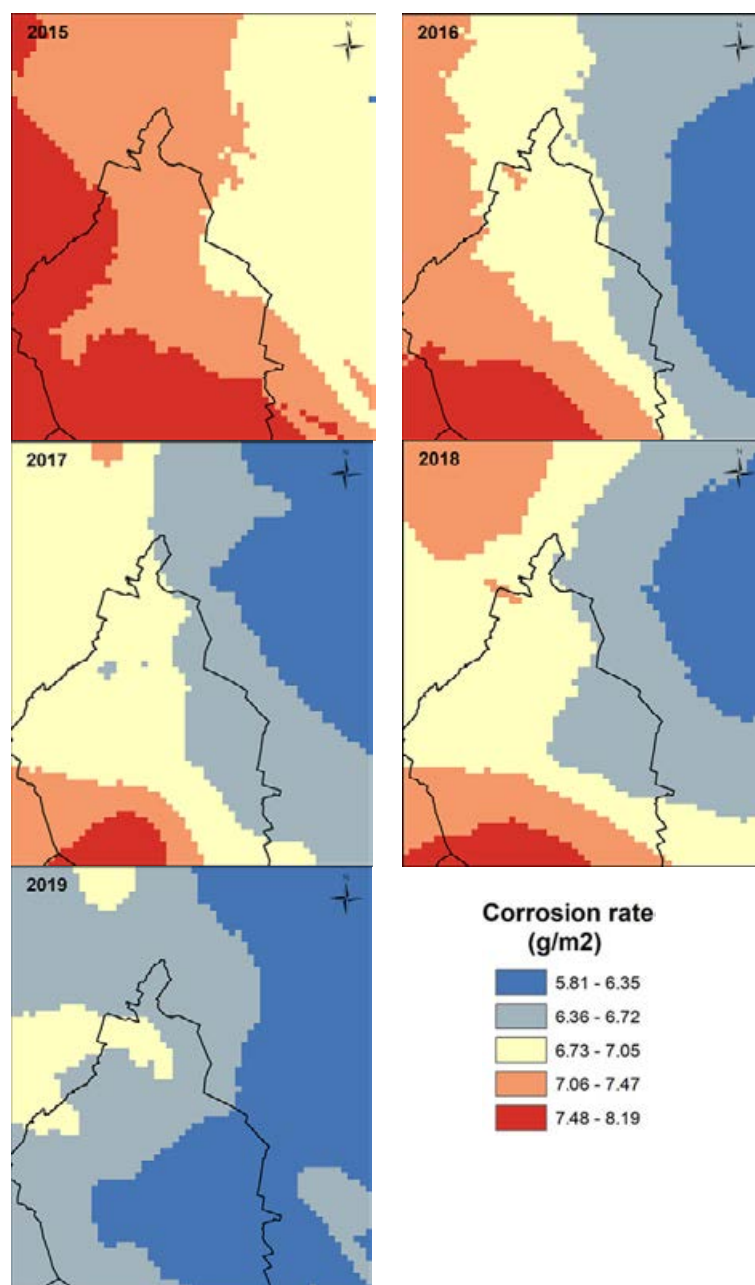


Fig. 4: Corrosion rate maps (g/m^2) of zinc, for a multi-pollutant situation for the period 2015-2019 in the MCMA

to other years, in percentage terms, has a significant influence on the corrosion rate for this year. The low determination coefficient values R^2 were obtained mainly in constructing the climatic parameters' layers, possibly due to the nature and variability of

the data obtained from the monitoring networks. The HNO_3 percentage contribution layers to the annual zinc corrosion rate are shown in Fig. 6. The impact is determined by combining the layers of corrosion rates of Figs. 4 and 5 in a percentage relationship

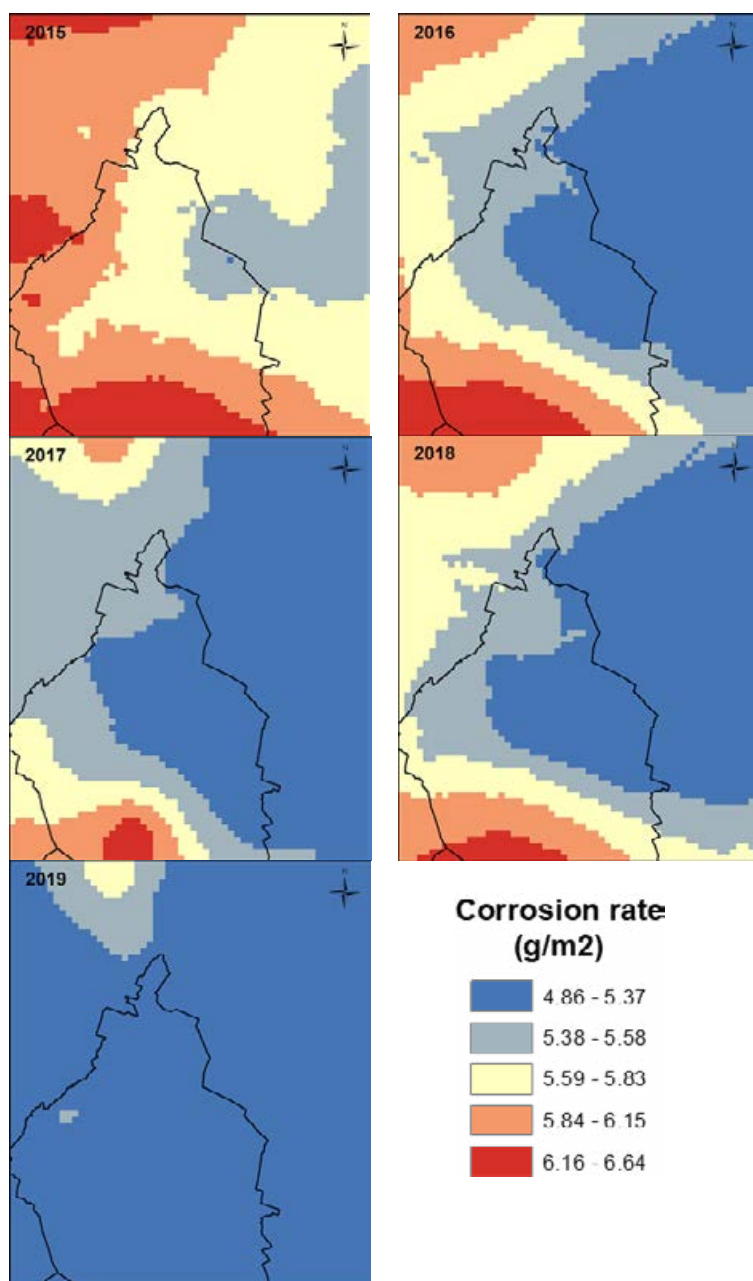


Fig. 5: Corrosion rate maps (g/m^2) of zinc, for a multi-pollutant situation without the HNO_3 component for the period 2015-2019 in the MCMA

for 2015-2019. The images indicate a % contribution interval of 12.23-32.15. Spatially, the areas with the greatest contribution are located in the “center”, and the areas with the least contribution are to the “east” of the MCMA. Temporarily the areas with the

highest contribution are presented in the years 2015, 2016, and 2019. This is mainly due to the decrease in $[\text{SO}_2]$ levels and the oxidation process of NO_2 to HNO_3 that occurs in the complex urban atmosphere of the MCMA, while the areas with the lowest contribution

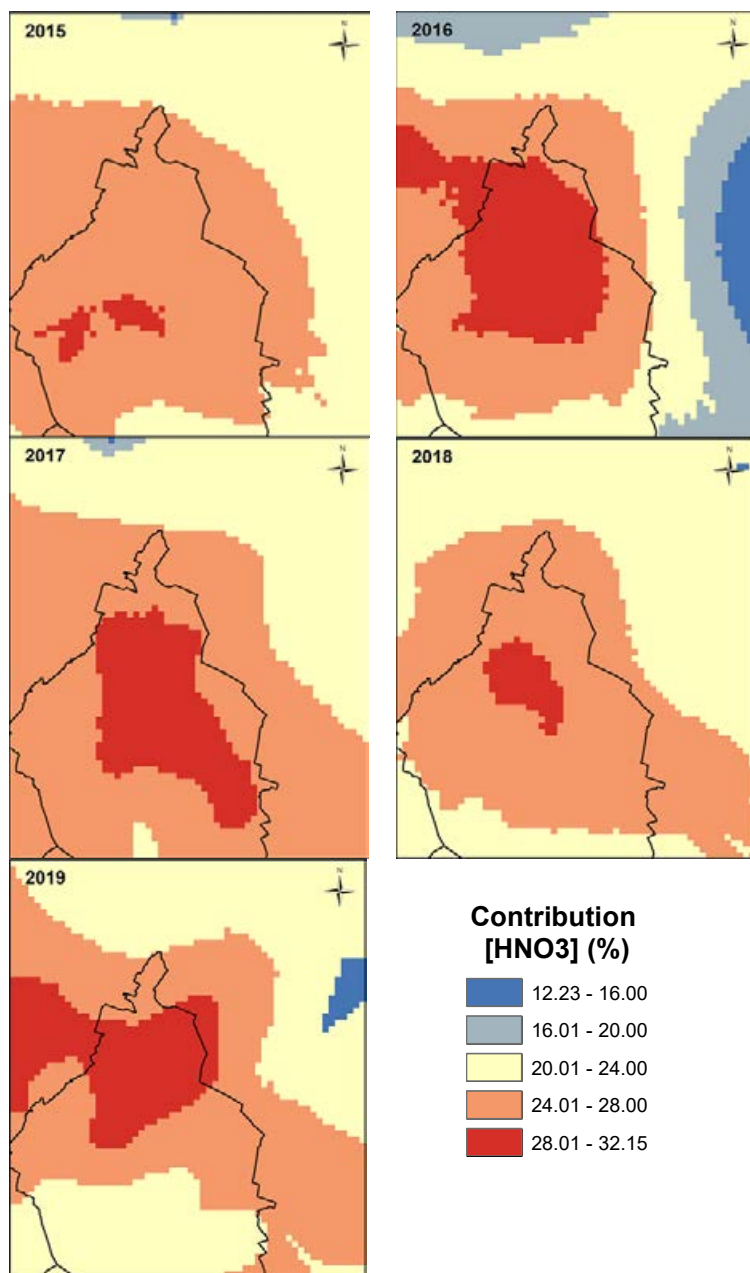


Fig. 6: Maps of the contribution (%) of HNO_3 to the annual zinc corrosion rate (g/m^2) of zinc, for a multi-pollutant situation for the 2015-2019 period in the MCMA

Table 2 Environmental data including temperature (T), relative humidity (RH), precipitation (Rain and pH) and gaseous pollutants (HNO₃ and SO₂). (Tidblad *et al.*, 2007)

Country	Test site name	SO ₂ (µg/m ³)	Temperature (°C)	Relative humidity (%)	Rain (mm)	pH	HNO ₃ (µg/m ³)
India	Bhubaneswar-u	4	26.5	69	425	6.0	1.3
India	Bhubaneswar-r	3	26.5	69	425	6.0	1
Thailand	Bangkok	11	29.3	76	1,371	6.8	2.3
Thailand	Phrapradaeng	59	29.3	73	1,335	6.2	1.5
Vietnam	Hanoi	15	24.7	79	1,556	5.8	0.8
Vietnam	Ho Chi Minh	21	28.3	74	1,441	6.2	0.9
Vietnam	Mytho	2	27	81	1,222	6.4	0.3
China	Chongqing	99	18.5	70	1,162	4.5	1.3
China	Tie Shan Ping	51	18.5	90	1,133	4.2	1.8
China	Hong Kong	16	22.9	78	2,092	4.6	1.8
Malaysia	Kuala Lumpur	12	28	78	2,776	4.3	3.8
Malaysia	Tanah Rata	0	18.1	91	2,433	5.1	0.1
South Africa	Johannesburg	18	17.2	78	417	4.8	2.1
Zambia	Kitwe	92	22.6	58	1,083	4.7	0.9
Zambia	Magoye	0	22.2	62	826	7.0	0.5
Zimbabwe	Harare	16	18.9	63	798	6.6	0.7

Table 3. Estimation of the % contribution of HNO₃ to the zinc corrosion rate (g/m²) for different test sites

Country	Test site name	ML (g/m ²) of zinc, for a multi-pollutant situation	ML (g/m ²) of zinc, for a multi-pollutant situation without the HNO ₃ component	Contribution % HNO ₃
Malaysia	Tanah Rata	4.46	4.32	3.17
Vietnam	Mytho	5.61	5.20	7.90
Zimbabwe	Harare	6.73	5.77	16.61
Zambia	Kitwe	8.43	7.19	17.14
Vietnam	Hanoi	7.33	6.23	17.58
Zambia	Magoye	4.22	3.53	19.39
China	Chongqing	10.63	8.85	20.12
Vietnam	Ho Chi Minh	7.17	5.94	20.75
China	Tie Shan Ping	13.66	11.19	22.04
India	Bhubaneswar-r	6.39	5.02	27.31
China	Hong Kong	10.84	8.38	29.44
Thailand	Phrapradaeng	8.49	6.43	31.96
India	Bhubaneswar-u	6.89	5.11	34.84
South Africa	Johannesburg	9.79	6.91	41.61
Malaysia	Kuala Lumpur	16.71	11.50	45.25
Thailand	Bangkok	8.78	5.63	55.98

% are located in the year 2016. The difference is due to the annual concentration levels of HNO₃ compared to SO₂ and the other climatic parameters of the dose-response function.

On the other hand, in the latitudinal position of the MCMA, solar radiation is intense and accelerates photochemical reactions of pollutants in the atmosphere such as ozone and aerosols. Since ozone is involved in the formation of nitric acid, empirical equation 3 derived for other conditions

must be reevaluated. There are limitations modeling atmospheric corrosion by empirical equations obtained in other situations. For example, in many cases, the weather when the exposure starts is relevant. It is different to start an atmospheric exposure in summer than in winter. Also, using annual mean temperature values for the MCMA has temperatures below 10 °C for many periods throughout the year. This effect is probably related to the time of wetness (TOW), an important variable that is not explicitly

considered in the MULTI- ASSESS equation. Table 2 shows gaseous pollutant and environmental data from a network of test sites that includes 12 sites in Asia (India, Vietnam, Thailand, Malaysia, and China, including Hong Kong) and four test sites in Africa (South Africa, Zambia, and Zimbabwe). Implemented by the RAPIDC program and funded by the Swedish International Development Agency (SIDA), in order to obtain corrosion values for materials including zinc after one year of exposure (2002-2003) (Tidblad *et al.*, 2007).

Table 3 shows the estimate of the % contribution of HNO_3 to the zinc corrosion rate (g/m^2). This is possible, using the values from Table 2 in equation 2 and combining in a percentage relationship the corrosion rate (g/m^2) of zinc in a multi-pollutant situation with the multi-pollutant situation without the HNO_3 component. Table 3 shows a contribution interval % HNO_3 of 3.17 - 55.98 with a great difference in the climatic and pollution values of the test sites. Johannesburg 41.61%, Kuala Lumpur 45.25% and Bangkok 55.98% show the highest HNO_3 contribution % values corresponding to the highest measured HNO_3 values. While Tanah Rata 3.17% and Mytho 7.90% have the lowest % contribution values of HNO_3 in relation to the lowest values of HNO_3 measured.

Comparing the interval of % contribution of HNO_3 of 12.23-32.15 observed in fig. 6 with the interval of values of 3.17-55.98 of Table 3, it is highlighted that it is possible to place it at an intermediate level with respect to the values of Table 3. However, the values in Table 3 are from more than a decade ago; there was already evidence of a significant contribution % of HNO_3 to the zinc corrosion process. However, at that time there was an important attention to SO_2 .

CONCLUSION

Most studies on atmospheric zinc corrosion and galvanized steel carried out in Mexico and the Mexico City Metropolitan Area have focused on contaminants such as sulfur dioxide and sodium chloride, coupled with meteorological factors such as those responsible for damage to urban infrastructure and objects made of zinc or galvanized Steel. This work estimates nitric acid's contribution to zinc corrosion in the Mexico City Metropolitan Area. This contribution is represented as a component that includes HNO_3 in the dose-response functions for a multi-pollutant situation. In the multi-pollutant case, other factors also intervene,

such as sulfur dioxide, relative humidity, amount of precipitation, temperature, and the acidity of the rain. The construction of corrosion rate maps using dose-response functions for a multi-pollutant situation in equation (2) provides a spatial and temporal estimate of the variation in corrosion rates in 2015-2019. This estimate allows visualizing areas where zinc materials are at risk corrosion due to the dispersion of atmospheric pollutants and climatic parameters. The construction of percentage maps shows the contribution (%) of HNO_3 . HNO_3 contributes at least 12% and up to 32% to the zinc corrosion rate. These layers show that the years 2015, 2016, and 2019 present the highest contribution percentages. The equation (2) is applicable in geographic areas without the influence of chlorides and countries with tropical and subtropical climates, characteristic of the MCMA. Therefore, it is likely that the corrosion rate values obtained with the multi-pollutant dose-response function in this work are overestimated. It is desirable to have specific multi-pollutant dose-response functions for the MCMA that adequately represent the dispersion of pollutants and climatic parameters of the study area's urban atmosphere. It would be possible to implement an Exposure Program on the effects of atmospheric zinc corrosion, with specific attention to HNO_3 . There is a relatively good correlation between the extrapolated loss of zinc mass resulting from laboratory exposures and the dose-response function based on statistical analysis. Therefore, the corrosion rates estimated for zinc in this work are very likely to represent an approximate value of the corrosion rates for zinc that can be found in field tests. This due to the absence of field tests.

AUTHOR CONTRIBUTIONS

J.O. Castillo-Miranda conceived the original idea, performed the literature review, analyzed and interpreted the data, prepared the manuscript text, and manuscript edition. F.J. Rodríguez-Gómez worked out the technical details, analyzed and interpreted the data, prepared the manuscript text, and manuscript edition. J. Genescá-Llongueras encouraged to investigate a specific aspect, helped in the literature review and manuscript preparation. L.G. Ruiz-Suárez developed the theoretical formalism and supervised the findings of this work. J.A. García-Reynoso helped shape the study, analysis and contributed to the interpretation of the results.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

%	Percentage
°C	Degree Celsius
µg/L	Micrograms per liter
µg/m ³	Micrograms per cubic meter
ArcGIS	Aeronautical reconnaissance coverage Geographic Information System
Ca ²⁺	Calcium ion
Cl ⁻	Chlorides
CLRTAP	Long-range Transboundary Air Pollution
cm/s	Centimeters per second
CO ₂	Carbon dioxide
DGAPA-UNAM	Dirección General de Asuntos del Personal Académico de la Universidad Nacional Autónoma de México
EMEP	European Monitoring and Evaluation Materials Program
Eq.	Equation
Fig.	Figure
FTIR	Fourier transform spectroscopy
g/m ²	Grams per square meter
GIS	Geographic information systems
h/y	Hour per year
H ⁺	Hydrogen ion

H ₂ O	Water
H ₂ S	Hydrogen sulfide
HCl	Hydrogen chloride
HNO ₃	Nitric acid
ICP materials	Co-operative programme on effects on materials, including historic and cultural monuments
IDW	Inverse distance weighting
ISO	International Organization for Standardization
ISOCORRAG	The International Testing Program
ISO/TC 156	International Organization for Standardization/Technical Committees 156
K ⁺	Potassium ion
km	Kilometres
m/s	Meters per second
MCMA	Mexico City Metropolitan Area
mg/L	Milligrams per liter
mg/m ² /day	Milligrams per square meter day
Mg/y	Megagrams per year
MICAT	Ibero-American Map of Atmospheric Corrosiveness
ML	Corrosion rate
mm	Millimeters
mmHg	Millimeters of mercury
mol/L atm	Mole per liter atmosphere
MULTI-ASSESS	Model for multi-pollutant impact and assessment of threshold levels for cultural heritage
Na ⁺	Sodium ion
NO ₂	Nitrogen dioxide
NO ₃ ⁻	Nitrate ion
NO _x	Nitrogen oxides
O ₃	Ozone
pH	Hydrogen potential
PP	Precipitation or rain
ppb	Parts per billion
ppm	Parts per million
R ²	Coefficient of determination
Rain	Amount of precipitation
RAMA	Automatic Atmospheric Monitoring Network
RAPIDC	Regional Air Pollution in Developing Countries

REDDA	Atmospheric Deposit Network
REDMET	Atmospheric Monitoring Network and the Meteorology and Solar Radiation Network
RH	Relative humidity
RMS	Root Mean Square
RMSE	Least mean square error
SD	Standard deviation
SIDA	Swedish International Development Agency
SO ₂	Sulfur dioxide
SUV	Sport utility vehicle
T	Temperature
TOW	Time of Wetness
UN ECE	Convention of the United Nations Economic Commissions for Europe
V _d	Deposition rate
Zn(NO ₃) ₂	Zinc nitrate
Zn ₅ (NO ₃) ₂ (OH) ₈ • 2H ₂ O	Basic nitrate or essential nitrate

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ORIGINAL RESEARCH PAPER

Simulation and analysis of marine hydrodynamics based on the El Niño scenario

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ABSTRACT

BACKGROUND AND OBJECTIVES: El Niño- Southern Oscillation is known to affect the marine and terrestrial environment in Southeast Asia, Australia, northern South America, and southern Africa. There has been much research showing that the effects of El Niño- Southern Oscillation are extensive. In this study, a simulation of an El Niño event is carried out, which is ideal in the vertical layer of the Pacific Ocean (0-250 meters). The fast Fourier transform is used to process the vertical modeling data so that the results can accurately represent El Niño.

METHODS: A non-hydrostatic 3-dimensional numerical model is used in this research. To separate the signal produced and obtain the quantitative difference of each sea layer, the simulation results are analyzed using the fast Fourier transform. Winds blow from the west to the east of the area in perfect El Niño weather, with a reasonably high wind zone near the equator (forming a cosine). Open fields can be found on the north and south sides, while closed fields can be found on the west and east sides. Density is uniform up to a depth of 100 meters, then uniformly increases by 1 kilogram per cubic meter from 100 to 250 meters.

FINDINGS: The results of the model simulation show that one month later (on the 37th day), the current from the west has approached the domain's east side, forming a complete coastal Kelvin wave. The shape of coastal Kelvin waves in the eastern area follows a trend that is similar to the OSCAR Sea Surface Velocity plot data obtained from ERDDAP in the Pacific Ocean in October 2015. In this period, the density at a depth of 0-100 meters is the same, while the density at the depth layer underneath is different.

CONCLUSION: Strong winds could mix water masses up to a depth of 100 meters, implying that during an ideal El Niño, the stratification of the water column is influenced by strong winds. The eastern domain has the highest sea level amplitude, resulting in perfect mixing up to a depth of 100 m, while wind effect is negligible in the lower layers. The first layer (0-50 m) and the second layer (50-100 m) have the same density and occur along the equator, according to FFT. The density is different and much greater in the third layer (100-150 m).

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INTRODUCTION

El Niño - Southern Oscillation (ENSO) is a manifestation of the inter-annual variability, but it is associated with major changes in the Pacific atmosphere through a phenomenon known as southern oscillation (Chen *et al.*, 2019a; Trenberth, 2019). Warming of the tropical Pacific Ocean surface and strengthening equatorial trade winds characterize El Niño events, which occur every few years. During El Niño years, such as 2015/2016, winds from the west are typically symmetrical (Sundararajan, 2020).

The El Niño outbreak in 2015/2016 was caused by a series of westerly wind events in boreal springs. There has been much research showing that the effects of ENSO are extensive. Doughty *et al.* (2021) reported that ENSO affects the tropical forests of the Amazon, which are the world's major CO₂ players, where the rates of photosynthesis, fluorescence, and greenness were above average during El Niño (2009/2010) and below normal during La Nina (2010/2011). Hao *et al.* (2018) shows that ENSO causes dry and hot summer events in such as northern South America, southern Africa, Southeast Asia and Australia. Meanwhile, in East Asia's subtropical forests, El Niño can increase the growth of individual trees through increased sunlight (Li *et al.*, 2020). In the fishing sector, El Niño can cause anomalous fish migration patterns, such as changes in latitudinal migration of sardinella along the African coast (López-Parages *et al.*, 2020). Pécastaing and Chávez (2020) also reported that ENSO makes coastal communities vulnerable to poverty, especially for communities in Peru's arid ecosystems. It must be taken into account that going forward, the potential for El Niño is projected to increase drastically due to the effects of global warming associated with the flattening of thermoclines in the equatorial Pacific (Yeh *et al.*, 2009; Chen *et al.*, 2019b). El Niño also has relatively strong connections to other oceans. Siswanto *et al.* (2020) imply that low and high trophic level marine organisms in the eastern Indian Ocean were also affected by ENSO. Yue *et al.* (2020) reported that the zone of anomaly induced by wind advection during El Niño affects the Walker circulation and sea surface salinity in the Indian Ocean. In addition, ENSO caused changes in the circulation of the tropical Pacific Walker due to warming in the Indian and Atlantic Oceans (Behera *et al.*, 2021). El Niño affects salinity so that one of the parameters to determine the occurrence of El Niño is sea surface salinity

(Sharma, 2018). So far, Sundararajan (2020) stated that the wind is an equinoctial symmetry in the 2015 El Niño year. This affects the magnitude of the tidal currents in the Pacific. According to Hu *et al.* (2013), ENSO behavior is influenced by the low-frequency variability of the tropical Pacific climate. These variations include increased warming and local wind variability in the Pacific Ocean's tropical parts (Hu *et al.*, 2013). Since the late 1970s, the ENSO frequency has shifted, with the ENSO frequency being high (2-4 years) in 1962-1975 and low (4-6 years) in 1980-1993 (An and Wang, 2000; Fedorov and Philander, 2000). In 1999-2000, the correlation between warm water volume around the equator and ENSO was also seen to be weaker. According to Horii *et al.* (2012) and McPhaden (2012), this condition relates to the divergence of winds in the central Pacific that inhibits the eastward spread of sea surface temperature anomalies. Weak east-west sea surface temperature gradient provides relaxation to trade winds. Consequently, with weak trade wind conditions, the equatorial upwelling, which carries cooler, high-density water to the Pacific surface, is also reduced (Kumar and Hu, 2014). Rizal *et al.* (2019; 2020) found that variations in density gradients influence the form of lee waves and baroclinic currents that contribute to the mainstream in a basin. According to recent studies, the ENSO regime continues to change spatially over time, and this is due to local wind variability. The aim of this study is to determine the properties of the sea level and density as a result of El Niño, which is best modeled using a non-hydrostatic 3D numerical model. The impact of the wind on the sea level and density at each layer is investigated in this paper. In this case, there are two methods for analyzing the data. To begin, graphically, by depicting the density in each layer. Second, by comparing the magnitudes of each layer density and sea level of first layer using the fast Fourier transform (FFT) process. To date, to the authors' knowledge, there is no research that has examined this issue in detail using the FFT. This study also contributes to the Pacific's subsurface investigation. This study has been carried out in the Ocean Modelling Laboratory, Department of Marine Sciences, Universitas Syiah Kuala, Indonesia in 2020.

MATERIALS AND METHODS

Winds from the west are generally symmetrical during El Niño years as was the case in 2015/2016.

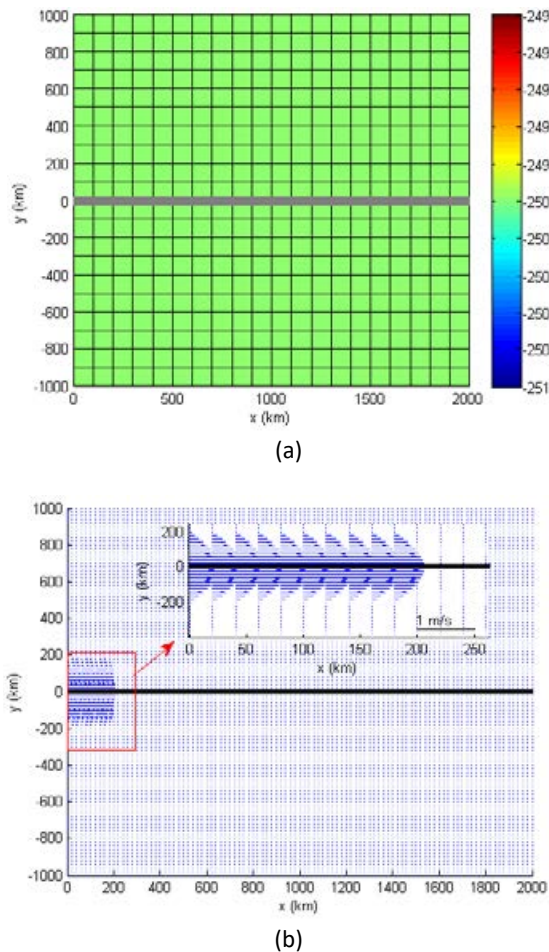


Fig. 1: Model setup (a) domain model, (b) wind stress force

A sequence of westerly wind events in boreal spring triggered the most recent El Niño outbreak in 2015/2016. Downwelling oceanic Kelvin waves were caused by the associated wind forcing, minimizing the upwelling of cold subsurface waters in the eastern Pacific cold tongue and resulting in surface warming in the central and eastern Pacific (Timmermann *et al.*, 2018). The emergence of Downwelling is a product of the effect of winds on the Pacific's surface. As a result, it is important that we obtain a model of the Pacific's surface layer. It's also crucial to understand how much wind is always blowing. Meanwhile, the most recent El Niño outbreak, which occurred in 2015/2016, was triggered by a series of westerly wind events in boreal spring. In the Pacific model, this condition is applied as a force. The three-

dimensional numerical model is efficient enough to represent low-resolution circulating currents. The current model is based on hydrostatic primitive equations, such as those proposed by Cha *et al.* (2018) and Peng *et al.* (2020). Cha *et al.* (2018) used a three-dimensional model to observe a distinct low-frequency mode of the tropical Pacific sea level and display its connection to global ocean warming. Meanwhile, this research employs a non-hydrostatic technique that has previously been used to model tides in the Malacca Strait (Haditir *et al.*, 2020), internal wave (Rizal *et al.*, 2019), and Lee waves (Rizal *et al.*, 2020). A non-hydrostatic method may be used to explain the mechanism of the Lee wave, as well as the internal wave. Numerical experiments were carried out based on Kämpf (2010). This study has a domain model with a horizontal boundary of $2,000 \times 2,000$ km and a uniform depth of 250 m (see Fig. 1(a)).

The lateral grid distance is $\Delta x = \Delta y = 20$ km, while the vertical grid distance is $\Delta z = 50$ m so that it forms five layers with time step $\Delta t = 120$ seconds. In the west and east, closed boundaries are defined, likened to a dry area (coast). Furthermore, the north and south sides of the model domain are given open boundaries. The sea's initial condition is at rest, and this model uses a free-surface so that the numerical time steps will last a long time. The seawater density at 100 m and above from the bottom is 1027 kg/m^3 , while below it is 1028 kg/m^3 . The internal gravity wave phase velocity associated with this density configuration is $c = 0.75 \text{ m/s}$. This value is obtained using the equatorial beta-plane approximation using Eq. 1.

$$f = \beta y \quad (1)$$

where, y is the meridional coordinate, and β is the set of $\beta = 2.5 \times 10^{-11} \text{ 1/ms}$. This model is given a wind force from the Westside, which consists of wind stress moving eastward in the form of cosine in the y -direction (Fig. 1(b)). The maximum magnitude is 0.1 Pa at the equator, and the wind stress force is applied within 200 km from the west side. Wind stress magnitude will be stopped after the first five days of simulation.

Cyclic boundary conditions are used in open boundaries on the North (ny and $ny+1$) and South sides (0 and 1). This cyclic boundary condition uses

Table 1: The procedure for achieving outcomes

Process	In-process information
1	To get the sea level and density performance, run the Kämpf model simulation in Fortran.
2	Display and retrieve/sample the desired performance data for sea level and density.
3	The output is displayed after searching for the FFT value.

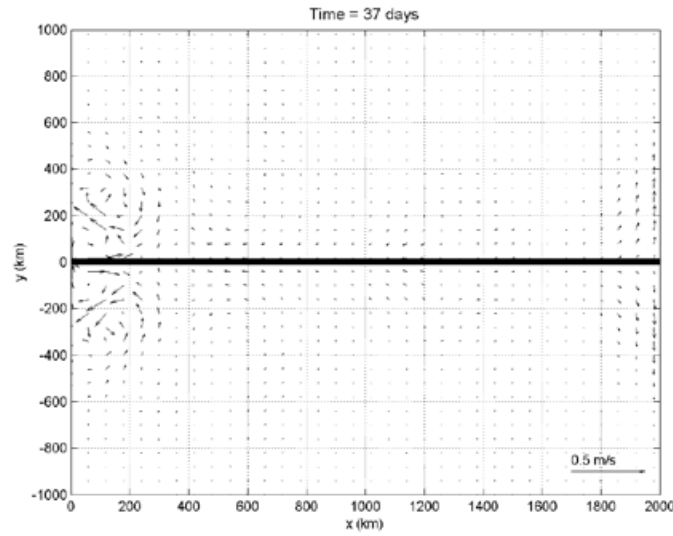


Fig. 2: Currents plot on the 37th day of simulation

the “mirror” condition for the meridional flow component (i -index) in Eq. 2 and 3 (Haditir *et al.*, 2019).

$$v(i, 0, k) = -v(i, ny, k) \quad (2)$$

$$v(i, ny+1, k) = -v(i, 1, k) \quad (3)$$

The advanced turbulence closure scheme introduced by Smagorinsky (1963) was applied to parameterize lateral eddy viscosity and diffusivity (A_h). Here is the equation using Eq. 4.

$$A_h = c_1 \Delta x \Delta y \sqrt{\left(\frac{\partial u}{\partial x}\right)^2 + \left(\frac{\partial v}{\partial y}\right)^2 + 0.5 \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x}\right)^2} \quad (4)$$

Where, c_1 is a parameter with a value between 0.1 and 0.2. The value of c_1 chosen in this study was 0.1. The simulation was run several times with the procedure listed in Table 1.

RESULTS AND DISCUSSION

This El Niño model simulation is run for 60 days with time step $\Delta t = 120$ seconds. This simulation generates current, sea level, and density data, which will be analyzed to determine sea level and density. Furthermore, the data is stored every 12 hours of simulation. In this article, the simulation output data for day 37 is analyzed because the coastal Kelvin waves have formed correctly on the East side (Fig. 2). The formation of coastal Kelvin waves in the eastern region follows a similar pattern as the OSCAR Sea Surface Velocity plot data obtained from ERDDAP in October 2015 in the Pacific Ocean (Fig. 3). Fig. 2 simulation and Fig. 3 observation are not identical. Since, Fig. 2 is based on ideal bathymetry, wind, and shoreline, whereas Fig. 3 is based on real bathymetry, wind, and shoreline.

The plot of sea level simulation results on the 37th day can be seen in Fig. 4. From the sea-level value in Fig. 4(a), the sea level value is taken at $x = 400$ km, called the western part, $x = 1200$ km is called

the middle part, and $x = 1980$ km is called the eastern part. In the three sections, the results are obtained, as shown in Fig. 4(b). Sea level in the western part is negative around the equator and is positive in areas far from the equator. Furthermore, in the middle part, the sea level value shows the same properties as the sea level value in the western part. However, in the western part, sea level has a steeper graph at the equator than in the middle. In contrast to the two sea-level values, the data in the eastern part can be observed that all values are positive, but at the

equator, it forms a peak with a value of 1.58 cm while on the North and South sides, which are far from the equator a value of 0.24 cm. The variation in sea level away from the equator is influenced by local and remote responses (Chang *et al.*, 2013).

Fig. 5 shows the density value of seawater at the equator. It can be observed that the densities at 0-50 m and 50-100 m depths show a similar pattern of change on the 37th day. The density at these two depths shows the same properties after being given a wind force until the end of the

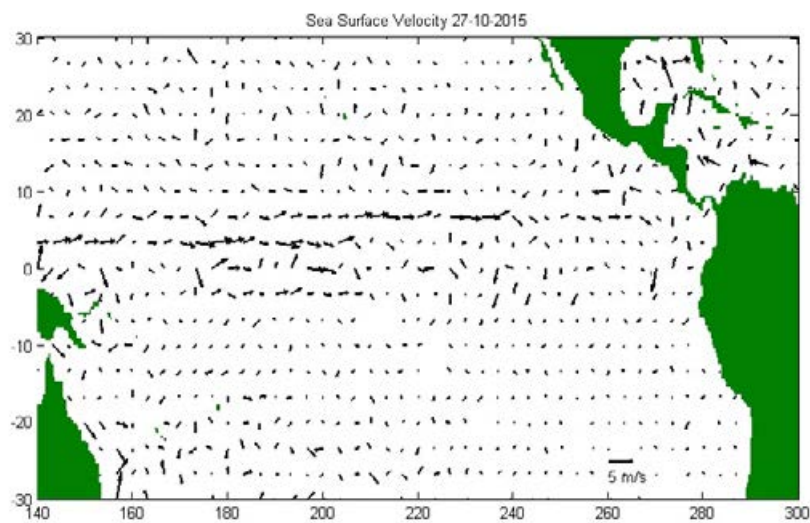


Fig. 3: Sea surface velocity on October 2015

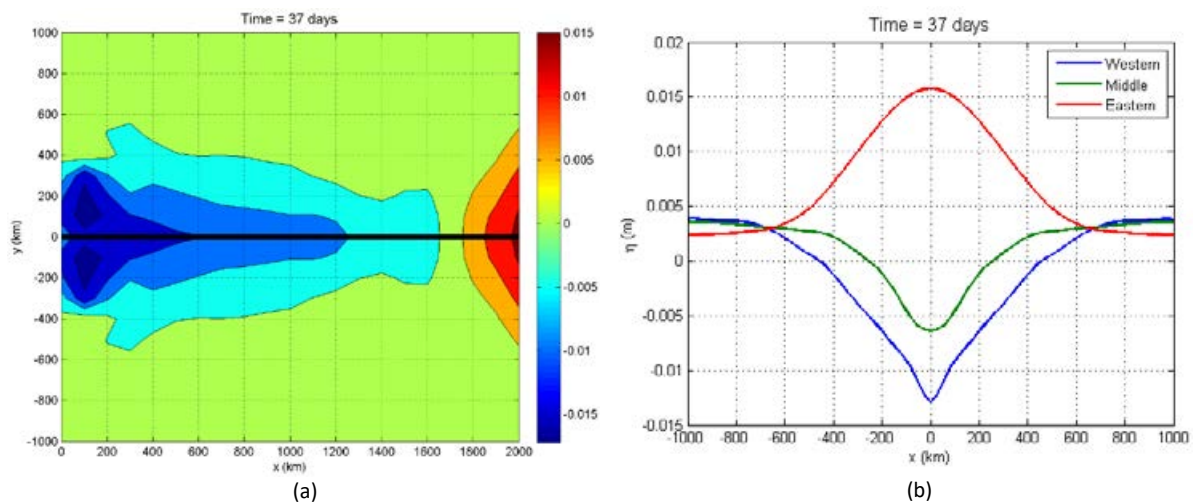


Fig. 4: Sea level (a) plot on the 37th day of simulation, (b) value at the equator on the 37th day

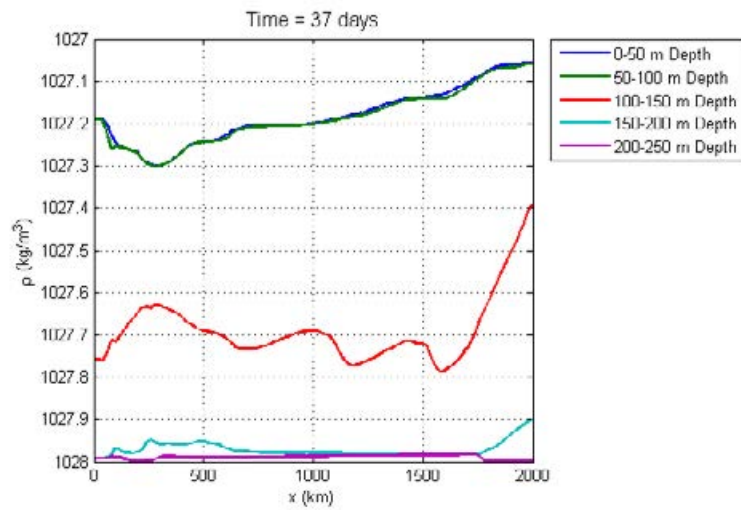


Fig. 5: The value of density at the equator at 37 days of simulation

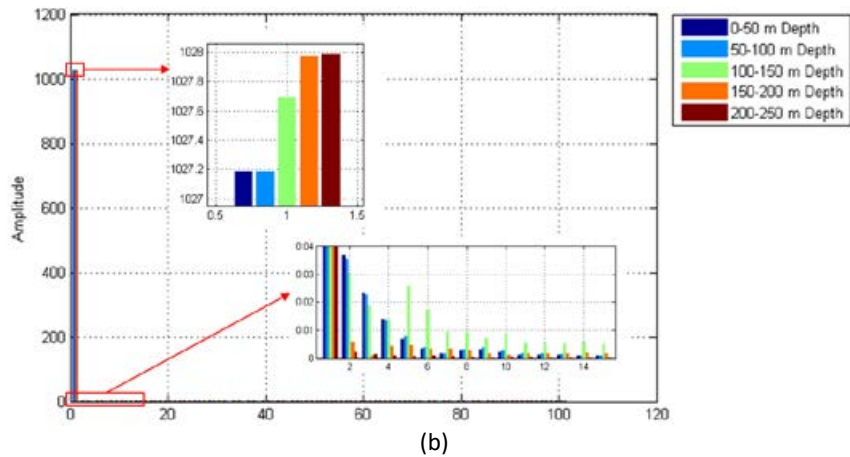
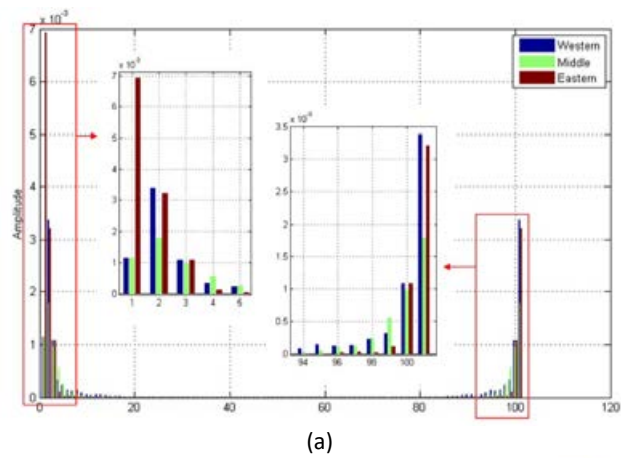


Fig. 6: The FFT value at the equator on the 37th day of (a) the sea level, (b) the density

Table 2: Amplitude, frequency, and phase of sea level values

<i>i</i>	Western		
	A_s	F	θ
1	0.00338	0.10000	0.03100
2	0.00338	10.00000	-0.03100
3	0.00115	0.00000	3.14159
4	0.00108	0.20000	-3.07910
5	0.00108	9.90000	3.07910
6	0.00032	0.30000	0.09086
7	0.00032	9.80000	-0.09086

<i>i</i>	Middle		
	A_s	F	θ
1	0.00179	0.10000	0.03096
2	0.00179	10.00000	-0.03096
3	0.00115	0.00000	0.00000
4	0.00096	0.20000	-3.07977
5	0.00096	9.90000	3.07977
6	4.00000	0.00055	0.30000
7	99.00000	0.00055	9.80000

<i>i</i>	Eastern		
	A_s	F	θ
1	0.00693	0.00000	0.00000
2	0.00321	0.10000	-3.11051
3	0.00321	10.00000	3.11051
4	0.00108	0.20000	0.06226
5	0.00108	9.90000	-0.06226
6	0.09305	0.00011	0.30000
7	-0.09305	0.00011	9.80000

simulation, in contrast to the density at a depth of 100-250 m. Densities at a depth of 100-150 m have begun to show changes in density values ranging between 1027.4 kg/m³ and 1027.8 kg/m³. At a depth of 150-200 m and 200-250 m, the density, in general, does not change significantly but shows a different pattern. This simulation generates current, sea level, and density data, which will be analyzed to determine sea level and density. The formation of Ekman and Kelvin waves is used to compare experiments theoretically. During the El Nino phase, Kelvin waves are thought to migrate into the eastern Pacific Ocean (Kämpf, 2010; Capotondi *et al.*, 2019). Meanwhile, the Ekman layer (D_E) can be approximated using Eq. 5 (Kämpf, 2010).

$$D_E = \sqrt{\frac{2\pi^2 A_z}{f}} = \frac{7.6}{\sqrt{\sin|\phi|}} U_{10} \quad (5)$$

The Ekman layer is related to a three-dimensional wind-driven circulation model, such as the one used in experiment. The Ekman layer is a thin layer that allows the wind's effects to continue to function. If the highest wind stress is wind = 0.1 pa and the wind speed is $U_{10} = 5.5$ m/s, the Ekman layer (D_E) at the equator is the deepest, while it can reach 220 meters at 200 km from the equator using Eq. 6.

$$(U_{10} = \sqrt{\frac{\tau^{wind}}{\rho_{air} C_d}}, \text{ where } \rho_{air} = 1.25 \text{ dan } C_d = 2.6 \times 10^{-3}) \quad (6)$$

Meanwhile, the most recent El Niño outbreak,

Table 3: Amplitude, frequency, and phase of the density values

i	0-50 m Depth		
	A_d	F	θ
1	1027.18267	0.00000	0.00000
2	0.03675	0.10000	-1.78340
3	0.03675	10.00000	1.78340
4	0.02330	0.20000	-1.82001
5	0.02330	9.90000	1.82001
6	0.01407	0.30000	-2.18821
7	0.01407	9.80000	2.18821
i	50-100 m Depth		
	A_d	F	θ
1	1027.18668	0.00000	0.00000
2	0.03566	0.10000	-1.76912
3	0.03566	10.00000	1.76912
4	0.02291	0.20000	-1.82199
5	0.02291	9.90000	1.82199
6	0.01371	0.30000	-2.14562
7	0.01371	9.80000	2.14562
i	100-150 m Depth		
	A_d	F	θ
1	1027.68937	0.00000	0.00000
2	0.03006	0.10000	2.86144
3	0.03006	10.00000	-2.86144
4	0.02565	0.40000	-1.70419
5	0.02565	9.70000	1.70419
6	0.01878	0.20000	-2.77093
7	0.01878	9.90000	2.77093
i	150-200 m Depth		
	A_d	F	θ
1	1027.97072	0.00000	0.00000
2	0.00600	0.10000	2.55146
3	0.00600	10.00000	-2.55146
4	0.00473	0.40000	-1.98551
5	0.00473	9.70000	1.98551
6	0.00452	0.30000	-1.79668
7	0.00452	9.80000	1.79668
i	200-250 m Depth		
	A_d	F	θ
1	1027.98959	0.00000	0.00000
2	0.00216	0.10000	-0.52145
3	0.00216	10.00000	0.52145
4	0.00136	0.20000	0.34275
5	0.00136	9.90000	-0.34275
6	0.00103	0.50000	2.48266
7	0.00103	9.60000	-2.48266

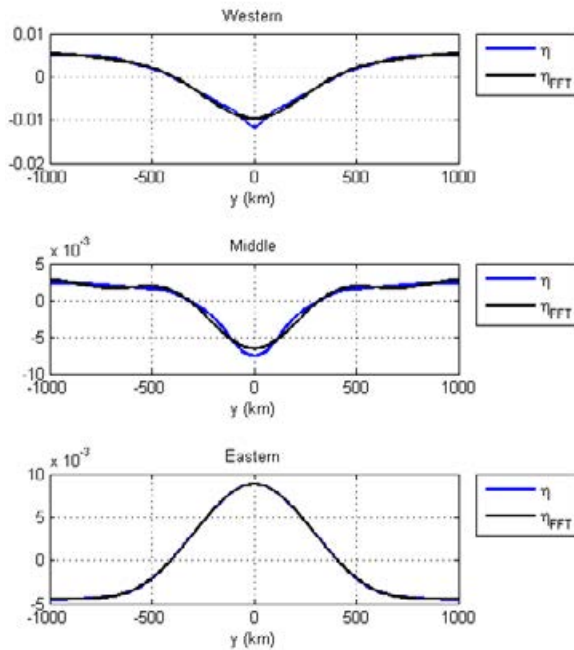


Fig. 7: Sea level plot from the FFT results

which occurred in 2015/2016, was triggered by a series of westerly wind events in boreal spring. In the pacific model, this condition is applied as a force. In this experiment, we attempted to evaluate the El Niño phenomenon described by ideal winds by examining the numerical effects of the pacific model (which included tides, sea level, and density changes). An analytical approach, namely FFT, was used to carry out further research. The variations and amplitudes of each fluid layer can be calculated analytically using FFT.

