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Microplastics in water, sediment and salts from traditional salt producing ponds

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ABSTRACT

Plastic pollution has universally known accumulated in all environment compartments and accelerating threat to the sustainability of earth. Field survey to examine the occurrence of microplastics in ancient sea water evaporation technology of ponds at Pallengu-Jeneponto, was conducted. From this sea salt producing ponds, samples of water, sediment and freshly harvested salts were collected. Sixteen samples each of water and sediments and 12 salts were collected. From 16 water and sediment samples there are 31 microplastics item discovered in 11 water samples (68.75% of total contamination) and 41 microplastics item observed in 10 sediment samples (62.5% total contamination), respectively. Interestingly, sampling points at sedimentation/heating pools were found to be the locations with highest occurrence of microplastics in both water and sediment. There are 7 salt samples positively contaminated with 29 microplastics or 58.3% of total contamination, which predominated by line and fragment forms. Fourier transform infra-red spectroscopy analysis has revealed polymers of polyvinyl acetate (41.7%), polyethylene (33.3%) and polystyrene (25%). There was no significant difference found on microplasticss occurrence from 3 kinds of samples collected, although there was a decreasing trend of total microplastics found from water, sediment and salt. Microplasticss abundance were ranged 7-55 items/L water, 14.6-50 items/kg sediments and 6.7-53.3 items/kg salt. With microplastics abundance reached over 53 microplastics items/kg salt, it is believed that continuous consumption by people will end up with possible accumulation of potentially absorbed of various toxic chemical pollutants which present in sea water as salt raw materials. The need for robust and practical strategy in water quality management for reduction of microplasticss contamination in consumed salts is a must.

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INTRODUCTION

The most conspicuous anthropogenic activities is the manufacture, the use and disposal of plastics, where inadequate management of the resulting waste have led to the accumulation of plastics in the environment. After the mass production began in 1950s, problems of plastics waste has emerged globally due to problems of degradation in the environment, both in terrestrial and aquatic. Plastic pollution has increased over the past decade, at least in part due to mismanagement of plastic waste from landfills to the ocean (Jambeck *et al*., 2015; Rochman, 2016). According to Jambeck *et al*. (2015), in 2010 the indices of plastic waste input to the world ocean from 192 coastal countries ranged from 4.9 to 12.9 million metric tons (MT) per year. Furthermore, without improvements in land based management, a 10 fold increase is predicted by 2025. Plastic has become an eminent pollutant in aquatic environment in water column, sediment and organisms. Plastic waste is a complex problem threatening all global aquatic ecosystems due to variation in its size, from nano, micro, macro, meso and mega-plastics (UNEP, 2016). Microplasticss (MPs) range in size from 1 µm to 5 mm (GESAMP, 2015) and primarily come from manufactured plastic products such as microbeads (cosmetics and personal care) (Napper *et al*., 2015; Tanaka and Takada, 2016) and pellets (Veerasingam *et al*., 2016) originating from landfill sites. Secondary MPs can result from the decomposition of larger plastic debris through ocean weathering processes' (UNEP, 2016; Masura *et al*., 2015). Due to the small size of MPs, organisms such as fish and small invertebrates can mistakenly ingest them as food. Through bioaccumulation, MPs ingested by zooplankton can end up in the guts and bodies of larger organisms (Au *et al*., 2017). One of the dangers of microplasticss is their ability to attract chemical pollutants (e.g. metals, POPs) present in the surrounding environment (Holmes *et al*., 2012; Rochman *et al*., 2013). Research by Li *et al*. (2018) revealed the accumulation of related toxic substances to MPs generating problems such as intestine injury and change in metabolic profile in fish. Furthermore, during the production of plastic products, chemicals are embedded within plastics to enable the desired form, composition, and texture to be achieved. Throughout the life-cycle of these products, these chemicals can and eventually will leach into the environment (Koelmans *et al*., 2014; Oehlmann *et al*., 2009). Microplasticss is well-known to be accumulated in sediments and water column of world oceans (Arthur *et al*., 2009) with estimated maximum concentration up to 100,000 item/m^3 (Wright *et al*., 2013). Microplasticss have been detected several years ago in marine organisms (Cole *et* al., 2013; Tahir and Rochman, 2014; Rochman *et al*., 2015), also in sea salts sold in market (Yang *et al*., 2015; Iñiguez *et al*., 2017; Karami *et al*., 2017). Eventhough new microplastics are constantly entering the oceans and some types of plastic have a relatively low density and therefore drift at the water surface, the microplastics concentrations at the surface of the oceans are often lower than expected. In addition, microplastics have repeatedly been found in deep-sea sediments in recent years (Michels *et al*., 2018). Recent finding by Kim *et al*. (2018) pointed out that sea salts from Indonesia was the worst with highest content of MPs ietmfrom 39 salt samples examined. Coupled with the fact that Indonesia is recently indicated as the second highest contributor of marine plastic pollution worldwide (Jambeck *et al*., 2015), this situation places Indonesia at high risk of environmental impacts from marine pollution, including marine plastic debris, and MPs in particular. With respect to world ocean currents, the location of Indonesia in particular with the Indonesian Through-flow II (also known as ITF II) which transported massive water mass through Makassar strait, having consequences that accumulated plastic debris can readily be carried into the nation's seas (Gordon, 2005; Cozar *et al*., 2014), with current condition of inadequate solid waste management. Since there is yet no study, investigating the presence of MPs in traditional salt producing ponds with ancient technology of sea water evaporation, research to look at the magnitude of MPs contamination on traditional sea salt production system was conducted. In 2017, Jeneponto produced some 93,000 tons of sea salt per-annum (BPS, 2018). Therefore conducting novel research about the current state of plastic pollution and the effect it can have on sea salts as human food can help mitigate future problems. This study was conducted at Pallengu Village, Jeneponto District of South Sulawesi Province, Indonesia in 2018.

MATERIALS AND METHODS

Samples of water and sediments were collected from traditional sea salt production system comprised with sampling points of: adjacent sea water (S-1 and S-2), primary canal (PC1-PC4), secondary canal (SC1-SC-4), tertiary canal (TC1-TC4), from here the sea water were pump into sedimentation/heating pools (HP1 and HP2). Sea water were left in heating pools for one day prior to be drained into production pools, where salts are harvested. Distance between each sampling spots were ± 150 m. There were 16 samples of each water and sediments, and 12 freshly harvested salt samples were collected from actively producing pools during the field sampling performed (Fig. 1).

Water samples (salinity 34 – 37 ppt) were grab two times with 10-L bucket and poured into nylon mesh sized 100 µm with cod-end containing 100 ml, also known as method of neuston net volume reduced (Covernton *et al*., 2019). In the laboratory, sea water in HDPE bottles were added with 30 ml of 10% KOH to digest any organic materials may present, and were left over night at room temperature. In the following day, water samples were filtered with cellulose Whatman 47 mm with mesh-size 0.45 µm (WCN Type 7141-104) through Buchi vacuum pump. Filtrates were then observed with stereo microscope Euromex SB1902. Any MPs found were pictured, counted and further subjected to Fourier Transformed Infra-red Spectroscope (FT-IR Shimadzu Prestice-21) for plastics polymer identification. Sediments were dried in room temperature for 4 days, left in oven at 65°C over night for complete dry, followed with sieving, using 5 mm and 63 µm mesh size. Remaining sediments on top of 63 µm were weighed to 120 g followed by MPs gravity separation in $ZnCl_2$ with density 1.4 g/m^3 (Hidalgo-Ruz *et al*., 2012), by shaking with a flatform-shaker at 150 rpm for 10 min. Supernatants were gently collected and subjected to cellulose filtration with vacuum pump. Filtrates were then observed under stereo microscope for MPs identification. As many as 150 g of salts were weighed and diluted in 1 L of double-distilled water with magnetic stirrer, filtered through the cellulose membrane with Buchi vacuum pump (Kosuth *et al*., 2018), the filtrate was then put in a clean petri dish, and directly observed with stereo microscope. While for the IR spectrum analysis, all MPs discovered were kept in between cover glass and further subjected to KBr then crushed and compressed in stainless steel dish (1 cm^2) to produce pellet. Pellets were then subjected to FT-IR which emitted infra-red radiation that turned out as spectrum values or wavelengths number. Percentage values of polymer types were obtained from grouping