The next step is to find FFT value from the sea-level value. Fig. 6(a) shows the magnitude of sea-level plots in the western, central, and eastern parts on the 37th day. From this graph, seven data were selected with the enormous output magnitude from FFT for sea-level values consisting of amplitude, frequency, and phase values (see Table 2). The enormous amplitude value of sea level in the western part is 0.00338 m. Furthermore, the enormous amplitude value from sea level in the middle is 0.00179 m and in the eastern part is 0.00693 m. Of the three regions, the eastern part of the domain has the largest amplitude, namely 0.00693 m. Table 2 shows two amplitude and frequency values of sea level values in the West and the center that have the same value,

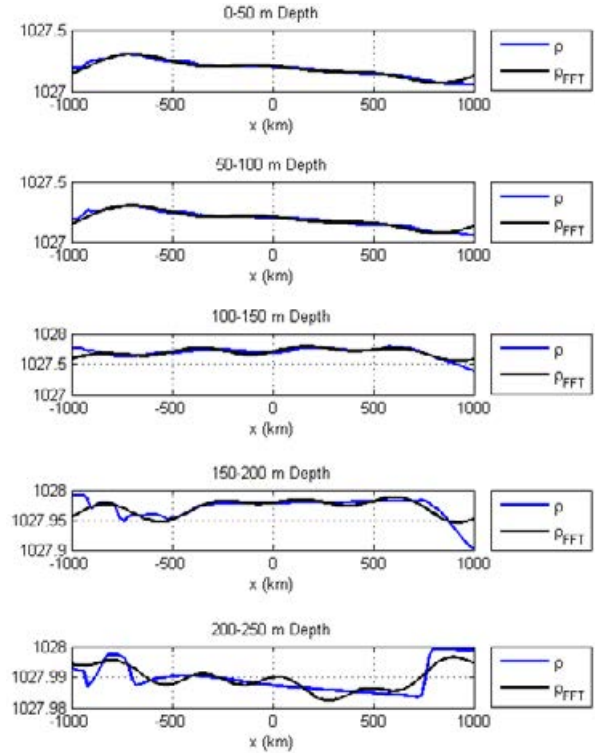


Fig. 8: The density plot of each layer depth from the FFT results

but the phase values are different. Furthermore, using Eq. 7 with $n = 7$ where i is the data index and the amplitude, frequency, and phase values in Table 2 and Fig. 7 can be obtained. This figure shows that with the seven data available in Table 2, most of them can represent the sea level data generated from the simulation using Eq. 7.

$$f(t) = \sum_{i=1}^n A_i \cos(2\pi F_i t + \theta_i) \quad (7)$$

Next, for density data, the amplitude, frequency, and phase values are also searched using the FFT. Fig. 6(b) shows a magnitude plot of the densities at depths of 0-50 m, 50-100 m, 100-150 m, 150-200 m, and 200-250 m at the equator on the 37th day. Seven data sets with an enormous magnitude are selected from the FFT output for density values (Table 3). An enormous amplitude value of the density at a depth of 0-50 m is 1027.18267 kg/m³. An enormous amplitude value of the density at a depth of 50-100 m is 1027.18668 kg/m³. An enormous amplitude value of the density at a depth of 100-150 m is 1027.68937

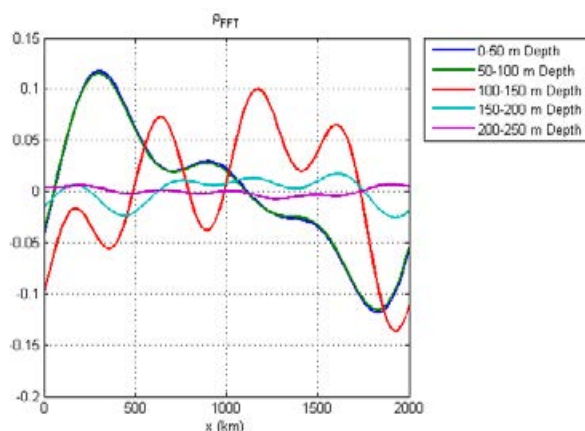


Fig. 9: The density value of the FFT results at $y = 0$

kg/m^3 . An enormous amplitude value of the density at a depth of 150-200 m is $1027.97072 \text{ kg/m}^3$. An enormous amplitude value of the density at a depth of 200-250 m is $1027.98959 \text{ kg/m}^3$. Of the five layers of depth, the depth of 150-200 m has the largest amplitude with a value of $1027.98959 \text{ kg/m}^3$.

Table 3 can also be observed that the amplitude, frequency, and phase at a depth of 0-50 m and 50-100 m have almost the same values. Then, using Eq. 7 with $n = 7$ and the amplitude, frequency, and phase values in Table 3, the graph is obtained in Fig. 8. This figure shows that with the seven data available in Table 3, most of them can already represent the density data generated from the simulation.

If the graph of each layer is shifted to $y = 0$, the result is shown in Fig. 9. From the figure, it can be seen that the density values of the FFT results for a depth of 0-50 m and 50-100 m are almost coincided, which means that the density at a depth of 0-50 m and 50-100 m has the same change pattern after being given a wind force on the sea surface.

CONCLUSION

In the 37 days of simulations of an ideal El Niño event in the Pacific Ocean, the current has entered the East and created a complete coastal Kelvin wave. In the eastern region, the shape of coastal Kelvin waves follows a pattern similar to the OSCAR Sea Surface Velocity plot data obtained from ERDDAP in the Pacific Ocean in October 2015. Around the equator, the sea-level value is negative in the western and middle regions, but positive in areas far from the equator. The western part's sea-level value, on the other hand, has

a steeper graph at the equator than the middle. The FFT findings also reveal that there are two amplitude and frequency values for the sea level values in the West and East that are the same, but the phase values are different. The eastern part of the domain has the highest amplitude of the three parts, according to FFT data. In addition, there are two amplitude and frequency values from sea level values in the West and East that are the same, but the phase values are different. Both graphically and from the FFT, densities at depths of 0-50 m and 50-100 m display a similar pattern of changes on the 37th day. Despite having given the wind force until the end of the simulation, the density at these two depth layers has the same properties as the density at the depth below, which is different from the density at the depth below. It's caused by wind forces operating at a depth of 0-100 meters. Meanwhile, wind impact is very weak or non-existent at depths of 100-150 m, 150-200 m, and 200-250 m. The density does not change significantly between 150-200 m and 200-250 m, but it does follow a different pattern. The vertical density stratification decreases from five to four layers in general. At depths of 0-50 m and 50-100 m, the amplitude, frequency, and phase are nearly identical.

AUTHOR CONTRIBUTIONS

M. Ikhwan performed the literature review, running the model, analyzed and interpreted the data, prepared the manuscript text, and manuscript edition. R. Wafdan performed the literature review, running the model, analyzed and interpreted the data, prepared the manuscript text, and manuscript edition. Y. Haditjar performed the literature review, prepared numerical code, prepared the manuscript text, and manuscript edition. M. Ramli performed the literature review, analyzed and interpreted the data, prepared the manuscript text, and manuscript edition. Z.A. Muchlisin performed the literature review, prepared the manuscript text, and manuscript edition. S. Rizal performed the literature review, experimental design, analyzed and interpreted the data, prepared the manuscript text, and manuscript edition.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

Δt	Time-step, s
$\Delta x, \Delta y, \Delta z$	Distance spacing, m
β	Meridional variation of Coriolis parameter, $\beta = 2.5 \times 10^{-11} \text{ 1/ms}$
ρ	Seawater density, kg/m^3
θ	Phase of density, degree
τ	Wind stress, pa
A_d	Amplitude of density, kg/m^3
A_h	Horizontal eddy viscosity, m^2/s
A_s	Amplitude of sea level, m
c	Phase speed of internal gravity waves, m/s
c_1	Free parameter for turbulence closure scheme, non-dimensional
D_E	The Ekman layer
ENSO	El Niño – Southern Oscillation
Eq.	Equation
f	Coriolis parameter, $1/\text{s}$
F	Frequency of density, Hz
Fig.	Figure
FFT	Fast Fourier Transform
Hz	Hertz
kg/m^3	kilogram per meter cubic
km	kilometers
m	meters
$1/\text{ms}$	one per meter second
m/s	meter per second
Pa	Pascal
U_{10}	Wind speed at 10 m above sea level, m/s

u, v	Currents, m/s
y	Meridional coordinate, m

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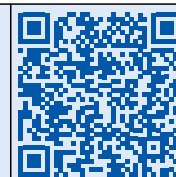


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ORIGINAL RESEARCH PAPER

Pre-sowing treatment of vetch hairy seeds, *vicia villosa* using ultraviolet irradiation

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ABSTRACT

BACKGROUND AND OBJECTIVES: Aiming to increase crop yield the antimicrobial/bacterial or fungicidal pre-sowing seed treatment received more attention in modern agronomy. Ultraviolet-C irradiation of pre-sowing seeds is an environmentally friendly method that became of great importance in recent years. It is, hereafter, being shown that, along with known antimicrobial use, there is additional important advantage of Ultraviolet-C irradiation of pre-sowing seeds. It was revealed that Ultraviolet-C radiation on Vetch Hairy seeds stimulates seeds germination and vigour.

METHODS: Various doses of Ultraviolet-C irradiation of seeds were used. The main sowing qualities of seeds were determined: seed vigour and germination, as well as the content of photosynthetic pigments in plant leaves and the main parameters of the kinetic values of hydration—moisture and hydration rate.

FINDINGS: It was found that ultraviolet-C radiation has a positive effect on sowing qualities and content of photosynthetic pigments in plant leaves of Vetch vary. The most effective dose of ultraviolet irradiation applied to vetch hairy seeds; *vicia villosa* was 1000 J/m². At this dose the seed vigour increases by 23.6%, germination by 15.1%, the mass of germinated seeds by 17.3%, the content of a- and b-chlorophyll by 12.4%, and 17.5%, respectively, the carotenoid content increased by 13.9%. The parameters of seeds hydration kinetics such as moisture content and hydration rate were determined. It was revealed that the hydration rate of seeds increased significantly in the first 100-minute time range. Later in time the hydration rate progressively decreased, achieving a saturated moisture content after 700 minutes. Additionally, it was found that Ultraviolet-C irradiation decreases the imbibition damage.

CONCLUSION: The results indicated that ultraviolet-C irradiation has a positive effect on sowing qualities of Vetch Hairy seeds, thus, could be proposed as a promising candidate for application in treatment pre-sowing agriculture seeds.

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INTRODUCTION

The main centre of origin of the hairy vetch (*Vicia villosa*) is Southwest Asia. However, this cultivar was found to be spread to the Mediterranean Sea and then to European countries, including England, Scandinavia and the Baltics. The widespread cultivation of Hairy Vetch in Ukraine makes it possible to obtain early spring forage during winter sowing and late summer forage during spring sowing. Hairy Vetch – a valuable legume, which on a number of biological and economic characteristics stands out among annual forage crops. There are twenty-two species of vetch in Ukraine, the most commonly used for the production of *Vicia sativa* L., *Vicia villosa* Roth and the Pannonian vulture – *Vicia pannonica* Grantz. Hairy vetch is a source of vegetable protein, one of the best precursors for winter and spring cereals. It is part of most annual legume-cereal mixtures grown for green fodder, hay, silage, haylage. Biomass with a high protein content is well digestible and is a valuable feed for all farm animals. From the point of view of accelerating productivity in colder climates and obtaining the needed biomass yield (Wilke and Snapp, 2008; Campiglia et al., 2012), Vetch seeds are unreliable and therefore there are not many studies regarding the requirements of this species (Aarssen et al., 1986). At first stage of germination for seed multiplication Vetch species faces problems associated with low sowing qualities of seeds (Jacobsen et al., 2010). The change of various parameters such as dormancy, temperature, humidity, as well as methods of pre-sowing stimulation of seeds may improve the sowing quality of seeds in various environmental conditions. Commercially available widespread chemical methods of pre-sowing seed treatment have environmental impact and only physical methods (i.e. use of electromagnetic radiation) are proved to be eco-friendly. In some studies, a limited number of chemicals were used, but majority of them were replaced with appropriate physical methods of processing, such as magnetic field, gamma radiation, electric field, laser radiation, ultrasound and optical irradiation (Govindaraj et al., 2017; Thakur et al., 2019; Ri et al., 2019; Lazim and Ramadhan, 2020). The optical irradiation became of great interest in recent years that stimulate the growth and increase the tolerance of plants towards negative external factors therefore increasing the crops yield. Particularly, the pre-sowing treatment of agricultural

seeds with ultraviolet radiation attracted significant attention (Neelamegam and Sutha 2015; Mariz-Ponte et al., 2018). Thus, the study (Thomas and Puthur, 2017) described the effect of ultraviolet radiation on seeds during pre-sowing treatment to increase crop productivity, yield quality, as well as plant resistance to various biotic and abiotic stresses. Backer et al., 2018 discussed the mechanisms of plant growth stimulation by inoculating plants with rhizobacteria that stimulate plant growth, or treating plants with microbial-to-plant signalling compounds. Other authors of publication (Brown et al., 2001) described the treatment of cabbage seeds with Ultraviolet-C (UV-C) radiation at doses of 3.6 kJ/m², which ensured resistance to bacteriosis (black rot) and improved quality and growth of cabbage. In study (Kacharava et al., 2009), an increase in the germination of seeds of black beans (*Phaseolus vulgaris*) under the influence of UV-C radiation was investigated. UV radiation at a dose of 460 mW/cm² for 60 minutes enhanced the growth and accumulation of biomass in beans. The application of UV-C radiation led to an increase in plant height by 11–39%. All these effects of pre-treatment of seeds with optical radiation in the UV-C range are associated with the changes in biological properties (Semenov et al., 2017). The mechanisms of such effects themselves were investigated using various methods and models. However, no unambiguous explanation of these actions has been found until today. Temperature and moisture content (hydration) are the most important factors influencing seed germination. The water absorption kinetics during seeds hydration affects the biological properties during growth and the quality of the final product (Turhan et al., 2002). The positive effect of various doses of pre-sowing UV-C irradiation on the sowing quality of seeds has been considered in many studies (Turhan et al., 2002; Neelamegam and Sutha, 2015; Rocha et al., 2015; Thomas and Puthur, 2017; Mariz-Ponte et al., 2018; Sadeghianfar et al., 2019; Korotkova et al., 2020; Hernandez-Aguilar et al., 2021). It was found that the optimal doses of irradiation of winter wheat (Semenov et al., 2020), carrots (Semenov et al., 2019) and rapeseed (Semenov et al., 2018) are 400–600 J/m², 120–150 J/m² and 120 J/m², respectively. Reviewing literature in this area it is possible to note that despite an extensive study (Neelamegam and Sutha, 2015; Thomas and Puthur, 2017; Korotkova et al., 2020),

as it was understood, this study is dealing with pre-sowing treatment of vetch hairy seeds (*Vicia villosa*) by use of Ultraviolet-C radiation has not been reported yet. Aiming to fill this research gap the objective of this study was focused on determination of hairy seeds (*Vicia villosa*) sowing quality (i.e. vigour and germination) by pre-sowing treatment of seeds using different doses of ultraviolet radiation. In addition, the hydration rate was evaluated, for treated and not treated pre-sowing seeds by ultraviolet radiation. The study was performed in the scientific and technical centre of Poltava University of Economics and Trade, Ukraine in 2019–2020.

MATERIALS AND METHODS

For conducting research, Hairy Vetch (*Vicia villosa*) of Poltava-77 variety was used, which is determined by the national standard for the conditions of the Forest-Steppe and Steppe of Ukraine. During pre-sowing treatment, Hairy Vetch seeds were irradiated using low-pressure ZW20D15W type ultraviolet lamps with a power of 20 W at an emission spectrum the C region: 200–280 nm. Irradiation was carried out with the following doses of 50, 120, 500, 1000 and 3000 J/m² and controlled using a Tensor-31 radiometer. Seed vigour and germination of seeds of control (without irradiation) and irradiated samples were determined by germinating 4 samples of 50 seeds each in Petri dishes on moistened filter paper in accordance with the [International Safe Transit Association \(ISTA\)](#) standard. The content of photosynthetic pigments in vetch leaves in the phase of three true leaves was established using a spectrophotometer, determining the optical density of the chlorophyll and carotene extract at 665, 649, and 440 nm (absorption maxima of chlorophyll a (Ca), chlorophyll b (Cb), and carotenoids (Ck), respectively). The concentration of pigments was calculated using Eqs. 1 to 3 ([Porra, 2002](#)).

$$C_a = 11,63D_{665} - 2,39 D_{649}, \text{ mg/L} \quad (1)$$

$$C_b = 20,11 D_{649} - 5,18 D_{665}, \text{ mg/L} \quad (2)$$

$$C_k = 4,695 D_{440} - 0,268 C_{a+b}, \text{ mg/L} \quad (3)$$

To determine the moisture content (hydration), samples of Hairy Vetch seeds in an amount of 10 ± 0.5 g were soaked in 200 ml of distilled water at each

dose of UV-C radiation. The soaked samples were removed from the water at intervals of half an hour and an hour. The samples were placed on absorbent paper to remove excess water, and then weighed using a precision analytical balance, model WLC 0.2, TM Radwag, Poland with an accuracy of 0.001 g. Eqs. 4 to 5 were used for determination of such parameters as moisture content (MC) and hydration rate (HR) ([Shafaei et al., 2016](#)):

$$MC = \left(\frac{W_f - W_i}{W_i} \right) \times 100\% \text{ (d. b. \%)} \quad (4)$$

$$HR = \frac{MC_{(t+\Delta t)} - MC_{(t)}}{\Delta t} \text{ (d. b. \% / min)} \quad (5)$$

At each value of the UV-C radiation dose, a hydration procedure was carried out, which lasted 12 hours. At this stage, the change in weight and, accordingly, the content of saturated moisture in the sample were determined. In order to reduce the measurement error of hydration indicators, the studies were carried out in triplicates. The appropriate statistical processing was used for interpretation of obtained experimental data.

RESULTS AND DISCUSSION

Determination of seed vigour and germination

Analysis of the seed vigour and germination of Hairy Vetch indicates the dependence of its sowing qualities on the energy dose of ultraviolet radiation ([Table 1](#)). At an irradiation dose of 500 J/m², the following results were obtained: in comparison with the control samples, the seed vigour was increased by 10.8% and the germination was increased by 6.6%. The maximal changes were observed at irradiation doses of 1000 J/m²: the seed vigour was increased by 23.6% and the germination was increased by 15.1%.

Less significant effect was observed at an irradiation dose of 3000 J/m²: in comparison with the control samples, the seed vigour was increased by 18.2% and the germination was increased by 9.6%. The obtained regularities of the pre-sowing effect on biological indicators (seed vigour and germination) are in good agreement consistent with the results of study ([Kalsa and Abebie, 2012](#)), where the effect of hydro- and osmotic priming in pre-sowing treatment on Hairy Vetch seeds was investigated.

Determination of biometric indicators (mass of germinated seeds)

The positive effect of UV-C irradiation on Hairy Vetch's growth processes was also noted. An effective change in their biometric parameters was observed at high doses of UV-C irradiation of seeds - 1000 J/m² and 3000 J/m² (Table 2).

The maximum stimulating effect of UV-C irradiation on growth processes was obtained with a dose of 1000 J/m²: in comparison with the control samples the mass of germinated seeds increased by 17.3% (Table 2). At the same time, the root length and seedling length were increased by 18.1%, and 6.1%, respectively. At an irradiation dose of 3000 J/m²: the mass of germinated seeds was 5% less than the maximum value obtained, and the root length and seedling length were decreased by 5.1% and 1.3%, respectively. At doses up to 500 J/m², changes in the biometric parameters of Hairy Vetch seedlings were insignificant and varied within the experimental error.

Determination of Chlorophyll

The intensity of photosynthesis is determined by the amount of plant pigments – chlorophylls and carotenoids, and an increase in these parameters

lead to an increase in the vegetative mass of plants. Pre-sowing UV-C irradiation of seeds affects the content of photosynthetic pigments in Hairy Vetch leaves (phase of three true leaves) (Fig. 1).

The concentrations of chlorophyll a and chlorophyll b in the leaves of the shaggy branch maximally increase (by 12.4% and 17.5%, respectively, compared with the control samples) under UV-C irradiation of seeds with doses of 1000 J/m². The content of carotenoids also increased by 13.9%. The obtained results agree with a number of works (Salama *et al.*, 2011; Geeta *et al.*, 2014; Li, *et al.* 2020). In (Li *et al.*, 2020), the amount of chlorophyll in the leaves of Hairy Vetch was determined. It was found that, depending on the year of sowing, fluctuations in the amount of chlorophyll can reach 10% or more. It follows from (Geeta *et al.*, 2014) that the content of total chlorophyll and chlorophyll a and b increased upon UV-B irradiation of germinated seeds for 1 h by 7.31%, 4.52%, and 14.92%, respectively. At the same time, the chlorophyll content decreased by 5.85%, 3.29%, and 11.19% compared to the control samples after an additional 3 hours of irradiation. A decrease in the content of chlorophylls was observed in a study (Salama *et al.*, 2011), where, after irradiation with 254 nm, the content of chlorophyll a and b

Table 1: Influence of UV-C irradiation on the sowing quality of vetch hairy

Radiation dose (J/m ²)	Seed vigour, (%)	Germination (%)
Control samples (without irradiation)	74, 0	83,0
50	75,0	83,0
120	76,5	85,0
500	82,0	88,5
1000	91,5	95,5
3000	87,5	91,0
LSD ₀₅	3,8	4,1

Table 2: Biometric indicators of seedlings of sowing Hairy Vetch after 14 days

Radiation dose (J/m ²)	Root length (cm)	Shoot length (cm)	Weight 50 pcs Germinated seeds (g)
Control samples (without irradiation)	3,97	6,43	4,90
50	4,00	6,35	4,86
120	4,02	6,50	4,98
500	4,15	6,63	5,16
1000	4,69	6,82	5,75
3000	4,45	6,73	5,46
LSD ₀₅	0,21	0,30	0,28

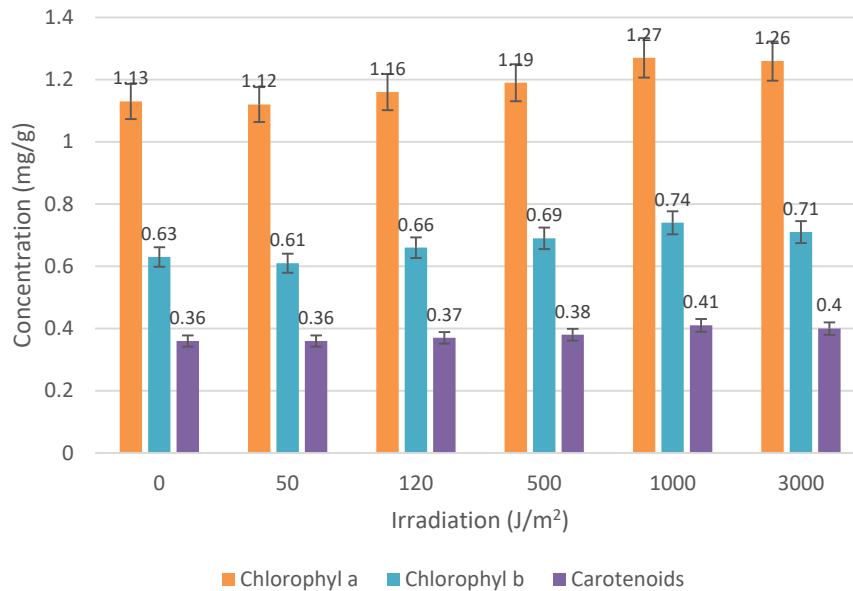


Fig. 1: Content of chlorophylls a, b and carotenoids in hairy vetch leaves (phase of three true leaves)

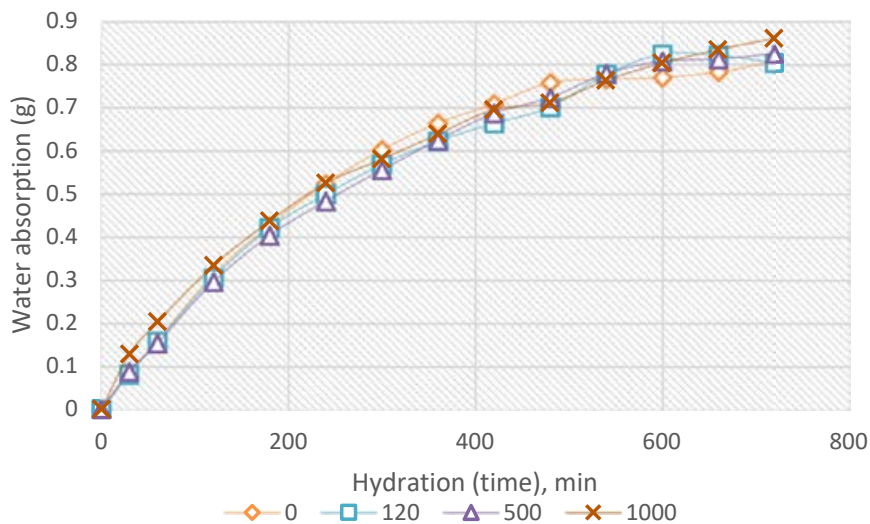


Fig. 2: Dependence of hydration on time at radiation doses: 0; 120; 500 and 1000 J/m²

and total chlorophyll decreased by 79, 43, and 74%, while the content of carotenoids increased by 91%. The results showed that the content of chlorophyll a, chlorophyll b and total chlorophyll decreased with UV-C irradiation.

Determination of hydration kinetics

Besides, the hydration characteristics that affect the quality indicators of production were studied (Turhan *et al.*, 2002). According to the literature (Kumar *et al.*, 2021; Turhan *et al.*, 2002; Shafaei *et*

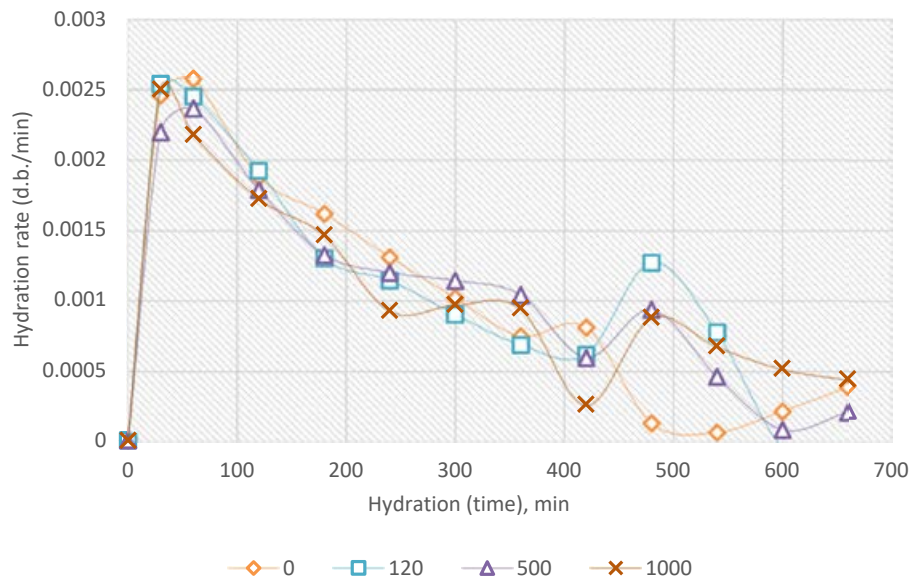


Fig. 3: The dependence of the change in the rate of water absorption on time at doses: 0; 120; 500 and 1000 J/m²

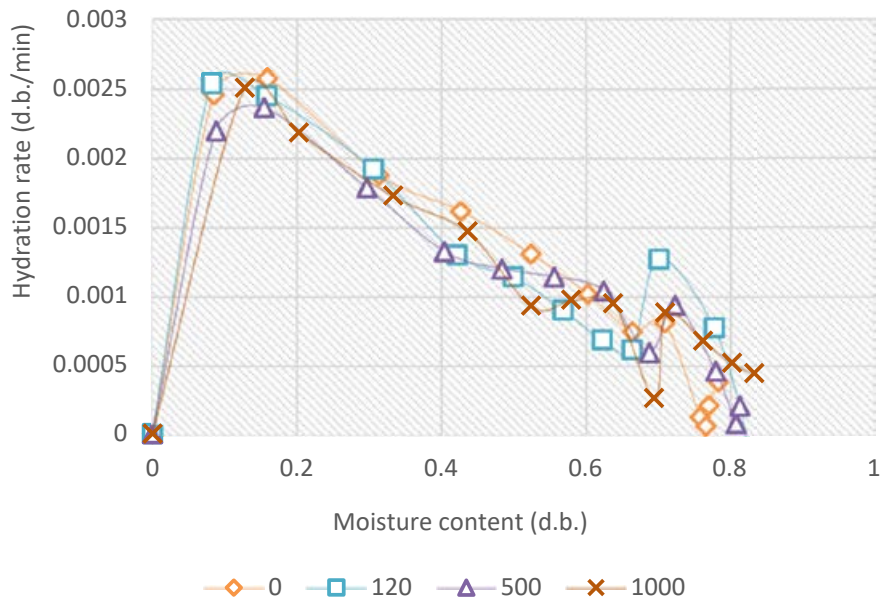


Fig. 4: Dependence of the change in the rate of water absorption on humidity at irradiation doses: 0, 120, 500 and 1000 J/m²

al., 2016, 2021), it can be concluded that a better understanding of hydration kinetics is achieved through accurate modelling. It helped to initiate the development of several mathematical models for describing hydration characteristics (Shafaei *et al.*, 2016). The dependence of the kinetics of

swelling of Hairy Vetch seeds on the dose of UV-C irradiation limits the process of their awakening and germination, and therefore, germination and yield. The time dependence of water absorption at various irradiation doses is shown in Fig. 2.

When hydrated, water slowly diffuses into

the granular structure of the grain and after a certain period of time reaches a constant level. It is understood that as the hydration time increases, so does the moisture content of the Hairy Vetch grain. It was also found that water absorption decreases over time with increasing hydration time and radiation dose. Moreover, this dependence is broken after 9 hours of hydration. The study of the dependence of the rate of hydration is associated with the fact that often too rapid water absorption occurs due to a high gradient of water potential, which can lead to a decrease in seed germination after soaking (Semenov *et al.*, 2021). The results are presented based on the forecast of an artificial neural network (ANN) model, where the input parameters are the radiation dose and the hydration time, and the output parameters are the moisture content and the rate of hydration. Figs. 3 and 4. Irradiation dose and hydration time were used as input parameters, and moisture content and hydration rates were used as output parameters.

The forecast of the ANN model shows (Figs. 3 and 4) that in the initial phase of hydration the rate sharply increased and gradually and slowly decreased in the middle and until the saturated moisture content was reached. As can be seen from Figs. 3 and 4, the rate of hydration decreases with increasing radiation dose under the same experimental conditions.

Pre-sowing irradiation with various doses of UV-C leads to a decrease in water absorption, which reduces damage to the seed coat during rapid water absorption (damage by imbibition) and ultimately leads to an increase in germination energy and seed germination. Thus, UV-C irradiation in most cases reduces the rate of water uptake by the seeds as the dose is increased, which prevents damage to the seed membrane during rapid water uptake and thus allows a higher percentage of Hairy Vetch seeds to germinate.

CONCLUSIONS

Pre-sowing treatment of seeds with UV-C radiation has a positive effect on sowing qualities and biometric indicators. UV-irradiated seeds germinate 1.5-2.0 times faster than non-irradiated seeds. In addition, UV irradiation promotes seed germination both under optimal conditions and under stress. The most effective dose of UV radiation of Vicky seeds is 1000 J/m². At this dose vigour and laboratory germination increased by 23.6%

and 15.1%, respectively. The weight of germinated seeds increased by 17.3% while the content of chlorophyll, chlorophyll b and carotenoids increased by 12.4%, 17.5% and 13.9% respectively. The kinetic parameters of hydration of irradiated seeds were defined by moisture content and hydration rate. This in turn, causing faster water absorption and thus more efficient seed hydration, positively affecting sowing qualities and other agronomic traits. The results of the analysis showed that the rate of seed hydration increased significantly in the initial phase and gradually and slowly decreased in the middle and end of the hydration procedure. Water absorption increased significantly with increasing hydration time from 0 to 700 minutes, following increase of irradiation dose, water absorption decreased, limiting this pattern after 9 hours. Improved water absorption after UV-C irradiation during germination cannot be considered as the only factor responsible for enhanced germination. It is established that one of the reasons for the rapid germination of seeds is the initiation of processes associated with germination during pre-hydration. The increase in germination energy during UV-C irradiation of seeds is the result of many mechanisms caused by irradiation, including efficient water absorption. Rapid absorption of water at an early stage can lead to the so-called seeds imbibition, which is manifested in the disruption of structural integrity of the membranes. In addition, the results of the study showed that pre-sowing treatment of seeds with UV irradiation reduces the rate of hydration thereby suspending seeds imbibition during rapid water absorption. However, at higher doses of UV irradiation the germination potential decreases. These studies demonstrated that UV-C radiation of pre-sowing seeds increases germination not only by UV degradation of pathogens originally present on seeds surface but also explains the nature of seeds hydration rate that changes with time of treatment. Since the report shows clear evidence that UV treated seeds at given dose do not promote seeds biodegradation while contrarily, increase germination, it may potentially lead to expansion of this research to other cultivars.

AUTHOR CONTRIBUTIONS

A. Semenov developed experimental design of equipment of ultraviolet action, conducted experiments, analysed and interpreted the data. T.

Sakhno performed the literature review, participated in experiments and translated. O. Hordieieva performed some of the remained experiments, compiled data. Y. Sakhno prepared the manuscript text, and manuscript edition.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

%	Percent
Δt	Hydration time min
ANN	Artificial neural network
Ca	Chlorophyll a
Cb	Chlorophyll b
Ck	Carotenoids
cm	centimetre
<i>D</i>	Optical density or absorbance at three wavelengths: 440, 649, 665 nm
<i>d.b.</i>	Content dry basis
Eqs.	Equations
<i>et.al</i>	others
<i>Fig.</i>	Figure
<i>Figs.</i>	Figures
<i>g</i>	Gram
<i>h</i>	Hour
<i>HR</i>	Hydration time (d. b./min)
<i>J</i>	Joules
<i>J/cm²</i>	Joules per Square Centimetre
<i>J/m²</i>	Joules per Square meters
<i>kJ/m²</i>	Kilojoule per Square meters
L	litre

<i>LSD₀₅</i>	Least Significant Difference
<i>m²</i>	Square meters
<i>MC</i>	Moisture content
<i>MC(t)</i>	Moisture content at time (t) (d. b. %)
<i>MC(t+Δt)</i>	Moisture content at time (t + Δt) (d. b. %)
mg	Milligram
mg/L	Milligram per litre
<i>min</i>	Minute
<i>MR</i>	Moisture ratio
<i>mW/cm²</i>	Mill watts per centimetre squared
<i>nm</i>	Manometer
<i>pcs</i>	Pieces
<i>UV-C</i>	Ultraviolet C irradiances are defined as the wavelength range of (100 u λ < 280) nm.
<i>W</i>	Watts
<i>W_f</i>	Mass of wet seed of Hairy Vetch (grams)
<i>W_i</i>	Initial mass seed of Hairy Vetch (grams)

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CASE STUDY

Mitigation of environmental impacts in ornamental rock and limestone aggregate quarries in arid and semi-arid areas

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ABSTRACT

BACKGROUND AND OBJECTIVES: Open-pit mining is an important activity to obtain mineral resources that supply society with raw materials to improve people's quality of life. However, this extractive activity causes negative environmental impacts and, it is therefore necessary to identify and evaluate these impacts in order to design preventive and control measures to reduce them and thus safeguard the environment and natural resources. In the Region of Murcia, in Spain, as well as other Mediterranean areas with similar climatic conditions, there is a great deal of mining activity linked to the building sector, in which mainly ornamental rock (marble and marble limestone) and limestone aggregates are used. All of this has given rise to numerous active and abandoned mines, where no restoration process has been carried out, generating strong impacts on the environment.

METHODS: In this study, 8 environmental impact assessments studies of ornamental rock and aggregate quarries in the Region of Murcia were analysed to identify the negative impacts on the abiotic and biotic environment, landscape, socio-economic and socio-cultural environment, and infrastructures and analysing preventive and control measures.

FINDINGS: According to the environmental impact assessment studies analysed, the importance of the most significant environmental impacts has been calculated, indicating whether the impacts are critical, severe, moderate or compatible, and based on it, preventive and corrective measures are proposed together in an impact mitigation management system based in flow charts that will serve to more easily apply and control these measures, in order to prevent them from causing significant or irreversible damage to the environment. Analysing these measures, it has been observed that 90% of the measures applied to control the different negative environmental factors in this type of quarry are the same.

CONCLUSION: Open-pit mining extraction systems have a series of similar characteristics that allow a systematic approach to be established when analysing the impacts. With the use of flowcharts, it becomes easier to apply measures to reduce environmental impacts and in addition, these diagrams, allow at the same time the easy incorporation of updates due to changing regulations.

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INTRODUCTION

Open-pit mining have been considered as an activity that represents a social and economic benefit for the regions and improve the quality of life of the inhabitants, thus constituting an important element for development. However, quarrying, like other mining-related works, causes negative impacts on the environment and strong ecological changes in the affected ecosystems (Sheoran *et al.*, 2010; Sort and Alcañiz, 1996). The level of destruction caused by open pit mining is ten times greater than that caused by other types of mining (Zhou *et al.*, 2018). It is important the identification and assessment these risk in order to design strategies that avoid, mitigate and compensate for these impacts (Luna, 2015). The increase in human capacity to transform the natural environment has led to an imbalance between the damage caused and the capacity of the environment to recover from it. It is clear that this type of exploitation cannot be dispensed with, as it supplies society with the raw materials necessary to improve its quality of life (Parrota and Knowles, 2001). The abandonment of open-pit quarries, without any kind of rehabilitation or recovery of their initial state, is the main source of impacts (Darwish *et al.*, 2010; Khabali and Kamal, 2013). Open-pit mining negatively affects the environment in a variety of ways, from exploration and blasting, transport and disposal of waste rocks (Lad and Samant, 2014). In addition, the exploitation of open-pit quarries causes other environmental impacts: affectation and disappearance of vegetation, fauna, soil and landscape degradation; disruption of animal habitats; changes in air quality, dust pollution; increase of particulate matter in the air; loss of water resources due to degradation of aquifers, diversion of underground streams, changes in the water table and water pollution; contamination of rivers; diversion and blockage of natural drainage systems; production of large volumes of highly polluting waste; noise and vibration; landscape alteration, visual pollution of waste dumps and degradation of large areas that take thousands of years to restore; land use conflict (Maponga and Munyanduri, 1998; Stehouwer *et al.*, 2006; Fierro, 2012; ; Karbassi and Heidari, 2015; Karbassi, and Pazoki, 2015). Moreover, it can also affect the health conditions of the surrounding population. Today, awareness of the limitation of natural resources, as well as that of the various elements which constitute ecosystems, makes it

necessary to solve the problems for the demand of raw materials in accordance with the conservation of nature, in such a way as to safeguard the environment and natural resources in order to be able to pass them on to future generations (Montes de Oca and Ulloa, 2013). To achieve greener mining with fewer impacts, it is necessary to improve the pollution produced by activities in the air and water and those produced by noise, as well as the need to recover the land and its uses after the activities on the exploitations (Zhou *et al.*, 2020). For all these reasons, Spain created the Law 21, (2013), on environmental assessment, amended by Law 9, (2018), which states that an environmental impact assessment is essential for the protection of the environment. This is an adaptation of the national legislation to the existing legislation in the European Union. It facilitates the incorporation of sustainability criteria in strategic decision-making and guarantees adequate prevention of the specific environmental impacts that may be generated, while at the same time establishing effective mechanisms for correction or compensation of the damage. In Spain, the mining activity achieve account 3,280 million euros (M€) in 2017 (MET, 2018). In the Region of Murcia, the exploitation of geological resources has always been linked to the extraction of minerals and rocks. Specifically, it is represented by the extraction of marble, marble limestone and limestone aggregates, which are used by building companies in multiple applications, ranging from the production of concrete, mortar and asphalt agglomerates, to the construction of bases and sub-bases for roads, ballast and sub-ballast for railway tracks, or breakwaters for the defence and construction of ports. The consequence of obtaining them is linked to the damage caused to rural areas and the natural environment during the different phases of exploitation, concerning for the correct development, conservation of the environment and, obligations under legislation have evolved over the years (Casas, 2018). Quarries are found in all the territories of the Region of Murcia, but the Norwest area and the Mula basin stand out due to their great variety of ornamental rock, and the Fortuna-Abanilla area due to its large production of limestone aggregates. The aim of this study is to create an impact mitigation management system based in flow charts with corrective and preventive measures that can be easily applied in ornamental rock and aggregate quarries in arid and semi-arid areas. To this

end, several environmental impact assessment (EIA) studies of quarries in the Region of Murcia have been analysed, the importance of the impacts they produce has been calculated and the measures used to mitigate these impacts have been taken into account. The conventional methods used to mitigate impacts are characterized by being a cumbersome, slow and expensive process. So, in this study, a decision support system is proposed that minimizes these problems. This study has been carried out in the Region of Murcia, Spain during 2020-2021.