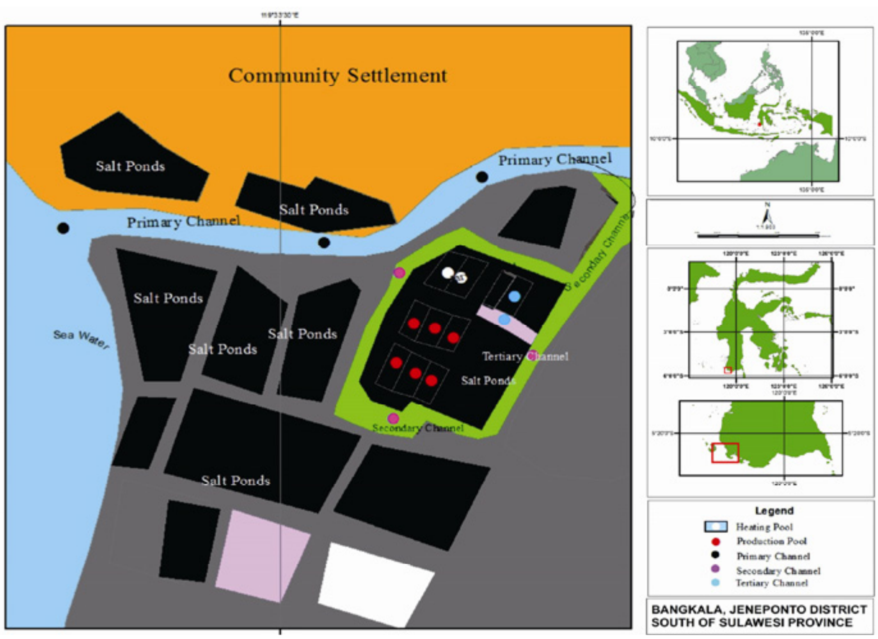

Fig. 1: Geographic location of the study area along with the sampling points at Pallengu village, Bangkala sub-district, Jeneponto, South Sulawesi, Indenesia

similar spectrums of all MPs samples processed. One-way analysis of variance was performed to look at any differences at MPs content in the three samples.

RESULTS AND DISCUSSION

From 16 water samples examined, there are 31 MPs item discovered from 11 water samples (or equal to 68.75% of total contamination) with fiber as the most predominated MPs form (83.9%), sized between 0.39 – 7.02 mm. Based on microscope observation, MPs numbers reached 55 item/m^3 water (Fig. 2a and b). Microplasticss color in water was predominated blue (38.71%) and red (16.13%) in the forms of line (83.87%) and fragment (16.13%). Meanwhile, there were 41 MPs item discovered from 10 sediment samples with line (50%), fragment (44.43%) and film (3.57%) forms (size between 0.39 – 4.29 mm), with blue (44.64%) and transparent (25%) as predominant colors. Total percentage of sediment contamination level was equal to 62.5% from the total samples. The range of MPs itemnumbers/kg sediments found was 14.6-50 (Fig. 3a and b). Variation on MPs distribution in water and sediment sampling points was detected,