European and national legal framework

The evolution of the regulations up to the current situation implies the need to study and define a systematic methodology for impact assessment and mitigation. The environmental assessment is a well-established instrument that accompanies mining development, ensuring that it is sustainable and inclusive. At the international level, environmental assessment regulation is under the [Espoo Convention, \(1991\)](#), and its Protocol on Strategic Environmental Assessment, ([UN, 2003](#)). In the European Union, it is regulated by [Directive 42, \(2001\)](#), by [Directive 92, \(2011\)](#), and by [Directive 52, \(2014\)](#). At the national level, in Spain, it is regulated by [Law 21, \(2013\)](#), on environmental assessment, modified by [Law 9, \(2018\)](#). This regulation includes the obligation to carry out an EIA of the projects. The EIA, aims to incorporate environmental aspects in decision making and seeks sustainable development and environmental protection ([Enriquez-de-Salamanca, 2021](#)). In addition, it is considered as an instrument to support decision making ([Retief et al., 2020](#)). According to [Pchalek \(2019\)](#), the objective of the EIA is to point out suitable alternatives, minimizing and compensating impacts. According to [Glasson and Therivel, \(2019\)](#), the EIA is a process in which the possible environmental impacts of a project are evaluated in a phase prior to decision-making to promote healthy environmental management. Therefore, the minimization of environmental impacts is a standard procedure in mining operations ([Falck, 2016](#)). Environmental management is the key to sustainability in mining ([Yildiz, 2020](#)), it is necessary to carry out a sustainable management of natural resources to create the minimum negative results on the environment ([Elvan, 2013](#)). Mining has an important role in the sustainable development

of natural resources, given by the important environmental and social impacts that it can generate ([Ghorbani and Kuan, 2017](#)). [Directive 52, \(2014\)](#). This indicates that EIA has to identify, describe and assess the significant effects of a project on the following factors: population and human health; biodiversity, paying particular attention to species and habitats protected under [Council Directive, \(1992\)](#) and [Directive 147, \(2009\)](#); land, soil, water, air and climate; material assets, cultural heritage and landscape; and the interaction between the factors referred previously. In addition, [Law 9, \(2018\)](#) makes compulsory to carry out an environmental impact study in the ordinary EIA. The EIA study is intended to prevent possible natural damage, establishing corrective measures and ensuring the possibility of compatibility of the extractive activity with the conservation of the environment and developing methods of recovery and monitoring of impacts on soil and water ([Casas, 2018](#)). The objective of EIA in mining is to identify, predict and prevent environmental alterations caused by extractive activities, from research and mining exploitation to the processing of the minerals to be processed ([Astorga et al., 2003](#)). The main objective is to ensure that environmental considerations are explicitly expressed and included in the decision-making process and to anticipate and avoid, minimise and compensate for negative effects on the environment ([Mora-Barrantes, 2016](#)). Among the main objectives are: to ensure that environmental considerations are explicitly expressed and included in the decision-making process; to anticipate and avoid, minimise and compensate for significant negative biophysical, social and other relevant effects of the development proposal; to protect the productivity and capacity of natural systems and their ecological processes; to promote sustainable development by optimising the use of resources and management opportunities ([Johnson and Bell, 1975](#)). In addition to the EIA, a complementary environmental assessment must be carried out in accordance with the "Methodological Guide for Environmental Impact Assessment in Natura 2000 Network" ([MET, 2019](#)), for Sites of Community Interest (SCI) and Special Areas of Conservation (SAC), which are protected areas integrated in the Natura 2000 Network (RN2000) designated for hosting an area of one or more types of natural Habitats of Community Interest (HIC) and/or habitats of the

species listed in [Law 42, \(2007\)](#), modified by [Law 33, \(2015\)](#), which transposes the Habitat Directive of the European union.

MATERIALS AND METHODS

In order to know the environmental impacts and the measures to mitigate them, a methodological procedure is used to identify mining actions, such as environmental impacting components and environmental factors susceptible to receive impacts. The environmental impacts were identified, characterised, assessed and evaluated. In this way, premises are established to ensure that quarries carry out environmentally friendly mining operations. In order to identify impacts, it is necessary to analyse the different environmental factors that undergo variations. In most studies for the identification and assessment of impacts, the impact assessment method of [Conesa et al. \(2010\)](#) is used, this is an *ad hoc* method described in their book "Methodological guide for environmental impact assessment". However, this methodology predates the latest legislative amendments. Nevertheless, it provides a solid basis on which to develop EIA studies. [Castelo et al. \(2019\)](#), after having analysed various methodologies to assess risks in open pit mining, has verified that evaluating these risks depends on a number of data that are difficult to obtain or on general quantitative methods or the data are of uncertain reproducibility, in addition, they depend on multiple occasions of the evaluator and their experience and if more reliability is required, experienced teams or expensive processes are needed, so there is no method to respond quickly and that is reproducible and reliable to date. So, following the method described above, the project

actions that may cause impacts are identified, and then the impacts are assessed in a qualitative manner, characterised by a series of attributes such as synergy, effect, accumulation, periodicity and recoverability. This technique is based on the cause-effect matrix method, which according to [Garmendia et al. \(2005\)](#) is the most widely used tool for determining impacts, derived from the Leopold matrix ([Leopold, 1971](#)) with qualitative results. This matrix is based on relating project actions with the environmental factors of the project, identifying the magnitude and importance of the potential effects on a certain environmental factor and the one generated for a certain project action. Once the impacts have been identified in the impact identification matrix and their causes, each of the impacts identified are evaluated according to a series of parameters to determine their importance in an impact assessment matrix. The importance will take values between 13 and 100 depending on the scores given to each parameter. The definition of these impacts together with their importance is shown in [Table 1](#) and it will be concluded in the EIA study whether the development of the project is acceptable or not.

The methodology followed in this article is based on the analysis of EIA studies by several authors ([Handjaba, 2012](#); [Khabali and Kamal, 2013](#); [Luna, 2015](#); [Gómez, 2016](#); [Villalba, 2017](#); [Miñarro, 2018](#); [Casas, 2018](#); [Moreno, 2021](#)), in order to apply techniques to mitigate and reduce the negative effects caused by the extraction of ornamental rock (marble and marble limestone) and limestone aggregates in open-pit quarries and their abandonment without any type of restoration. In this study the EIA studies of 8 quarries found in the Region of Murcia, which is an arid and semi-arid region, have been analysed. To identify

Table 1: Classification of impacts and its importance

Classification of impacts		Importance
Compatible	It is the one whose recovery is immediate after the cessation of the activity and does not require preventive or corrective measures.	13-25
Moderate	It is one whose recovery does not require intensive preventive or corrective measures and in which the achievement of the initial environmental conditions requires a certain amount of time.	25-50
Severe	It is one in which the recovery of the environmental conditions requires the adoption of preventive or corrective measures, and in which, even with these measures, recovery requires a long period of time.	50-75
Critic	It is one whose magnitude is higher than the acceptable threshold. It results in a permanent loss of the quality of environmental conditions, with no possibility of recovery, even with the adoption of preventive and corrective measures.	75-100

the most significant impacts on these quarries, the importance of each activity in the different phases of a quarry with respect to the different environmental factors has been calculated. For this, the lowest and highest data have been collected on the importance that each mining activity can take in each of the different phases of exploitation in relation to each environmental factor in each of the 8 analysed quarries. The components usually considered in this inventory are the abiotic and biotic environment, the perceptual environment, the socio-economic environment and the socio-cultural environment and infrastructures. The different phases in a quarry are: phase I, of preparation, in which the roads and accesses are adapted and the clearing of the vegetation and the drainage and sewers are carried out; phase II, of operation, in which the start-up is carried out by loading and blasting, the transport of materials, the creation of fissures, the auxiliary and processing operations and the maintenance of machines, an occupation and change of use is carried out of the land, the fencing and enclosure of the quarry and spills and dumps are generated; and phase III, of restoration, in which in which remodelling and revealing is carried out and there is vehicle traffic. Once the most important impacts that occur in quarry operations have been obtained, preventive and control measures are proposed to reduce or eliminate these impacts based on the different components of the environment (these measures have also been obtained from impact studies analysed) and flow diagrams are created to facilitate the application of corrective and preventive measures with respect to the pollutants existing in the quarries (dust, noise, oils and lubricants, reject materials and other quarry waste). In addition, a diagram will be created to control the topsoil, another to improve the environment and human health and one to improve the health and safety of workers and another to improve the surfaces altered by the quarrying activity and reduce the impact on the landscape, fauna and flora. This detailed study establishes a methodology that can be applied generally in other regions and open-pit mining operations, facilitating systematic monitoring and the proposal of impact mitigation measures.

Study area

The Region of Murcia is located in the Southeast

of the Iberian Peninsula, and forms part of the eastern part of the Betic Mountain Range. The area, according to the National Geographic Institute (IGN, 2020), is structurally divided into two zones: External Zones, subdivided into the Prebetic and Subbetic domains, and the Internal Zone, which is divided into three domains that rode on top of each other but were later transformed into extensional detachment faults. These domains are, from bottom to top, Nevado, Filábride, Alpujárride and Maláguide. All these domains are represented in the Region of Murcia. In addition to the materials of the Betic Mountain Range linked to the main tectonics, there are other post-orogenic materials that are well developed in the inner depressions and alluvial valleys: the tertiary basins of Campo de Cartagena, Mula, Fortuna, Calasparra syncline, Moratalla, Lorca and Rambla de Tarragoya stand out, and among the latter, the Plio-Quaternary valley of the Guadalentín-Segura. Mining in Murcia is mainly represented by the ornamental rock and aggregate sector. According to the Mining Service of the Autonomous Community of the Region of Murcia (CREM, 2019), the number of existing mines in the Region of Murcia is 227, all of them carried out by open-pit mining methods. Of these, 98 are still active. Fig. 1 shows the municipalities in which there are ornamental rock quarries and the most representative ones in which there are aggregate quarries.

There were 122 ornamental rock quarries in the Region, of which 42 are active (CREM, 2019). Specifically, these ornamental rocks are different varieties of marble and marble limestones. Their location is shown in Table 2, which shows the type of ornamental rock that exists in each municipality. The ornamental rock, in the Northeast and the Altiplano, is associated with the Jurassic layers of the Subbetic and Prebetic Eocene, large thicknesses of limestones and dolomites, with a wide variety of colours: creams, browns and reds. Marbles, strictly speaking, appear in the Betic alignments of the Alpujárride of the Cabezo Gordo in Torre Pacheco. Aggregate quarries are distributed throughout all the districts of the Region, there are 74 quarries and 32 of them are active. The most representative limestone aggregate quarries are located in the Fortuna-Abanilla district and in the district of the metropolitan area of Murcia, in Santomera, with 44 percent (%) of production, and in the district of Campo de Cartagena, in Fuente Álamo,

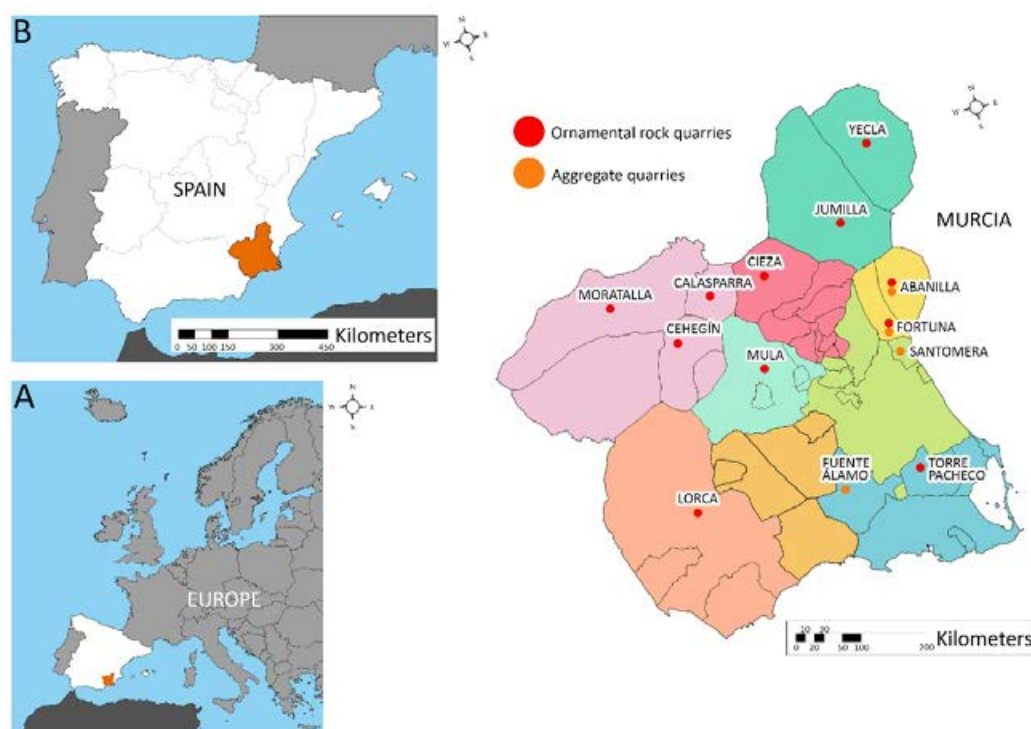


Fig. 1: Geographic location of the study area in the most representative municipalities in the Murcia region of Spain with ornamental rock and aggregate quarries

with 14% of production. Of the total production in quarries in the Region, approximately 74.8% corresponds to the aggregate extraction subsector.

The Region of Murcia has a Mediterranean climate with arid features: hot, dry summers, mild winters, although with frequent frosts in the interior, and rainfall in spring and autumn. The general characteristic of this climate is its scarcity of rainfall, concentrated in a few days of the year, with maximum rainfall in autumn. These rains, usually torrential, are produced when a mass of warm, humid air from the Mediterranean Sea enters the area and, on colliding with the coastal mountain ranges and rising, comes into contact with another mass of cold air and precipitates. These rains must be considered precisely because of the high erosive power they can unleash when it comes to recovering mining areas. According to Köppen's climate classification, (Köppen, 1936), the following climate types can be distinguished in the region: warm semi-arid (BSh), cold semi-arid (BSk) and Mediterranean (Csa). In the BSh climate, the mean annual temperature is above 18 degrees

Celsius (°C) and precipitation is low, solar evaporation exceeds precipitation, it is a hot and dry climate; in the BSk climate, the mean annual temperature is below 18°C, precipitation is also low and evaporation, like BSh, also exceeds precipitation, this climate is cold and dry; in Csa, the average temperature of the coldest month is below 18°C and above -3°C and that of the warmest month is above 10°C, the temperature of the warmest month is above 22°C and average temperatures above 10°C occur in less than four months of the year. Precipitation exceeds evaporation and there is seasonal rainfall. The summer is dry, so the minimum rainfall is quite marked and coincides with the period of highest temperatures. The rainiest season is not necessarily winter. Table 3 shows the average annual temperatures, in °C, and rainfall, in litres per square meter (l/m²), for 2019 in the study sites, the table is based on data obtained from the Murcia regional statistics portal (CREM, 2020). With regard to the winds, the west/northwest-south/east orientation (WNW-SE) of the great Betic relief lines channels the winds from the Atlantic, while the winds

Table 2: Types of ornamental rock currently exploited in the Region of Murcia and their geographical distribution. Source: own elaboration according to data obtained from the Ministry of Business, Industry and Spokesperson of the Region of Murcia

District	Municipality	Ornamental rock
Region of Lorca	Lorca	Oolitic limestones Crinoid limestones
Campo de Cartagena-Mar Menor	Torre Pacheco	Greyish-white or greyish-white limestones, with white and ochre veining Light and grey massive dolomites White to pinkish coloured limestones
Mula basin	Mula	Reddish nodular limestones Limestones with Nummulites Limestone with algae Limestones with large Nummulites
	Calasparra	Dark brown dolomites Greyish and brown dolomites Greyish limestones
	Caravaca and Moratalla	White and cream massive limestones Massive sandy limestones Calcarenites Cream-coloured massive limestone
	Cehegín	Brecciated and massive limestones varying from red to dark grey
Vega del Segura	Cieza	Brown dolomites White limestone
Abanilla-Fortuna basin	Fortuna and Abanilla	Reddish massive limestone Tertiary limestone Dark-coloured massive dolomite
Altiplano	Jumilla and Yecla	Honey-coloured dolomite White limestone

from the north/northwest orientation (N-NW), which predominate during the winter, are dry and cold due to their long route. The Mediterranean squalls associated with convection phenomena give rise to a regime of easterly winds, with humid characteristics on the eastern flank of the region, which gradually dry out towards the interior. This situation predominates in spring and summer, extending into autumn. Wind speed is generally moderate, except on the coast, where it is higher due to its exposure to easterly winds. Relative humidity varies according to proximity to the sea. Inland it varies between 52 and 63% and on the coast between 71 and 76%. These climatic characteristics create a high rate of evapotranspiration which results in a permanent water deficit. This situation is generalised throughout the Region of Murcia.

The Region of Murcia is the most arid area of the Iberian Peninsula and is part of the Segura Hydrographic Basin. The drainage network of the Region is structured around the Segura river and its tributaries, the Guadalentín, the Argos and the

Quipar. Groundwater comes from the fraction of precipitation that infiltrates due to the action of gravity. The Segura basin has been one of the first in the exploitation of groundwater, which has allowed unirrigated areas to be transformed into irrigated areas. There are currently ten different aquifer systems in the Region: Jumilla-Yecla, Calasparra, Cieza-Abanilla, Mula-Aledo, Caravaca-Moratalla, Bullas-Coy, Puentes-Valdeinfierno, Segura-Guadalentín, Cartagena and Mazarrón-Águilas (SITMURCIA, 2019). In general terms, in the Region of Murcia, there are more than two thousand plant species, which represents approximately 33% of the total number of species on the Iberian Peninsula, giving rise to a wide regional spectrum and making it one of the richest in Spain. The area occupied by crops exceeds 50% of the total and the rest is dominated by scrubland and tree species. With regard to scrubland, there is noble scrubland, with species such as Pistacia, Quercus, Rhamnus, Chamaerops, Arbutus, etc., and the characteristic scrubland of regressive stages such as rosemary (*Salvia imantopus*), esparto grassland

Table 3: Average annual temperature and annual rainfall by region with quarries

District	Municipality	Approximate annual average temperature 2019 (°C)	Approximate annual rainfall in 2019 (l/m ²)
Region of Lorca	Lorca	17,5	318
Campo de Cartagena-Mar Menor	Torre Pacheco and Fuente Álamo	18,6	419,5
Metropolitan area of Murcia	Santomera	18,5	523,9
Mula basin	Mula	18	455
Northwest	Calasparra, Caravaca, Moratalla, Cehegín	15,4	493,2
Vega del Segura	Cieza	17,3	475
Fortuna-Abanilla	Fortuna and Abanilla	18,8	575
Altiplano	Jumilla and Yecla	16	430

(*Stipa tenacissima*), thyme (*Thymus*), etc. From the coast to the inland mountain ranges, in a south-east-north-west direction, the wooded area increases and, therefore, the diversity of forest systems. The tree formations consist of frugal species, such as pines, junipers and xerophytic oaks. The wooded hills are made up of conifers and the broadleaved forests are mainly composed of kermes oaks, although they also appear in mixtures with other broadleaved and resinous species. The mid-mountain and foothill areas in the centre, north-west and north-east of the Region, include quercines (*quercus*), although these areas are highly altered. The intensive use of vegetation cover has led to significant deforestation, accentuated by climatic conditions. The fauna presents a very heterogeneous territory with a great variety of habitats, which contributes to maintaining a significant biodiversity of species such as Phartet (*Aphanius iberus*), Great Bustards (*Otis tarda*), Otters (*Lutrinae*), Shelducks (*Tadorna*), Vultures, Birds of Prey, Salamanders (*Caudata*), etc. Linked to the forest ecosystems, there are unique species such as the mountain goat (*Capra pyrenaica*), the Trompet Bullfinch (*Bucanetes githagineus*) and Dupont's Lark (*Chersophilus duponti*), as well as other species included in the annexes of the Habitats Directive and the Birds Directive, especially bats. There are also 51 species of mammals, both terrestrial and marine, many of them threatened or under some kind of conservation status. On the other hand, there are not many reptile and amphibian species, there are only 20 reptile species and 11 amphibian species and they are decreasing due to several negative factors that are influencing the region such as the aridity of the area, the use of water resources, the intensive

agriculture, the introduction of invasive species, etc. Regarding to birdlife, there are almost 300 species, only one third of which are sedentary. According to the Council Directive, (1992), 42% of the regional area is classified as HIC (SITMURCIA, 2019).

RESULTS AND DISCUSSION

Main impacts from ornamental rock and limestone aggregate quarries in the Region of Murcia

In EIA studies, the description of the Environmental Inventory must take into consideration the components of the environment that intervene or may potentially be affected by the activities of the project to be environmentally assessed (Garmendia, 2005). The following tables show the most significant impacts that are usually produced by ornamental rock and limestone aggregate quarries in the Region of Murcia, in arid and semi-arid areas, according to the EIA studies analysed (Table 4, 5 and 6). These tables indicate the values that the impacts usually take, these values are previously defined in table 2 according to the classification of the impact. The impacts represented in Tables 4, 5 and 6 are those that generate a negative impact.

Analysing this phase, it is obtained that the environmental factors that can cause a severe impact on quarries in arid and semi-arid areas are noise and vibrations in the phase of adaptation of roads and accesses, the loss of soil quality in the clearing of the land, the loss of water quality when creating drains and aquifers, the alteration of flora and vegetation in the adaptation of roads and accesses and the clearing of vegetation and a visual impact on the creation of roads and accesses. In the elimination of the existing vegetation to carry out work in the

Table 4: Importance of the most significant environmental impacts in ornamental rock and aggregate quarries in the Region of Murcia in Phase I of preparation

Environmental factors			Phase I preparation		
			Adequacy of roads and accesses	Drains and sewers	Vegetation clearing
Physical environment	Atmosphere	Noise and vibration	35-55		
		Air quality	35-40		15-30
	Terrestrial environment	Soil quality	25-30	25-40	35-55
		Geomorphology	20-35		
	Water	Water quality		45-60	30-40
Biotic environment		Hydrogeology		35-45	
	Flora and vegetation		45-60		25-60
	Fauna	Biotypes	25-50		
		Species of interest	25-50		25-30
Perceptual environment	Landscape	Intrinsic quality	40-50		25-40
		Visual impact	45-60		25-40
Socio-economic and socio-cultural environment	Productive use	Agricultural forestry and livestock	40-45		

quarry; the surrounding vegetation is affected by the dust particles generated on the exploitation, these particles are deposited on the stomata, interfering with the chlorophyll function and intervening in growth. High concentrations or long exposures of nitrogen oxides (NO_x) from vehicles and machinery cause leaf pigmentation, necrosis and reduced growth. The high concentration of sulfur dioxide (SO₂), released from the fuels used, prevents an assimilable transformation for the metabolism of the vegetation and initiates a cellular rupture (Villalba, 2017). With the elimination of the vegetation the habitat of some species is destroyed, which leads to the emigration of the fauna, and the edaphic fauna is also eliminated. The rest of the impacts are moderate (with values of the importance between 25 and 50) or compatible (with the importance between 13 and 25) and in the gaps in which there is no importance it is because they are insignificant.

In this phase, there may be a critical impact on the activity of occupation and change of land use, since it creates a modification of the landscape and a qualitative visual impact. There may also be severe impacts (with values of importance between 50 and 75) such as noise and vibrations to the atmosphere produced by the use of heavy machinery and in drilling, blasting and mechanical preparation of the raw material. Loss of air quality caused by emissions of polluting substances due to the use of heavy machinery, the use of explosive substances

for blasting, and the use of fuels and lubricants for machinery. Among the polluting substances that are released are nitrogen oxides (NO_x), which contribute to the generation of photochemical smog and acid rain and, in addition, in areas where there is surface water, can cause eutrophication. Another polluting substance is carbon dioxide (CO₂), which increases the greenhouse effect, in addition, it also contributes to the loss of air quality, the generation of dust in extraction operations, drilling, blasting, accumulation, charging, transport, crushing and classification of the resource and that produced in the creation of accesses to the quarry (Villalba, 2017; Jiskani, 2021). Another severe impact would be the alteration of the geology and the loss of the quality of the soil when carrying out the work in the quarry. There is also the risk of groundwater contamination due to accidental discharge of polluting substances such as oils or other substances from vehicles and machinery used. The voids generated in the quarries, in addition, can create irreversible contamination in the water table due to the creation of tailings, especially if these voids occur in karst areas with a high degree of cracking and in which the waters flow rapidly, without being by filtration processes, reaching more or less deep aquifers (depending on the topography) with their respective discharge and catchment areas (Khabali and Kamal, 2013). With regard to surface waters, there may be contamination of rivers due to erosion by rainwater currents and

Table 5: Importance of the most significant environmental impacts in ornamental rock and aggregate quarries in the Region of Murcia in Phase II of exploitation

Environmental factors	Phase II of operation							
	Noise and vibration	Blast start and charge	Transport of materials	Auxiliary and processing operations	Creation of fissures	Occupation and change of land use	Landfill of tailings, waste rock dumps	Fencing and enclosures
Medium physical	Atmosphere	35-65 20-25	40-55 50-55 25-30	45-60 40-50	40-45 40-60 25-60 25-30 30-40 35-45	40-45	45-50 40-50	
	Terrestrial environment	25-35 25-35				25-30		
	Water	30-35	30-40 40-50				30-55	
	Flora					25-30 25-35 25-30 45-50		40-45 35-45 40-45
Biotic environment	Fauna	25-35 50-55	35-40 30-40	30-35		55-90		25-30
	Landscape Visual impact Recreational use: Tourism, hunting and sporting activities					35-50		
Socio-economic and socio-cultural environment	Productive use: Forestry, agriculture and livestock Human health and safety Communication routes: mobility Land use		50-65 40-50		40-45	40-50		

Table 6: Importance of the most significant environmental impacts in ornamental rock and aggregate quarries in the Region of Murcia in Phase III of restoration

Environmental factors			Phase III restoration		
			Vehicle traffic	Remodelling	Revegetation
Medium physical	Atmosphere	Noise and vibration	45-55	25-30	
		Air quality	35-45	25-35	
	Terrestrial environment	Soil quality	45-50	40-50	40-45
		Geomorphology		30-35	
Medium biotic	Water	Water quality	45-50		
		Plant formations	30-40	25-35	25-30
	Flora	Species of interest	45-50	30-40	25-35
		Biotypes		30-35	40-45
Medium perceptual	Fauna	Species of interest	45-50	30-35	30-40
		Intrinsic quality	45-50		25-30
	Landscape	Visual impact	35-45	30-40	30-35
	Productive use:				25-30
Socio-economic and socio-cultural environment	Forestry, agriculture and livestock				
	Human health and safety		40-50	30-40	
	Communication routes: mobility		45-50		
	Land use		40-45		

the wind, since they can carry substances from the vehicles and machinery used and can lead to the loss of ecosystems. Due to the noise, vibrations and dust produced in the quarries, fauna is displaced and redistributed and habitats can be altered due to the possible contamination of groundwater and rivers. In the generation of holes and slag heaps, the visual quality of the landscape is lost and, in addition, visual intrusion occurs due to the use of machinery. The emission of noise, vibration, dust and water pollution can cause harmful effects on workers, those generated cause eye irritation, congestion and lung diseases and SO₂ and dust particles in suspension, produce allergies and infections in the respiratory system. The rest of the impacts are moderate (with values of the importance between 25 and 50) or compatible (with the importance between 13 and 25) and in the gaps in which there is no importance it is because they are insignificant.

In this phase, all the impacts obtained would be moderate, except for the noise and vibrations produced by vehicle traffic, which could become severe. The rest of the impacts not indicated would be compatible or insignificant. In this phase, the contribution of land is of interest, which can generate occasional air pollution due to suspended particles generated by the unloading of trucks. Dust has a negative impact on air pollution in the atmosphere, but also on water and land resources, as it interrupts the natural processes of flora and fauna and causes

irreversible effects on human health (Timofeeva and Murzin, 2020). Reducing the noise and vibrations produced in many operations such as removal of the surface soil, drilling, blasting, excavation, crushing, handling and transport and other operations with machinery is important to the health of people and their well-being and that of the ecosystem, as high levels negatively influence species by increasing stress levels, modifying their habitat and masking other sounds they need for their survival (Jain et al., 2016). Another factor that negatively influences the environment is mining waste, because if they are not treated properly they contaminate the water, the atmosphere, the soil and the occupied land. (Chen et al., 2020).

Preventive measures to control and mitigate environmental impacts

Once the impacts that usually occur in ornamental rock and aggregate quarries in the region of Murcia have been obtained and analysed, the measures that are usually applied to control and reduce these impacts have been analysed on the basis of the environmental impact studies studied and it has been found that 90% of the measures that are applied to control the different negative environmental factors are always the same. These measures are described below, and in relation to them, flow diagrams will be proposed to simplify their control and application in the quarries. In order to control the visual impact

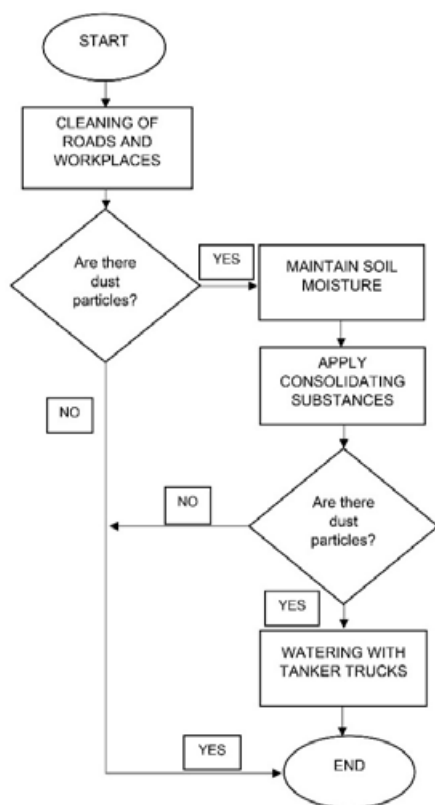


Fig. 2: Flowchart to reduce dust on roads and workplaces

and the modification of the landscape, the original forms of the accesses that are not essential for access to the restoration of the land must be restored by means of disabling and environmental recovery. In addition, as work on the slopes is completed, a slope ageing product should be applied that simulates the chromatic characteristics of the environment. Efforts should also be made to work in areas of low visibility from main roads and urban centres. The measures that are usually applied to reduce dust are: clearing land and opening roads, as far as possible, on days when the force of the wind does not imply a high risk of suspension of materials; carrying out effective maintenance of access roads to avoid the formation of dust and the accumulation of mud on the roads due to the transit of lorries; maintaining tracks and squares with a sufficient degree of humidity to avoid the dust deposited on them becoming suspended, using, if necessary, substances that consolidate and maintain the humidity of the soil. Another option is to irrigate with water tankers, as water is not normally

available in mining areas and rainfall in the Region of Murcia is scarce; workplaces should be kept clean to prevent the accumulation of dust that can later be put into suspension. When there are accumulations of dust in different parts of the quarry, these shall be removed as soon as possible; in addition, the provisions of [Order ITC 2585, \(2007\)](#) on protection of workers against dust, in relation to silicosis, in the extractive industries. The suspension of dust in vehicle and machinery transit operations must also be controlled by irrigation, paying special attention to the quarry site, access tracks and unpaved areas, in order to affect human beings and the surrounding flora and fauna as little as possible. In order to make it easier to control dust, a flow chart has been drawn up with some proposed measures ([Fig. 2](#)).

To control noise and vibrations in quarries, the engines of machines that are going to be on standby for long periods of time should be switched off, the noisiest operations should be planned so that they do not coincide in time to avoid a large amount of noise and vibrations, and noise should be made on working days and intermittently. Except in emergency situations, work should not be carried out at night. The machinery used must comply with the permitted noise emissions, in this case regulated by [Royal Decree 212, \(2002\)](#). Blasting must be carried out in the middle of the day. In addition, to reduce the effect of air waves and vibrations from blasting, bottom-loading cartridges should be primed with non-electric detonators, detonating cord should not be used in the open air and the connection and sequencing between blast holes on the surface should be done with non-electric connectors so that the sequencing time is different from one another. In addition, if mechanical preparation plants are present, noise protection screens should be provided. The correct operation and commissioning of the vehicles on the operation must be checked, carrying out the corresponding gas emission controls and equipment checks established by the manufacturers. This will reduce noise and the emission of polluting gases, as well as reducing the risk of breakdowns and potential accidental spillage of polluting liquids. Periodic inspections of machinery must comply with [Law 34, \(2007\)](#), with respect to air quality and protection of the atmosphere and with [Decree 1439, \(1972\)](#) with respect to noise. In order to control oil and lubricant spills, the use of vehicles should be optimised to allow for the maximum

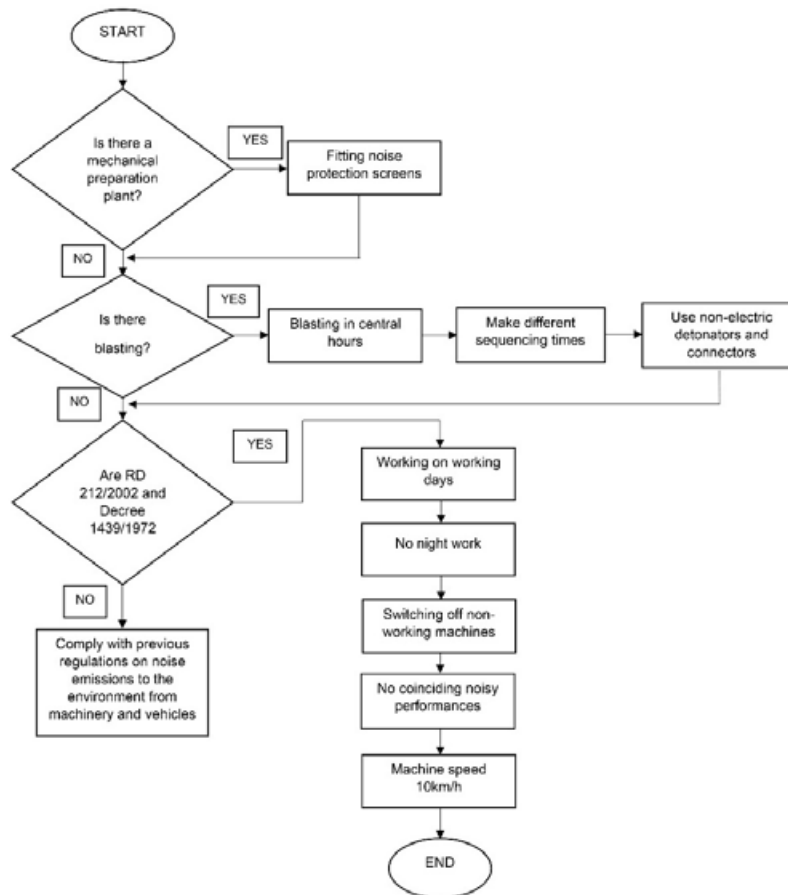


Fig. 3: Flow chart to reduce noise and vibrations in mining

operationally possible fuel savings. Equipment and vehicles must be more efficient and less polluting, more efficient vehicles and heavy equipment produce less pollution and reduce greenhouse gas emissions since their engines are cleaner, so older equipment must be replaced (Jiskani *et al.*, 2021). Repairs or oil changes of machines should be carried out in specialised workshops. In the event of accidental spillage of these materials, they must be cleaned and collected, deposited in containers for subsequent removal by an authorised waste manager, so that they do not affect runoff water or water that may infiltrate. Used oils and any other waste qualified as such from the exploitation must be compulsorily removed by an authorised hazardous waste manager. To reduce SO₂, CO₂ and NO_x emissions as far as possible, the speed of machinery will be reduced to 10km/h in the area, in addition to choosing machinery

that is equipped with catalytic converters that reduce emissions as much as possible, and the days on which the atmospheric phenomenon of thermal inversion occurs will also be taken into account so that accumulations of emissions do not form in the area due to a lack of ventilation (Villalba, 2017). To make it easy to implement these measures, a flow chart for noise and vibration reduction (Fig. 3) and a flow chart for fuels and lubricants (Fig. 4) have been developed.

A simple flow chart (Fig. 5) is proposed to control topsoil. The measures to be applied for this would be as follows: the topsoil should be removed, stockpiled and adequately maintained for later use in restoration. This removal must be carried out with care to avoid its deterioration by compaction and thus to preserve the soil structure, the existence of microorganisms, etc. For this reason, the passage of machinery over this soil should be avoided. During storage, it must

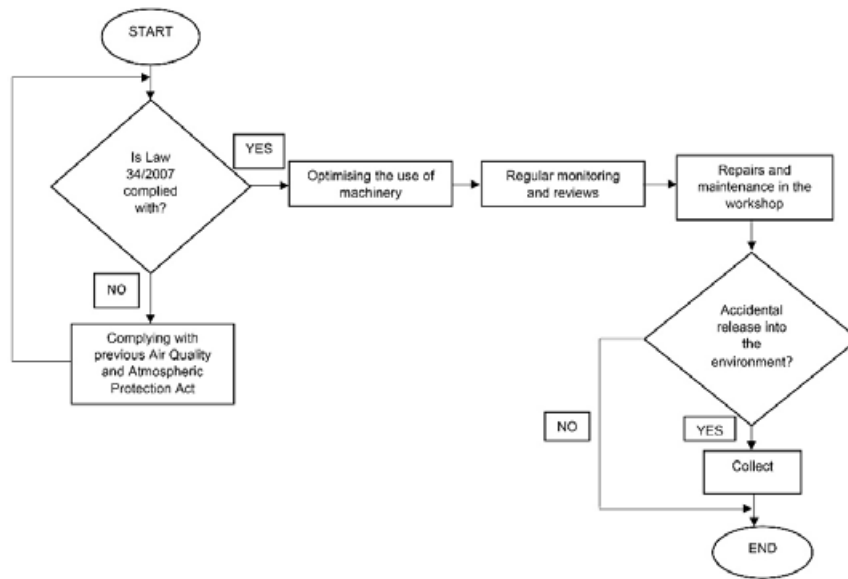


Fig. 4: Flow chart for reducing fuels and lubricants in mining

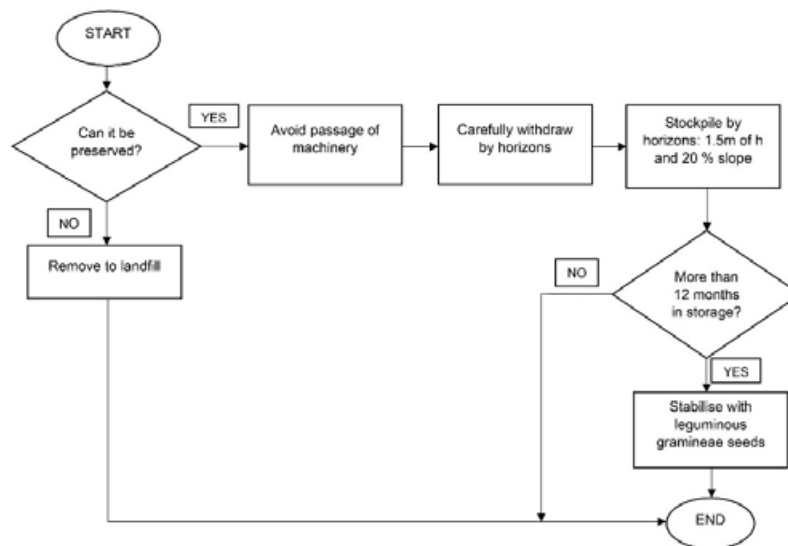


Fig. 5: Flow chart to control topsoil in mining

be protected from wind, water erosion and pollutants that alter the vegetative capacity. Stockpiles should not exceed 1.5m in height and slope gradients should not exceed 20° to avoid erosion (Casas, 2018; Moreno, 2020). The removal and stockpiling should be done differentiating the different soil horizons in order to be able to restore them in the restoration with the same original arrangement. Where feasible,

the original soil flora will be preserved, in order to try to maintain a fertile layer on the surface to facilitate vegetation growth and control runoff erosion in the short term in sloping areas. If topsoil is to be stockpiled for more than 12 months, it should be stabilised with a mixture of leguminous and grass seeds to protect it from erosion and preserve its soil characteristics.

In order to control waste material in quarries, an inner spoil heap must be created to deposit the tailings generated in the exploitation, which will progress as the exploitation pit progresses. The pit must be filled to the level of the surrounding ground surface with the tailings from the limestone rejects. The topsoil, previously removed and reserved, shall be spread over the tailings from the rejects located inside the shaft, in order to subsequently proceed with the revegetation of the land. If there is no topsoil from the mining activity, a topsoil with similar characteristics to the existing one must be placed. When work is finished in the mining areas or work is stopped for more than one year, all types of material, machinery, waste and remains that may be left in the area must be collected and taken to a landfill site, leaving the area in a perfectly clean condition. Revegetation should be attempted with native and fast-growing species. Exotic species should be avoided, as they are susceptible to becoming invasive. Planting will include, when the soils require it due to insufficient stockpiles or inadequate quality, topsoil, fertilisers and amendments, and of course,

the necessary tillage. When the revegetation work is completed, care of the topsoil has to be carried out, including replacing dead plants during the following years, installing protective structures to prevent trampling and additional foot watering. A simple flow chart has been proposed to control the reject material (Fig. 6).

In quarries there are reject materials that must be removed from the work areas so that they do not contaminate the environment; these can produce, among other things, water, soil and atmospheric pollution. For this purpose, the most common measures that are usually applied are shown together with a flow chart (Fig. 7) to make it easier to control them. Solid waste should be disposed of in controlled landfills. Leftover materials and non-hazardous waste should be deposited in authorised landfills. If there is a layer of debris prior to the removal of usable soil, this should also be taken to authorised landfill sites or put to an appropriate use. Perimeter channels can be planned to prevent water from the outside from entering exploitation. Small mammals and other vertebrates that may fall into ditches or holes created

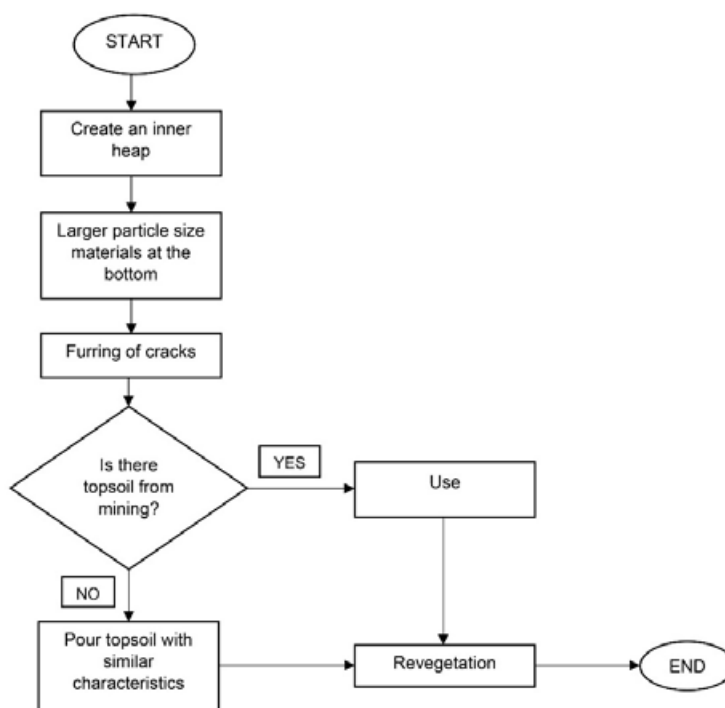


Fig. 6: Flow chart for controlling reject material from mining

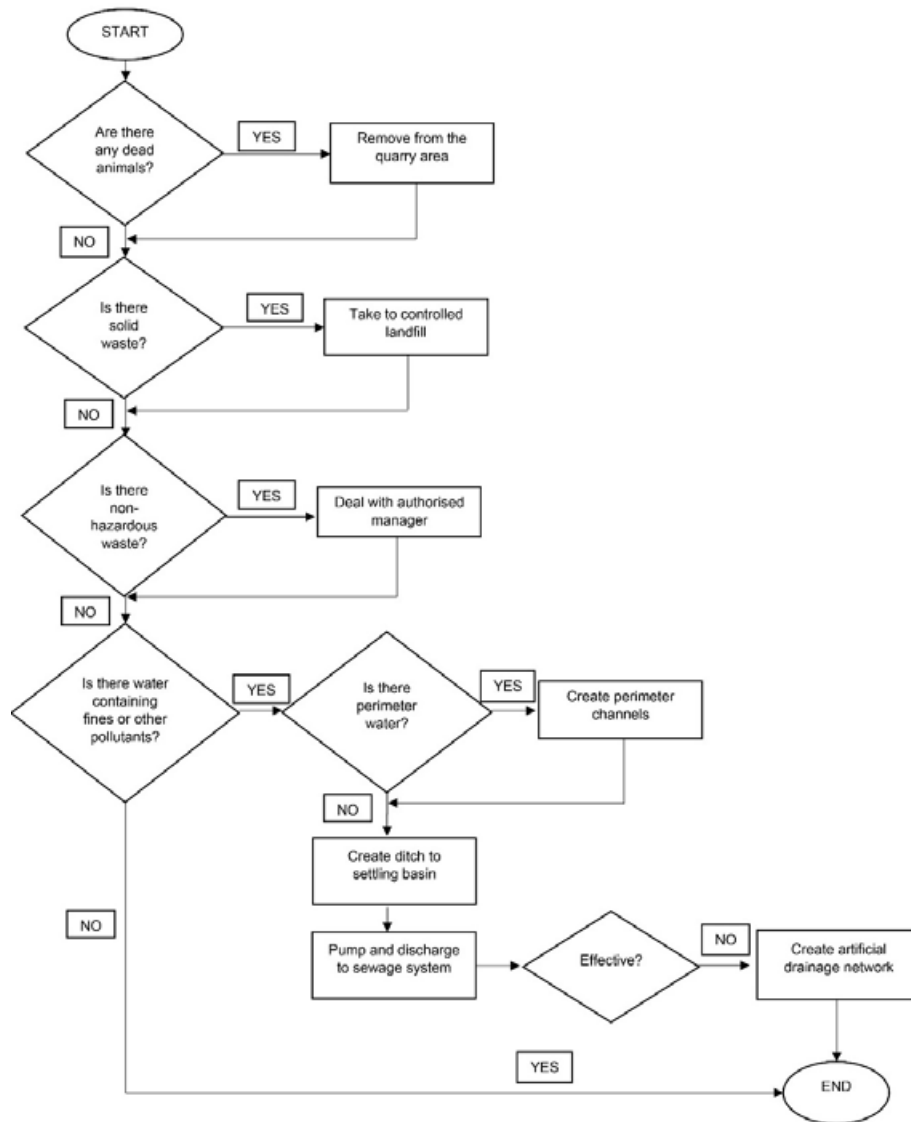


Fig. 7: Flow chart to control and reduce other quarry waste in quarries

on the exploitation must be released after daily checks prior to the start of exploitation work. For the collection of water from the interior, a ditch can be built to collect the water in a settling basin, so that the water laden with fines does not reach the natural drainage network. The clean water from the settling basin will be pumped and discharged directly into the drainage network. If the water table is below the excavation level and with the above measures, the mining activity will not affect the drainage network or any aquifer. If necessary, an artificial drainage network

must be created to ensure trafficability and to channel the resulting runoff. The processing and use of waste, in addition to not polluting the environment, serves to compensate for the scarcity of resources, which means that I have great social, environmental and economic benefits. In addition, there is a strategic commitment to the circular economy that supports waste minimization (Jiskani et al., 2021).

It is crucial to consider the environmental and human health, one to improve the health and safety of workers for which the measures given below are

usually used and based on them a flow chart has been created for easy application (Fig. 8) and another to improve the surfaces altered by the quarrying activity and reduce the impact on the landscape, fauna and flora for which other measures also described below have been established and have been collected in Fig. 9. Beacons and barriers must be placed indicating danger zones on the site, access points, speed limits, etc. Workers must wear the appropriate work clothes and personal protective equipment necessary for

the performance of their tasks. The collaboration of forestry agents must be ensured so that the works are carried out with the least possible risk of fire. The evolution of the slopes must be monitored as the work progresses. The bottom layer of the backfill must be made up of the materials with the highest granulometry, in order to favour the stability and drainage of the whole deposit. No people or material should be in the vicinity of the working slope during the removal of the material. Fire must not be used in

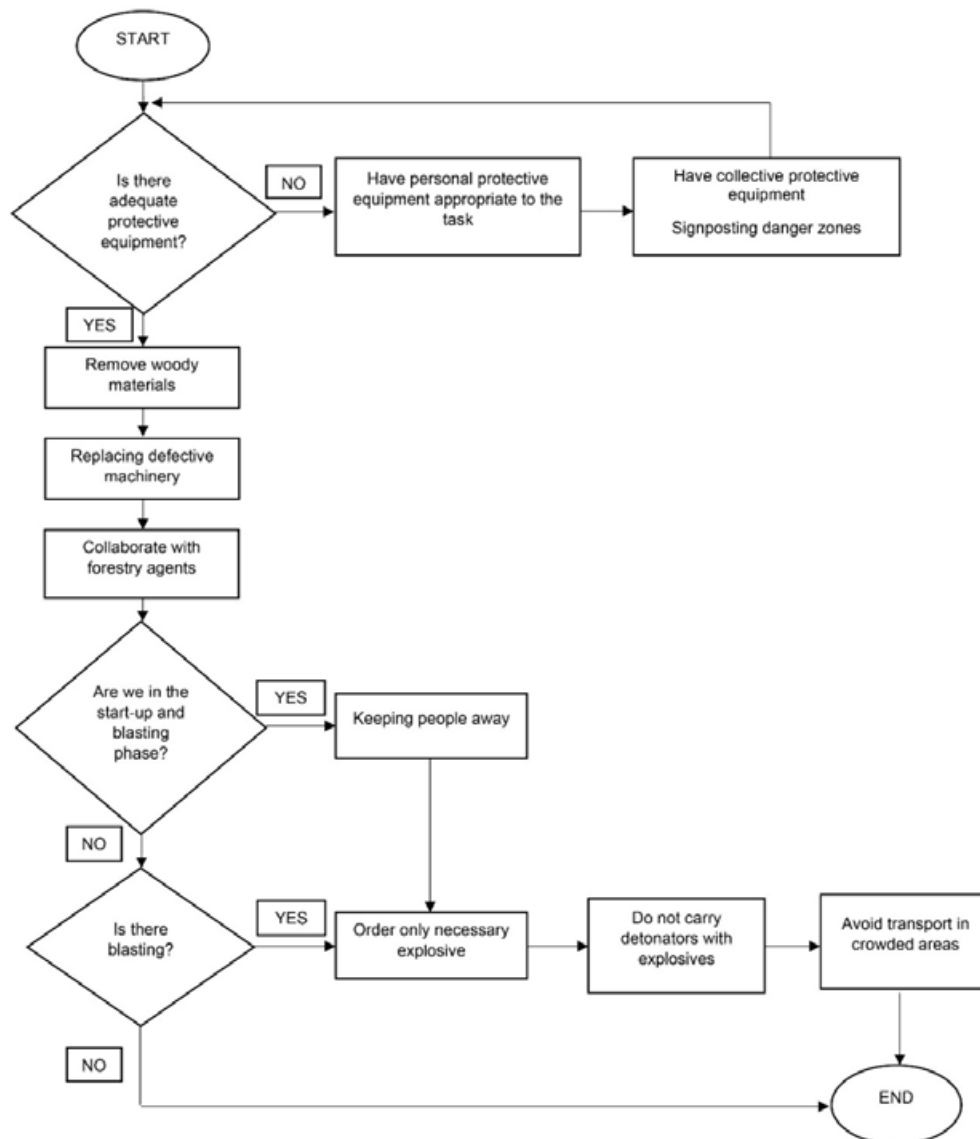


Fig. 8: Flow chart for improving the health and safety of workers in quarrying

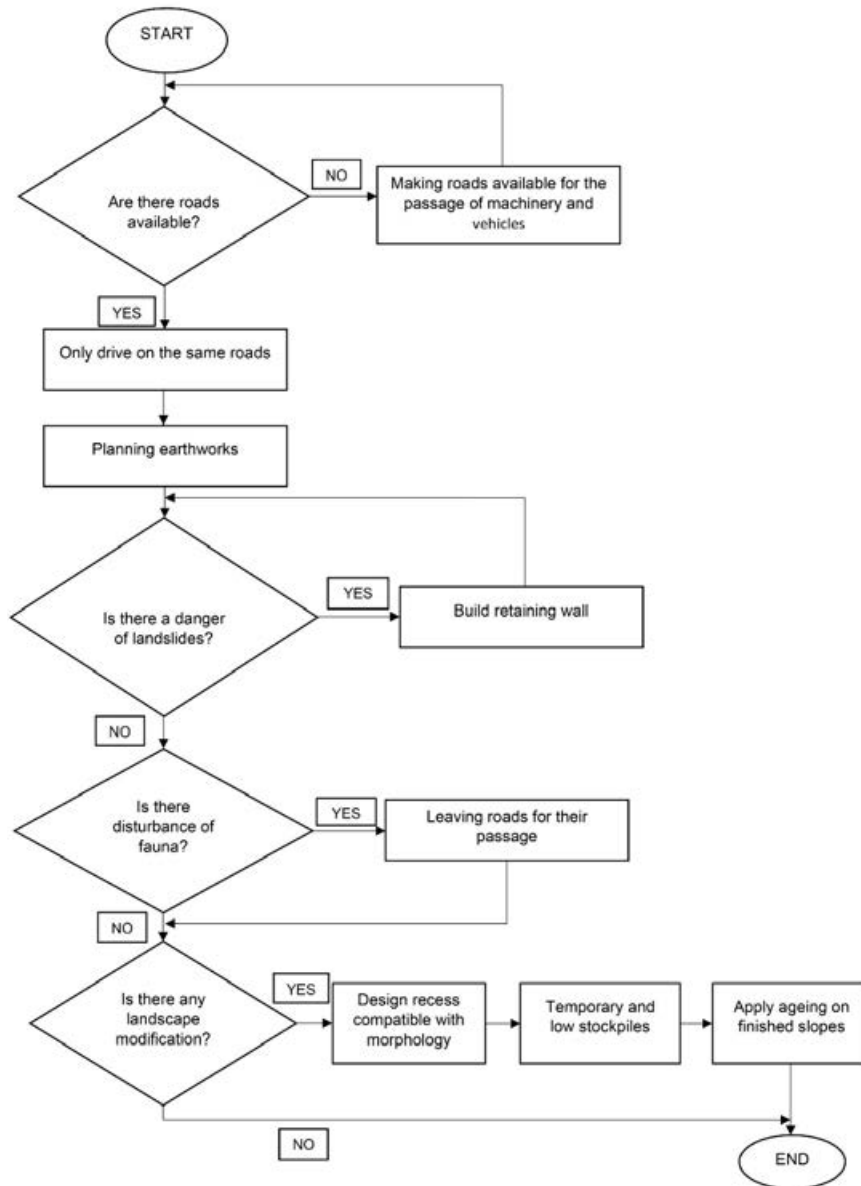


Fig. 9: Flow chart to improve disturbed areas in mining and reduce the impact on landscape, fauna and flora

the area during the mining phase. Woody materials from the opening of roads and tracks must be removed so that they do not pose a fire risk once dry. In addition, to avoid sparks, malfunctioning machinery must be replaced. The necessary extinguishing media must be provided to prevent the spread of fire. Non-flammable species should also be selected from among the species suitable for revegetation in this area.

For the preservation of the fauna, the roads must be used by both vehicles and people and not use areas not designed for traffic. In order for the fauna species to gradually adapt to the changes in their habitat, the removal of soil and vegetation must be done progressively and slowly. Every day before work begins, it must be checked that there are no animals in the ditches or holes. If an area is to be enclosed, small mammals must be allowed to pass through

at points where areas with natural vegetation are interconnected. Design of a hollow compatible with the morphology of the environment both during the exploitation phase and in the final phase of restoration. In order to foster natural regeneration over time, conservative as well as preservative actions must be carried to protect biodiversity through the greening of areas (Jiskani *et al.*, 2021). Existing roads and tracks should be used, opening new roads only if strictly necessary. It is important to plan earthworks to reduce disturbed areas. If necessary, retaining walls should be built to prevent soil slides and possible landslides.

The use of this flow charts, which can be widely applied in open-pit mining activities, allow to reduce the impacts of this activity. Moreover, they serve as tools before starting the exploitation and aid to prevent possible effects.

CONCLUSION

Open-pit mining extraction systems have a series of similar characteristics that allow a systematic approach to be established when analysing the impacts. Many authors use different methodologies to assess, prevent and mitigate risks, so there is no single and simple method to respond quickly and that is easy to reproduce and reliable to date, so an impact mitigation system has been presented that allows evaluating the impacts of quarries in arid and semi-arid areas (most of the time the impacts are the same in areas with similar climatic conditions), but at the same time it includes proposals established through the use of flow charts that allow the development of actions to mitigate the negative effects of the exploitation during the active phase and once it finishes. In addition, the use of these charts before starting mining allows to prevent some possible environmental impacts. In this study, were found that 90% of the preventive and corrective measures applied in environmental impact studies in quarries in arid and semi-arid zones are the same. Several environmental impact studies have been first analysed to create these charts, taking into account the importance of the most significant environmental impacts in this type of quarries in the different exploitation phases (preparation, operation and restoration) in order to identify the impacts, the negative effects that they produce. The flow charts propose in a simple way measures to reduce

dust on roads and workplaces, noise, fuels and lubricants in mining activities, measures to control topsoil and reject materials in open-pit mines and measures to control other wastes. In addition, some measures have been created to improve the safety and health of workers in quarrying and to improve disturbed areas affected by mining, reducing the impact on landscape, fauna and flora. In this study, a number of preventive and corrective measures have been taken in order to avoid or, when appropriate, moderate the impacts indicated above, considering the components of the environment that intervene or could potentially be affected by mining: physical and biotic environment, perceptual environment, socio-economic and socio-cultural environment. However, as it can be deduced from the preliminary analysis of the legislative framework, the mitigation of environmental impacts is subject to possible new modifications that will have to be incorporated into the methodological framework of the EIAs. In this sense, the establishment of flow charts allows their easy incorporation and updating in a simpler way, as well as their subsequent application and evaluation during and after exploitation. Moreover, future actions will achieve the development of a software, modelling the environmental impact studies based on the framework established by these flow charts. Another future action is to establish the Environmental Surveillance Plan (ESP), since in it, it is about establishing a system that guarantees compliance with the indications, preventive and corrective measures, it tries to define the fundamental elements that must be controlled to meet its objectives. In this specific case, the function of the ESP is to establish a control system, that is, to monitor during the preparation, exploitation and restoration phases of the project.

AUTHOR CONTRIBUTIONS

M.A. Peñaranda Barba performed the literature review, experimental design, analysed and interpreted the data, prepared the manuscript text, compiled the data and manuscript preparation and manuscript edition. V. Alarcón Martínez compiled the data and manuscript preparation, helped in the literature review and manuscript preparation and helped in the manuscript text. J. Navarro Pedreño performed the literature review, manuscript edition, helped in the literature review and manuscript

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

<i>BSh</i>	Warm semi-arid
<i>Bsk</i>	Cold semi-arid
<i>CREM</i>	Statical portal of the Region of Murcia
<i>CO₂</i>	Carbon dioxide
<i>Csa</i>	Mediterranean
<i>EIA</i>	Environmental impact assessment
<i>ESP</i>	Environmental Surveillance Plan
<i>HIC</i>	Habitats of community interest
<i>IGN</i>	National Geographic Institute
<i>l/m²</i>	Liters per square meter
<i>MET</i>	Ministry for Ecological Transition
<i>M€</i>	Million euros
<i>N-NW</i>	north/northwest orientation
<i>NO_x</i>	Nitrogen oxides
<i>RN2000</i>	Natura 2000 network
<i>SAC</i>	Special areas of conservation
<i>SCI</i>	Sites of community interest
<i>SITMURCIA</i>	Territorial information system of the Region of Murcia
<i>SO₂</i>	Sulfur dioxide
<i>WNW-SE</i>	west/northwest-south/east orientation
<i>°C</i>	Degrees Celsius
<i>%</i>	Percent

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CASE STUDY

Analysis of legislative acts in water management

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ABSTRACT

BACKGROUND AND OBJECTIVES: This paper focuses on the development of Czech laws of water resource protection. The presented research examines the statistical data of the number and type of legislative acts concerning to water protection issued in the Czech Republic during the period 1990-2019. Several types of legislative acts are followed in administrative law and statistically compared by the development in time and its type. The survey focuses on general water protection acts, water sewage management, agriculture sector, hygiene standards, and the protection of the basins of Czech rivers (e.g., blue water and gray water).

METHODS: The analysis firstly concerns to the development of the number of legislative acts during 1990-2019 and secondly discusses a diversification of the legislative acts types (laws, decrees, resolutions, regulations, and strategic plans). A total of 12,272 legislative acts is analyzed during three phases of Czech modern history: 1990-1992 (Czechoslovakia), 1993-2003 (Czech Republic before its accession to the European Union), and 2004-2019 (Czech Republic in the European Union).

FINDINGS: Statistical elaboration of legislative acts proves that it is possible to determine different types of water management over time. Protection of water resource management in the Czech Republic was forming from crisis management (1990-1992), via operational management (1993-2003) to strategic management (2004-2019). Current trends after 2020 show a new trend towards integral management.

CONCLUSION: Findings provide better understanding of changeable importance of water protection and management attitudes in the Czech Republic in reaction to the development of society.

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INTRODUCTION

Environmental state protection can be monitored from the volume of individual laws and their impact compared to other regulations that accompany them (Keizer and Shapiro, 2019). The nature of the regulations reflects the quality of government decision-making (Povitkina and Bolkvatze, 2019). The statistical comparison of laws reflects the long-term quality of the government approaches to water conservation issues. The presented case study focuses on the development of Czech laws of water resource protection and statistically observes all Czech administrative and legislative acts about water protection (laws, decrees, regulations, resolutions, strategic plans) focusing on blue water (i.e., freshwater surface and groundwater), and gray water (i.e., polluted water, sewerage problematics). The study primarily focuses on the quantitative evaluation of legislative acts. The examined period of legislative acts (1990-2019) is divided according to the milestones represented by major political events in Czech history: 1989 (the end of the socialist regime), 1993 (the establishment of an independent Czech Republic), and 2004 (Czech accession to the EU). A comparison of the legislative processes in the surveyed stages reveals a different approach to water protection issue. A research question (Q1) asks whether there are differences in the relative statistical evaluation of all Czech adopted legislative acts documenting different approaches to drinking water protection management in individual periods (1990-1992, 1993-2003, 2004-2019). Statistical analysis of publicly available data has been chosen as a main criterion for evaluation of all administrative and legislative regulations associated with the protection of drinking water in the Czech Republic (Czechoslovakia) from public sources. The final

evaluation of administrative law is based on 12,272 legislative acts. Table 1 presents the classification of legislative acts from 1945 till 2019 according to administrative sectors.

Relevant data for the observed period (1990 - 2019) were selected from the set of legislative acts of the Czech Law. According to political changes of the country, such as a split of Czechoslovakia in 1993 and the entry to EU in 2004, the presented approach uses a differentiation of three periods (1990-1992, 1993-2003, 2004-2019) for a relative data comparison. The international Water Exploitation Index (WEI) stands for a dependent indicator (Dikovitsky and Shishaev, 2019). The analysis is considering a relation between individual laws, e.g. how many laws has an EU Member State adopted from the EU administration. This kind of data analysis is applied much more often in political science, sociology of law and also in management. The applied method is based on modern trends in the sociology of law (DeGroot *et al.*, 1994). Statistics in legislative acts and openly accessible meta data can be used to identify new multilevel dimensions of law (Dikovitsky and Shishaev, 2019). A quantitative survey of legislative acts and their comparison by the legislative type and the WEI indicator in observed periods is used for the analysis. Different approaches to water protection have been demonstrated by quantitative statistics. According to the mutual ratio of individual legislative acts in three periods, different approaches to water protection can be interpreted by using management tools. Discussions about functional models that are implemented in the management of public institutions evoke another issue. According to the current studies (Boin and Christensen, 2008), the public sector is focusing on new ways of managing and defining “public interest.” One of the factors

Table 1: Legislative Acts Classification Based on Administrative Sector

Administrative Sector (1945-2019)	Laws (including amendments)	Public regulations	Governmental decrees	All legislative acts
Environment	135	1036	470	1641
Water	58	198	60	316
Waste	42	112	50	204
Agriculture	289	894	572	1755
Forestry	39	131	85	255
Healthcare	256	736	511	1503
Other Sections	458	3456	2684	6598
Total	1277	6563	4432	12 272

shaping new management approaches is based on written documents guaranteeing a central role in the performance of the governmental power to the administration. Public management is inspired from private management in a way of “New Public Management”. A view in one specific sector can survey several diverse goals and methods of public management. There is also a trend of empirical accounts of public institutions consistently identifying leadership as a crucial explanatory variable (Deverell, 2012). The division of management into crisis, operational, and strategic components means that a sustainable and responsible organization can be maintained. A similar management process in the number of decisions on a specific topic can be observed in a legislative act within the EU. One of the criteria for dividing management into crisis, operational or strategic is the volume and type of legislative acts issued in the specified sector. Therefore, this approach also applies to political decisions professionally driven to solve real problems. Water supply as well as the policy of protection of drinking water resources in today’s state are subjects to professional supervision. The protection of water resources in democratic states is not burdened by the ideology but rather by maximalization of the “public good” protection (Schmidt and Matthews, 2017). The aim of the current study is to find out whether there are statistical differences in all adopted legislative acts regarding to water protection topics demonstrating different approaches towards drinking water protection management. The study was performed in the Czech Republic for three time periods (1990-1992, 1993-2003 and 2004-2019).

Background of water protection in postwar Czechoslovakia (1945-1989)

A specific political interest for protection of water resources did not exist in Czechoslovakia after 1945. Subordination of water protection to economic and industrial development was expressed after 1948 by so-called Five-Year Plans where water was primarily defined as a secondary raw material for industry or agriculture. Furthermore, the socialist system did not deal with this topic in any detail (Palát *et al.*, 2010). Several partial decrees of the Czechoslovak Government were the first impulses of water protection in the early 1950s. The construction of waterworks and water supply municipal and

agricultural networks was massively supported as an indicator of improving the local economy and life in a centrally planned economy. In line with this upward trend, the need for conceptual management of natural resources arose in the middle of the 1950s (Nesiba and Smolik, 2019). The first specific laws (Czechoslovakia Act No. 1/1955, Act. No. 11/1955, Act. No. 25/1955) regarding to nature and water protection were approved that time together with the declaration of the first National Nature Parks of Czechoslovakia (e.g., “National Park of High Tatras” proclaimed by the Slovak National Council in 1948). Subsequently, the National Assembly of the Republic of Czechoslovakia issued the State Nature Protection Act No. 40/1956 on August 1, 1956. Only nineteen paragraphs however dealt with the protection of nature and the water management of Czechoslovakia insufficiently. The law continuously introduced specific sanction mechanisms for the control. This first approach towards protection can be called as a “passive” legislation, with a very small strategic framework. Other national parks and a network of protected landscape areas were gradually created. Nature protection was subsequently issued in the Constitutional Act of the Czechoslovak Republic in 1960 where it was defined by the public property – primary forest fund, watercourses, and natural healing resources (Czechoslovakia Act. No. 100/1960). Several ecological disasters, such as the release of poisonous substances into water or air, are associated with the lack of control at that time. It became apparent from the 1960s that the laws and institutions of the 1950s were no longer sufficient to water protection. Simultaneously, new scientific findings were made on the ecosystems function (Carson, 1962). Even the United Nations began to establish nature protection departments. These ideas became popular political themes across national borders so-called “Iron Curtain.” Environmental problems that required a comprehensive approach have been gradually accepted as a topic of international meetings during the Cold War. After the first environmental conference in Stockholm in 1972, the strategic document “Action Plan” concerning to the environmental protection and defining the framework for the future joint action of the international community, including the Czechoslovak Socialist Republic, was adopted. The UN General Assembly followed up the Stockholm Conference by adopting several resolutions to

address among other things the absence of a more substantial institutional background. That is why the first-ever UN coordinating, consulting, and expert body dedicated to environmental protection, the UN Environment Program, was created. Water protection became a public issue across the international environment. The environment began to be so damaged by massive production that the public started to control political actions and approved laws in this regard. New scientific findings and models of development also served as a basis for the Stockholm Conference (Meadows *et al.*, 1972). In connection to this development, some associations were naturally engaged in nature conservation in Czechoslovakia (e.g., the “Slovak Union of Nature and Landscape Conservation” was established in 1969, and the “Czech Union for Nature Conservation” was established in 1979). In the 1970s and 1980s, the Government established majority of protected landscape areas and national parks (in function till nowadays). In 1978, the “Ecological Society” was established at the Academy of Sciences. The protection zones of drinking water were increasing together with the development of towns and their supply management. In the second half of the 1980s, water protection has become a society-wide standard with more explicit rules and sanctions of limited managerial instruments under the socialist regime. The Five-Year Plan of the Czechoslovak Socialist Republic in 1986 committed to expand and protect all water sources as well as to ensure water protection against pollution (Czechoslovakia Act. No 81/1986) were however insufficient. The actual strategic nature and water conservation planning did not exist. Therefore, it was necessary to pass completely new laws to protect water after the fall of the communist regime in 1989. The emergence of completely new laws after 1989 demonstrates a deep change in the water management style.

The phase of crisis management (1990-1992)

The period 1990-1992 asked for a big change in the approach to water protection. Proposition of new laws representing a complete change the way water protection is approached quickly emerged. Crisis management is defined as the identification of threats to an organization and its stakeholders and it requires decisions to be made within a short time frame (Bundy *et al.*, 2016). It should prevent the crisis itself

or respond to the first signals of the coming problems (Kouzmin, 2008). Another definition evaluates crisis management as a process of crisis identification and the enterprise’s subsequent stabilization (Pedersen *et al.*, 2020). Crisis management manifests in more practical steps rather than theory and it is therefore necessary to look for clear criteria of crisis and crisis management (Deverell, 2012). During the period of crisis, increasing communication, and one-way direction of orders to solve problems can be observed from top to bottom (Frandsen and Johansen, 2011). The crisis means that new and precisely targeted legislative acts are being issued. Czechoslovak legislative acts were not sufficient to protect nature and water which resulted in increasing volume of approved legislative acts. In order to protect nature as quickly as possible, there was a pressure to intensively adopt many laws in the shortest possible time. During 1990-1992, a political representation, a civic movement, and a specialized institution for nature protection were established. After an insufficient approach of the socialist government, pressure on water protection standards in Czechoslovakia emerged. Many laws, regulations, or restrictions had been created to replace the unsatisfactory socialist norms. Specialized environmental institutions were established, e.g. the Ministry of the Environment. From all legislative acts of that time, the “Nature and Landscape Protection Act” (Act No 114/1992) is the most significant document. It defined protected areas as well as sanction mechanisms. Its quality is demonstrated by the variations of this law till nowadays. During the whole measured period (1990-2019), the most legislative measures on average per year were carried out.

The phase of operational management (1993-2003)

Before the establishment of the independent Czech Republic (1.1.1993), it was directly settled into the principle of the Czech Constitution that this supreme law protects natural resources and their careful use (Czech Republic Act No. 1/1993). The Charter of Fundamental Rights and Freedoms in the Czech Republic states that everyone has the right to a favorable environment and the right for complete information on the state of the environment and natural resources (Czech Republic Act No. 2/1993). A new conceptual direction of nature and water protection began to emerge. This approach can be

called as operational management, generally capable of rapid change. Operational management is involved in coordinating and developing new processes while reevaluating current structures. Operational management enable a gradual transition from one management style to another, depending on the conditions, within a time frame of three to five years (Ansoff *et al.*, 2019). This situation occurred with the Czech Republic's accession negotiations to the EU. Due to the adopted legislative acts, Czech legislative acts moved to a harmonization stage with European law. Therefore, this transition can be observed as operational management. In the second half of the 1990s, there were two political tendencies in the Czech Republic concerning to water protection. On the one hand, privatization of waterworks took place, on the other hand, the adoption of European laws happened. Mass privatization of Czech municipal (regional, state) waterworks took the effective control instrument away. Public political power lost the opportunity to effectively manage waterworks in large part of Czech cities (Nesiba and Smolík, 2018). This situation was overwhelmed by the Czech future membership in the EU. The politicians focused on operational management and Czech and European law needed to be operationally harmonized. Therefore, at the end of the 1990', the gradual implementation of operative management in the whole system of legislative standards could be observed. Operational management manifested itself in several European operational regulations that were growing. The effort to unify with European water protection law was also based on the experts analyzes of the time (McAdam *et al.*, 2011). Several legislative acts concerning to the EU requirements (Council Directive 98/83/EC, 2000/60/ES, Czech Republic Act No 258/2000) have been approved. It was a parallel process together with harmonization of European legislative standards. The obligation to change basic national standards for wastewater management upon accession to the EU was a typical example that required changes in principal legislative Acts in the country (Council Directive 91/271/EEC, Czech Republic Act No. 185/2001, Act No. 254/2001, Act No. 274/2001). These laws were followed by several changes that clarified the paragraph wording and responded to the needs of water protection (especially with the development of new technologies and industrial risks) (Czech Republic Act No. 20/2004).

Among other dozens of standards concerning to water protection, it is necessary to mention the Act introducing protection into the Czech legislation within the framework of the Natura 2000 system (Czech Republic Act No. 218/2004). Generally, the Czech Republic has committed itself to complete the harmonization of water protection into EU legislation as a condition for Czech accession to the EU which also concerned the unification of water protection conditions, namely Annex V, Sections 7b-7c.

The emergence of strategic management (2004-2019)

Strategic management relates to long-term planning. An important factor in strategic management is the precise setting of goals and timeframe. Strategic planning is considered as the cornerstone of any organization, and the institution's strategy also forms the entire structure of the organization (Menguc *et al.*, 2010). It is common for all strategic management theories to define objectives and goals and to determine how to achieve them (Gleeson, 2019). Empirical studies confirm that strategic management is the most comprehensive management tool in SME enterprises (Zeemering, 2018), corporations (Monday *et al.*, 2015) as well as public sector (Joyce, 2015). In the case of strategic management, specific and measurable goals are set because they can be compared with the actual situation after the specified time has elapsed (Ansoff *et al.*, 2019). The stability of the main legislative documents in terms of a larger space for their regulations and interpretations is one of the criteria (Menguc *et al.*, 2010). This approach is called as sustainable and responsible management (Bryson and George, 2020). After the Czech Republic accessed to the EU in 2004, water protection has been gradually becoming a public topic. It is also a phase of decline of legislative acts and the growth of strategic plans. The number of legal standards has gradually increased (Directive 2006/11/ES Directive 2006/118/EC). Compared to previous periods, a more significant number of legislative measures concerning to the strategic interests of the Czech Republic were approved. Discussed topics of environmental security, food security or strategic national security relate to the protection of water resources and water systems. The surveyed period (2004-2019) is characterized not only by law-making but mainly by national government decrees, public regulations, and strategic plans (Palát

et al., 2010). Several impulses have contributed to the policy of strategic decision-making for water protection management in the Czech Republic as an EU member from 2004. The European Commission sets goals and plans for next five years for each policy sector. Moreover, a multiannual financial framework of the EU (so-called seven-year budget) was set for periods 2000-2006, 2007-2013, 2014-2020. So far, in each programming period, one line was devoted to water protection issues. The second impulse came from scientific research published to investigate the global impact of climate change and the rise of green policy; e.g. "Stern Report on Global Warming on the Impact of Climate Change on the World Economy" and "Fourth Assessment Report of the Intergovernmental Panel on Climate Change" received considerable public attention. For this purpose, many Czech expert platforms have been created to inform the general public about ongoing measurements. The third impulse can be seen in the framework of the United Nations global policy. The UN documents such as the "Paris Climate Change Agreement" of 2015 and the subsequent commitment approved by the UN General Assembly entitled "Transforming our world: the 2030 Agenda for Sustainable Development" are among the most critical decisions. At the heart of this document, the indicators embodied in seventeen "Sustainable Development Goals" are calling for strategic planning of water protection. The number of laws is declining but the number of government regulations, public regulations, and strategic plans is increasing. Strategic plans are the sub-legal norm as an effective instrument for private and public institutions. The goal-setting theory proposes that organizations with set goals perform better as their activities and resources are focused on addressing core issues and employees can understand the firm priorities (Jung and Lee 2013; Locke and Latham, 2002). Studies confirm managerial strategic planning as the most effective instruments (George *et al.*, 2019). Based on the UN decision, the Czech Republic adopted a strategic framework for strategic planning until 2030 (Government Resolution No.292/2017). It had been followed by a long-term preparation process (consultations between political representatives, experts, and the general public) that resulted in the "Implementation Plan of the Czech Republic Strategic Framework 2030" (Government Resolution No.669/2018) and "Implementation the 2030

Agenda for Sustainable Development" (Government Resolution No.670/2018). The "Agenda 2030" was therefore adopted in the Czech Republic. Currently, the Czech Government has approved several types of following strategic plans:

- umbrella strategy papers (12),
- strategic plans and concepts (185),
- implementing strategy papers (82),
- following strategy papers (5)

The number of the strategic documents is increasing as observed by the State Program of Nature Conservation and Landscape Protection of the Czech Republic (2009), Waste Management Plan 2015-2024 (2015), Environmental Security Concept 2016-2020, with a view to 2030 (2016), Czech Republic Action Plan for the Development of Organic Farming 2016-2022 (2016), Concept for the protection against drought consequences for the territory of the Czech Republic (2017). Since 2019, water protection has affected 24 strategic plans, although many of them collide with strategic plans. For the last years, the construction of the Danube-Oder-Labe canal is being prepared in the Czech Republic. It is a case where strategic water protection documents and transport strategies directly contradict each other (Palát *et al.*, 2010). A new type of management is needed to set priorities after 2020.

Situation after 2020

The situation in this area is changing rapidly due to the trends in social and political development in the Czech Republic. Global climate change, temperature, drought, soil quality and ecological problems affect water management in the Czech Republic (Kavan *et al.*, 2021). Restrictions resulting from the pandemic Covid-19 have only frozen this situation for a while, but development trends start to show the need of the Czech government to move to legal action again (towards crisis management). This situation can be documented not only by political decisions. From the regional level, there is a tendency to buy out privatized waterworks back from private sector (municipality, regions, e.g. statutory city Plzeň in 2018, Liberec in 2019, etc.). A number of parliamentary political parties in the Czech Republic are supporting the possibility of amending the entire Constitution of the Czech Republic where water protection rights

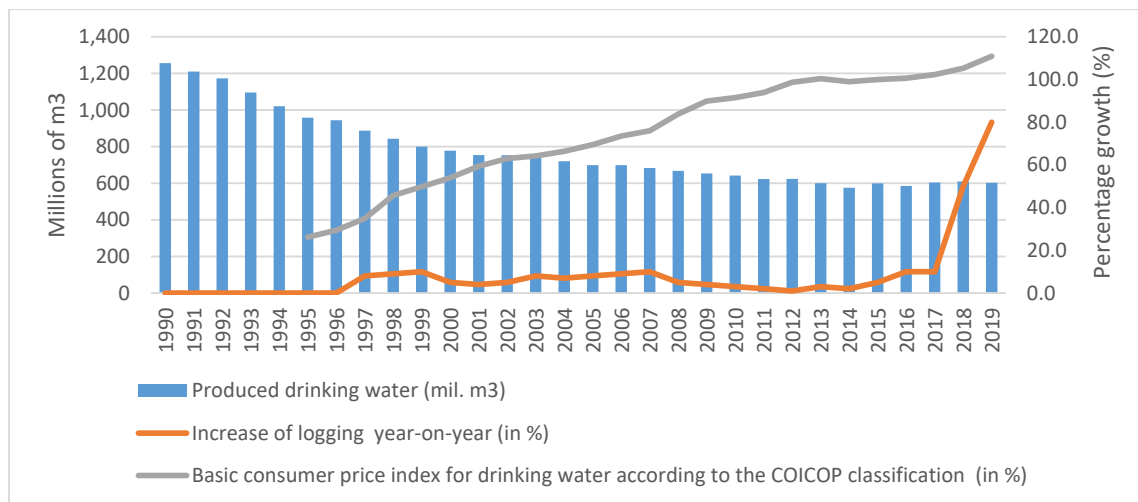


Fig 1. Proportion of drinking water production, prices index and logging

would be defined more strictly. Public regulations and governmental decree are no longer enough, in fact, many of these lower legislative acts are stopped by the Czech Constitutional Court (Judgment of the Czech Constitutional Court 261/2018, 2018). A tendency towards new legislative is accelerated by the current issues in the Czech Republic, namely crisis in forestry called a “bark beetle calamity”. The entire Czech Republic must preventively cut down the forest fund against the danger of “bark beetles”. The area of forests in the Czech Republic therefore decreases dramatically since 2019. This pushes the cost of water management up because natural cleaning is no longer sufficient (Křeček *et al.*, 2021). As demonstrated below (Fig. 1), the share of drinking water decreases but the price increases (due to the consumer price index according to the specific COICOP methodology). Simultaneously, production of drinking water is decreasing due to lower economical demand.

Developments are moving forward political solutions through new and stricter laws that address the cost and scarcity of water. More than the issue of wastewater treatment itself, the attention is shifting towards the protection of the forest fund and nature overall.

MATERIALS AND METHODS

Survey design and data collection

The research is set to analyze the legislative measures adopted between 1990-2019. These

analyzed legislative acts include:

- Laws passed by the Czech Parliament
- A government decree issued by the Czech Government (according to article 78 of Czech Constitution)
- Public regulations issued by the Ministry (according to article 79 of Czech Constitution)
- Governmental resolutions
- Czech National Strategic (Action) plans

The presented study of the legislative measures related to water protection was carried out based on online public sources (collection of laws of Czech Parliament, collection of government resolution and decrees from official websites of Government of Czech Republic, Collection of Public regulation from non-profit organizations’ web pages). Firstly, this study gathered all relevant administrative laws dealing with water protection in the Czech Republic. The time frame of 1990-2019 was divided into three stages based on significant political events in past that influenced the legislative action specified for this framework, e.g., period 1990-1992 (after the end of the socialist regime and the current Czechoslovak state), period 1993-2004 (establishment of independent Czech Republic) and the period 2004-2019 (after accession to the EU). The research focused on two sub-questions (SQ):

SQ1: How does the total number of legislative measures in the Czech Republic in each period change?

SQ2: How do the ratios among laws, government decrees, and public regulations at the administrative, and legislative levels change?

The hypotheses are set as:

H1: When the number of laws in the given period increases, it refers to crisis management principles.

H2: When the number of laws decreases, it refers to the principles of strategic management as the number of government decrees, public regulations, and strategic plans (with a clearly defined time frame) increase.

The presented study does not examine water quality (such as contamination or protection methods). The survey is methodologically incorporated with Law and Management Approach that explores legislative opportunities and managerial strategy. Societies can optimize their management legally and develop strategies that give them a competitive advantage (Roquilly, 2009).

RESULTS AND DISCUSSION

Overall, all types of legislative acts can be evaluated on the resulting figure that is compiled from 1948 to 2019. Fig. 2 illustrates the gradual change of the legislative acts.

Apart from these absolute numbers, a different comparison can be observed by a look at the relative numbers, i.e. with respect to the yearly averages. Following Fig. 3 demonstrates the gradually increased number of legislative measures. The statistic sum represents the total number of all legislative measures

(laws, government decrees, public restrictions) and the year average, i.e., how many water protection standards in the Czech Republic were approved in relative numbers.

To answer the Q1, the number of legislative acts was increasing gradually on yearly average. In the first examined period 1990-1992, the average is three legislative acts per year. In the second period 1993-2003, 3.4 legislative acts are calculated as the average per year. In the last period 2004-2019, the average increased to 4.7. It can be stated that the adoption of legislative acts in 2004-2019 was mean the most often. Different situation occurs if having a look on type and character of legal norms (Dikovitsky and Shishaev, 2019). Administrative laws are issued as the highest regulatory conduct of the state. The number of laws in one area indicates the urgency of solving an issue and it can be deduced that the area shows the need for a crisis management approach. Therefore, in a longitudinal comparison, if more laws are issued for a specific sector in a certain period, it represents an attitude of crisis management. In the case of ambiguity of specific laws interpretation, insufficient law, or preparation of new laws, government decrees are issued. That is a situation of an operational solution (e.g., harmonization of the interconnection of several management sectors). If the number of laws decreases but the number of Government Decrees increases, it is a situation of operational management (McAdam *et al.*, 2011). To continue, if the number of government regulations in a given

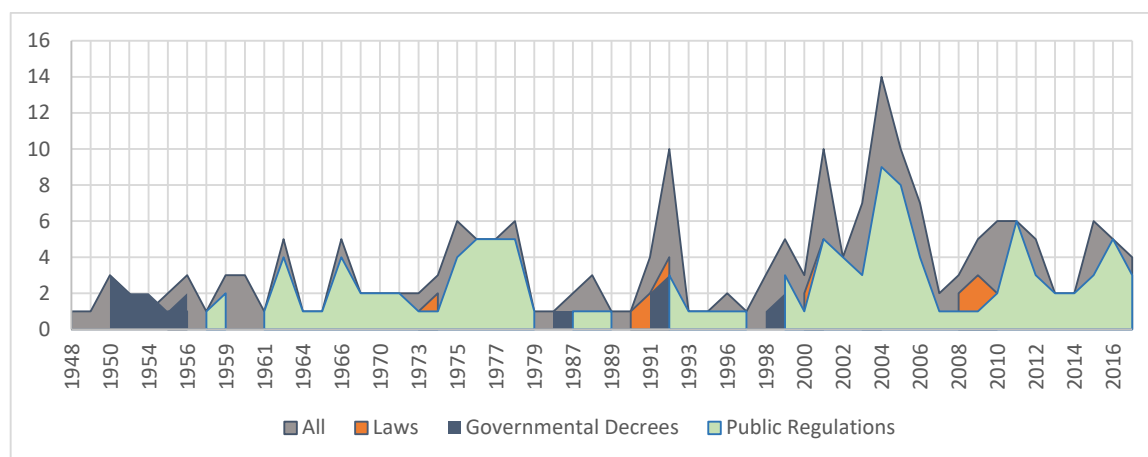


Fig. 2. Legislative acts about water protection in Czech Republic 1948-2019

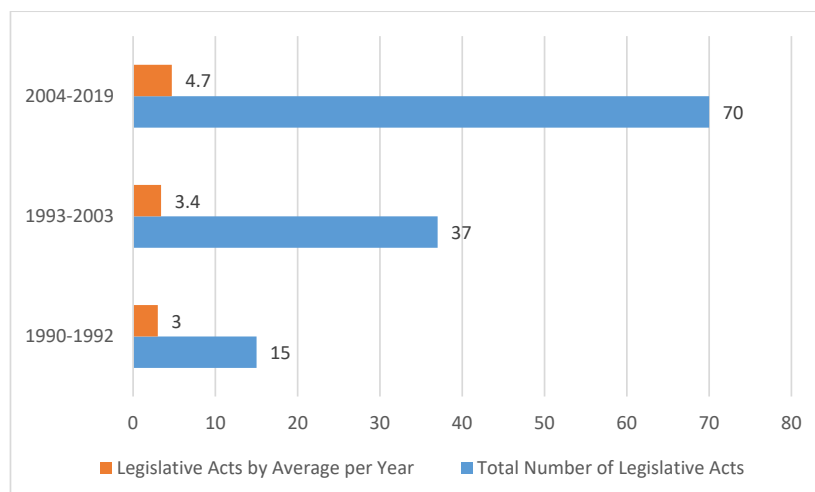


Fig. 3. Increasing number of all legislative acts (laws, government decree, public regulations)

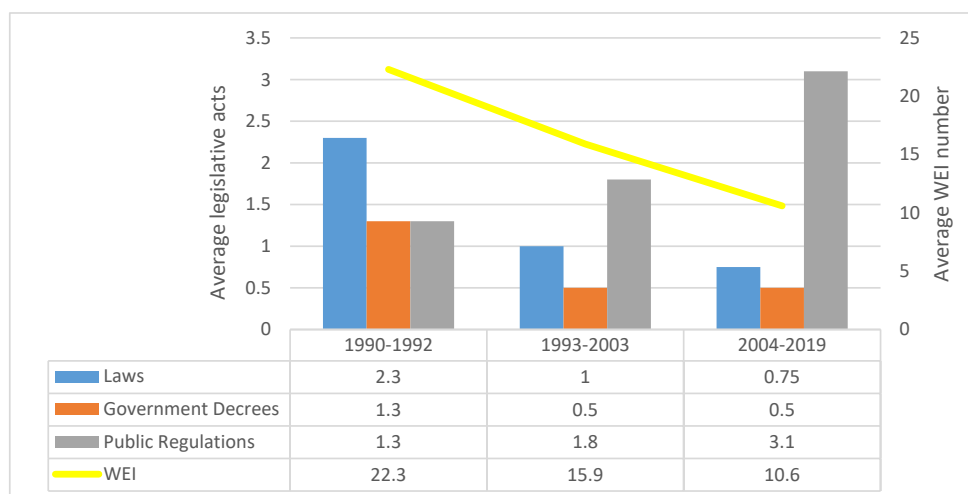


Fig. 4: Changes in legislative acts (average per year) and Water Exploitation Index (WEI)

area decreases but the number of Public Regulations of individual ministries regulations increases, this is another level of management. If the number of national strategic documents grows in this situation, strategic management is demonstrated. The type of legal norms varies according to the political attitude to water protection. It shows the result of the proper legislative acts diversification among other acts in every period and their comparison. The change in the legislative approach is confirmed by the WEI which has been measured by European institutions

since 1990. Decreasing WEI indicators identify more responsible and strategic approach to water use in the Czech Republic and correspond to presented average-per-year number of legislative acts (Fig. 4).

Laws

The findings show the most significant number of laws in the period 1990-1992. The lowest number of other legislative acts (decrees, regulations) was passed. Contrary, in the years 2004-2019, the number of laws decreased, and the number of implementing

regulations, government decrees, and public regulations increased. The ratio of laws to regulations and decrees was high in period of crisis management. The function of law is more valid than in terms of decrees or regulations (Schmidt and Matthews, 2017). There is no need to implement the law accurately and adapt it to the changing conditions in a crisis. In the period 1993-2003, the number of laws is declining. These laws were passed in line with EU interests as Czech laws were operationally adapted to European norms. This attitude towards water protection can be called as operational management. In the period 2004-2019, the situation was changing and laws were not issued so often. However, the regulations and decrees gained importance as part of strategic (action) plans.

Government decrees

The government decrees were 1.3 at the yearly average from 1990 till 1992. From 1993 till 2003, the average was reduced to 0.5. This average volume remained the same in the following period 2004–2019. As defined above, this trend illustrates the operational management. Czech legislative acts were implemented with the interest of harmonization with the European law upon the Czech Republic's accession to the EU.

Public regulations

The average number of public regulations in the monitored area proliferated due to both laws and governance decrees. Between 1990 and 1992, the average was 1.3. It gradually increased to 1.8 in the next monitored period 1993-2003 and the average reached even 3.1 in the last period after 2004. That indicates a change in the approach to water protection towards the operational management. However, combined with the fact that strategic national plans are growing in the same period, the role of strategic management should be considered. It was confirmed that the total sum of legislative acts for water protection in the Czech Republic has been increasing from 1990 till 2019. The research positively answered the research question (Q1), whether other types of management for water protection can be found in the statistical evaluation of legislative acts on bird protection. Different management methods can be indicated in the three monitored periods - crisis, operational and strategic management approach. To answer how the number of legislative acts has changed (SQ1);

crisis management during 1990-1992 focused on the most potent policy instrument, namely the passage of laws made by Czech (Czechoslovakia) Parliament. Crisis management manifested itself in an attempt to politically change the overall public attitude to water protection in Czechoslovakia. The governmental laws were the common instrument (Roquilly, 2009). On the other side, the least sub-legal standards (Government Decrees, Public Regulations) were used (and any water protection strategy was not formulated at the same time at all). In the following period 1993-2003, the number of governmental decrees and public regulations increased due to the implementation of Czech laws. These were adopted due to the accession negotiations with the EU. Government regulations and public ordinances clarified the new laws to be harmonized with EU law. In the last period 2004-2019, the number of laws was decreasing but the number of strategies and strategic plans was simultaneously growing. It was also shown (SQ2) that in the monitored time, the ratio between individual legislative norms changed, and the number of implemented regulations (government decrees, public regulations) to the already existing laws increased. Moreover, the number of Czech strategic plans has proved to be presented. The research findings demonstrate the development of approved legislative acts. The relationship between laws, regulations and decrees can refer to three types of management - crisis, operational and strategic. These types of management can be assigned to the three stages of legislative development in the Czech Republic by a qualitative evaluation. Crisis management was characterized by the accelerated passing of laws in 1990-1992. Operational management was characterized by an effort to harmonize Czech and European standards in the years 1993-2003. That created an emphasis on the operational alignment of Czech laws with European legislation and the number of government decrees and public regulations increased. In the years 2004-2019, the number of laws decreased but contrary, the number of strategic plans as the basis of strategic management increased. During this period, there were tendencies towards strategic management of water protection in the Czech Republic. After 2020, there is a new period of legislative development in drinking water protection. The Czech Republic is a country with specific location in Europe. No river from foreign territory flows into the Czech Republic. Therefore, water protection is related to the overall protection of

nature (Kavan *et al.*, 2021; Křešek *et al.*, 2021). Forests in the Czech Republic are now being deforested due to the “bark beetle calamity”. Therefore, water management in the future should take into account the overall concept of nature and forest protection. The closest characteristic in the field of management is the so-called integral management that combines strategic plans with the creation of specific laws. The incorporation of the protection of drinking water into the Constitution of the Czech Republic is the first and currently happening step.

CONCLUSION

The types, quantities and mutual ratio of legislative acts dealing with water protection are changing in the Czech Republic. The presented analysis examined and evaluated 12,272 legislative acts. The changes in legislative acts correspond with different attitudes towards water protection and water management. As quantitative research standards in the Czech legislation has shown that there have been changes corresponding to three types of management (1990-1992, 1993-2003, 2004-2019). These legislative changes reflect the development of society and correspond to the overall social changes in the approach to nature protection. From a quantitative point of view, these approaches to nature protection can be linked to a qualitative approach where three different stages of water protection in the Czech Republic - crisis, operational and strategic management can be named. In conclusion, the recommendations for future development are delivered. A new type of integrated management should be implemented in water protection decision-making. This approach will link both strategic documents (so they do not contradict to each other), as well as these documents will be associated with the creation of specific laws. Public regulations and governmental decrees will serve as channels for the transfer of information between the strategy and the legislature.

AUTHOR CONTRIBUTIONS

J. Nesiba prepared the research design, analyzed and interpreted the data. R. Cuhlova performed the literature review and prepared the manuscript text.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

<i>COICOP</i>	Classification of Individual Consumption by Purpose
<i>e.g.</i>	For example
<i>et al.</i>	And others
<i>EU</i>	European Union
<i>Fig.</i>	Figure
<i>i.e.</i>	That is
<i>SQ</i>	sub-question
<i>SQ1</i>	Sub-question 1
<i>SQ2</i>	Sub-question 2
<i>Q1</i>	Question 1
<i>UN</i>	United Nations
<i>WEI</i>	Water Exploitation Index

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CASE STUDY

Resident-based learning model for sustainable resident participation in municipal solid waste management program

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ABSTRACT

BACKGROUND AND OBJECTIVES: Encouraging resident participation in the Municipal Solid Waste Management system still becomes a challenge for cities in developing countries. Previous studies showed that existing education strategies ineffective and insufficient to change resident behavior sustainably. Therefore, this study aimed to develop a resident-based learning model to encourage sustainable resident participation in waste management programs at the household level using the Community of Practice approach.

METHODS: This study was a conceptual model study using a case study as the research strategy. The case being analyzed was *Kawasan Bebas Sampah* (Zero Waste Area) program implemented in Bandung City, Indonesia. The primary data was collected through field observation on the 8 *Kawasan bebas sampah* and in-depth interviews with 63 key informants comprising 31 key actors in the educational programs and 32 informants as resident representatives, conducted from January 2020 – November 2020. Additional data from the quantitative method was gathered in April 2021, focusing on surveying resident's habits toward waste management as evidence of the program result.

FINDINGS: The findings showed that the education strategy implemented in Bandung City, Indonesia presented effective results, based on waste reduction rate reaching up to 0, 47% at the city level from 2019 to 2020. The survey provided evidence that the education program has succeeded in forming new habits for residents. Averagely 60% of respondents in each sample area have the habits and up to > 90% for the areas with more durable education program. Some critical points for education strategy implementations are identified. First, collaboration and supports from key stakeholders such as facilitators, local leaders, and educators become the enabler of the program. Second, key stakeholders need to identify knowledge and value needs before the program started. Third, the learning activities are conditioned to facilitate practice-based and dialogue-based learning through group and non-group learning activities. Fourth, local cadres are vital to sustaining the education program.

CONCLUSION: This study has succeeded in giving a new strategic approach to improve resident participation in municipal solid waste management. The resident-based learning model proposed in this study offered a more effective strategy for other cities in developing countries to improve the sustainable participation of residents in their waste management system. However, some adjustments may be required for residents with different characteristics. Future studies may focus on testing and refining the model to improve its applicability.

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INTRODUCTION

Household waste is to be the most dominant component of Municipal Solid Waste (MSW) in many developing countries (Esmailizadeh et al., 2020; Jouhara et al., 2017; Speier et al., 2018;), which reached up to 55 – 85 % of MSW composition (Banerjee and Sarkhel, 2019). It indicated the crucial role of resident participation in the MSWM system (Kamaruddin et al., 2017). Sustainable waste management at the household level remains a challenge due to high reliance on resident behavior (Jiang et al., 2021; Moh and Manaf, 2017; Mukama et al., 2016;), which has become the core problem in many cities in developing countries (Azevedo et al., 2021; Banerjee and Sarkhel, 2019; Gundupalli et al., 2017). Previous studies showed that education becomes one of the most critical factors to change resident behavior (Meng et al., 2019; Liu et al., 2019). However, existing education for residents in developing countries was considered ineffective and insufficient (Moh and Manaf, 2017; Wang et al., 2018). Education cannot directly influence resident behavior toward waste management (Wang et al., 2018). It is mediated by personal factor such as awareness (Chen and Gao, 2020; Lissah et al., 2021; Pierini et al., 2021), attitude (Lissah et al., 2021; Liu et al., 2019; Wang et al., 2018), and moral norms (Wang et al., 2018) to effectively nurture resident willingness to participate and change their behavior (Liu et al., 2019; Meng et al., 2019; Wang et al., 2018). It is expected that the waste management behavior finally becomes their habit (Liao et al., 2018; Moh and Manaf, 2017; Xu et al., 2017). Education strategy should be improved to be more effective and impactful to promote personal factors (Chen and Gao, 2020; Pierini et al., 2021; Wang et al., 2018; Zheng et al., 2020). This situation raises a question, what education strategy is more effective and impactful to nurture sustainable change on resident waste management behavior? Some studies offered several approaches for resident-based education improvements. Wang et al. (2018) recommended more informative and frequently shared information campaigns using posters and social media to reach more broadened communities, but Zheng (2020) found that media publicity has an insignificant impact and extensive publicity is not the solution. As indicated by Jiang et al. (2021), publicity was to be a communication channel to strengthen willingness.

So, it may become a supporting system, instead of being the primary education approach. Pierini et al. (2021) proposed a relatively new strategy for resident-based waste management education through the citizen science concept while Pei (2019) and Zheng et al. (2020) advocated neighborhood ties, community attachment, and social connections, which highly rely on community empowerment for effective education. Chikowore (2020) added active community involvement as a behavioral change driver for sustainable waste management practice. It is expected that education can build new habits (Liao et al., 2018; Xu et al., 2017). In Knowledge Management (KM), intensive interaction and community engagement for educational purposes can be facilitated by a Community of Practice (CoP). The CoP concept was firstly introduced by Wenger et al. (2002), who contended that CoP becomes the source of learning and knowledge sharing for a particular community. A CoP is underpinned by the notion of social learning theory emphasizing informal learning groups with intensive interaction to deal with shared problems of the members (Denscombe, 2008; Stein, 2005). CoP is developed based on knowledge domain, comprising community identity determined by shared problems among members (Dessne and Byström, 2015; Madsen and Noe, 2012) and the community objectives (Middlemiss and Parrish, 2010; Reed et al., 2014). So, CoP can be implemented in various domain, including waste management. Both shared problems and community objectives influence knowledge and value being shared, improved, and maintained by the communities (Li et al., 2009; Madsen and Noe, 2012). The main goal of the learning activity is to prevent knowledge gap among members, so that the shared knowledge should be transferred to all the members and improve their practice (Li et al., 2009). CoP enables social learning in two forms: collective learning and individual learning (Madsen and Noe, 2012; Tran et al., 2018). The learning group in CoP can be formed naturally or intentionally to reach specific group objectives (Wenger et al., 2002). As the learning interaction is conducted intensively (Stein, 2005), the members become experts on their problem (Ghazali et al., 2017). In the learning process, personal elements play a vital role, especially the leaders. Wenger et al. (2002) and Reed et al. (2014) indicated collaborative leadership for CoP success, meaning that the leadership should be

distributed to the whole community with different roles. Some roles are such as community coordinators, boundary spanners, thought leaders, knowledge experts, administrators, pioneers, and others (Wenger *et al.*, 2002). Community coordinators and thought leaders are central for successful CoP (Wenger *et al.*, 2002). Managerial element is related to regulations and norms required to encourage participation in the learning process (Abou-Setta, 2015). Participation level can vary from being the core group, active members, and legitimate peripheral participants (Wenger *et al.*, 2002). Other support systems are technology and infrastructure, required to facilitate more intensive knowledge sharing and practice (Li *et al.*, 2009; Middlemiss and Parrish, 2010; Stein, 2005). The previous studies about CoP implementation showed that the CoP framework is implementable for resident-based learning activities. CoP has been implemented in farming household communities in Vietnam (Tran *et al.*, 2018), an agricultural community in Denmark (Madsen and Noe, 2012), and resident-based waste management education program in Indonesia (Sunarti *et al.*, 2020). Tran *et al.* (2018) found that social learning facilitated by bonding and bridging relationship provided by CoP approach contributed significant effect to households' learning system. The study conducted by Madsen and Noe, (2012) showed that knowledge creation and knowledge implementation in the learning activities are important for CoP success. On the other hand, acquiring the relevant knowledge is vital to nurture the expected personal factors in waste management (Janmaimool and Denpaiboon, 2016) and implementing the waste management in the daily basis also crucial to nurture waste management habits (Xu *et al.*, 2017). Thus, applying CoP as the baseline concept of resident-based education can be an effective and sufficient way to nurture residents' personal factors and improve their behavior. Besides, resident-based education using CoP facilitates neighborhood engagement (Pei, 2019), social connections among community members (Zheng *et al.*, 2020), active community involvement (Chikowore, 2020) and direct practice as a part of learning system (Madsen and Noe, 2012). Thus, this study was aimed to investigate resident-based education program which implement CoP platforms. Based on study finding of Sunarti *et al.* (2020), A resident-based education program implemented in Bandung City,

West Java, Indonesia, *Kawasan Bebas Sampah* (KBS/ Zero Waste Area), can be considered as CoP. However, this study investigated only one KBS location while there were 8 KBSs available. Thus, this study aimed to complete the previous study by investigating more broadened KBS areas to get comprehensive understanding about its education strategy. The second objective of this study was to investigate the effects of the education strategy for the resident waste management habits. Finally, the third objective was to develop resident-based education model to allow other cities in developing countries implementing similar strategy to improve household participation in the waste management program. The primary contribution of this study is to give more detailed picture of resident-based education strategy using CoP approach to improve resident participation in waste management effectively. The study was conducted in Bandung City, West Java, Indonesia from January - November 2020. Additional data from quantitative method was gathered in April 2021.

MATERIALS AND METHODS

Study area description

Bandung City is one of the cities in Indonesia that has a concern to handle MSW problems through resident participation encouragement. The local government of Bandung city has launched a program called 'KBS' (*Kawasan Bebas Sampah*/Zero Waste City) aimed to campaign 3R (Reduce, Reuse and Recycle) at *Kelurahan* (sub-district) level (Sunarti *et al.*, 2020). The KBS program was the follow up of Bandung City Local Regulation No.9/2018 about Waste Management, where the resident must separate their organic waste at home. The KBS program was handled by DLHK (*Dinas Lingkungan Hidup dan Kebersihan*/ Service Office of Environment and Cleanliness) in collaboration with a local-owned enterprise, PD. Kebersihan. The DLHK placed their officers in the KBS areas who were responsible to educate the residents and other stakeholders, supervise the waste management activity, handle the organic waste recycling process at the communal level and organize separated waste transfer from the communal to the city level. PD. Kebersihan was responsible to pick up the separated waste from the area and handle the waste management system at the city level. The location of the study is presented in Fig. 1.



Fig. 1: Geographic location of the study area in Bandung City, West Java, Indonesia

The KBS program was initiated in 2015 by local NGOs at the 'RW' (*Rukun Warga*/community association) level. There were started from one RW and then expanded after a year. In 2019, the program was officially started as government's program and reaching more expansive areas (*kelurahan* level). It was chosen 8 *Kelurahans* to be KBS models named KBS Sukaluyu, KBS Cihaurgeulis, KBS Neglasari, KBS Kebon Pisang, KBS Babakan Sari, KBS Kujangsari, and KBS Gempolsari. There was no exact data about how many households living in the KBS areas. Public statistics mostly provided number of group family. One household sometimes contains 2 – 4 families with only one waste disposal system. Each KBS commonly consists of 1,000 – 9,500 families. In total, the family groups living in all KBS areas were approximately 34,877 families. Assumed that each household contain 3 families, the total households were about 11, 625 households, spreading into 8 – 12 RWs/KBS. The whole residents in KBS areas were supposed to be exposed by the education program. Due to limited time, personal resources, and local leader problems, some RWs within KBSs did not get proper education. Before the program was started, all chosen areas implemented Collect – Transfer – Dispose system, similar with the municipal waste

management system implemented in other cities in Indonesia. With the new system, the program's primary goal is to encourage residents' involvement, especially on waste separation. If the chosen areas have spaces, the organic recycling centers are built within the area to enable the waste collector recycling the organic waste. The residents were also allowed to utilize the recycling tools if they want to. If there is no space available, the separated waste will be transferred to a recycling center at the city level. The recycling center was built within an RW/several RWs, so waste collectors quickly put the separated waste into recycling tools. The government provided a transfer system to put the separated waste from the KBS to the recycling center at the city level. The waste management system in KBS areas is presented in Fig. 2.

Study design and data collection

The study's unit of analysis was the KBS program for residential in Bandung City, which comprised 8 KBSs areas. This study was a conceptual model study using case study as research strategy. There were employed multiple methods to gather the data for better understanding toward cases (Yin, 2014). This study's main method was qualitative, conducted from

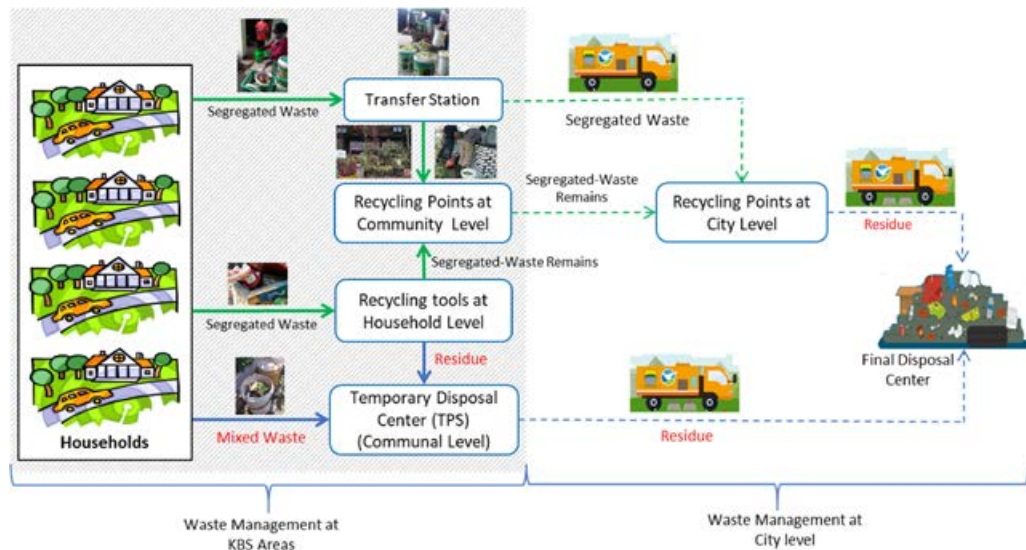


Fig. 2: Waste Management system in KBS areas

Table 1: Key Informants of the study

No	Data Collection	Position	Number of Informants for every KBS								Number of Participants
			Suka luyu	Cihaur geulis	Negla sari	Kebon Pisang	Suka miskin	Babakan Sari	Kujang Sari	Gempol Sari	
1	Interviews	Mentor	1	1	-	1	1	1	1	1	63
2		Supervisor	2	2	2	2	2	1	1	1	
3		Local leaders	2	1	3	1	-	1	2	1	
4		Residents	3	3	3	3	3	3	3	3	
5	Survey	Residents	40	103	71	60	89	70	27	30	490

January 2020 – November 2020. The quantitative data collection was gathered afterwards in April 2021 to get evidence about the effect of KBS program. It used abductive approach, as the data collection was applied to examine a phenomenon, recognize patterns and themes, locate the finding into a conceptual framework to modify existing theory which was subsequently tested through additional data collection (Saunders, 2019). The primary techniques to gather the data were in-depth interviews and field observation. It was added a survey about habits about waste management as education strategy evidence. For qualitative data analysis, the content

analyzed and synthesized was from the verbatim version of the interviews and field observation. The data was then triangulated and synthesized to build the resident-based education model. Therefore, this study can be considered as a conceptual model study (Jaakkola, 2020) using a directed Qualitative Content Analysis (QCA) approach (Assarroudi *et al.*, 2018). Data triangulation was conducted to improve the internal validity of the qualitative research. There were five types of data triangulation for internal validity in qualitative research: data source, method, researcher, theory, and data type triangulation (Miles *et al.*, 2014). For this study, the triangulation was based on the data

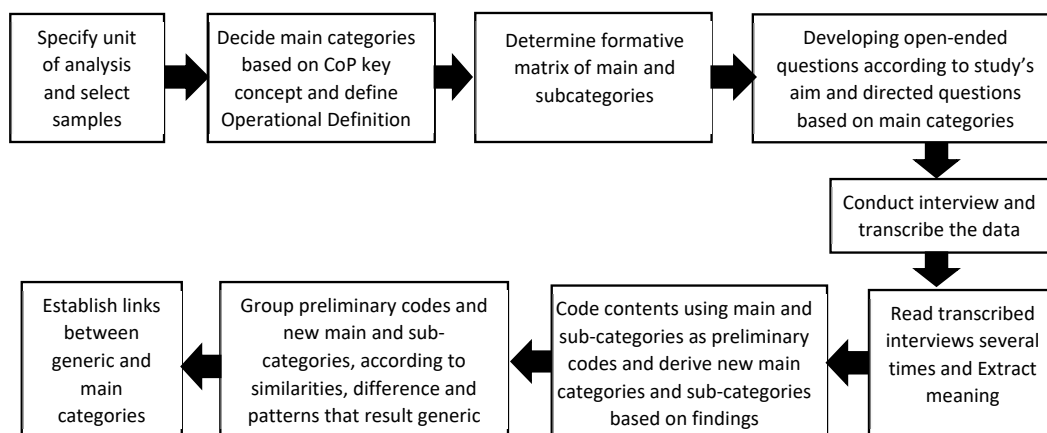


Fig. 3: Qualitative data analysis process using Directed QCA (Assarroudi et al., 2018; Miles et al., 2014)

source and method variation. The data source for qualitative method was from different persons with various roles, interviewed at different times and from different places. The data collection methods were from in-depth interviews, observation, and documents. For qualitative data, it was chosen 63 key informants consisting of 31 key actors in the educational programs and 32 informants as resident representatives. The sample selection technique was purposive sampling for the key actors in the educational programs and random sampling for the resident representatives. The informant selection from the key actors was based on the following criteria: 1) involve actively in the KBS activity for a considerable time, 2) have an essential role in the KBS activity, formally or informally. Therefore, the key actors from each KBS can be different because of the selection criteria consideration. The data collection was ended when the data gathered has been saturated. The survey respondents involved 490 residents as representatives of each KBS area which got sufficient educational exposure. The respondents were chosen using purposive-random sampling in which it was chosen residents who have been exposed by the education program. The key informants of the study are presented in Table 1.

Data analysis

According to Jaakkola (2020), a conceptual model study develops a model by identifying the existing elements or variables and then establishing the causal linkages of the elements. Main data was from qualitative method while quantitative data became

the evidence to support the qualitative findings. The main elements were identified based on generic categories resulted from the qualitative data analysis process. CoP main concepts become the preliminary codes as the initial categories/sub-categories. The data analysis process was started as the interview recordings were transcribed to get the verbatim version. The Directed QCA (Qualitative Content Analysis) approach was applied to code the verbatim data using A Computer-Assisted qualitative data software (CAQDAS) named NVIVO 12. While Miles et al. (2014) provided six classic steps of qualitative data analysis, Assarroudi et al. (2018) introduced 16 steps of directed QCA comprising three main phases: the preparation phase, the organization phase, and reporting phase to increase the trustworthiness of directed QCA approach. In this study, it is combined the two versions by grouping some steps altogether. The learning model in this study was developed by determining the input, process, and output elements leading to resident participation improvement in the MSWM program based on the findings. The whole process of research methodology was presented in Fig. 3.

RESULTS AND DISCUSSION

Personal elements and their roles

According to the interviews and field observation, stakeholders who have crucial roles in the KBS program were identified. All stakeholders involved and their roles in the KBS program are shown in Table 2.

There were 8 KBSs chosen based on their

Table 2: KBS Program stakeholders

Organization	Stakeholders	Roles in KBS Program	Status of the Roles
Government Party	DLHK	administrator, knowledge sharing and infrastructure facilitator	mandatory
	PD. Kebersihan mentors	waste management operator at city level official thought leader, knowledge expert supervising and monitoring the waste management activities, boundary spanner, knowledge expert	supporting function mandatory
	supervisors		mandatory
Structural local leaders under <i>Kelurahan</i>	(Lurah/KASIE EKBANG)	field representative of <i>Kelurahan</i> , facilitator, community organizer,	supporting function mandatory
	RW/RT Chiefs	facilitator, community organizer, pioneer	encouraged
	PKK members	community organizer, local cadre	voluntary
	<i>karang taruna</i>	voluntary educator, local cadre	voluntary
Local Residents	residents	Waste management operator at household level	encouraged
	waste Collector	Waste management operator at communal level	mandatory
NGOs	e.g. YPBB, GSSI	External (boundary spanner)	voluntary
Outsider	environmental communities,	temporary knowledge experts	voluntary (only found in KBS Kebon Pisang)
	visiting Students	voluntary educator	

achievement in various local championships conducted by local government for RW level. In 2019, the 8 KBSs had intensive guidance from DLHK as program organizer and owner. DLHK organized the program and collaborated with PD. Kebersihan to manage the waste further. For every KBS, DLHK provided a 'mentor' as a knowledge expert and a thought leader at the same time, accompanied by two 'supervisors' chosen from the residents. The mentors collaborated with the government representative in *Kelurahan* (Structural local leaders) including 'KASIE EKBANG' (*Kepala Seksi Ekonomi Pembangunan*/Economic Development Section Chief), 'PKK' (housemother organization), 'RW chiefs', 'RT chiefs' (*Rukun tetangga*/neighborhood/the lower level of RW), and 'Karang Taruna' (Youth Organization). They bridge the educators to the residents, organize the residents and also educate the residents. In some KBSs, there were also NGOs who support the program and other outsiders. Finally, the key actors who also become the target of education programs were residents and waste collectors. Residents should separate their waste at home, while waste collectors should be responsible for waste collection waste transfer and waste recycling at communal level. When the KBS first started, the

structural local leaders (RT/RW chiefs, PKK chief) played a vital role in the community because mentors were outsider, so they need to be introduced and allowed to interact with the residents. Once the local structural leaders were not active in bridging the education mission by mentors, there will be no learning activities in the related RW areas. These unexposed areas were found in many RWs in KBS Kujangsari, KBS Gempolsari and small number of RWs in other KBSs. It showed that structural local leaders played a role as community organizers and boundary spanner at the same time (Wenger *et al.*, 2002). This finding confirmed the importance of active community involvement to be change driver in the community as suggested by Chikowore (2020) and existence of boundary spanners to bridge difference between multi stakeholders as suggested by Reed *et al.*, (2014). Wenger *et al.* (2002) also emphasized the importance of community organizers at initial process of CoP development. To help mentors educating the residents, there were supervisors, recruited from the residents interested in joining the program. The supervisors stay in the area to supervise waste management practice conducted by the residents and waste collectors. Unlike the mentors, supervisors and waste collectors were mainly

joined because of occupation instead of enthusiasm toward the program. Local government and mentors should prepare their readiness by organizing formal knowledge sharing activities with all supervisors and waste collectors before the program started. This event was mainly to encourage their motivation in doing their task and give them basic skills on waste management practice. However, the study found that some waste collectors did not do their job, causing abandoned recycling infrastructure in the area. Some informants stated that it can be because either waste collectors had not enough skills to utilize the tools, or they did not want to recycle the organic waste. It is also implied by 7 informants that waste collectors mixed their separated waste, causing residents' reluctance in separating waste anymore. This finding confirmed the importance of internal motivation (passion) to join the CoP for successful learning process as suggested by [Abou-setta \(2015\)](#). As waste collectors and supervisors were recruited, they should be educated intensively to nurture their internal motivation. The other personal element involved was local cadres who helped the educator team and local leaders educating a vast number of residents. Local cadres played a role as knowledge experts and community organizers at the same time. This requirement is crucial, especially for KBS that covers a wide area. Moreover, the leaders who were active typically has a limited territory (e.g., KBS Sukaluyu, KBS Kujangsari, KBS Kebon Pisang), and limited time length of service, causing their inability to reach other areas outside their territory and disable them to conduct sustainable learning activities. Thus, the program needs more leaders in a smaller group (such as within an RW) to work effectively. Local cadres' existence confirmed [Wenger et al. \(2002\)](#)'s

suggestion about small groups to facilitate personal relationship among peripheral members. Thus, neighborhood ties, community engagement and social interaction as proposed by [Pei \(2019\)](#) and [Zheng et al. \(2020\)](#) can still be facilitated. However, local cadres were commonly chosen from the local organization members such as PKK, *Karang Taruna*. Some KBSs even obliged the organization members to be the cadres causing possibility of internal motivation absence on the local cadres as suggested by [Abou-setta \(2015\)](#). Improvement can be achieved by allowing any residents who have enthusiasm toward the domain to join. Thus, local cadres are chosen not due to their activeness in the local organization under the structural government body, but their enthusiasm toward the program. Moreover, they played as knowledge experts for the residents so that intensive knowledge sharing activities with primary educators are required. In 2020, DLHK only continued the guidance to 2 KBSs: KBS Sukamiskin and KBS Cihaurgeulis. The other KBSs can continue the program and PD. Kebersihan still handled the waste management at city level. Only *Lurah* at KBS Neglasari continued the program officially (sub-district level), while other KBSs mostly continued it independently without *kelurahan* guidance. As a result, only some RWs/RTs which stay active, if they have local leaders to organize them. This confirmed the vital role of community organizer as suggested by [Wenger et al. \(2002\)](#). However, most local leaders were from RW chiefs and or RT chiefs which have limited territory (RT or RW level). So, they cannot affect other RT/RWs outside their coverage, causing limited area to continue the program. The detailed information about leadership existence in each KBS is presented in [Table 3](#).

Table 3: Leadership existence in every KBS

KBS	Facilitators	Community Organizers	Thought Leaders	Knowledge Experts	Boundary Spanners	Pioneers
KBS Sukaluyu	√	-	-	-	-	√
KBS Cihaurgeulis	√	√	√	√	√	√
KBS Neglasari	√	√	-	-	-	√
KBS Kebon Pisang	-	√	√	√	√	√
KBS Sukamiskin	√	√	√	√	√	√
KBS Babakan Sari	-	-	-	-	√	√
KBS Kujangsari	-	√	√	√	-	√
KBS Gempolsari	-	-	-	-	-	√

Knowledge domain

Knowledge domain consisted of community shared problems, program objectives, and knowledge and value in the learning activities. According to the interviews and field observation, it is found that the shared problems for all KBSs were similar. First, most residents' mindset was not pro to the waste separation. Second, the residents had low awareness and no willingness to separate their waste. As a result, the participation rate on waste separation at the household level was low. The programs' objectives were to encourage sustainable residents' participation in waste separation at home by changing their mindset toward waste management, giving separation skills for the residents, and recycling skills for waste collectors and local leaders. The local leaders are encouraged to educate their residents by being role models to initiate waste separation and recycling at their homes. To reach the objectives,

educator team was mandated to educate residents, waste collectors and local leaders by disseminating several information, especially technical knowledge about organic waste separation. The educator team should also educate waste collectors and local leaders about recycling techniques of organic waste. Even though the main content of education was focused on technical knowledge, the educator team was permitted to implement other approaches to reach the objectives based on their initiatives. Therefore, it was found various other knowledge and value being shared as the educational contents. According to the observation and interviews, their decision to share specific additional knowledge and value were based on the educators' knowledge and value toward waste management and their consideration toward the residents' interest. For example, the educators who educate residents from low-income economic level will approach them by informing economic benefits

Table 4: The educational contents in the KBS

Type of knowledge and Value	KBS Sukaluyu	KBS Cihaurgeulis	KBS Neglasari	KBS Kebon Pisang	KBS Sukamiskin	KBS Babakan Sari	KBS Kujangsari	KBS Gempol Sari
1. Technical knowledge of waste segregation for residents, mainly for organic waste	√	√	√	√	√	√	√	√
2. Technical knowledge of organic waste recycling for waste collectors and local leaders	√	√	√	√	√	√	√	√
3. Practical tips to reduce and recycle organic waste		√	√					
4. Recycling technique of inorganic waste				√	√		√	√
5. Impacts of waste for environment and health	√	√	√			√		√
6. Benefits of organic waste for planting	√	√	√			√		
7. Economic benefits of inorganic waste				√	√		√	√
8. Local regulation about waste segregation	√	√			√			
9. Individual and social roles to environmental protection	√			√	√			
10. Social Religious value toward personal obligation to environment	√						√	
11. Cleanliness for aesthetics				√				

of waste. They also activate waste banks to facilitate valuable inorganic waste sale from the residents as found in KBS Kebon Pisang, KBS Sukamiskin, KBS Kujangsari, and KBS Gempolsari. Education in KBS Kebon Pisang were concerned about cleanliness for aesthetics because one of the RWs in the KBS has been famous for their Mural Art village. Some educators shared environmental values, religious obligations, and responsibility toward environmental protection and organic waste benefits because they were more into the contents. It showed educators' preference and their knowledge influence their decision to choose what content will be shared. The detailed information about knowledge and value being shared as the educational contents in KBSs is presented in Table 4.

Previous studies showed that some of mentioned knowledge and value in Table 4 were crucial to encourage residents to participate in waste management. The knowledge about harmful impacts of waste to environment and health, was found only in 5 KBSs. According to previous studies, this knowledge was crucial to improve personal awareness toward waste management (Moh and Manaf, 2017) which had significant effect to waste separation and recycling behavior of residents in other countries such as China (Fan et al., 2019; Meng et al., 2019) and Singapore (Fan et al., 2019). Sharing knowledge about waste management benefits was found in different places. KBSs which highlighted additional income from waste did not campaign benefits of organic waste for planting, and vice versa. It was probably because of educator consideration toward the residents' characteristic as suggested by Knickmeyer (2019) and Sunarti et al. (2021) who stated that educational approach should be adjusted to residents' typicality. Previous studies confirmed that economic profit became the powerful driver of waste management behavior (Chen and Gao, 2020), thought it may be more impactful for low-income residents (Sunarti et al., 2021). The combination of awareness to waste management and perceived benefits will nurture strong motivation to participation (Wadehra and Mishra, 2018). Other educational contents being shared were about individual and social roles to environmental protection and social religious value toward personal obligation to environment, found in 4 KBSs. These two contents highlighted

that knowledge about the individual and social responsibilities was associated with personal moral responsibility (Zebua and Suhardini, 2021; Wang et al., 2020). Previous studies, such as Mukama (2016) and Liu et al. (2019) prove that personal moral responsibility and motivation had crucial effect to attitude toward waste management. Combination among awareness, personal moral responsibility and attitude was also proven to be influential to encourage willingness to participate in waste management in China (Meng et al., 2019; Zhang et al., 2019). In fact, personal moral responsibilities which were nurtured through social-religious value have affected sustainable waste management behavior of community members in Malaysia (Tiew et al., 2015). As implied by Aboul-Enein (2017) and Zebua and Suhardini (2021), educating residents through a religious value approach is vital especially for the religious communities as Bandung City residents. Some educators in KBS Sukaluyu, KBS Cihaurgeulis and KBS Sukamiskin also socialized local regulation about waste separation obligation, while educators in other KBS areas stated that they did not want to encourage the residents through law enforcement because they want the residents to separate voluntarily. This consideration may be true, but past studies, for instance Xu et al. (2017) and Ma et al. (2018), found that regulation was influential for initial encouragement to improve their skill and form new habits. The importance of habituation for learning approach was also mentioned by 9 informants from 7 to build sustainable participation on waste separation. But habituation without enlightening them about the value are not strong enough to sustain the participation, as indicated by 5 of resident representatives, who stated that they only followed the rules. They intended to end the separation activity once the rules were not enforced anymore. Even though previous literature showed significant effects of educational contents being shared by educators initiatively, there was no obligation for the educators to share the whole. The primary educational contents they shared were about technical knowledge. It indicated no identification of knowledge needs being done before the KBS started. Wenger et al. (2002) stated that one of the basic strategies to enable the CoP to achieve its goals is by identifying knowledge needs at the first step based on the shared problems. Wenger's

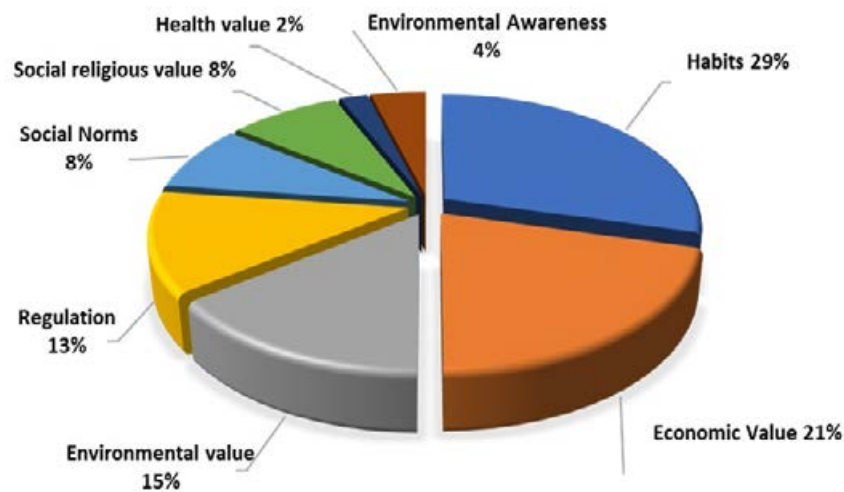


Fig. 4: The driving force of residents' participation in KBS program

statement was agreed by the study conducted by Janmaimool and Denpaiboon (2016) who indicated that knowledge relevance influence what personal factors will be nurtured. Thus, identifying knowledge and value needs should be done at first place. The findings from this study showed that the educational contents being shared by educators in the KBSs (Table 4) are vital to nurture waste management behavior. The crucial knowledge and value for waste management behavior improvement should be combined altogether to be effective (Sunarti *et al.*, 2021). To confirm the driving factors of resident behavior in KBS area, the informants from residents' representatives were asked to what factors causing them to keep participating in the waste management program for more than months. The result of driving factors is presented in Fig. 4.

The participants who have separated their waste for more than months mentioned at least 2 driving force that keep them participating in the waste separation program. For instance, a resident decided to participate in KBS program for an extended period because of three primary reasons: social value, regulation, and environmental awareness at the same time. Other resident representative stated that they stay participated because they get used to do it (habits) and they felt the benefits, especially for its cleanliness (environmental value). Beside the driving force resident participation, some insights about residents' rejection in participation were

also identified. Some supervisors stated that the residents often complained about the separation because they have paid for the waste disposal. Other reasons were because they think that waste separation was supposed to be waste collectors' job. These findings indicated low personal responsibility toward environment and their low understanding to the consequence of their behavior to the environment. These findings may indicate that the residents have not got the required knowledge in the knowledge sharing activities.

Knowledge sharing activities

The immediate concern in the KBS program is to improve resident mindset and awareness to waste management, so they will be willing to participate and change their habits sustainably. There are some scheduled knowledge-sharing activities conducted in all KBS to reach the objectives. According to the study findings, there were some characteristics of learning activities found: 1) some activities were formal, given to all/specific stakeholders, without regular schedule (eventual), 2) some activities need certain requirement to join (informal membership), 3) activities with certain requirements have regular meeting schedule and specific activities conducted, 4) All activities have dialogue-based and/or practice-based learning. Based on the characteristics of knowledge sharing activities in the KBS areas, the activities were classified into two main forms: non-

Table 5: Profile of Non-group Learning Activities in KBS Program

Organizers	Type of Learning	Educational Contents	Freq of Learning	Targets	Knowledge Sharing
Mentors, Supervisors, Kelurahan, NGOs	Recycle Training for organic waste	- Technical knowledge about waste recycling (household composters)	1 – 3 times/year	Local Leaders (RWs, RTs, PKK, Karang Taruna, Kelurahan)	Practice-based learning
Mentor and supervisors	1. Program Socialization in Kelurahan	- Technical knowledge about waste separation	1 – 3 times/year		Dialogue-based learning
	2. Program Socialization embedded in local leader meetings (RW meetings, RT meetings)	- Background of the program (waste issues) - Regulation socialization	Eventual (according to meeting schedules)		Dialogue-based learning
Mentors, Supervisors, Kelurahan, NGOs	1. Recycle Training for organic waste	- Technical knowledge about waste recycling and waste separation	1 time before the program started and then being monitored in the field by the supervisors	Waste Collectors	Practice-based learning
	2. Socialization about technical skill of waste separation and waste issues in Bandung City	- Background of the program (waste issues) - Regulation socialization			Practice-based learning
Mentor and Supervisors accompanied with PKK members	1. Door to Door Education (DTDE)	- Technical knowledge about waste separation - Impacts of waste for environment and health - Regulation socialization	1 – 3 times / household, each time took 10 – 15 minutes	Residents	Dialogue-based learning
	2. Socialization through community activities (<i>Senam Lansia</i> / elderly Workout, <i>arisan</i> , <i>pengajian</i> , <i>Gerakan Pungut Sampah</i> (GPS)/Trash Pick Motion)	- Technical knowledge about waste separation - Program socialization	At least every month	Residents	Practice-based and dialogue-based learning

group and group learning activities. Non-group and group learning activities were presented in Table 5 and Table 6.

According to Table 5, educational contents were dominated by technical knowledge of waste management. A minor portion has been given for sharing knowledge about waste impacts on the environment, social and personal obligation to environmental protection and other essential contents. Regulation socialization was conducted to encourage learning from habituation to enable the

residents sensing the benefits as they experience it directly. Sensing the benefits may be limited when they were not informed about the value of the waste management activity through dialogue. Therefore, the perceived value driving resident participation was dominated by environmental value (cleanliness) and economic value which they felt directly (Fig. 4). Non-group learning activities tended to lack intensiveness and iterative cycle because most activities were held 1-3 times/year (eventual). Other knowledge sharing activities were also too short (e.g., DTDE), causing

Table 6: Profile of group learning activities existing in KBS areas

No	KBS	Learning Groups	Member of Group	Educational Contents	Activities	Freq.	Knowledge Sharing
1	KBS Sukaluyu	Weekly cleaning days (Friday Clean Day and Sunday Clean Day)	Local leaders (PKK, RW, RT), Supervisors, Mentor	Waste separation habituation, waste impacts to environment	1. Trash Pick activities 2. Workout together 3. <i>Botram</i> (potluck)	Weekly	Practice-based and Dialogue-Based learning
2	KBS Chaurgeulis	RW Urban Farming	Local leaders (PKK, RW, RT), Supervisors, Mentor	Waste separation and organic recycling practice, economic benefits of organic waste	Planting vegetables, fruits, and Fish farming by using recycled organic waste (BSF larvae and compost)	Weekly	Practice-based and Dialogue-Based learning
3	KBS Neglasari	<i>Kelompok Berkebun/Gardening Group</i> , collaborated with other government department Trash Hero Kids program (Pasgeber <i>Pasukan Gerakan Bersih</i> /Cleanup-Motion Troof)	Kelurahan, PKK, RW, RT, Karang Taruna, residents who are interested in gardening NGOs, Students, Children	Waste separation and organic recycling practice, economic benefits of organic waste Waste separation habituation, waste impacts to environment, individual and social roles of environmental protection	Planting vegetables, fruits, and fish farming and make sellable products 1. Cleanup activities, 2. Habituation on waste separation, 3. Trash Pick activities	Weekly/bi-weekly Eventual	Practice-based and Dialogue-Based learning
4	KBS Kebon Pisang	<i>Cibunut Berwarna</i> /Cibunut Colored	Karang Taruna, RW 7 Cibunut residents	Environmental Cleanliness campaign	1. Coloring RT area with the same specific color 2. Mural Art containing persuasive messages for waste management campaign	Eventual	Practice-based and Dialogue-Based learning
5	KBS Sukamiskin	KSM (<i>Kelompok Swadaya Masyarakat/Resident self-subsistent Group</i>) Waste Bank for kids and housewives A learning group for each RW 1 day/week to learn about inorganic recycling waste (Soap, candle, handy crafts)	RW 7 members, active residents who are concern about environment Kids and active residents who are concern about environment PKK and Housewives who are interested in learning in inorganic recycling	Educative tourism to campaign environmental cleanliness Inorganic waste separation practice, economic benefits of inorganic waste Inorganic recycling practice, economic benefits of inorganic waste	Manage Mural Art Village for tourism destination buy-sale inorganic waste and save their point to get exchange to money every year 1. Learn about creating handy drafts made of inorganic waste 2. How to recycle organic and inorganic waste (Eco Enzyme, making soap from used oil, and so forth)	Monthly Weekly	Practice-based and Dialogue-Based learning Practice-based and Dialogue-Based learning
6	KBS BabakanSari	RW Urban Farming GPS (<i>Gerakan Pungut Sampah</i> / Trash Pick Motion)	Local leaders (PKK, RW, RT), Mentors, Supervisors Residents based on RWs	Waste separation and organic recycling practice, economic benefits of organic waste Waste separation habituation, waste impacts to environment	Planting vegetables, fruits, and Fish farming by using recycled organic waste (BSF larvae and compost) Cleanup the home yard together every week in the morning	Weekly	Practice-based and Dialogue-Based learning
7	KBS Kujangsari	PPO (<i>Pilah Pilih Olah/Separating Recycling</i>) every Saturday in Waste Bank in 1 RW (buy-sell inorganic valuable waste)	Housewives who are willing to earn money from their inorganic waste	Inorganic separation and recycling practice, economic benefits of inorganic waste	Cleanup valuable inorganic waste, weight the waste, buy-sale inorganic waste and save their point to get exchange to money every year. Cooking together for bonding, making handy crafts from inorganic waste	Weekly	Practice-based and Dialogue-Based learning
8	KBS GempolSari	RW Urban Farming	Local leaders (PKK, RW, RT) at RW 9	Organic recycling practice	Planting vegetables, TOGA (Tanaman Obat keluarga/Herbal medicine for Family)	Eventual	Practice-based learning

little chance to share more knowledge, no possibility to have intensive interaction as typical activity in CoP (Tran et al., 2018; Wenger et al., 2002). Such learning activities also did not meet the requirements of expected education strategy as suggested by Chikowore (2020); Zheng et al. (2020) and Pei (2019). Meanwhile, group-based learning provided more intensive interaction with regular activities to allow bonding establishment as expected.

Table 6 presented some group-based joint activities conducted regularly. Weekly group activities allowed intensive and iterative learning processes, building bonding, community engagement and intensive communication among members which are impactful to encourage collective participation in waste management in other cities (Chen and Gao, 2020; Pei, 2019). Intensive and iterative learning activities enabled the members to sense their community identity (Li et al., 2009; Madsen and Noe, 2012; Wenger et al., 2002), nurture their sense of belonging, and build trust among members (Reed et al., 2014) which are essential to enable effective knowledge sharing. Moreover, most activities combined practice-based and dialogue-based learning for habituation and sharing required knowledge and value which are suggested by previous study for CoP success (Tran et al., 2018). In the learning group, the members are encouraged to learn by doing intensively enabling the members to sense the benefits of the waste management activities directly in the long term. According to the interview, 9 informants from 7 KBSs agreed that habituation is the best way to build new behavior for the residents related to waste management. The habituation is required to allow the residents to learn by doing, making the behavior familiar to them, and eventually building their new behavior by transforming the iterative practice into their new habits (Sunarti et al., 2021). According to literature, habituation through learning by doing practice is significant in affecting waste management behavior (Ulhasanah and Goto, 2018) and has been proven in changing residents' behavior toward waste separation in many countries such as in China (Liao et al., 2018; Xu et al., 2017). Learning by doing was also suggested by Madsen and Noe (2012) for successful CoP based on their study about CoP in agricultural farmers. The group learning activities which were based on economic orientation such as Waste Banks and PPO are required but

cannot stand independently without non-economic-oriented activities. Dependence on extrinsic benefits such as only expecting money is disfavored because earning money from waste may have some limitations, such as marketing and product quality, as indicated by some informants. Moreover, the income was relatively low, which encouraged only low-income residents as indicated by previous studies (Almasi et al., 2019; Meng et al., 2019). In fact, the obstacles faced often cause low interest for several low-income residents as well, as found in KBS Kebon Pisang, KBS Sukaluyu and KBS Neglasari. Once they found a better way to make money, they will leave the activity. It indicated the vital role of balanced motivation between intrinsic motivation and extrinsic motivation for sustainable participation (Chen and Gao, 2020; Sunarti et al., 2021). Waste bank can be very useful when it becomes education approach for children as conducted in KBS Kebon Pisang. Through Trash Hero Kids and PASGEBER program, KBS Kebon Pisang has educated their youth since their childhood. This approach was positive for program sustainability because the children can be cadres in the future. Besides several positive impacts found in the group learning activities, some weaknesses were also identified. First, some learning groups are only accessible for local structural leaders (e.g., *Kelompok Berkebun* in KBS Neglasari and KSM in KBS Kebon Pisang). The learning groups were supposed to allow other members (including peripheral members) to join as they have enthusiasm toward the knowledge domain to regenerate more local cadres. Local cadres may not only from the structural organization under local leaders but also peripheral members who have readiness toward the domain. Second, the educational contents of the existing group learning activities were mostly still focused on technical knowledge of waste separation and waste recycling. More portions should be provided for other important contents such as waste impacts to environment and health, social and personal behavioral consequences that result responsibility to environmental conservation in dialogue-based learning activities. Less attention to fundamental knowledge being shared causes residents' mindset focusing solely on extrinsic factors they sense (e.g., to obey the rules, to meet social norms, expect economic benefits), which was not enough to encourage sustainable participation (Issock et al., 2020; Lawrence et al., 2020; Zhang et al.,

2019). Third, the group learning was varied between KBSs. Combining several types of group learnings in every KBS to cover diverse groups of residents can be beneficial, such as groups for kids, housewives, youth, and any other possible groups, according to the potency of the area.

Managerial elements

Managerial elements were related to regulation and norms to support mutual interactions and leadership with various roles to complement each other. Based on study findings, there is no obligation for the residents to involve in the joint activities even though waste separation practice is mandatory for all residents, according to the local regulation. The involvement in the community activities is only voluntary, while it is mandatory for local leaders and educators. Low enthusiasm and less perceived value toward the knowledge domain caused a few residents willing to join the learning activities. Therefore, non-group learning activities are still required to reach all residents for early campaign including for disseminating regulation about waste separation mandatory. Previous study found that dissemination of regulation was significant in encouraging participation in the waste management, if the participation mandatory (Liu *et al.*, 2019). When the regulation about waste management practice was not mandatory, regulation publicity was proven to be insignificant in encouraging participation (Meng *et al.*, 2019). As the mandatory for direct practice will encourage participation and habituation, the follow up should be involvement in the group learning activities to share knowledge and value more deeply and improve their practice. Social norms can be the tool to encourage their participation in the group learning activities. According to research findings, when residents within the same areas are close enough, they tend to feel shame when they were different from others. This cultural value was adopted by local cadres and educators when encouraging the residents in waste separation. This finding confirmed the study conducted by Chen and Gao (2020) who found that neighborhood influenced other people's decision-making to participate in the recycling activity of households in China. This social norm should also be utilized to encourage group-learning participation for more effective learning result.

Infrastructure elements

There were two primary facilities required in waste management: organic waste bins and recycling facilities. According to the field observation, some KBS programs provided an organic waste bin for each house for the residents who have separated their waste (e.g., KBS Cihaurgeulis, KBS Neglasari). KBS Sukamiskin provided big-sized bins that cover several houses. Most residents' representatives implied that they did not depend on waste bins to decide whether they want to separate or not. As presented in Fig. 4, most driving forces of their behavior beside habits were related to perceived value such as economic, environmental, social-religious, and health value. There were only 2 residents who stated that they would be more motivated to separate their waste if they get specific bins. It indicated that waste bins were not the primary driving force of the behavior. Instead, it may become the support system to moderate the separation behavior. Educators informed that residents did not have to provide brand-new waste bins for separation. They can use anything, such as secondhand buckets for the bins. In terms of recycling facilities, all informants stated no problems because DLHK provided all the required facilities for the recycling process including vehicles to carry the waste. Some recycling facilities were placed at recycling points at the communal level, while others were placed between housings. Some recycling facilities have different products, according to residents' interests and or personal resource recycling skills. When the RWs or RTs had no space to put the facilities in the KBS areas, the separated waste will be handled by the government to be recycled further at the city level. The existing recycling facilities are presented in Table 7.

Commonly, all the KBSs (except KBSs guided by DLHK) relied on waste collectors to manage the recycling process unless the areas had local experts as found in KBS Kebon Pisang. The role of waste collectors was crucial if there were recycling facilities available in the area. However, most informants agreed that recycling facilities did not become the primary facilities they should have. Residents who sense the internal benefits, such as social-religious, environmental, and health value, implied that it does not matter if they cannot get concrete benefits from their separation activity; they will continue the separation activity

Table 7: Recycling facilities, the placement, and the end products

No	Recycling facilities	Placement	Middle Products	End products/utilization
1	Bata Terawang (Aerob Composter)	Recycling Point	compost	Vegetables, flowers
2	BSF (Black Soldier Fliers)	Recycling Point	Animal feed	Fish
3	Biopore/Losedra (organic waste holes)	Between housings		Compost directly to land
4	Takakura (portable Composter)	Household	compost	Vegetables, flowers
5	Biodigester	Recycling Point	Gas	Electricity
6	Waste Bank	Recycling Point		Earn money
7	Windrow	Recycling Point	compost	Vegetables, flowers
8	hydroponic	Recycling Point		Vegetables

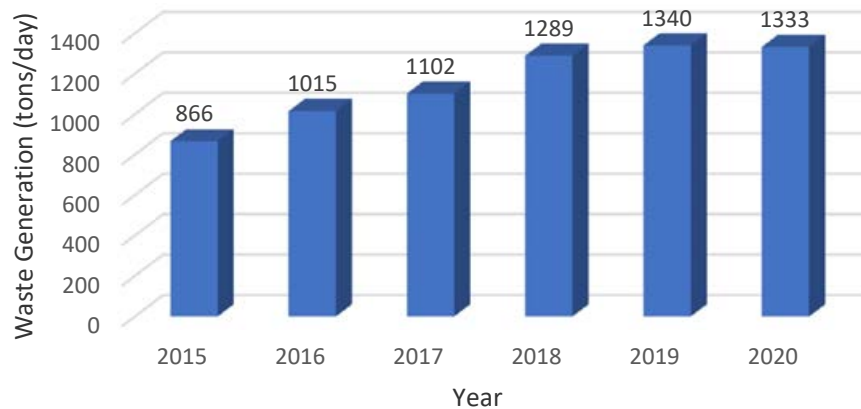


Fig. 5: Waste generation at Bandung City which was relatively decreased after education implementation (Source: PD. Kebersihan Archives)

because they have got the benefits directly. In contrast, residents who were mainly motivated by economic benefits for the activity will expect recycling products or money from selling/utilizing the inorganic recycling products. Different finding was found by the study conducted by [Meng et al. \(2019\)](#) and [Trihadiningrum et al. \(2017\)](#) who found that accessibility to separation and availability of recycling facilities were significant factors of waste management activities of residents in China and Indonesia. However, it was supported by previous study conducted by [Lawrence \(2020\)](#) who found that strong intrinsic motivation was enough to keep long-term participation of residents in Trinidad-

Tobago without external driver. Thus, recycling facilities in KBSs are optional if it is feasible, and it is suitable as residents' expectation. The primary point is that the education should improve residents' intrinsic and extrinsic perceived value of waste management to avoid dependency on facilities. When the intrinsic perceived value was higher, residents tend to be more effortless in supporting the program. In terms of technology utilization, there was no found technological involvement, except *Whatsapp* group. The group was utilized merely for information dissemination among local leaders. According to the interviews with the local leaders, most members were

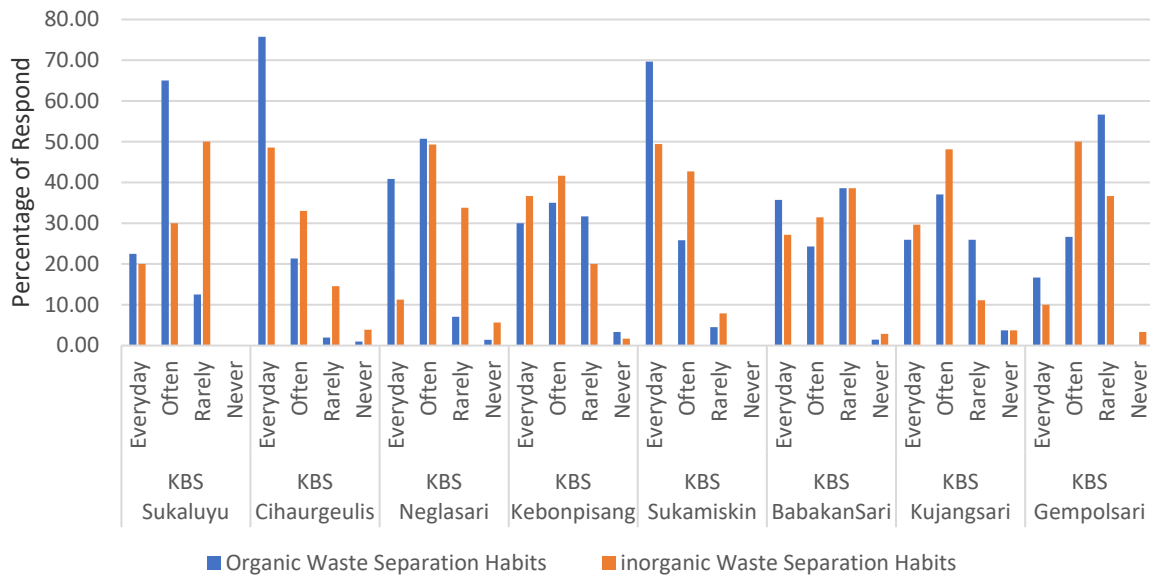


Fig. 6: Waste separation habits of residents in KBS area

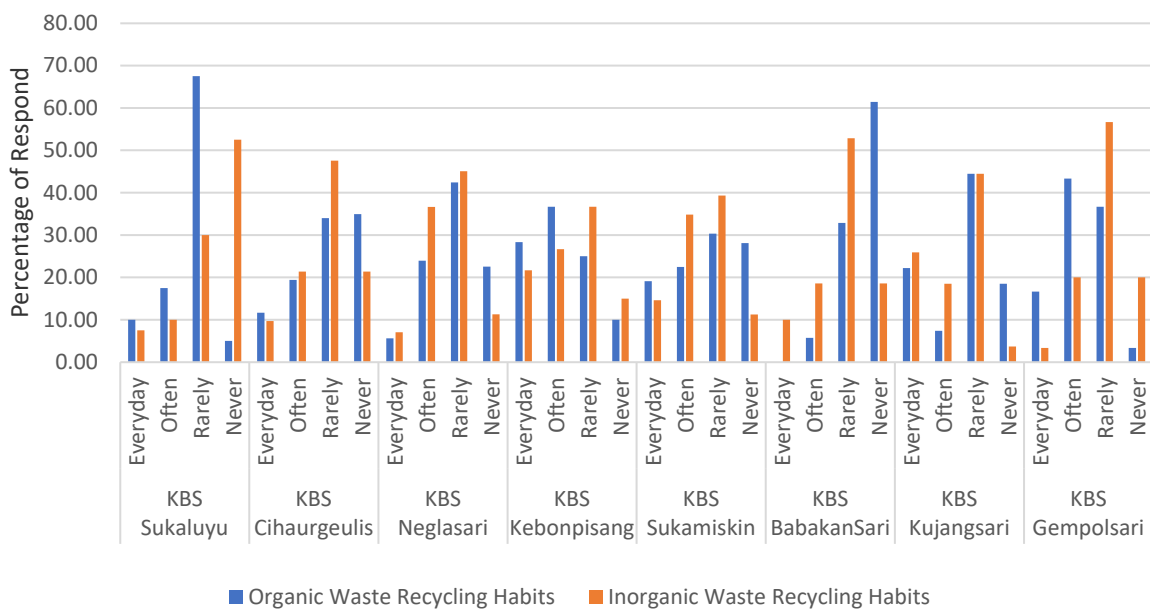


Fig. 7: Waste recycling habits of residents in KBS area

passive in knowledge sharing in the platform. Instead, face-to-face and direct interaction are more dominant and more required because they live in the same areas. There were also no knowledge repositories found as the storage for the activity documentation, regulations of the communities required to be easily retrieved

anytime to support knowledge sharing (Koeglreiter *et al.*, 2006). Knowledge repositories are required, especially for educators, to enable knowledge retrieval and performance re-evaluation for improvement. Thus, knowledge repositories can be crucial for the core members.

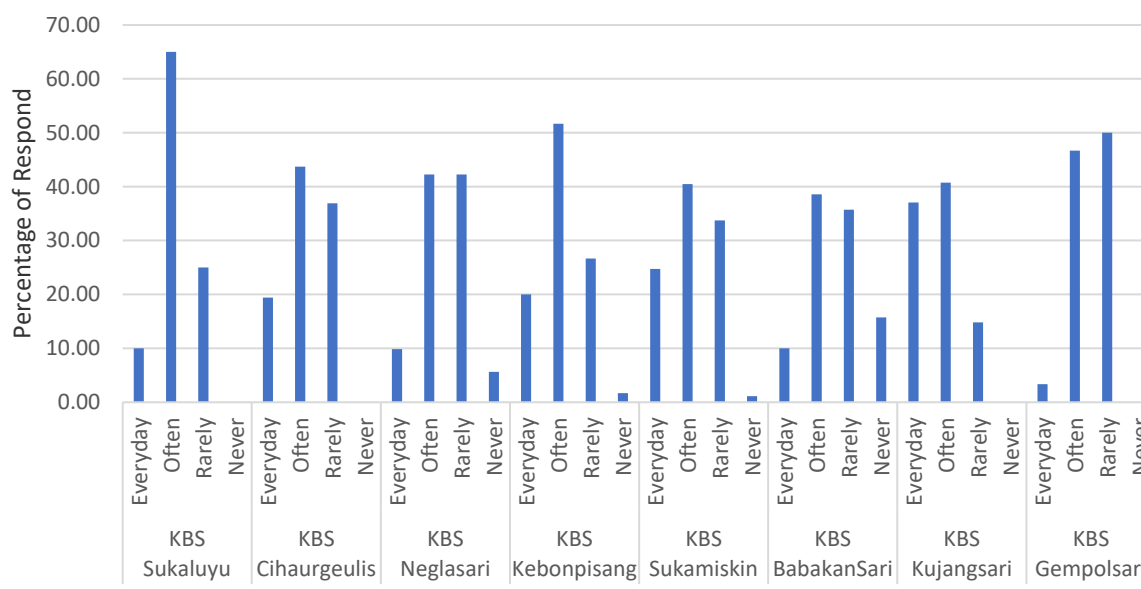


Fig. 8: Waste reduction habits of residents in KBS area

Resident waste management behavior after education strategy Implementation

The effects of the resident-based education using CoP approach was gathered through secondary documents from the governments and survey to get information about resident waste management behavior. The information become the evidence to show the effectiveness of the resident-based education to improve resident changing behavior effectively. The profile of waste generation at city level from 2015 to 2020 is presented in Fig. 5.

Waste generation showed in Fig. 5 indicated waste reduction from 2019 to 2020 up to 0,47 % after slowing down from 2018 to 2019. Further evidence is given related to waste management habits of respondents who become resident representatives within KBS areas as presented in Fig. 6.

Data from the survey as presented in Fig. 6 showed that some KBSs presented high percentage of waste separation habits, averagely more than 60%. KBS Cihaurgeulis and KBS Sukamiskin showed higher percentage than other KBSs (>90%) because the two KBSs still get guidance from the government so that they get sufficient, intensive, and regular education with infrastructure supports. However most other KBSs (except KBS Gempolsari) which had no official guidance from government still showed relatively

positive habits. Organic separation habits in all KBSs are higher than inorganic waste separation habits because the program focused mainly on organic waste separation. Inorganic waste separation was allowed and facilitated but not the main campaign, similar with recycling activities. The data of recycling habits of residents in KBS areas was presented in Fig. 7.

Recycling habits (Fig. 7) were relatively low for both organic and inorganic waste, except KBS Gempolsari. This was because the program' key objectives focusing on waste separation. The recycling process was mainly handled by waste collectors or *PD. Kebersihan*. KBS Gempolsari showed quite higher percentage on organic waste recycle because the local leader encouraged residents to dispose their vegetable waste on recycling tools he provided around their neighborhood. The resident representatives implied that they get used to dispose vegetables in the recycling points without thinking about waste management. It indicated that they did not know the value of their activity, proven from the low percentage of the separation habits which are supposed to be higher as well. The local leaders played as a pioneer and community organizer but there is no thought leader available so there were no learning activities once DLHK ended the

guidance. This finding confirmed the vital role of two key leaders' existence for effective CoP: community organizers and thought leaders, as indicated by [Wenger et al. \(2002\)](#). The thought leader is needed to facilitate proper knowledge and value sharing. Another waste management activity being surveyed was waste reduction habits as presented in [Fig. 8](#).

Even though waste reduction is not the main campaign in the KBS program, most KBS areas showed higher percentage of waste reduction habits. KBS Sukaluyu showing higher percentage of waste reduction can be due to their long experience in waste management. All informants from KBS Sukaluyu stated that their area was the pilot for KBS program since 2015, initiated by an NGO named YPBB. This finding confirmed the requirement of long-term education as suggested by [Azevedo et al. \(2021\)](#) who indicated that one of weaknesses of waste management in developing countries was the absence of regular agenda of public campaigns. CoP approach meets this requirement because CoP suggested long-term and regular learning activities ([Wenger et al., 2002](#)). The findings about waste management behavior of residents in KBSs become the evidence that the educational strategy was effective and powerful in changing resident behavior sustainably. The percentage can be higher if all required educational contents got sufficient portions and the education is lengthened (not only one year) to allow intensive interaction. A longer period of education enables the educators to nurture more local cadres from the learning activities, add personal elements and nurture key leaders. A long-term education also provides more chance for the educators to share important educational contents sufficiently. If these requirements are met, zero waste area can be achieved.

Resident-based learning model development

According to the analysis to critical elements of KBS program using CoP approach, it was generated generic codes which were developed through grouping the codes based on their similarities, differences, and roles in the input-process-output and support system. The main and sub-main elements were grouped and result new main categories. To determine the causal links among categories, the roles of each new main elements are determined. Based on the discussion section, it was found that

some key stakeholders' support become the input because they played a vital role in initiating the program. The key stakeholders refer to government support as facilitator, and educators (mentors and supervisors), local leaders and waste collector who should be ready in the beginning. The readiness for facilitators included providing supporting elements such as infrastructures to support learning, educators, and operational support. The facilitators may also need 3rd parties such as NGOs and environmental communities to help conducting learning activities for educators and local leaders' readiness. The readiness for educators and local leaders included ownership of knowledge and value being shared which were changing their personal factors to encourage their waste management behavior. Those referred to understand personal responsibility, environmental awareness, perceived value of waste management activity, attitude, willingness to participate and waste management behavior. They also need to have waste management skills to enable practice. Waste management readiness can be called waste management literacy, rooted from environmental literacy concept introduced by [Roth \(1992\)](#). Environmental literacy comprises four dimensions: knowledge, skills, affect and behavior ([Coyle, 2005](#); [Roth, 1992](#); [Scholz, 2011](#)). In the waste management context, the value is added as part of knowledge, consisting of economic, environmental, social, and religious value, which are vital to encourage waste management behavior. Environmental literacy involved behavior as the observable form of the learning result. To build the resident-based learning model using CoP approach, this study referred to the basic model of CoP proposed by ([Scarso et al., 2009](#)). Because knowledge domain contains community shared problems, community objectives, knowledge, and value needs for the educational contents, it is placed to be CoP baseline. The core process is the learning activities which should be conducted both group and non-group using two learning approaches: practice-based learning and dialogue-based learning. The group learning is mainly separated into two major groups: groups for key stakeholders and local cadre development from residents. Since residents commonly contain thousand households, it requires subgroup division, based on residents' characteristics. The group can be based on interest on certain waste management option, age group or profession, such

Table 8: Content synthesis process for model development

No	Preliminary Codes	Sub-preliminary Codes	New main categories	Group of codes	Generic Categories	Roles
1	Personal Resources	<ul style="list-style-type: none"> Leaders Leaders with various roles Local Cadres Waste Collectors as operator and education target Residents as education target 	<ul style="list-style-type: none"> Supports from leaders and waste collectors to the program initiation Facilitator Community Coordinators Thought leader Knowledge expert Boundary spanner Pioneer Knowledge expert readiness to support education program sustainability Waste Collector readiness to support program initiation Resident readiness for sustainable program 	<ul style="list-style-type: none"> Key stakeholders' support Facilitator Community coordinators/organizers Thought leader Knowledge expert Local cadres Pioneers Local cadres' learning system Waste collectors' learning system Residents' learning system 	<ul style="list-style-type: none"> Facilitator readiness Educators' waste management literacy Local leaders' waste management literacy Local cadres' learning system Local cadres' waste management literacy Waste collectors' learning system Waste collectors' waste management literacy Residents' learning system Residents' waste management literacy 	Group-learning activities (Main Process) Group-learning activities (Initial phase Process) Group and non-group learning activities (Main Process)
2	Knowledge Domain	<ul style="list-style-type: none"> The shared problems of the CoP that determines the community identity The objectives of the CoP 	<ul style="list-style-type: none"> Negative residents' mindset to waste separation Residents' Low willingness to waste separation Low participation rate on waste separation Sustainable residents' participation in waste separation at home 	<ul style="list-style-type: none"> Problems to be solved by learning Residents' waste management literacy 	Knowledge and value needs Residents' waste management literacy	Learning Baseline Output

Continued Table 8: Content synthesis process for model development

No	Preliminary Codes	Sub-preliminary Codes	New main categories	Group of codes	Generic Categories	Roles
		<ul style="list-style-type: none"> Educational contents 	<ul style="list-style-type: none"> Technical knowledge of waste management Impacts of waste for environment and health Economic, environmental, social, and religious value of waste management Individual and social roles to environmental protection Local regulation about waste management 	<ul style="list-style-type: none"> Waste management skills Environmental awareness Personal responsibility Perceived value of waste management activity Attitude Personal responsibility Willingness to participate Personal responsibility Waste management skills Habituation 	<ul style="list-style-type: none"> Knowledge and Value needs Waste management literacy 	Learning Baseline
3	Knowledge Sharing activities	<ul style="list-style-type: none"> Knowledge sharing activities to enable knowledge flow Mechanism of bonding, intensive interaction, community engagement 	<ul style="list-style-type: none"> Group learning Non-group Learning Practice-based learning Dialogue-based learning Group Learning to nurture local cadres Effective knowledge sharing Practice-based learning Dialogue-based learning 	<ul style="list-style-type: none"> Type of learning activities Learning approach Core process 	<ul style="list-style-type: none"> Habituation Learning process Local cadre development 	Learning Process
4	Managerial Element	<ul style="list-style-type: none"> Identified regulation and norms that support joint interactions 	<ul style="list-style-type: none"> Waste Separation practice through law enforcement Social norms of joint activities 	<ul style="list-style-type: none"> Learning approach Regulations and Norms Habituation 	<ul style="list-style-type: none"> Knowledge toward regulations and norms Practice-based learning 	Supportive System
5	Infrastructure	<ul style="list-style-type: none"> Social Media for knowledge repositories Waste bins for waste separation practice Recycling tool facilities Recycling product utilization facilities 	<ul style="list-style-type: none"> Knowledge sharing support Practice-based learning Encourage willingness to participate Encourage attitude 	<ul style="list-style-type: none"> Technology for Knowledge repositories Practice-based learning Waste management literacy Waste Separation facilities Waste Recycling facilities 	<ul style="list-style-type: none"> Technology for Knowledge repositories Learning process Waste management facilities 	Supportive System Learning process Supportive System

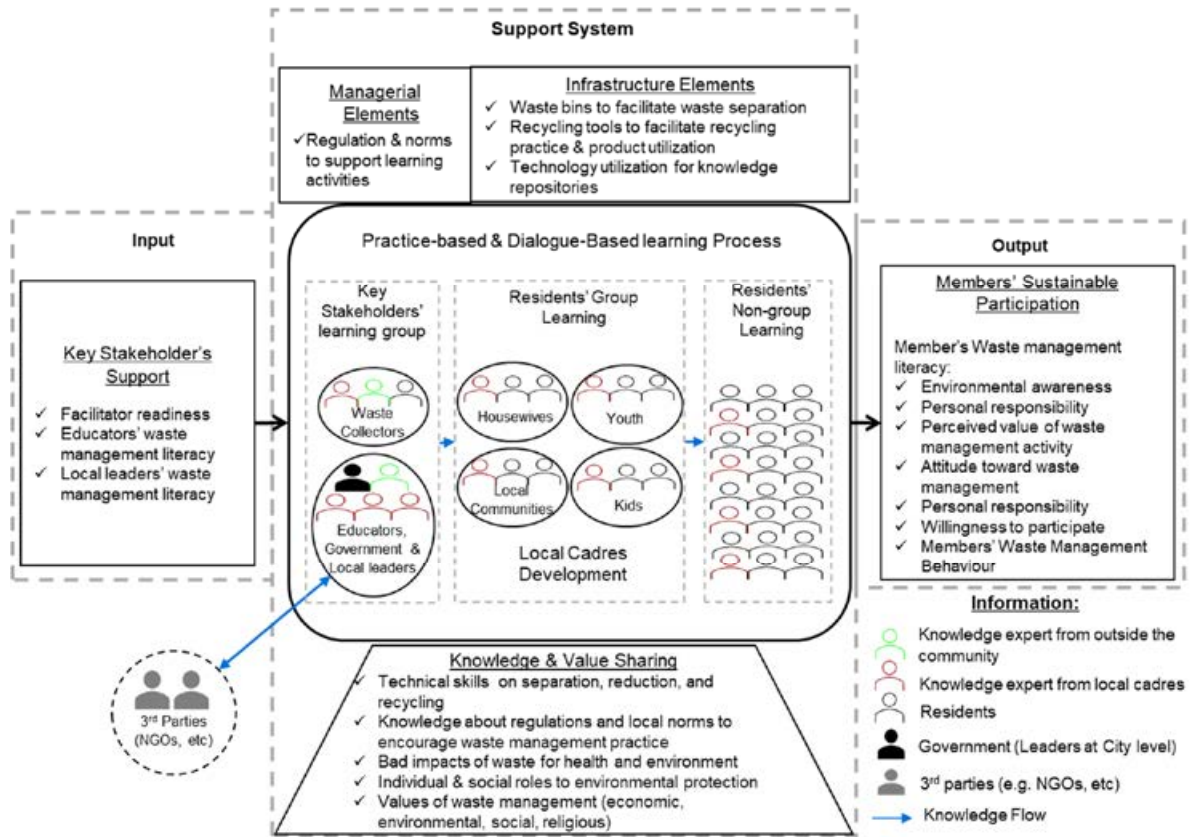


Fig. 9: The proposed design of resident-based learning model for waste management participation

as kids, youth, housewives or local communities. The output of the learning process in the CoP is represented by residents' waste management literacy. The whole process of model building through coding categories is presented in Table 8.

According to the model development process through content synthesis as presented in Table 8, the proposed model of resident-based learning can be presented as Fig. 9.

CONCLUSION

Resident participation in MSWM system is vital to ensure its effectiveness because residential are considered the most dominant waste producers in MSWM. It requires education to encourage sustainable participation from the residents. Previous studies have failed to provide effective and impactful learning strategy to promote personal factors that eventually nurture their

sustainable behavior. This study offered a new learning strategy by developing a resident-based learning model using CoP approach. Using case study on resident-based education implemented in Bandung City, Indonesia, this study gave a feasible and implementable resident-based learning model for other cities in the developing countries which faced the same problems. This study was successful in conceptualizing the resident-based education model. The local government, policy makers or other facilitators who were struggling in campaigning household participation in their waste management system can implement this educational strategy. This study provided some key findings. First, the educational strategy is feasible and implementable when there are facilitators and local leaders collaborated to provide educators, operators, and support systems. Second, facilitators and local leaders need to identify the main problems of the

residents related to waste management practice before the program started to determine the learning main objectives. Second, key stakeholders together with educators should identify knowledge and value needs based on their residents' characteristics to reach residents' waste management literacy. Third, support systems are required to facilitate and support practice-based and dialogue-based learning for effective resident learning activities. Fourth, local cadres are vital to sustain the waste management program. Fifth, educational strategy using CoP approach highlighted a long-term small group learning to facilitate intensive interaction and engagement among members for impactful learning outcomes. This study provides essential insights to understand how to build a resident-based educational program as a learning activities platform for residents to result sustainable participation in the MSWM process. There are some limitations which need improvement for further research. First, the proposed model was still hypothesis. It needs further studies to test the relationship among elements in the model. Second, the findings were highly based on cultural characteristics of Bandung city, Indonesia. Other cities with different culture may need some adjustments to implement the model. Third, samples for quantitative data were chosen purposively based on their history being exposed by the learning program. So, the survey finding may not represent the entire residents in Bandung city and Indonesia. Future research can focus on investigating other type of resident-based learning programs which meet CoP characteristics for comparison.

AUTHOR CONTRIBUTIONS

A. Ghazali conducted the literature review, research design, analyzed and interpreted the data, and developed the model, J.H. Tjakraatmadja analyzed and interpreted the data, developed the model, review the manuscript. Sunarti conducted the literature review, collected, analyzed the data, and prepared the manuscript text, Eka Yunita Dian Pratiwi collected and analyzed the data.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

3R	Reduce, reuse, recycle
BSF	Black soldier fly
CoP	Community of practice
CAQDAS	Computer assisted qualitative data software
DLHK	Dinas Lingkungan Hidup dan Kebersihan
DTDE	Door to door education
GPS	Gerakan pungut sampah
GSSI	Generasi semangat selalu ikhlas
K3	Kebersihan, ketertiban, keindahan
KANG PISMAN	Kurangi pisahkan manfaatkan
KASIE EKBANG	Kepala seksi ekonomi pembangunan
KBS	Kawasan bebas sampah
KM	Knowledge management
KSM	Kelompok swadaya masyarakat
MSWM	Municipal solid waste management
MSW	Municipal solid waste
NGO	Non-government organization
NVIVO	A qualitative data analysis software provided by QSR International
PASGEBER	Pasukan gerakan bersih
PD Kebersihan	Kebersihan perusahaan daerah kebersihan
PKK	Pemberdayaan dan Kesejahteraan Keluarga

PPO	<i>Pilih pilih olah</i>
P3MI-ITB	<i>Penelitian, Pengabdian kepada Masyarakat dan Inovasi Institut Teknologi Bandung</i>
QCA	Directed content analysis
QGIS	Quantum Geographic Information System
RT	<i>Rukun tetangga/hamlet</i>
RW	<i>Rukun warga/ community association</i>
TOGA	<i>Tanaman obat keluarga</i>
YPBB	<i>Yayasan Pengelolaan Biosains dan Bioteknologi</i>

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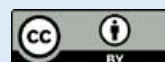
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ORIGINAL RESEARCH PAPER

Characteristics and combustion kinetics of fuel pellets composed of waste of polyethylene terephthalate and biomass

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ABSTRACT

BACKGROUND AND OBJECTIVES: The needs of fuel pellets from varied feed stocks have opened up opportunities and challenges for pellets production from non-woody biomass. Wastes of plastic recycling and wood sawing contained a high potential for energy source and suited for pelletizing as a solid fuel.**METHODS:** The characteristics and combustion kinetics of fuel pellets made using a mixture of waste of polyethylene terephthalate and biomass (*Tectona grandis* Linn.f) with a polyethylene terephthalate to biomass ratio of 9:1. The investigation covered physico-chemical properties and their functional group analysis, heavy metal concentration and ionic leachability testing, and ash analysis. In this context, thermogravimetric analysis was used in an atmosphere of oxygen gas, over a temperature range of 50-800 °C and at different heating rates. The work ends with discussion of the kinetics study via three comparative evaluations and the feasibility of fuel pellets for energy utilization.**FINDINGS:** Pelletizing with this ratio (9:1) was present the durability of PET/biomass pellets, a uniform dimension, ease handling, storage, and transportation common as woody pellets. Some technical challenges such as low moisture content and high volatile matter content were feedstock dependent. The major characteristics were a combination of those from both the constituent materials. Functional groups of the pellets were contributed by terephthalate and lignocellulose. The addition of a small amount of biomass in pellets could improve their thermal decomposition behavior. The properties of the polyethylene terephthalate/biomass pellets indicated that were fit for combustion with a high heating value equal to 19.20 MJ/kg. Heavy metals and ionic contaminants were below the maximum limits of the standards because of the cleanliness of the raw materials. However, the minor effects of earth materials and a caustic soda detergent were resulted in the alteration of residue chemicals. The pellets had lower ignition, devolatilization, and burnout temperatures than the original polyethylene terephthalate waste; likewise, the peak and burnout temperatures shifted to a lower zone. The activation energy values obtained using the Kissinger-Akahira-Sunose, Ozawa-Flynn-Wall, and Starink models were similar and in the range 142–146 kJ/mol.**CONCLUSION:** These findings may provide crucial information on fuel pellets from blended polyethylene terephthalate/biomass to assist the design and operation of a co-combustion system with traditional solid fuels. Such modifications of fuel pellets suggest the possibility of operating in large-scale furnace applications and can further be upgraded to other fuels production via modern bioenergy conversion processes.DOI: [10.22034/gjesm.2021.04.09](https://doi.org/10.22034/gjesm.2021.04.09)

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INTRODUCTION

The use of plastics has increased tremendously around the world due to their compatible properties in diversified applications, as well as being relatively cheap and safe. Post-consumption plastic is regarded as a major waste source and creates unhealthy conditions for humans and the environment (Patnaik *et al.*, 2020). The ever-rising amount of plastic waste has tended to increase polyethylene terephthalate (PET) waste. PET usage has rapidly grown in daily life applications such as bottling carbonated soft drinks, beverages, and other liquids and as food containers, microwave trays, and food packaging films. Furthermore, it is appropriate for lightweight, large-capacity, and shatter-resistant containers which are mainly found in the shipping industry (Sinha *et al.*, 2010). Used PET bottles can be recycled using a mechanical recycling process that uses shredding and crushing to convert a bottle into flakes (Jabłońska *et al.*, 2019). In this process, purification and decontamination of the PET-flake are important methods followed by washing (with a detergent) and water soaking, centrifuging, drying, and separating foreign plastics. PET-flake from these processes can be utilized as raw-PET for producing goods with both up- and down-cycling processes (Tolinski, 2011). Nevertheless, secondary waste from a mechanical recycling plant occurs as the process releases undesired materials such as dust, sludge, debris, tiny pieces, and microplastics, as mentioned waste of PET. These amount to 3-5% by weight (%wt) of the PET bottle in a mechanical recycling plant and it is feasible to recover energy and importantly reduce the amount of waste landfill (Beata *et al.*, 2019; Surenderan *et al.*, 2018). Biomass is considered one of the most important renewable sources and is produced in large amounts in industry, agriculture, and forestry (Becidan *et al.*, 2007). It can be directly used or used cooperatively with coal and other solid fuels (Ahn *et al.*, 2014; Basu *et al.*, 2011; Nussbaumer, 2003; Sajdak *et al.*, 2019). Teak sawdust (*Tectona grandis* Linn.f) provides as a waste of wood industry in the sub-regional area of northern Thailand. It is used as a fuel for firing in pottery furnaces and biomass power plants. Furthermore, it is used as a mixture material for mushroom and vegetable plantation. Pelletizing is one process for handling powder-like materials such as dust, sludge, debris, tiny pieces, and microplastic as well as biomass and sawdust to become fuel

pellets. Moreover, it has been widely used for mass and energy densification to overcome the disadvantages associated with raw material use (Mostafa *et al.*, 2019). To avoid the resource wastage and environment contamination, fuel pellets production is proposed to utilize the waste of PET and biomass. Currently, waste of PET and teak sawdust for alternative use is mainly received from local factories. Efforts are being made to understand as the study novelty in the co-densification of the mixtures of dusty PET waste and sawdust invite new challenges to pelletization process. In addition, the detailed studies of fuel pellets from varied feedstocks with different morphology and characteristics are opened up opportunities and challenges (Pradhan *et al.*, 2018). Thus, the co-densification of waste of PET and teak sawdust can be discussed. The use of this type of fuel in co-firing processes reduces greenhouse gas emission and cheaper than traditional fuels (coal, lignite) and sometimes get income by a surcharge (Jabłońska *et al.*, 2019). Previous works, the pyrolysis-gasification of biomass/PET feedstock has been extensively studied by numerous researchers (Abnisa and Wan Daud, 2014; Bu *et al.*, 2018; Madadian *et al.*, 2017; Narobe *et al.*, 2014; Robinson *et al.*, 2016). They reported that the composition and fraction of the feedstock have an important influence on the distribution, composition, and characteristics of the gas, liquid, and solids (Block *et al.*, 2019; Çepelioğullar and Pütün, 2013). Chattopadhyay *et al.* (2009), reported a significant interaction or synergistic effect between plastics and biomass (paper) under pyrolysis in a thermogravimetric analyzer (TGA). Paper-biomass started to decompose at a lower temperature (below 100 °C) than plastics and rapidly degraded at 350 °C. At higher temperature, PET started with an initial degradation temperature of 200 °C followed by a curve of rapid degradation at around 420-490 °C and a small amount at around 500-550 °C. Since the chemicals in plastic easily break up at higher temperatures, increased mass loss is observed more rapidly at a high temperature. Plastics/biomass composited with a mixed plastics (HDPE, PP, PET) to biomass ratio of 9:1 had the best performance with a slightly decreased mass loss during thermal degradation of 5% at a lower temperature and significantly decreased mass loss of over 95% at 500 °C. This may be due to the higher ratio of plastic in the mixture contains lower moisture, ash, and fixed

carbon contents than for a low ratio of plastics in fuel pellets (Martín-Gullón *et al.*, 2001). Energy required for the pyrolysis of multicomponent mixtures of waste PET and solid biomass increased with increase in amount of plastic content in the mixture due to the pyrolysis of PET plastic needs more energy than wood (Wang *et al.*, 2021). Combustion of multicomponent mixtures acquired lower the activation energy than pyrolysis, therefore the leads of a minimum biomass supplied as part of plastic wastes instead of a minimum plastic waste supplied as part of biomass needs to be examined further. It is noticed that the study in fraction of these materials (PET/biomass) has not fully explored. The approach has to account for the behavior of PET/biomass during combustion. TGA is a worldwide method to analysis thermal behavior of materials such as food, feedstock, chemical, soil, fertilizer, plastic, advance polymers, biomass, solid fuels such as coal, lignite, shale oil, and so on. TGA is one of the most extensive practices for the preliminary prediction of thermal behavior and kinetics for complex solid fuels (Govindan *et al.*, 2018; Yuan *et al.*, 2017). In TGA, the thermal decomposition of the sample under a selective environment is analyzed by determining its mass loss during an increased temperature duration (Chandrasekaran *et al.*, 2017; Xie *et al.*, 2019). The mass loss is used to determine kinetic parameters including the activation energy, pre-exponential factor, and reaction mechanism. Two common approaches applied for kinetic analysis are the model-fitting and model-free (iso-conversional) methods. The analysis is accurate and convenient for iso-conversional methods compared to model-fitting methods (Bu *et al.*, 2018). Only few data on heating rates assuming negligible mass transfer are required to estimate the activation energy in terms of the degree of conversion (Vyazovkin and Wight, 1999; Xu and Chen, 2013). Many iso-conversional methods, such as the Friedman, Ozawa-Flynn-Wall (OFW), Kissinger-Akahira-Sunose (KAS), and Starink methods have been used for the kinetic analysis of oxidative and non-oxidative processes in various types of solid fuels, including coal and biomass (Chen *et al.*, 2017; Sharara and Sadaka, 2014; Słopiecka *et al.*, 2012). There is a need for research into the utilization of waste of PET by pelletizing in combination with local biomass that could be applied in process of waste to energy. It is a new study pathway revealed with the novel materials, techniques of analysis, and

evaluation. The important properties and crucial data of characteristics of fuel pellets from blended PET/biomass with a view towards energy recovery in the combustion process are also considered. Moreover, this study aims to determine their functional group, heavy metals and ionic contaminants, and residue composition as well as evaluation of the kinetic parameters by iso-conversional methods (OFW, KAS, and Starink methods). It is expected that the knowledge of the characteristics and combustion behavior of the PET/biomass pellets and in particular the precise estimation of kinetics, will provide essential guidance for further work. This study has been carried out in Phitsanulok province, Thailand in 2019.

MATERIALS AND METHODS

Raw materials and pelletizing

The waste of PET was sourced from a bulk of waste plastics at a PET bottle recycling plant in Surin province, Thailand. The similar case of mechanical recycling processes for used PET bottles was demonstrated by Jabłońska *et al.* (2019). The sample of PET waste contained different sizes of PET plastic from <0.5 to 1.5 mm (sieve size no. 14-35). Biomass (teak sawdust; *Tectona grandis* Linn.f) was sourced from a saw mill in Phrae province, Thailand. Both the PET waste and biomass were dried before pellet production. The fuel pellets of PET/biomass was produced using a flat-die pelletizing machine. Prior to pelletizing, the machine was warmed using a mixture of PET/biomass as a pre-pelletizing material. Then, the mixture of PET and biomass (9:1, w/w) was fed at a continuous rate of 1 kg/min. About 10%wt of water was added during the process to prevent materials from sticking. Pellets were approximately 1.0 cm in diameter and 2.4 cm in length, as shown in Fig. 1. Then, pellets were dried in an air oven at 65 °C for 12 hours.

Proximate, ultimate, and calorific value tests

The samples of PET waste, biomass, and PET/biomass pellets were analyzed to determine their physical and chemical properties with three replications. The moisture content (MC) was measured by weighting mass before and after heat in a hot-air oven at 105 °C for 24 hours. Volatile matter (VM) of sample was measured by weighting mass before and after the covered sample-containing crucible in



Fig. 1: A flat-die pelletizing machine and the PET/biomass pellets

a muffle furnace at 900 ± 10 °C for 4 minutes (CEN/TS 15148). The sample in uncovered crucible was heated at 550 ± 10 °C for 2 hours to determine the ash content (CEN/TS 14775). The fixed carbon (FC) was found by subtraction; $FC = 100 - (MC + VM + \text{ash})$. The carbon (C), hydrogen (H), nitrogen (N), and sulfur (S) contents of samples were determined using a TrueSpec Micro (Leco, CHNS628) and the oxygen (O) content was calculated based on dry ash-free basis by using the formula; $O = 100 - (C + H + N + S + \text{ash})$, more details found as Samaksaman *et al.* (2021). In addition, the high heating value (HHV) was carried out based on analysis of the calorific value using a bomb calorimeter (Leco, AC-500) in an atmosphere of oxygen.

Fourier transform infrared spectrometry (FTIR)

Raw materials and PET/biomass pellets were tested for the determination of the functional groups of polyethylene terephthalate (polyester group) and biomass components (lignocellulose group) using FTIR technique (Perkin Elmer, Spectrum GX). The IR spectra were recorded at wavenumbers in the range 4,000 to 400/cm at a resolution of 1/cm. The data were interpreted using the Perkin Elmer Spectrum version 10.5.2 software.

Heavy metal and ionic leachability tests

The PET waste and biomass may become contaminated with heavy metals and minerals during the process of mechanical recycling and sawing. These metals may cause scratching and corrosion of machinery. Heavy metals concentration such as copper (Cu), zinc (Zn), chromium (Cr), cadmium (Cd), and lead (Pb) were investigated. The procedure was detailed in a previous work of Samaksaman *et al.* (2015). Samples of PET waste, biomass, and PET/biomass pellets were digested using an acidic solution (a mixture of hydrochloric and nitric acids), followed by dilution with deionized (DI) water and filtration. Atomic absorption spectrometry (GBC, Avanta PM) was used in the subsequent analysis. The standardized curves with coefficient of determination (R^2) values of 0.95-0.99 were used to evaluate heavy metal concentrations. The leachability test was used to determine the concentration of ionic leachates from the samples. The test applies quantity analysis of elements for seven anions leachate (AnL): fluoride (F^-), chloride (Cl^-), nitrite (NO_2^-), bromide (Br^-), nitrate (NO_3^-), phosphate (PO_4^{3-}), and sulfate (SO_4^{2-}); and for six cations leachate (CaL): lithium (Li^+), sodium (Na^+), ammonium (NH_4^+), potassium (K^+), magnesium (Mg^{2+}), and calcium (Ca^{2+}). One gram of each sample

was placed in a centrifuge tube and mixed with 10 ml DI water. A shaking incubator (JSR, Jssi-300C) was used for extraction at 250 rpm at room temperature for 12 hours. Then the sample was filtered twice using a paper filter before passing through a nylon filter. Prior to measurement, dilution of the sample (1:100 v/v) with ≥ 18 (m Ω) DI water was carefully conducted in a clean room. The sample solution was analyzed for anions and cations using ion chromatography (Dionex DX500). All experiments were done three replications.

Ash composition analysis

X-ray fluorescence (XRF) spectroscopy is a rapid method used to determine the composition in solid samples like ash and residue. It is a non-destructive technique and suitable for ash samples from the combustion test. The ash samples were the as-receive samples from the combustion of raw materials and pellets within a muffle furnace at 815 °C for 2 hours (Xing *et al.*, 2016). The test was employed using an energy dispersive XRF (EDXRF, Horiba XGT-5200 X-ray Analytical Microscope) with Rh X-ray tube, 1.2 mm of XGT diameter, and 30 kV of X-ray tube voltage.

Thermogravimetric analysis

A simultaneous thermal analyzer (Mettler-Toledo, TGA-DSC II) was used to measure the thermal decomposition of samples. Thermogravimetric analysis and derivative thermogravimetry (DTG) were used to analyze samples of PET waste, biomass, and PET/biomass pellets. Raw materials were analyzed in an atmosphere of oxygen, over a temperature range of 50-800 °C at a heating rate of 15 °C/min. The PET/biomass pellets were analyzed under the same conditions as the raw materials but various heating rates of 5, 10, 15, and 20 °C/min.

Kinetic analysis

The combustion of carbonaceous material is considered in term of a gas-solid heterogeneous reaction. The conversion (X) of PET waste, biomass, and pellets during the combustion process can be defined using Eq. 1.

$$X = \frac{m_0 - m_t}{m_0 - m_f} \quad (1)$$

Where, m_0 , m_t , and m_f are the initial sample mass, the sample mass at time t , and the sample mass left

after ending process, respectively. The combustion rate of the materials can be represented using Eq. 2.

$$\frac{dX}{dt} = k(T)f(X) \quad (2)$$

Where, k is the combustion rate constant and $f(x)$ is the reaction model expressing the dependence of the combustion rate. The combustion rate constant (k) is temperature-dependent and generally is described using the Arrhenius formula using Eq. 3 (Mishra *et al.*, 2019).

$$k = Ae^{-E_a/RT} \quad (3)$$

Where, E_a is the activation energy (kJ/mol), A is the pre-exponential factor (s^{-1}), T is the absolute temperature (K), and R is the universal gas constant, 8.314 kJ/(kmol·K). A heterogeneous function of the uniform kinetic reaction of the first order ($n = 1$), $f(x)$ can be written using Eq. 4.

$$f(X) = (1 - X) \quad (4)$$

Thus, the rate constant can be expanded using Eq. 5.

$$\frac{dX}{dt} = Ae^{-E/RT}(1 - X) \quad (5)$$

In non-isothermal analysis, the heating rate varies with the reaction time and temperature at a constant heating rate, $\beta = \frac{dT}{dt}$ that can be rewritten as Eq. 6.

$$\frac{dX}{dT} = \frac{A}{\beta} e^{-E/RT}(1 - X) \quad (6)$$

The integral form of $f(X)$ can be represented as $g(X)$ by integrating Eq. (6) with respect to temperature as shown in Eqs. 7 and 8.

$$g(X) = \int_0^X \frac{dX}{f(X)} = \frac{A}{\beta} \int_{T_0}^T e^{-E/RT} dT \quad (7)$$

$$g(X) = \frac{AE}{\beta R} \int_0^X u^{-2} e^{-u} du = \frac{AE}{\beta R} p(X) \quad (8)$$

Kissinger-Akahira-Sunose method

The Kissinger-Akahira-Sunose (KAS) method (Akahira and Sunose, 1971) is an iso-conversional method that uses an approximation of $p(X) = x^2 e^{-x}$ in Eq. (8) to determine E_a as shown in Eq. 9.

$$\ln\left(\frac{\beta}{T^2}\right) = \ln\left(\frac{AE_a}{Rg(X)}\right) - \left(\frac{E_a}{RT}\right) \quad (9)$$

Plotting $\ln\left(\frac{\beta}{T^2}\right)$ versus $1/T$ will obtain the slope and intercept that can be used to estimate E_a and A , respectively.

Ozawa-Flynn-Wall method

The Ozawa-Flynn-Wall (OFW) method is a model-free method applying Doyle's approximation in Eq. (8) to evaluate the activation energy (Tran *et al.*, 2014) as shown in Eq. 10.

$$\ln(\beta) = \ln\left(\frac{AE_a}{Rg(X)}\right) - 5.3305 - 1.052\left(\frac{E_a}{RT}\right) \quad (10)$$

E_a was determined using the least square regression line as the slope of $1.052E_a/R$ by plotting the graph between $\ln(\beta)$ on the y-axis versus $1/T$ on the x-axis.

Starink method

The Starink method is a model-free method like the KAS and OFW methods; however, the value it produces for E_a is more accurate than from using the other two methods (Gai *et al.*, 2013). The Starink equation is shown in Eq. 11.

$$\ln\left(\frac{\beta}{T^{1.8}}\right) = C_s - 1.0037\left(\frac{E_a}{RT}\right) \quad (11)$$

Plotting a linear graph of $\ln\left(\frac{\beta}{T^{1.8}}\right)$ and $1/T$ enables the values of E_a and A to be determined from the slope and intercept, respectively.

Thermodynamic analysis

Thermodynamic parameters can be presented by the change in the enthalpy (ΔH), Gibbs free energy (ΔG), and entropy (ΔS) (Manatura, 2019). In brief, ΔH represents an endothermic or exothermic reaction which is the nature of the reaction process. ΔG indicates the energy related to a chemical reaction

that can be used to do work and ΔS measures the irreversibility of the system. The parameters can be calculated using Eqs. 12-14.

$$\Delta H = E_a - RT \quad (12)$$

$$\Delta G = E_a + RT_p \ln\left(\frac{K_B T_p}{hA}\right) \quad (13)$$

$$\Delta S = \frac{\Delta H - \Delta G}{T_p} \quad (14)$$

Where, K_B , h , and T_p are the Boltzmann constant = 1.381×10^{-23} J/K, Plank constant = 6.626×10^{-34} J.s and peak temperature, respectively.

RESULTS AND DISCUSSION

Properties and fuel characteristics

Table 1 shows that the proximate and ultimate analysis for entire PET waste, biomass, and PET/biomass pellet samples. The PET waste had lower moisture and ash content compared to the biomass. The PET waste contained large amounts of volatile matter due to the nature of the plastic material (Luo *et al.*, 2018; Zhao *et al.*, 2016). The PET/biomass pellets also contained a large amount of volatile matter (83.19%). However, it had higher ash content (4.62%) and fixed carbon (6.83%) than the biomass. These changes in the physical properties of the pellets were due to the mixing ratio and the pelletizing process. Subsequently, the HHV of pellets was 19.20 MJ/kg via the calorimetric test and was slightly different from the HHV of pellets via calculation (Huang and Lo, 2020). Mass and energy densification by pelletizing could overcome the disadvantages of PET waste and biomass. The H/C and O/C atomic ratios of PET waste, biomass, and pellet samples were in the range of 0.08–0.13 and 0.85–1.47, respectively. The H/C value for the PET waste was different compared to the biomass and was linked to the H/C value of the PET/biomass pellets. The PET/biomass pellets had a

Table 1: Properties of the raw materials and PET/biomass pellets

Sample	Proximate analysis (%wt)				Ultimate analysis (%wt)					H/C	O/C	HHV (MJ/kg)
	MC	VM	FC	Ash	C	H	N	O	S			
PET	5.09	87.80	5.02	2.09	38.41	3.02	0.08	56.40	ND	0.08	1.47	21.77
Biomass	7.78	78.33	8.26	5.63	47.58	6.32	0.06	40.41	ND	0.13	0.85	17.39
Pellets	5.36	83.19	6.83	4.62	39.33	3.35	0.07	49.43	ND	0.09	1.26	19.20

ND: not detected

higher H/C value mainly due to the PET composition. The percentage of N showed similar results in the range 0.06-0.08% for the raw materials, while the percentage of S was untraceable in all samples. Previous works reported that C and H contents in PET samples were higher compared to the current study (Luo *et al.*, 2018; Surenderan *et al.*, 2018; Zhao *et al.*, 2016). The N and S content was substantially low. Low emissions of oxides of nitrogen (NO_x) and sulfurdioxide (SO_2) could be expected when this PET/ biomass pellet was used as fuel in the combustion or co-combustion processes (Edo *et al.*, 2016). The current results from the proximate and ultimate analysis suggested that the purity of the PET waste was an important factor in determining their characteristics. Compared to PET waste from bottles, the pure-PET had less contaminants and decay materials from the mechanical process, it would be clearly resulted as a previous work by Jabłońska *et al.* (2019). In the pelletizing of PET waste and biomass into a fuel pellet, the PET plastic played a key role in the compaction and made the pellets harden, as seen in Fig. 1. In conclusion for this sub-section, the PET/biomass pellets could be used as alternative sources and in combination with solid fuels in the same manner as refuse-derived fuel (RDF) and solid recovered fuel (SRF).

FTIR results

The FTIR technique allowed the identification of the functional groups present in the polyethylene terephthalate (polyester group) and biomass components (lignocellulose group). Fig. 2a-c illustrates the IR spectra of samples of PET waste, biomass, and PET/biomass pellets. The identified vibrational groups are summarized in the Table 2. The bands were in agreement with previous works (Chen *et al.*, 2013; Lopes *et al.*, 2018; Manatura, 2020; Pereira *et al.*, 2017; Taleb *et al.*, 2020). The IR spectrum of PET waste (Fig. 2a) contained bands at 2,916/cm (C-H, stretch), 1,714/cm (C=O, stretch), and 1,338-1,408/cm (C-O, stretch). The main observation at 1,247/cm and 1,125/cm affirmed the terephthalate group ($\text{OOC}_6\text{H}_4\text{-COO}$). The same results for the PET functional groups were suggested by previous researchers (Chen *et al.*, 2013; Pereira *et al.*, 2017). In addition, the IR bands of the PET waste at around 1,090, 950-1,016, 796, and 439/cm, represented groups of Si-O asymmetric, Si-O(H) asymmetric, Si-O symmetric, and Si-O-Si bending, respectively (Capeletti and Zimnoch, 2016; Pereira *et al.*, 2017). These results were due to the characteristics of soil and sand that remained in the PET waste from the mechanical recycling process of the PET bottles. The IR spectrum of the biomass sample in Fig. 2b shows

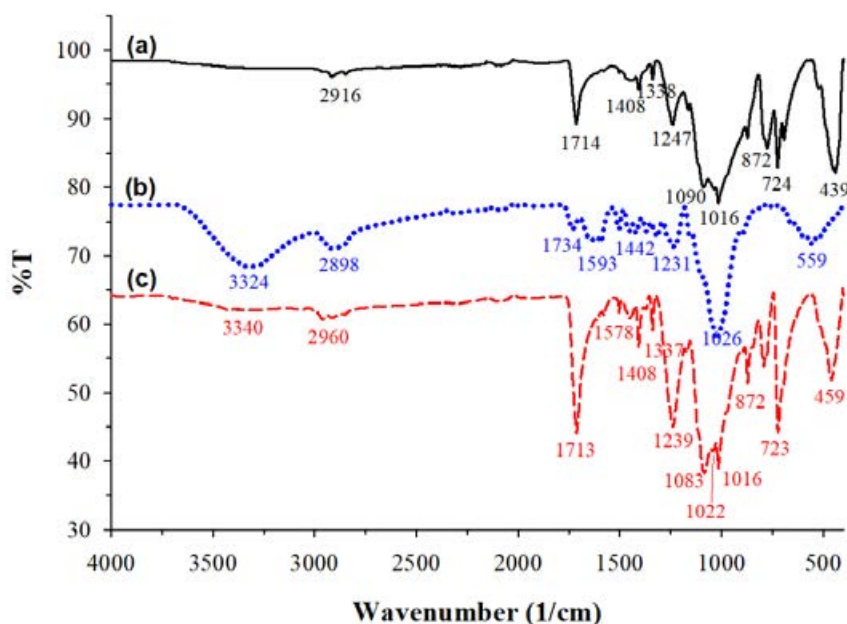


Fig. 2: IR spectra of samples; (a) PET waste, (b) biomass, and (c) PET/biomass pellets

Table 2: Characteristics bands of the PET waste, biomass, and pellets obtained by FTIR

Wavenumber (1/cm)			Characteristics bands
PET	Biomass	Pellets	
-	3,324	3,340	Hydroxyl (O-H) groups
2,916	-	2,960	C-H stretching in aromatic methoxyl groups and methylene groups
-	2,898	-	C-H stretching in the methyl groups
-	1,734	-	Stretching of C=O of the carbonyl, carboxyl, and acetyl groups, and of xylans
1,714	-	1,713	The non-conjugated carbonyl group stretching
-	1,593	1,578	Aromatic skeleton vibration (C=C) of lignin
-	1,442	-	C-H deformation in lignin and carbohydrates, CH ₂ stretching, and CH ₃ asystematic
1,338-1,408	-	1,337-1,408	Stretching of C-O group deformation of the O-H group and bending and wagging vibrational modes of the ethylene glycol segment
1,247&1,125	-	1,252-1,239	Terephthalate group (OOC ₆ H ₄ -COO)
-	1,231	-	C-C, C-O, and C=O stretching
1,090	-	1,083	Si-O asymmetric
-	1,026	1,022	Stretching of C-O of the ester-methyl group of lignin
950-1,016	-	1,016	Si-O(H) asymmetric
796	-	723	Si-O symmetric
-	559	-	C-H bending
439	-	459	Si-O-Si bending

a band at 3,324/cm (O-H group) due to the water content in the biomass sample. In addition, there were bands at 2,898/cm (C-H stretch), 1,734/cm (C=O, stretch), 1,593/cm (C=C, aromatic skeleton vibration), 1,442/cm (C-H, CH₂, stretch, and CH₃, asystematic), 1,231/cm (C-C, C-O, and C=O, stretch), 1,026/cm (C-O, stretch), and 559/cm (C-H, bend). Similar results for the functional groups of lignocellulosic materials have been reported using FTIR in teak hardwood, coffee grounds, and sugarcane bagasse (Lopes *et al.*, 2018; Manatura, 2020; Taleb *et al.*, 2020). The PET/biomass pellets characterization interpreted the mixture between PET waste and biomass as seen in Fig. 2c. While functional groups were contributed by both raw materials, the mixture was mainly characterized by the functional groups from the PET waste at around 1,252-1,239/cm, represented groups of terephthalate. Even though the biomass made up a small proportion of the pellets, some biomass characteristics were evident in the band at 1,578/cm that showed vibration of the aromatic skeleton with stretching (C=C) of lignin and at 1,022/cm representing C-O stretching of the ester methyl group of lignin. However, silica group occurred bands at 1,016, 723, and 459/cm in the sample of PET/biomass pellets due to the contamination was resulted from earth materials as found in the sample of PET waste.

Heavy metal concentration and ionic leachability

Table 3 summarizes the results of heavy metal concentration and ionic contaminants of the raw materials and PET/biomass pellets. The concentrations of Pb, Zn, Cr, and Cu in all tests of raw material samples were below regulatory levels and none of the tests yielded evidence of Cd. These results confirmed that the heavy metals (Cd~Cu<Pb<Cr<Zn) content in PET/biomass pellets were below the limits set by some European countries (Finland, Italy, France and the Netherlands) for alternative solid fuels such as RDF and SRF (Zhao *et al.*, 2016). In addition, the contaminant leaching of AnL and CaL was also investigated and reported in Table 4. The PET waste had a high content of chloride (Cl⁻) of around 3.99 ppm and the other six tested anions were recorded in the range from non-detectable to 0.33 ppm, while for the cations, sodium (Na⁺) was the highest (28.35 ppm). The biomass sample had a substantially amount of the phosphate (PO₄³⁻) anion at 58.58 ppm with lesser amounts of the other three cations-potassium (K⁺) at 28.57 ppm, sodium (Na⁺) at 16.21 ppm, and magnesium (Mg²⁺) at 7.42 ppm. The experimental results showed that pellets characterization regarding ionic leachability was based on the combined properties of the PET waste and biomass. The major ionic elements in

the pellets were not higher than in the original biomass for PO_4^{3-} , Cl^- , F^- , SO_4^{2-} , K^+ , Mg^{2+} , and Ca^{2+} and furthermore, were not higher than in the PET waste for Cl^- and Na^+ . The high sodium (Na^+) cation level in the PET waste and the pellets might have been affected by the caustic soda (NaOH) detergent used during the cleaning step in the mechanical recycling process for the PET bottles.

Ash composition

The ash composition analysis obtained by EDXRF technique showed in Table 5. The ashes from combustion of biomass, PET, and PET/biomass pellets would consist of the most common oxides which formed during the sintering process (Lu *et al.*, 2015). Silicon oxide (SiO_2), iron (III) oxide (Fe_2O_3), calcium oxide (CaO), magnesium oxide (MgO), and aluminium oxide (Al_2O_3) were the major components of all ash samples. The content of SiO_2 in biomass ash was 48.88%wt or more suggested by Xing *et al.* (2016) for the raw materials of biomass, which was much lower

than PET (78.24%wt) and pellets (80.68%wt). While the lower SiO_2 (10.32%wt) composition of waste from PET bottles washing has been reported by Jabłońska *et al.* (2019). SiO_2 composition occurred in the samples of waste PET and PET/biomass pellets due to the contamination was mainly resulted from earth materials. The biomass ash had a substantially amount of phosphorus (V) oxide (P_2O_5), and potassium oxide (K_2O) were 6.14% and 4.23%, respectively. Chromium (III) oxide (Cr_2O_3), copper oxide (CuO), zinc oxide (ZnO), titanium dioxide (TiO_2) were found in the ash of pellets sample. These metal-oxides fractions in the ash were supposed the effect of aggregation of metal elements with oxygen during combustion process (Samaksaman *et al.*, 2015). The appearance of metal elements such as CrO , CuO , ZnO , and TiO_2 might be released during the process of PET-bottle grinding and PET/biomass pelletizing. Sulfur trioxide (SO_3) was also found in the low value. Trace metals such as manganese (IV) oxide (MnO_2), strontium oxide (SrO), zirconium dioxide (ZrO_2), and tantalum (V) oxide

Table 3: The results of heavy metal concentration

Sample	Heavy metal concentration (mg/kg)				
	Pb	Zn	Cd	Cr	Cu
Limits ^a	200	500	5	100	300
PET	ND	3.5	ND	9.4	5.0
Biomass	ND	0.7	ND	1.6	1.4
Pellets	0.8	7.4	ND	5.2	ND

^a The limits set by European countries for the RDF and SRF.

ND: not detected

Table 4: The results of anionic and cationic leachability

Sample	ANL (ppm)							Cal (ppm)					
	F ⁻	Cl ⁻	NO ₂ ⁻	Br ⁻	NO ₃ ⁻	PO ₄ ³⁻	SO ₄ ²⁻	Li ⁺	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺
PET	0.06	3.99	ND	ND	ND	ND	0.33	ND	28.35	ND	0.55	1.52	1.82
Biomass	0.32	4.61	ND	ND	ND	58.58	1.75	ND	16.21	ND	28.57	7.42	3.88
Pellets	0.06	3.25	ND	ND	ND	16.96	0.62	ND	20.88	ND	10.70	2.71	2.03

ANL: Anions leachate; Cal: Cations leachate; ND: not detected

Table 5: Composition of the different ash samples

Sample	Ash composition (%wt)														
	MgO	Al ₂ O ₃	SiO ₂	P ₂ O ₅	SO ₃	K ₂ O	CaO	TiO ₂	Cr ₂ O ₃	MnO ₂	Fe ₂ O ₃	CuO	ZnO	SrO	ZrO ₂
PET	3.29	4.23	78.24	0.15	0.27	0.75	3.93	0.95	0.08	0.10	7.88	0.05	0.04	ND	0.03
Biomass	9.21	10.02	48.88	6.14	0.64	4.23	16.08	0.49	0.06	0.08	4.11	0.01	0.01	0.02	0.01
Pellets	2.86	2.63	80.68	1.27	0.13	0.65	3.80	0.53	0.45	ND	6.94	0.03	0.03	ND	ND

ND: not detected

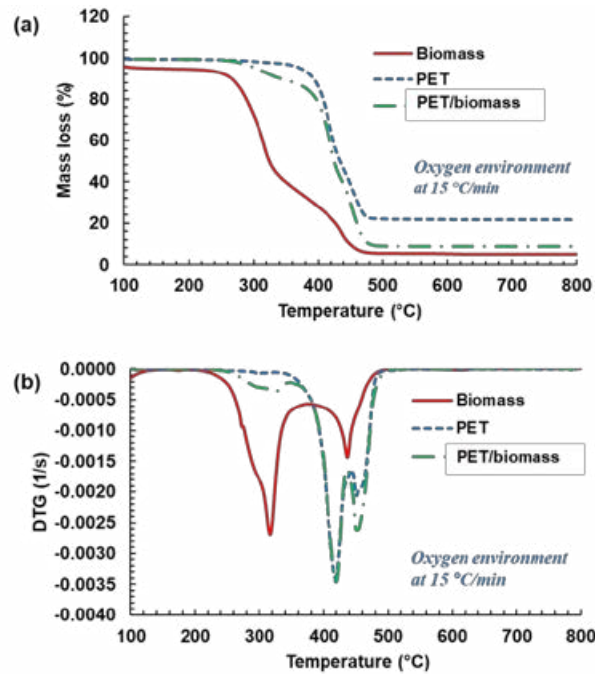


Fig. 3: (a) TGA and (b) DTG for combustion of waste of PET (PET), biomass (Biomass), and pellets (PET/biomass) at 15 °C/min

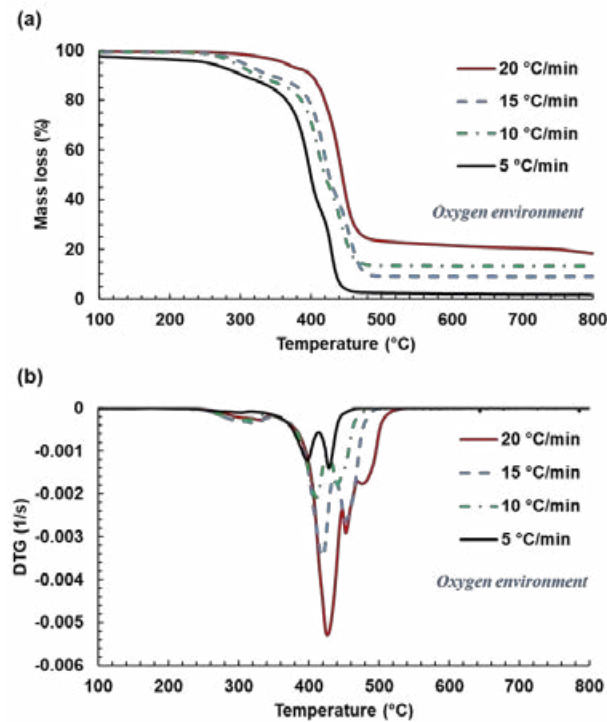


Fig. 4: (a) TGA and (b) DTG for combustion of pellets at 5-20 °C/min

(Ta₂O₅) were untraceable in the ash of pellets sample. It is proven that the ash characterization of pellets exhibited unique characters based on the combined properties of the PET waste and biomass. Overall, the results of properties and fuel characteristics, FTIR, heavy metal concentration and ionic leachability, and ash composition made the PET/biomass pellets useful in a co-firing process with traditional solid fuels. Moreover, the PET/biomass pellets can further be upgraded to other fuels production via modern bioenergy conversion processes.

TGA and DTG results

The combustion characteristics of PET waste and the raw biomass at 15 °C/min are shown in the TG and DTG curves in Fig. 3. Both waste of PET and biomass are different in chemical structure, but the tests could be compared because used the same condition of oxygen atmosphere and a heating rate. The reaction temperature varied in the range 100 to 800 °C. It was clear that the biomass characteristics differed noticeably compared to those of the PET. For the biomass, below 150 °C represented removing vapor and light volatile matter (Ahmad *et al.*, 2017). The mass loss of about 5.80% was represented as the water content in biomass (Fig. 3a). The clear mass loss of the biomass commenced from 190 up to 500 °C. Two clear peaks were observed during combustion with maximum mass loss rates of 0.0027 s⁻¹ at 317 °C and 0.0014 s⁻¹ at 437 °C, respectively. The first and second peaks were caused by the release of volatile matter in the temperature range from 148 to 372 °C and combustion of the remaining char,

respectively (Ahn *et al.*, 2014). For all samples of the PET waste and PET/biomass pellets, no obvious mass loss was detected below 266 °C. However, a small peak was observed with 0.0003 s⁻¹ at 325 °C for the mixed PET with biomass related to the level of higher volatiles in the biomass. The distinct mass loss and rate of mass loss of the PET waste and the pellets started at 278 °C and finished at around 458 °C. Two peaks were observed in Fig. 3b, with the first peak being for PET waste and pellets that were very similar to each other with a maximum mass loss rate of 0.0034 s⁻¹ at 419 °C. The second peak had the same peak temperature (T_p) at 454 °C with the mass loss rate for pellets of 0.0026 s⁻¹ and for PET waste of 0.0020 s⁻¹, respectively. It was clear that the biomass blending supported reactivity of the process. In addition, the experimental results affirmed that the decomposition of material with complex structure depended on the loss of chemical bonding and properties of materials. The combustion properties of the PET waste, biomass, and pellets are listed in Table 6. The ignition temperature (T_{ig}) and peak temperature (T_p) of PET were consistent with Das and Tiwari (2019). The results indicated that a low content (10%wt) of biomass mixed with PET improved T_{ig} by lowering it. There were no obvious changes in other properties such as the burnout temperature (T_b), higher reactivity (R_{avg}), and maximum derivative thermogravimetric value (DTG_{max}). The effects of the heating rate (5, 10, 15 and 20 °C/min) on mass loss and the rate of mass loss in terms of combustion temperature are shown in Fig. 4. The mass loss and rate of mass loss curves

Table 6: Thermal decomposition of samples in combustion at 15 °C/min

Sample	T _{ig} (°C)	T _b (°C)	T _p (°C)	DTG _{max} (1/s)	R _{avg} (1/(s·K))
PET	390	475	419	-0.0034	-4.92E-06
Biomass	278	458	317	-0.0027	-4.56E-06
Pellets	380	471	420	-0.0035	-4.99E-06

Table 7: Combustion characteristics of pellets in combustion at various heating rates

Hearing rate (°C/min)	T _{ig} (°C)	T _b (°C)	T _p (°C)	DTG _{max} (1/s)
5	360	460	398	-0.0012
10	378	460	411	-0.0021
15	380	471	420	-0.0035
20	398	499	426	-0.0053

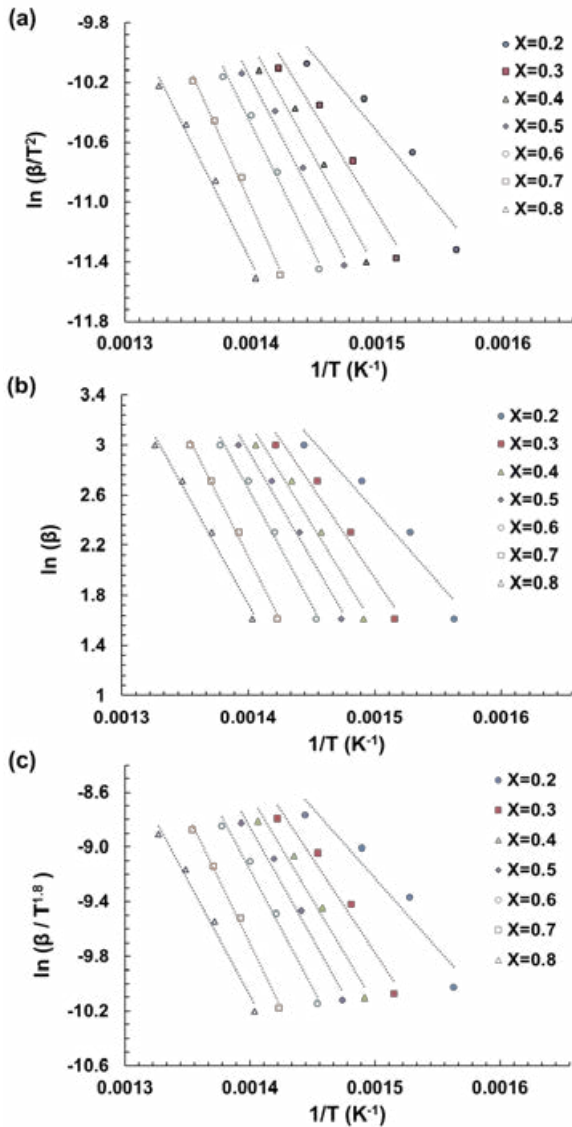


Fig. 5: Linear regression lines; (a) KAS, (b) OFW, and (c) Starink at various conversions (X)

were parallel and similar. A higher heating rate required a larger temperature range for complete combustion because there was not sufficient time for the biomass to combust (Manatura *et al.*, 2018). Table 7 shows the combustion characteristics of pellets for the four heating rates. With an increased heating rate, the values of T_{ig} , T_b , and T_p increased as did DTG_{max} , indicating that the lower heating rate produced more effective combustion than the higher heating rate (Lu and Chen, 2015).

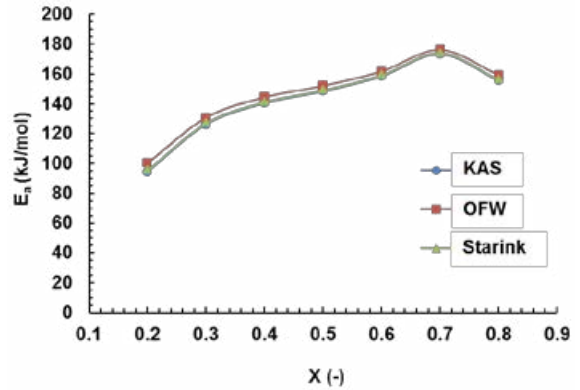


Fig. 6: E_a vs X for the KAS, OFW, and Starink

Kinetic analysis

The iso-conversional KAS, OFW and Starink methods from Eq. (9)–(11) were used to determine the kinetic factors (E_a and A) in this study. The typical linear regression lines of the KAS, OFW, and Starink methods fitted with conversion (X) in the range 0.2 to 0.8 are shown in Fig. 5. The estimates of E_a and A from the models were very close in value, as shown in Table 8. The quality of linear fitting is shown by R^2 , with the values being mostly higher than 0.96 which implied the simulations had a good fit with the experimental data. Fig. 6 illustrates the variation of E_a in terms of X. The kinetic factors E_a and A showed similar trends by increasing first for X values of 0.2 to 0.7, then decreasing at 0.8. The minimum and maximum E_a values were at X values of 0.2 and 0.7, respectively. It was noticed that a rapid increase in E_a occurred between X values in the range 0.6–0.7 because almost all the volatile matter was expelled from the char residue. Moreover, E_a rapidly dropped for X values in the range 0.7–0.8 due to the sudden char combustion at the higher temperature in the oxygenated atmosphere (Das and Tiwari, 2019). The average E_a value for pellets was in the range 142 to 146 kJ/mol for all prediction models, with these values in accordance with previous work of Das and Tiwari (2019), as was the average value of A represented in the range 6.70×10^{12} to $1.83 \times 10^{15} \text{ min}^{-1}$.

Thermodynamic parameters

To design a high performance macro-scale combustor requires not only choosing suitable bio-fuels and kinetic parameters but also requires

Table 8: Thermokinetic factors of pellets for three models

X (-)	KAS			OFW			Starink		
	E _a (kJ/mol)	A (min ⁻¹)	R ² (-)	E _a (kJ/mol)	A (min ⁻¹)	R ² (-)	E _a (kJ/mol)	A (min ⁻¹)	R ² (-)
0.2	94.31	2.76E+07	0.92	100.16	8.21E+09	0.93	95.06	7.48E+09	0.92
0.3	126.15	1.07E+10	0.96	130.65	1.90E+12	0.97	126.80	2.91E+12	0.96
0.4	140.41	1.53E+11	0.97	144.34	2.26E+13	0.97	141.03	4.15E+13	0.97
0.5	148.37	6.66E+11	0.98	152.01	9.02E+13	0.98	148.97	1.81E+14	0.98
0.6	158.46	3.83E+12	0.99	161.72	4.66E+14	0.99	159.04	1.04E+15	0.99
0.7	173.42	4.06E+13	1.00	176.14	4.29E+15	1.00	173.97	1.11E+16	1.00
0.8	155.58	1.62E+12	0.99	159.35	2.17E+14	0.99	156.21	4.44E+14	0.99
Avg.	142.38	6.70E+12	-	146.34	7.26E+14	-	143.01	1.83E+15	-

Table 9: Thermodynamic analysis of pellets at 15 °C/min

X (-)	A (s ⁻¹)	ΔH (kJ/mol)	ΔG (kJ/mol)	ΔS (J/mol·K)
0.2	7.63E+04	88.73	204.07	-166.50
0.3	2.57E+07	120.45	202.40	-118.29
0.4	3.40E+08	135.09	201.78	-96.27
0.5	1.43E+09	142.55	201.46	-85.05
0.6	8.82E+09	152.56	201.08	-70.04
0.7	1.30E+11	167.41	200.56	-47.85
0.8	5.25E+09	149.48	201.19	-74.64
Avg.	2.08E+10	136.61	201.79	-94.09

investigating the thermodynamic parameters (Xu and Chen, 2013). Thermodynamic factors such as A, ΔH, ΔG, and ΔS evaluated using the OFW method under the TGA conditions of a heating rate at 15 °C/min, are shown in Table 9. The A value increased from 7.63×10⁴ to 1.30×10¹¹ for X values in the range from 0.2 to 0.7, respectively, and then decreased to 5.25×10⁹ at an X value of 0.8. The lowest and highest values of A were 7.63×10⁴ and 1.30×10¹¹ at X values of 0.2 and 0.7, respectively. The higher values of A were due to elevated numbers of molecular collisions so more heat was generated which was related to the values of E_a (Fong et al., 2019). On the other hand, the lowest A indicated a restriction in particle rotation of the activated complex compared to the initial reagent, indicating a large surface reaction (Zhang et al., 2016). ΔH is the deviation of energy between the reagent and the activated complex depending on the activation energy (Xu and Chen, 2013). ΔH increased from 88.73 to 167.41 kJ/mol and decreased to 149.48 kJ/mol with an average of

136.61 kJ/mol during the conversion. It was noticed that the deviation between E_a and ΔH was very low (about 10 kJ/mol) which implied that this reaction was simple to achieve product formation (Ahmad et al., 2017). Positive values of ΔH represent an endothermic reaction which requires an external heat source to break and form new chemical bonds. The ΔG value refers to the amount of available energy for the formation of activated complexes (Laougué and Merdun, 2020). It was quite steady for X values from 0.2 to 0.8 with an average ΔG of 201.79 kJ/mol which was consistent with Das and Tiwari (2019). This indicated that the combustion of the mixed PET waste with biomass consumed more energy compared to chicken manure (163.37-165.39 kJ/mol) (Yuan et al., 2017). The high ΔG value showed that the PET/biomass pellets were an optimal choice for converting the PET waste and biomass to energy. The values of E_a, ΔH, and A were the highest at an X value of 0.8 for combustion. The ΔS value measures the disorder or randomness of energy and matter in

a system and shows how near of far the state of the sample is from its own thermodynamic equilibrium. Entropy also shows the degree of arrangement of the carbon layers in samples (Xu and Chen, 2013). Positive values indicate a high affinity of the sorbent and negative values indicate that the adsorption process is mainly driven by ΔS . In the current study, the ΔS values were negative regardless of the model and process used. The higher values of ΔS occurred between X values of 0.2 and 0.3 (-166.50 and -118.29 J/(mol.K)), respectively. It was observed that the trend for ΔS was opposite that of E_a , ΔH , and ΔG .

CONCLUSION

Increase in fuel pellets demand coupled with rising environmental issues of the secondary waste from the plastic recycling system have attracted researcher to explore. Fuel pellets production by pelletizing is an efficient method for manipulating waste of PET from a mechanical recycling plant and local biomass like teak sawdust. It is a useful tool to manage the waste of PET and biomass in terms of dusty forms (macro-, meso-, micro-, and nano-sizes) and plays a role on reduce the loads of plastic waste pollution that releases into the soil, public water, as well as air emissions. Notice, the reduction of loads of plastic waste dumps into landfills and to prevent the improper disposal of plastic waste are considered. In further, it can adapt into a concept of waste to energy following by converting to fuel pellets and using with industrial furnaces i.e. using as substituent fuels in cement kiln, boiler, power plant, incinerator, and so on. Pelletizing was employed to convert waste of PET and biomass into a potential solid fuel with the fit ratio of 9:1 (PET:biomass). This ratio gives the durability of PET/biomass pellets, a uniform dimension, ease handling, storage, and transportation common as woody pellets. Some technical challenges such as low moisture content and high volatile matter content were feedstock dependent. The PET/biomass pellets had characteristics and physico-chemical properties that were a combination of those of the PET waste and biomass. A high heating value of the PET/biomass pellets (HHV = 19.20 MJ/kg) was obtained from the results of pelletizing the complex solid fuels. The HHV value was slightly decreased by adding biomass. The complex structure of ethylene terephthalate and lignocellulose was clearly identified by FTIR technique. Heavy metals ($Cd \sim Cu < Pb < Cr < Zn$) and

ionic (7 AnLs and 6 CaLs) contaminants had lower concentration than the limitation of the regulations. The results of ash composition affirmed that the ash of PET/biomass pellets was a non-hazardous residue which was unique characters based on feedstock. Therefore, the PET/biomass pellets can be used without harms of the environment and health impacts. Earth materials (SiO_2 composition) and a detergent (Na^+ element of caustic soda) from step PET flake cleaning had a slightly effect on the alteration of residue chemicals. However, biomass supplied as part of PET plastic waste improve combustion kinetics such as T_{ig} , T_b , R_{avg} , and DTG_{max} . Lower ignition temperature, burnout temperature, reactivity, and DTG_{max} were affected by the blended biomass in the PET/biomass pellets that could be attributed to the combined effect of terephthalate and lignocellulose groups present in the pellet matrix. Subsequently, the combustion characteristics and kinetics of PET/biomass pellet samples were examined via the iso-conversional methods of KAS, OFW, and Starink. The results exhibited similar trends and values for the activation energy (E_a) of 142.38, 146.34, and 143.01 kJ/mol, respectively, with the values of conversion ranged from 0.2 to 0.8. The thermodynamic factors of ΔH , ΔG , and ΔS were 136.61 kJ/mol, 201.79 kJ/mol, and -94.09 J/mol \times K, respectively. These findings may provide comprehensive knowledge to assist the design and operation of a combustion system for adding PET/biomass pellets in a co-firing process with traditional solid fuels. Moreover, PET/biomass pellets can further be upgraded to other fuels production via modern bioenergy conversion processes.

AUTHOR CONTRIBUTIONS

K. Manatura performed the literature review, writing original draft, conceptualization, methodology, investigation, data analysis, visualization, and manuscript edition. U. Samaksaman performed the literature review, experimental design, analyzed and interpreted the data, research summary and recommendation, and manuscript edition.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

%	Percentage
% wt	Weight percentage
°C	Degrees celsius
°C/min	Heating rate (degree of temperature per time)
ΔG	Gibbs free energy
ΔH	Enthalpy
ΔS	Entropy
A	The pre-exponential factor
Al_2O_3	Aluminium oxide
AnL	Anions leachate
β	The heating rate varies with the reaction time and temperature at a constant heating rate
Br	Bromide
cm	Centimeter
1/cm	The unit of wavenumber of IR spectrum
C	Carbon
CaL	Cations leachate
CaO	Calcium oxide
Ca^{2+}	Calcium
Cd	Cadmium
Cl	Chloride
Cr	Chromium
Cr_2O_3	Chromium (III) oxide
Cu	Copper
CuO	Copper oxide
CEN/TS 15148	Method for the determination of the content of volatile matter

CEN/TS 14775	Method for the determination of ash content
DI water	The deionized water
DTG	Derivative thermogravimetry
DTG_{max}	Maximum derivative thermogravimetric value (1/s)
E_a	The activation energy (kJ/mol)
Eq.	Equation
EDXRF	Energy dispersive X-ray fluorescence spectroscopy
$f(x)$	The reaction model expressing the dependence of the combustion rate
F ⁻	Fluoride
FC	Fixed carbon
Fe_2O_3	Iron (III) oxide
FTIR	Fourier transform infrared spectrometry
$g(X)$	The integral form of $f(X)$
h	Plank constant 6.626×10^{-34} (J.s)
H	Hydrogen
HDPE	High density polyethylene
HHV	High heating value
H/C	Hydrogen to carbon atomic ratio
IR	Infrared radiation
k	The combustion rate constant
kg/min	Kilogram per minute
kV	Kilovoltage
K	Degrees Kelvin
K^+	Potassium
K_B	The Boltzmann constant 1.381×10^{-23} (J/K)
KAS	The Kissinger-Akahira-Sunose method
K_2O	Potassium oxide
Li^+	Lithium
mg/kg	Miligram per kilogram
ml	Milliliter
mm	Millimeter
m Ω	Milliohm
min ⁻¹ .	The unit of A interm of minute (pre-exponential factor)
m_0	The initial sample mass

m_f	The sample mass left after ending process
m_t	The sample mass at time t
MC	Moisture content
MJ/kg	Megajoules per kilogram
MgO	Magnesium oxide
Mg^{2+}	Magnesium
MnO_2	Manganese (IV) oxide
N	Nitrogen
Na^+	Sodium
$NaOH$	Sodium hydroxide (caustic soda)
NH_4^+	Ammonium
NO_x	Oxides of nitrogen
NO_2^-	Nitrite
NO_3^-	Nitrate
O	Oxygen
OFW	The Ozawa-Flynn-Wall method
O/C	Oxygen to carbon atomic ratio
ppm	Part per million
Pb	Lead
PET	Polyethylene terephthalate
$PET/biomass$	The mixture of PET and biomass
PO_4^{3-}	Phosphate
PP	Polypropylene
P_2O_5	Phosphorus(V) oxide
R	The universal gas constant 8.314 (kJ/(kmol·K))
R^2	The coefficient of determination value
R_{avg}	Higher reactivity
RDF	Refuse derived fuel
$Rh\ X-ray$	Rhodium X-ray tube
s^{-1}	The unit of A interm of second (pre-exponential factor)
S	Sulfur
SiO_2	Silicon oxide
SO_2	Sulfurdioxide
SO_3	Sulfur trioxide
SO_4^{2-}	Sulfate
SrO	Strontium oxide

SRF	Solid recovered fuel
<i>Starink</i>	The Starink method
T	The absolute temperature (K)
Ta_2O_5	Tantalum (V) oxide
TiO_2	Titanium dioxide
T_b	Burnout temperature
T_{ig}	Ignition temperature
T_p	Peak temperature
TGA	Thermogravimetric analysis
v/v	The volume ratio
VM	Volatile matter
X	The conversion of samples
XRF	X-ray fluorescence spectroscopy
$XGT\ diameter$	The X-ray irradiation diameter
Zn	Zinc
ZnO	Zinc oxide
ZrO_2	Zirconium dioxide

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REVIEW PAPER

Wastewater-based epidemiology for novel Coronavirus detection in wastewater

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ABSTRACT

The entire world is reeling under the worst pandemic of last 100 years. Over 128 million people have been infected with it and 2.8 million deaths have already taken place, till 30th March 2021. The identification of materials positive cases is the first step towards its containment and treatment. However, testing of individuals is an extensive, expensive and time-consuming exercise. In addition, societal taboos are also associated with infected individuals resulting in very few people volunteering for testing, esp. in the developing and under-developed world. An alternative approach that circumvents individual testing is the wastewater-based epidemiology. A state-of-the-art review of this method is provided in context of its utility for COVID-19 detection. This technique relies on collecting and testing samples from sewers and/or wastewater treatment plants for the presence of pathogens and then using that data to determine and predict the spread of the infection, thereby allowing the provision of appropriate containment and treatment steps. The study covers key aspects of wastewater-based epidemiology application for COVID-19 detection including its need, detailed process of detection and assessment, data analysis, economics and challenges to its application. Findings from a number of case studies are presented to elucidate the utility of this technique. It is clearly seen that WBE-based approach is a much better strategy as compared to individual testing and can be adopted to prevent further spread of Covid-19. The work is expected to further emphasize the application of this method for COVID (and other pandemic) detection and implementing containment strategies. This is clearly a much more economical and non-intrusive approach as compared to the individual testing.

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INTRODUCTION

Coronaviruses are “nonsegmented, enveloped, positive-sense, single-strand ribonucleic acid viruses”, and are part of the *Coronaviridae* family (Weiss and Leibowitz, 2011). There have been seven types of coronaviruses discovered which causes human sickness. The names of known coronaviruses till date are the alpha coronaviruses HCoV-229E and HCoV-NL63, the beta coronaviruses HCoV-OC43 and HCoV-HKU1, MERS-CoV which causes Middle east respiratory syndrome and SARS-CoV which causes Severe acute respiratory syndrome. SARS-CoV-2 is the 7th type and the most recent one to be found out (Lima, 2020). Four of these viruses i.e., alpha coronaviruses and beta coronaviruses induce (mild) respiratory symptoms. The other two viruses (Middle East respiratory syndrome or MERS, and Severe Acute Respiratory Syndrome or SARS) have been a cause for pandemics with a high fatality rate (Kooraki *et al.*, 2020). This is the third time, in last two decades, that a coronavirus infection has taken place (Guarner, 2020). The upsurge of SARS-CoV occurred in 2002 (Zhong *et al.*, 2003), MERS-CoV in 2012 (Nassar *et al.*, 2018), followed by the SARS-CoV-2 in 2019. SARS-Cov-2 was declared a pandemic by the World Health Organization (WHO) on 11th March, 2020 (Cucinotta *et al.*, 2020). As of March 8, 2021 – 117.47 million cases were recorded globally, in which 2.61 million fatalities were reported (Worldometer, 2021a). The SARS-CoV-2 is extremely infectious as compared to SARS-CoV and MERS-CoV. While, the mortality rate associated with the MERS-CoV was higher than that of both SARS-CoV and SARS-CoV-2, the SARS-CoV-2 spread extremely rapidly and infected a very large number of people globally (Meo *et al.*, 2020). Respiratory droplets and direct contact are the major pathways for the spread of SARS-CoV-2 (WHO, 2020). With the median of 2.79 and interquartile range (IQR) of 1.16, the R_0 was calculated to be around 3.28, which implies that an infected person will spread to another 3.28 cases (Liu *et al.*, 2020). The main strategies for control of this pandemic include isolation of positive cases, tracing of contacts (people who were in physical contact with the infected individual), and social/physical distancing (MacIntyre, 2020). The essence of wastewater-based epidemiology (WBE), known as Wastewater based epidemiology is that infected people shed the virus through excreta and the virus ends up in the wastewater treatment plant where the virus can be detected. This is a community-based method

that allows early and reliable detection of infections, and can help in intervention and control of infection spread. Not only viruses, the illegal use of drugs also can be monitored through the sewage. Some countries like China (Cyranoski, 2018), Spain (Huerta-Fontela *et al.*, 2008), France (Karolak *et al.*, 2010), and Canada (Metcalf *et al.*, 2010) use sewage to monitor the illicit use of drugs. Some research studies (Young *et al.*, 2020; Chen *et al.*, 2020; Xiao *et al.*, 2020) have demonstrated that RNA of SARS-CoV-2 is perceivable in the feces of COVID-19 patients. Chen *et al.* 2020 showed that RNA of SARS-CoV-2 can be diagnosed in human feces a few days to a week ahead of the symptoms onset. This is not the first-time wastewater-based epidemiology has come into rescue to monitor epidemic outbreaks. It proved as a promising approach in Brazil and Israel for detecting diseases like polio (Michael-Kordatou *et al.*, 2020). Sewage surveillance system of Israel, developed in 1989 by their national health authorities to notice poliovirus in wastewater, allowed them to track polio in urban wastewater treatment plants and sewage trunk lines at the time of the re-emergence of polio in 2013, and enabled them to react quickly (Brouwer *et al.*, 2018). It can also be used to access a situation when a new virus enters the community (Sinclair *et al.*, 2011). The biggest advantage of the wastewater-based surveillance for Covid- 19 is the unbiased results. It detects the virus even when the person is asymptomatic or presymptomatic. In addition, testing of sewage ensures that a composite of different human excretions (including, urine, feces, sweat, dead cells, saliva, etc.) is investigated thus providing a broader coverage of infections. Netherlands was one of the first countries to detect the traces of Covid-19 in sewage treatment plants (Hart *et al.*, 2020). Other countries which adopted the wastewater surveillance for routine Covid-19 detection are Australia, New Zealand, Netherlands (Aguar-Oliveira *et al.*, 2020). Nonetheless, WBE is not without its share of disadvantages, for example, it cannot help in finding specific infected individual in a community. Currently, wastewater treatment facilities serve only 27% of the global population hence decreasing the chances of surveillance of Covid by WBE alone (Mandal *et al.*, 2020). Even with these issues, WBE can be used, esp. in countries/cities with better sewage treatment facilities, as an economic technique for detection and trend analysis. This study provides a state-of-the-art comprehensive review of the application of

WBE-based approach for detection of coronavirus in wastewater. The necessity for this approach is first established, followed by detailed description of the process of detection of Coronavirus in the water. This work was conducted in Greater Noida, Uttar Pradesh, India during November 2020 to March 2021.

Need of WBE for COVID-19 Surveillance

The rapid spread of Covid-19 around the world warranted extensive and rapid testing of the suspected cases. However, the testing rate was very slow as compared to the rate of spread and lagged it by a big margin. This is especially true for developing and underdeveloped countries. Further, the test results are not a current indicator, but rather a lagging indicator of the infection spread. Testing was originally only carried out for the symptomatic individuals. Unfortunately, it may take up to 2 weeks' post-infection for the symptoms to show and there is delay between testing and results. This means that an infected person (whether symptomatic or asymptomatic) may already have infected a large number of individuals even before the detection if carried out. WBE offers a measure of determining the presence and spread trend of the infection, without disturbing anyone. It should be noted that in the initial phases, there was a stigma associated with the Covid infected individuals, which deterred many potentially infected people from getting tested or revealing the test results. On January 30, 2020, India recorded its first case of Covid-19 (Perappadan, 2020). In the

last 13 months, 11.2 million cases were recorded, in which 0.15 million fatalities were reported, in India (Worldometer, 2021b). The testing rate should rapidly increase in a country like India where the disease can spread very quickly. There are presently two types of testing methods which are under use: A viral test which shows positive if a person has a current infection, and an antibody test, which shows a positive result if a person had the infection in the past (CDC, 2021a). In general, people only get themselves tested when they start showing symptoms like fever, cough, and headache. So, people who are asymptomatic and presymptomatic can spread the disease unintentionally which is a major concern. This suggests that there is a need of testing method which detects small loads of virus in the body and give results without a need for symptoms to show. Moreover, the surveillance of Covid-19 in wastewater will provide a picture of the community as a whole and help in better hotspot detection. And this type of detection methods does not need the approval of the infected person, as people may be cynical about getting themselves tested due to fear of social stigma.

Process of testing COVID-19 in wastewater

Even though there is no standard testing methodology, different studies have followed different methods to detect Covid-19 in the wastewater treatment plants. A summary of the generic steps involved in the process of COVID-19 testing in wastewater is presented in the Fig. 1.

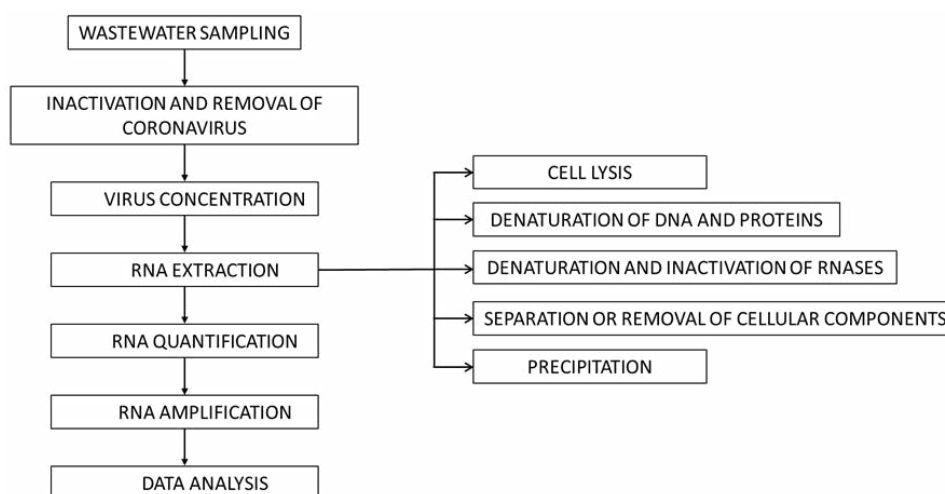


Fig. 1: Process of testing COVID-19 in wastewater

Wastewater sampling

Water sampling is the first step towards detection of Novel Coronavirus in STP. Sampling wastewater for SARS-CoV-2 is used to evaluate trends of infection within the citizenry contributing water to the sewer system (CDC, 2021b). There are two types of wastewater sample collection methods commonly used for this purpose: Grab sampling and Composite sampling. The grab sampling, as the name suggests collects the sample at a time. In comparison, the Composite sampling, involves collecting various individual discrete samples over a specified period of time – usually 24 hours (CDC, 2021b). The composite sampling can be done both manually and automatically. For detection of SARS-CoV-2, both grab and composite sampling methods were used. Sherchan *et al.*, 2020 collected the samples by grab sampling method early in the morning, from 7-11 am. In a study in the Gujrat state of India, Kumar *et al.*, 2020 used the grab sampling method to collect the samples at 11:30am on 8th and 27th May 2020, using sterile bottles. In Netherlands, Medema *et al.* 2020 collected the samples by using composite sampling method in February and March in different selected cities such as Amsterdam, Den Haag, Utrecht, Apeldoorn, Amersfoort, Schiphol. The first Covid-19 case was reported officially after 3 weeks from the first sample round. Nemudryi *et al.*, 2020 used the automatic composite sampling method. The researchers collected the samples with the use of Liquistation CSF34 (Endress+Hauser), an automatic flow proportional sampler located at their facility doorway. In a study in Australia, Ahmed *et al.* 2020 collected samples using the Composite sampling method.

Inactivation and removal of Coronavirus

Getting to know the inactivation or removal process of Coronavirus in the samples can help in establishing better steps to decrease the probability of spread. WHO stated that Covid-19 has a fragile outer membrane as it is known to be an enveloped virus (WHO, 2020). And generally enveloped viruses are environmentally unstable and known to be more vulnerable to oxidants like chlorine. Wang *et al.* 2020 suggested some effective disinfection technologies for deactivating SARS-CoV-2 virus from sewage. The disinfectants included chemicals such as, Chlorine (Cl_2), Sodium hypochlorite (NaOCl), Ozone (O_3), and

Chlorine dioxide (ClO_2) and also physical scheme like, UV irradiation. Each disinfection method has its own merits and demerits, so the selection of appropriate disinfection method depends upon various factors like cost, energy consumption, operating parameters, and disinfectants' toxicity. Wang *et al.* 2005 suggested that chlorine is a better disinfectant when compared to chlorine dioxide. It states that SARS-CoV inactivation is completed with 10 mg/L chlorine after 30 minutes of disinfection whereas chlorine requires 40 mg/L in 30 min. Pasteurization is a process where samples are treated with mild heat usually less than 100°C in order to inactivate any live pathogens. Heat pasteurization of wastewater samples was conducted to decrease the biosafety risk from procedures that generate bioaerosols during the processing of wastewater samples (CDC, 2021c). Wu *et al.* 2020 pasteurized the samples at 60°C for 90 minutes as a preliminary step before opening the sample containers to increase the protocol safety. Former studies on SARS-CoV-1 suggest that a heat inactivation of 30 minutes at 60°C is adequate for virus inactivation by more than 6 log units (Rabenau *et al.*, 2005). Extra precautions were taken by increasing the pasteurization time.

Virus concentration

Virus concentration is the process of enumerating the quantity of virus in a specified sample volume. The methods which were used in various studies are polyethylene glycol (PEG) precipitation (Lewis *et al.*, 1988; Bibby *et al.*, 2013), skimmed milk flocculation (SMF) (Calgua *et al.*, 2013), ferric chloride precipitation – (FeCl_3) (Falman *et al.*, 2013), ultracentrifugation (Prata *et al.*, 2012), ultrafiltration (Hill *et al.*, 2007), and glass wool filtration (Cashdollar *et al.*, 2007). The effective method of virus concentration should ideally be technically simple, rapid and should have the ability to process large volume of water, repeatable, reproducible and predominantly cost and time effective. As yet, there is no concentration method which meets all these criteria (Michael-Kordatou *et al.*, 2020). Ahmed *et al.* 2020 used two methods for the process of concentration, including direct RNA extraction from electronegative membrane and ultrafiltration. For the first step, the researchers used a RNeasy power microbiome kit®. For ultrafiltration, centrifugation was followed by a QIAcube Connect platform. Westhaus *et al.* 2020 started their process of concentration by centrifugation at 4700g for 30

minutes without a break. Then by using centrifugal ultrafiltration units, the purified wastewater was concentrated. 15 mL of wastewater sample was then added to the filter unit, followed by centrifugation at 3500g for 15 minutes. The concentrated supernatant was then harvested. The step was repeated twice until the quantity of 45 ml of sample was completely concentrated. The pellet was then cleaned to remove any aqueous remains, by deionized water and was centrifuged at 4700g for 5 min. [La Rosa et al. 2020](#) used the PEG – polyethylene glycol precipitation method to concentrate the sample. The researchers followed the WHO guidelines for environmental surveillance of poliovirus ([WHO, 2003](#)), by modifying the protocol for the enveloped virus. The sample was first centrifuged and, then mixed with dextran and PEG. It is then left in a separation funnel at 4° C. The interphase and base layer were collected dropwise, and the concentrate was added from the initial centrifugation to the pellet. WHO protocol intends to follow the chloroform treatment at this stage and was left out to conserve the integrity of the enveloped viruses. [Sherchan et al. 2020](#) used two sample concentration methods to enhance the possibility of SARS-CoV-2 occurrence in wastewater. Ultrafiltration, the first process, started with 250 ml sample centrifugation for 30 min. at 3000 g to extract large particles. A centrifugal filter (Centricon® Plus-70) was used to concentrate 70-140 mL of the 250 mL supernatant with a nominal molecular weight limit (NMWL) of about 100 kDa through centrifugation. In order to recover 350 µL of the viral concentrate, the filter unit was inverted and then centrifuged for 2 min at 1000g. Using a pipette, the viral concentrate was then extracted from the sample reservoir. Adsorption-elution was the second method and was carried out with the use of electronegative membrane. 2.5 M MgCl₂ was used to attain a final concentration of 25mM MgCl₂ in all the samples. Then samples were then passed through an electronegative filter (90 mm diameter and 0.45-µm pore size) attached to a glass filter holder assembly. The viruses were eluted with 10 mL of 1.0 mM sodium hydroxide (pH 10.8). The eluate was recovered for neutralization, in a tube consisting of 50 µL of 100 mM sulfuric acid and 100µL of 100× Tris-EDTA buffer. Later 10 mL of this eluate was centrifuged, containing an ultrafiltration membrane (NMWL = 30 kDa) to obtain approximately 650 µL of final volume. However, the viral recovery

yield can be affected by the volume of wastewater to be concentrated. According to [Bibby et al. 2013](#), PEG precipitation was proven to be a highly efficient method for virus concentration. [Falman et al. 2019](#) suggested that for recovery of poliovirus type 1, SMF was found to be more efficient with a recovery of 106 ± 24.8% while the PEG yielded 59.5 ± 19.4% recovery in wastewater.

RNA extraction

The main aim of method of RNA extraction is to acquire the RNA without any damage from the sample matrix. The most widely used methods for RNA extraction are organic extraction using Phenol-guanidine isothiocyanate, silica-membrane dependent spin column techniques and the use of paramagnetic particles. The fundamental steps in RNA extraction using organic solvents are as follows ([Johnson, 2021](#)):

1. *Cell lysis*: Cell lysis is the process which releases the RNA of the cell by breaking down the cell membranes' outer boundary ([Michael-Kordatou et al., 2020](#)). It is carried out by using viral, enzymatic, or osmotic mechanisms ([Listwan et al., 2010](#)). Through the use of chaotropic agents containing buffers or reagents like guanidinium isothiocyanate, guanidinium chloride, sodium dodecyl sulphate, sarcosyl, urea, phenol or chloroform, the aim of cell lysis is accomplished. And to maintain the integrity of RNA during lysis, solutions such as TRIzol or RNeasy or Qiazol ([DiToro et al., 2018](#)) may also be used.
2. *Denaturation of DNA and proteins*: Denaturation refers to the three-dimensional structure loss of the biological molecule viz. DNA or proteins. To break down DNA, DNase can be used whereas proteinase K can be enumerated to digest proteins. Another way to remove the proteins is through repeated organic extraction through the use of phenol and chloroform. This can be achieved by sample dissolution in buffers containing guanidinium salts.
3. *Denaturation and Inactivation of RNases*: By using chaotropic agents such as phenol and chloroform, RNases can be denatured and inactivated.
4. *Separation or Removal of cellular components*: Addition of chloroform and subsequently centrifuging the solution can be done in order to

isolate RNA from other cellular components. This divides the solution within two phases known as aqueous and organic phases where, RNA is present in aqueous phase.

5. *Precipitation*: Isopropyl alcohol is often used to recover RNA from aqueous phase whereas by using ammonium acetate preferential precipitation of RNA can be done. And Lithium chloride can also be used to for selective precipitation of RNA from DNA and proteins.

RNA extraction can be done through various methods like Magnetic bead technology, Silica technology, Lithium chloride, and urea isolation (Johnson, 2021). But during the process of identification of SARS – CoV – 2, mostly commercial kits have so far been used. Some of the kits used for RNA extraction are RNeasy kits, ZR Viral RNA kit, Combination of RNeasy Power Microbiome Kit and RNeasy Power Water Kit. RNA extraction can also be done by combining two specific kits: RNeasy Power Water Kit and RNeasy Power Microbiome Kit (Ahmed *et al.*, 2020). To accommodate the electronegative membrane, a 5 ML bead tube from RNeasy Power Water Kit was used and a Precellys 24 tissue homogenizer to integrate the samples differing from 3 x 20 s at 8000 rpm at an interval of 10 seconds. From there on the RNeasy Power Microbiome kit was used to extract RNA, as per the manufacturer's instructions. NucleoSpin RNA Virus kit was used to isolate RNA by following the manufacturer's instructions (Westhaus *et al.*, 2020). Nemudryi *et al.* 2020 used RNeasy mini kit to extract RNA. RNA extraction was carried out from samples of concentrated sewage sample using viral RNA kit to get the final volume (Sherchan *et al.*, 2020). Apart from the above kits, the CDC has given a list of viable RNA extraction kits (CDC, 2021d). Some of them are QIAcube® QIAcube, Roche MagNA Pure LC®, Roche MagNA Pure 24, Roche MagNA Pure 96.

RNA quantification

Some detection approaches and quantification of viruses in sewage are epifluorescence microscopy, transmission electron microscopy, throbbling gel electrophoresis, immunofluorescence test, flow cytometry, conventional cell culture and molecular techniques. Molecular methods include conventional polymerase chain reaction (cPCR), Reverse transcription polymerase chain reaction (RT-PCR),

Real-time quantitative polymerase chain reaction (RT-qPCR), Multiplex PCR and sequencing (Corpuz *et al.*, 2020).

For SARS-CoV-2 detection in sewage and its quantification, PCR-based methods have been predominantly used. All these methods use the same principle except for RT- qPCR, where the process starts with RNA material in place of DNA, and RT- PCR requires a reverse transcription step before qPCR (Corpuz *et al.*, 2020). The “q” in qPCR stands for quantitative, which means that qPCR tests not only detect the virus's genetic information but also quantify the amount of genetic information (Bustin and Nolan, 2020). Several studies in Australia (Ahmed *et al.*, 2020), France (Wurtzer *et al.*, 2020a), Spain (Randazzo *et al.*, 2020), and Italy (La Rosa *et al.*, 2020) have detected and quantified SARS-CoV-2 by qPCR. With N_Sarbeco and NIID_2019-nCoV_N primers both being nucleocapsid protein gene specific, RT-qPCR was conducted for specific detection and quantification (Ahmed *et al.*, 2020). For reverse transcription and the QPCR, a single-step RT-QPCR was used. Raw sewage and wastewater treatment plant (WWTP) samples were acquired in Israel, and when tested using the RT-qPCR assay, a few of the samples were found positive. The Ct value of less than 40 was obtained in one out of two sewage samples taken from the sewer networks of hospitals treating Covid-19 patients, three out of three isolation facilities (Bar-Or *et al.*, 2020). Wurtzer *et al.* 2020b examined the incidences of SARS-COV-2 in Paris by collecting samples from three different WWTP inlets during the period of 5 March 2020 to 23 April 2020 and tested using RT-qPCR assay. It was found that all the tested samples were positive. The researchers also attempted to associate a relation between quantified number of SARS-CoV-2 genomes in wastewater and carrier numbers. They noticed a significant link between genome units' number of COVID 19 cases reported. They also indicated that monitoring wastewater would serve as a backup and early identification method for SARS-CoV-2 spread. Kumar *et al.* 2020, in India, examined RNAs of SARS-CoV-2 with the use of TaqPath™ Covid-19 RT-PCR Kit for detection of ORF1ab, N gene and S gene and s gene. Medema *et al.* 2020 from Netherlands quantified by performing qRT-PCR on serial 10 – fold dilutions of quantified synthetic double stranded DNA.

RNA amplification

The nucleic acids PCR amplification and detection of the products is concurrently achieved in qPCR method (Cobo *et al.*, 2012). This is a quantitative method because it measures the target sequence unlike conventional PCR methods that provide qualitative data through gel electrophoresis (Kadri *et al.*, 2020). qPCR not only provides quantitative data but also shows high sensitivity and is mostly independent of the amount of reagent concentration (Watzinger *et al.*, 2006). RT-qPCR can also be used for RNA amplification (Ahmed *et al.*, 2020). Kumar *et al.*, 2020, applied amplification in a reaction mixture of 25 μL which consists of 7 μL to extract nucleic acids of each sample. 2 μL TaqPath™ COVID-19 Control and purified 5 μL sample were used as the positive and negative controls, respectively. For no template control, nuclear free water has been used. One of the other molecular methods used for amplification is Reverse transcription loop-mediated isothermal amplification (RT-LAMP). The core aspect of the RT-LAMP is a reverse transcription step (RNA to DNA), after which 6 primers attach to the target gene of interest. These 6 primers get attached to the target DNA at an appropriate temperature, and they loop around to create circular constructs that help in extending the DNA. For primers to bind, each loop opens up a new site further amplifying the gene. RT-LAMP allows a rapid and exponential increase in the gene of interest. When probes, dyes, or a fluorophore are added to the mixture, a visible change during the reaction can be detected by eye or with use of special apparatus (Watzinger *et al.*, 2006). LAMP method has an upper hand over RT-qPCR in the point of temperature. LAMP reactions take place at a single isothermal temperature range of 63 °C to 65 °C, whereas rapid cycling of multiple temperatures is needed in RT-qPCR method to amplify nucleic acids. This method has also shown substantial potential results by producing them in less than an hour, and on some occasions within 11 minutes (Augustine *et al.*, 2020). Another digital PCR method known as ddPCR - Droplet Digital PCR technology works by using a water-oil emulsion droplet system. In such system, droplets are formed to create the partition that separates the template DNA molecules. In the plate where PCR reaction takes place, the droplets serve the basic purpose as individual test tubes or wells but in smaller space. In the ddPCR technique, the mass

sample key partitioning is a critical part. The nucleic acid samples are divided into several thousands of droplets (nanoliter-sized) and PCR amplification is carried out in each of these droplets. The sample requirement is one of the merits for this method. It requires smaller sample size as compared to other digital PCR systems which are commercially available. Reduced cost and preserving precious samples are some of the other merits of this method. This method has shown to have less consumable costs and more accuracy than RT-qPCR (Bio-Rad, 2021).

Data analysis

The results of the studies which were conducted have been reported in one of the following ways (Michael-Kordatou *et al.*, 2020).

- “Absence or presence of the virus in the form of Ct values reported directly by the qPCR instrument”
- “Gene copies/volume of sample, with the use of a quantitative calibration curve of Ct values against known concentrations of the virus for the calculation of the gene copies present in a certain sample volume (relative quantification)”.

The Ct (cycle threshold) is defined as the number of cycles required for the fluorescent signal to cross the threshold. Ct levels and the amount of target nucleic acid are inversely proportional to each other (i.e. the lower the value of Ct level, the greater is the amount of target nucleic acid) (Yagci *et al.*, 2020). A patient is considered to be Covid-19 positive if the Ct value is below 40. However, the Indian council of Medical Research (ICMR) does not recommend reliance on Ct values to determine the infectious status of a Covid-19 patient (ICMR, 2020). Some factors which can adversely affect the Ct values are the temperature of transportation, the time between collection and receipt in the lab. Ct values of asymptomatic/mild cases are similar to that of Ct values of severe cases. The number of viral genomes of SARS-CoV-2 per ml of human fecal material are between 6,00,000 to 30,00,000 assuming fecal load of 100-400 g feces/day/person with a density of 1.06 g/mL (Hart *et al.*, 2020). The number of persons infected can be found by using Eq. 1 (Ahmed *et al.*, 2020).

$$\text{Persons Infected} = \frac{\left(\frac{\text{RNA copies}}{\text{liter wastewater}} \right) * \left(\frac{\text{liters wastewater}}{\text{day}} \right)}{\left(\frac{\text{g feces}}{\text{person} - \text{day}} \right) * \left(\frac{\text{RNA copies}}{\text{g feces}} \right)} \quad (1)$$

Eq. 1 has been utilized to calculate the potential number of cases in Brisbane North and Brisbane South primary health networks. The Monte Carlo simulation estimated the median number of infections, which ranged from 1,090 on 27 March 2020 to 171 on 1 April 2020 in the catchment basin. The sensitivity analysis indicated a strong correlation (-0.977) between the number of infections and prevalence with \log_{10} SARS-CoV-2 RNA copies in human feces samples.

Accuracy of WBE-based analysis

The accuracy of the results depends upon some of the key variables like seasonal temperature, per person water use, community demographics, average in-sewer travel time and degradation rate of biomarkers as these variables affect the accuracy of data (Hart *et al.*, 2020). These factors influence the precision of the results because the survival of SARS-CoV-2 is influenced by these factors. Chin *et al.* 2020 revealed that when the stability of SARS-CoV-2 was determined at different temperatures, it was found that at 4°C the virus was highly stable and on day 14, there was only ~ 0.7 -log unit reduction of infectious titer. But when the incubation temperature was increased to 70°C, the time taken for inactivation of virus was brought down to 5 Minutes. Chin *et al.* 2020 also states that SARS-CoV-2 is highly stable over a wide-range of pH values (3-10). Noble *et al.* 1997 which conducted studies on survival of viruses in wastewater discovered that higher molecular weight of dissolved matter has an impact on their survival. Hart *et al.* 2020 also stated that when calibrated carefully, the wastewater surveillance method at worst can detect 1 infected person in 114 individuals and at best can detect one infected case in 2 million uninfected individuals. This shows the advantage of this detection method over the traditional testing methods as they would require significant amount of time and resources. Medema *et al.* 2020 highlighted the ability of wastewater surveillance when spread of the disease was low. An investigation was carried out in Netherlands, for the SARS-CoV-2 presence in wastewater of both domestic and airport, at the start of the pandemic, to find out the effectiveness of sewage surveillance for monitoring. No samples of February 6, 2020 were tested positive i.e. three weeks before the first case was reported. And a N1 fragment was found in the samples of March 5. It indicates that the wastewater

can help in finding out the entry of new type of virus or can raise an alarm before the wide spread of virus in the community.

Economics of Covid-19 testing

Per assay costs of both clinical and wastewater-based epidemiology may be different among different countries due to government regulations, labor costs and medical/testing infrastructure. The estimated cost of a test kit which contains key reagents that are essentially identical (qRT PCR buffers, probes, etc.) is available for \$10 - \$20 USD (Hart *et al.*, 2020). Expenses for reagents required for primary checkup of the community through municipal wastewater was calculated by multiplying the total number of wastewater treatment plants and cost of reagents. The study concludes that countries like Germany can test the people by using 0.014% of the cost required for clinical testing, and in America by 0.00006% of cost required for clinical testing (Hart *et al.*, 2020). These calculations show that WBE costs way less than the cost of clinical test. The price of a RT-PCR test in all states across India ranges from INR 400 to INR 2100, whereas the test was made free in the state of Assam but in order to obtain the report in 24 hours, INR 2200 has to be paid (Timesnow News, 2020). The costs initially ranged between INR 4500 to INR 6000, but once the number of testing labs increased, the cost was brought down significantly. In India approximately 222 million people were tested as of March 8, 2021 (ICMR, 2021), which means that 16.30% people have been tested. Assuming the present cost, it would cost approximately between INR 54,000 Crores and INR 2.83.500 Crores, i.e. USD 7.4 Billion and USD 38.7 Billion to get every person in India tested. WBE-based detection offers a much more economical alternative that will allow the analysis of infection spread without the necessity of testing of all the individuals. In general, the WBE-based approach is to be applied at a broader scale and then subsequently narrowed down to sub-areas of interest (generally locations with higher infection levels). For example, when applied over a city, this approach will indicate the state of spread and its trend (increasing or decreasing). By sub-dividing the city into various sections (or zones), areas with higher occurrence or faster rate of spread can be determined. Targeted individual testing can then be

carried out to isolate the cases and prevent further spread. This is certainly much less expensive than testing all individuals within the city.

Challenges

The foremost challenge to the application of WBE-based approach is the locations from where sampling can be done. These locations are generally the sewage treatment plants and in some cases, the sewer network. Unfortunately, the developing and underdeveloped countries have low prevalence of sewers and STPs. For example, there are only 920 sewage treatment plants in India. Furthermore, out of 61,754 MLD sewage generated, 38,791 MLD remains untreated (International Institute of Health and Hygiene, 2021). Approximately only 37% of the total sewage is passing through the wastewater treatment plants and only those places can be used for community testing through WBE. This leaves a large fraction of the Indian population out of the loop of community-level testing. Another challenge would be the process and test protocol. There are studies being conducted to find out the best and accurate process for detection of virus (Brouwer *et al.*, 2018). Nevertheless, the process is sensitive to key variables like temperature, in-sewer travel time, season, rainfall, sampling time, etc. and data may be affected if not calibrated carefully (Brouwer *et al.*, 2018). For Covid-19 detection through WBE, there are several tests available with varying level of results. A standardized protocol is needed for Covid-19 detection for community based assessment. In addition, the RT-PCR based testing is expensive and requires skilled personnel, both of which are roadblocks for successful implementation in underdeveloped world. Biosensors may be a better approach for real-time monitoring in the sewer networks. Finally, there are still some concerns over the correlation between the results obtained from WBE and actual spread. This may be a function of the population being catered by the sampled WWTP, movement of tourists (esp. for smaller towns), testing method used, etc. (Zahedi *et al.*, 2021). Further studies in these directions are needed.

Case Studies_of WBE application for Coronavirus detection

In different case studies and research conducted

around the world, the observations of viral load were made and, in some instances, WBE helped in detecting Covid-19 infected cases in real time (Table 1). In a study conducted in Paris, the concentration of vRNA in raw sewage was found to be 5.104 genome units /L during an early stage when total confirmed cases in Paris were 91 (Wurtzer *et al.*, 2020b). Wu *et al.*, 2020 suggested that the concentrations indicated a much higher COVID-19 prevalence (0.1%–5%) than confirmed clinical cases (0.026%). The study implies that WBE can help in finding the hotspots of spread in a much better way than the clinical testing. In another study in India, the estimated maximum concentration was 3.5×10^2 copies/L in the month of May 2020 (Kumar *et al.* 2020). According to Foladori *et al.*, 2020, the analytical data does not give the correct information of the viral load as it is variable to number of cases. It is also suggesting that the load can be up to 4 orders of magnitude, from $5 \cdot 10^3$ to 107.6 copies/mL and there is a need of further research to get the exact values. There are cases when the testing of wastewater for detection of Covid-19 helped prevent the outbreak of the disease. In University of Arizona, daily screening of sewage was conducted (Pineda and Leingang, 2020). On 25 August 2020, the official found the traces of coronavirus. They immediately got the 311 people in the dorm tested on 26 August 2020 and found two students who were infected with the virus (although they were asymptomatic). They sent the infected students into isolation and the next day the samples were clear of the traces. It helped the authorities to stop the outbreak of virus to other students living in the same dorm. Two recent developments have taken place: the availability of the vaccines and the detection of new strains. Multiple vaccines have been approved, in different countries, and mass-vaccination programs have also been started to protect the frontline workers and people at-risk as a priority. On the other hand, new strains have emerged and have led to an upsurge in the new cases. Globally, around half million new cases are being reported daily. This shows that while human beings have come up ways to tackle the virus, its mutations have allowed it to continue the infection. This calls for a concerted effort in detection, containment, treatment and prevention. WBE-based testing is a cornerstone of such efforts.

Table 1: A summary of case studies of WBE application for Covid-19 detection in wastewater

Place of study	Location of sample collection	Process of Sample collection and Dates of collection	Virus concentration method	RNA extraction method	RNA quantification & amplification method	Genome value / C _v value	Reference
Ahmedabad, Gujarat India	Wastewater treatment plant	Composite and Grab, 8 th May 2020 – 27 th May 2020	Use of PEG	Commercial available kit (NucleoSpin® RNA Virus)	RT-PCR using TaqPath™ Covid-19 RT-PCR Kit	Maximum concentration of 3.5×10^2 copies/L	Kumar et al., (2020)
Australia	One suburban pumping station and two WWTPs	Conventional refrigerated autosampler and Submersible in-situ high frequency autosampler as well as grab sampling; 24 th February 2020 – 5 th April 2020	Direct RNA extraction from electonegative membranes And Ultrafiltration	Combination of two kits (RNeasy power water Kit and RNeasy power microbiome Kit.	RT-qPCR	Maximum of 12 copies / 100ml	Ahmed et al., (2020)
Brazil	Sewage treatment plants, hospital wastewater and sewers network	Ten-hour composite samples; 15 th April 2020	Ultracentrifugation Method	Using QIAamp® Viral RNA Mini kit and a QIAcube® automated system.	RT-qPCR	SARS-CoV-2 RNA was detected in 5 of 12 samples, Ct values: 36.3 to 39.8	Prado et al., (2020)
Italy	Three WWTPs	Composite; 3 rd February 2020 – 2 nd April 2020	PEG-dextran method	NucliSENS miniMAG semi-automated extraction system.	PCR using Kit Platinum™ SuperFi™ Green PCR Master Mix	6 of 12 WWTP'S tested were positive.	La Rosa et al., (2020)
Paris, France	Three WWTPs	Composite; 5 th March 2020 – 23 rd April 2020	Ultra centrifugation and extraction	Using PowerFecal Pro kit, QIAasympfony automated extractor	Using Fast virus 1-step Master mix 4x	3x10 ⁶ GU/L on peak	Wurtzer et al., (2020b)
Israel	Hospital treating SARS-COV-2 patients, isolation facility, Sewer network, Wastewater treatment plant	Using Automatic sampler; 10 th March 2020 – 21 st April 2020	Centrifugation, and using polyethylene glycol (PEG)	Using viral RNA extraction kit (RNeasy mini kit- QIAGEN and EasyMAG)	Reverse transcribed and qPCR and Step One Plus real-time PCR system	10 out of 26 collected samples tested positive with Ct values: 32.76 to 38.5	Bar Or et al., (2020)
Japan	Five times from a wastewater treatment plant and Three times from a river	Grab sampling; 17 th March 2020 – 7 th May 2020	Electronegative membrane-vortex (EMV) method and adsorption-direct RNA extraction method	Using QIAamp Viral RNA Mini Kit in a QIAcube automated platform	Using Thermal Cycler Dice Real Time System TP800	Ct value: 39.96, 2.4x10 ³ copies/L in the water sample	Haramoto et al., (2020)
Istanbul, Turkey	WWTPs and Man holes	Composite and Grab sampling; 21 st April 2020 – 25 th April 2020	Centrifugation	Using Pathogen Mini kit	RT-qPCR	Five out of Seven samples collected from WWTP were positive	Kocamemi et al., (2020)

Table 1: A summary of case studies of WBE application for Covid-19 detection in wastewater

Place of study	Location of sample collection	Process of Sample collection and Dates of collection	Virus concentration method	RNA extraction method	RNA quantification & amplification method	Genome value / C _t value	Reference
Louisiana, USA	Two WWTPs	Composite and Grab; 13 th January 2020 – 8 th April 2020	Ultrafiltration and Adsorption-elution method using an electronegative membrane	Use of <i>Pseudomonas</i> bacteriophage	RT-qPCR using a CFX96 RealTime PCR instrument	3.1x10 ³ - 7.5x10 ³ copies/L	Sherchan <i>et al.</i> (2020)
England	WWTP	Composite; 14 th January 2020 – 12 th May 2020	Centrifugation	RNA extracted from the National Institute for Biological Standards and Control	RT-qPCR	3.50 - 4.20 Log ₁₀ gc/L	Martin <i>et al.</i> (2020)
Southeastern Virginia, USA	Nine WWTPs	Composite and Grab; 9 th March 2020 – 26 th July 2020	n InnoVaprep Concentrating Pipette Select and electronegative filtration	Using NucleiSENS easyMag	RT- digital droplet PCR	98 out of 198 water samples were found positive.	Gonzalez <i>et al.</i> (2020)
North Rhine-Westphalia Germany	Nine WWTPs	Composite; 8 th April 2020	Amicon® Ultra-15 Centrifugal Filter Unit	NucleoSpin RNA Virus kit	OneStep RT-qPCR using Luna Universal Probe One-Step RT-qPCR Kit or LightCycler® Multiplex RNA Virus Master and the CFX96 Real-Time System, with a C1000 Touch Thermal Cycler.	3 - 20 gene equivalents per mL in the inflow, and 2.7 - 37 gene equivalents in the WWTP effluent	Westhaus <i>et al.</i> (2020)
Santiago, Chile	Two WWTPs	Composite; March 2020 – June 2020	Ultracentrifugation	Using the QIAamp® Viral RNA Mini kit	Using the TaqMan 2019-nCoV Assay Kit v1	Ct values: 28.1- 37.7	Ampuero <i>et al.</i> (2020)
Netherlands	WWTPs	Composite; 7 th February 2020 – 25 th March 2020	Ultrafiltration	Using the RNeasy PowerMicrobiome Kit and Biomerieux NucleiSens kit	RT-qPCR	14–30 gene copies (gc)/mL	Medema <i>et al.</i> (2020)
Southern Nevada, USA	WWTP and from lake	Grab and Composite; March 2020 – May 2020	Primary concentration with hollow fiber ultrafiltration (HFUF) and secondary concentration with Centricon ultrafiltration	PureLink Viral RNA/DNA Mini Kit	q-PCR	4.5 x 10 ⁵ gc/L	Gerrity <i>et al.</i> (2020)

RECOMMENDATION

Given the unique advantages offered by the WBE-based approach, it should be the preferred choice for Covid-19 detection and spread analysis, globally. This pandemic has severely affected the entire world and hence a global approach is required to its containment and remediation. It is expected that countries with more resources will come forward to help the lesser-endowed nations in terms of scientific and technical know-how, testing kits and other accessories needed for testing and control. Formation of a collaborative group (Covid19WBEC) is a welcome step in this direction. WBE-based detection, followed by human testing in areas with high concentration of cases and mass-scale vaccination is likely to be the key to overcome this pandemic.

CONCLUSION

A global pandemic has engulfed the humanity and has infected 128 million people (so far), killing millions in a very brief span of 15 months. Unfortunately, despite the best efforts, it is yet to be controlled. The first step in the fight against Covid-19 is its detection. Unfortunately, the targeted human testing is expensive, intrusive and a lagging indicator of the infection. An alternate approach such as, wastewater based epidemiology may act as a potential solution. Here, a comprehensive literature review focusing on the application of wastewater based epidemiological approach to the detection of Covid-19 is presented. This method obviates the need for extensive, non-targeted individual testing and helps in determining the spread and its trend at a much lower cost, in a non-intrusive and timely manner. It has been established, through various studies, that WBE-based Covid-19 detection can provide an early indication of the spread as compared to direct-human testing, thus providing significant lead time. These results can then be used to focus, in a targeted manner, on areas with high level of viral spread leading to human testing, and containment and treatment efforts. This approach has been applied in different geographies (Table 1) and has proven to be an asset in containing Covid-19 and preventing further spread of the infection, saving lives and resources in the process. Different variations of PCR-based assays have been utilized in these studies to a varying degree of success. With the availability of multiple vaccines globally,

this approach can be coupled with the targeted vaccination program to prevail over this pandemic. The biggest challenge to this approach is the non-availability of representative sampling locations, especially in developing and underdeveloped countries. This brings the focus back on the lack of extensive sanitation programs in such countries. Standardized protocols and knowledge/resource sharing, through concerted global efforts, is expected to enhance the application of this technique in different parts of the world and is likely to aid humanity's fight against Covid-19.

AUTHOR CONTRIBUTIONS

P.S. Deepak wrote the first draft of the manuscript and helped with updates (as needed), including the post-review activities. G. Saini developed the concept, supervised the manuscript preparation and edited it.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

ABBREVIATIONS

<i>CDC</i>	Centers for Disease Control and Prevention
<i>Cl₂</i>	Chlorine
<i>ClO₂</i>	Chlorine dioxide
<i>cPCR</i>	Conventional polymerase chain reaction
<i>Ct</i>	Threshold Cycle
<i>ddPCR</i>	Droplet Digital polymerase chain reaction
<i>DNA</i>	Deoxyribonucleic acid
<i>FeCl₃</i>	Ferric chloride
<i>ICMR</i>	Indian council of Medical Research
<i>IQR</i>	Interquartile range

<i>kDa</i>	Kilodaltons
<i>MERS</i>	Middle East respiratory syndrome
<i>MLD</i>	millions of liter per day
<i>NaOCl</i>	Sodium hypochlorite
<i>NMWL</i>	Nominal molecular weight limit
<i>O₃</i>	Ozone
<i>PCR</i>	Polymerase chain reaction
<i>PEG</i>	Polyethylene glycol
<i>qPCR</i>	Quantitative polymerase chain reaction
<i>RNA</i>	Ribonucleic acid
<i>RT-LAMP</i>	Reverse transcription loop-mediated isothermal amplification
<i>RT-PCR</i>	Reverse transcription polymerase chain reaction
<i>RT-qPCR</i>	Real-time quantitative polymerase chain reaction
<i>SARS</i>	Severe acute respiratory syndrome
<i>SARS-CoV-2</i>	Severe acute respiratory syndrome coronavirus 2
<i>SMF</i>	Skimmed milk flocculation
<i>vRNA</i>	Viral Ribonucleic Acid
<i>WBE</i>	Wastewater based epidemiology
<i>WHO</i>	World health organization
<i>WWTP</i>	Waste water treatment plant

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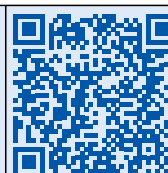


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