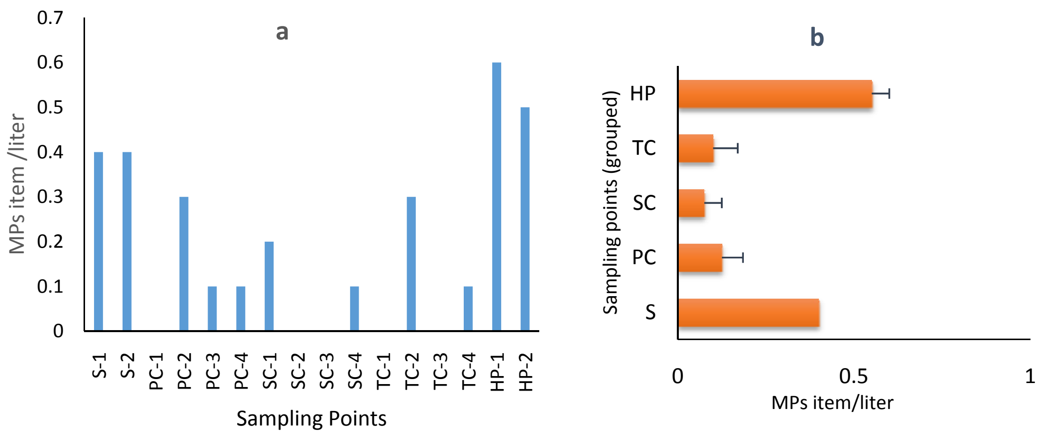

Fig. 2: MPs item abundance per- liter of water (a is value per-liter at each sampling point and b is average values from grouped sampling points ± SE, where S: sea; PC: primary channel; SC: secondary channel; TC: tertiary channel; HP: heating pool).

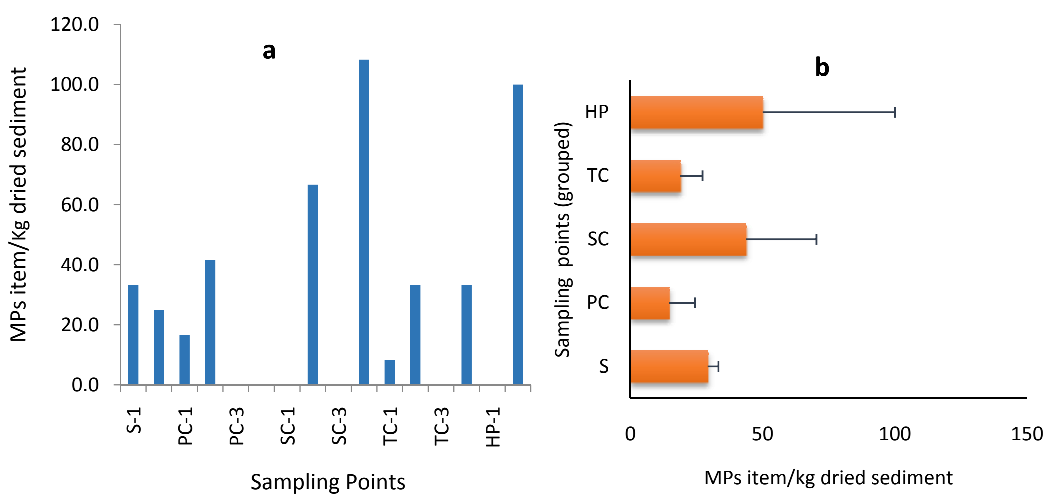

Fig. 3: MPs item abundance per- kg dried sediments (a is value per-kg sediments at each sampling point and b is average values from sampling points ± SE, where S: sea; PC: primary channel; SC: secondary channel; TC: tertiary channel; HP: heating pool).

and there was a trend in several sampling points, particularly at sedimentation/heating pools, showing high content of MPs items compared to other points although with no statistical significance found ($p=0.1$). Most likely due to high sedimentation rate of still water condition resulting in higher presence of microplastics. Of the 12 freshly harvested sea salts at production pools which were actively producing during the sampling, 7 samples were positive contained microplasticss or equal to 58.3% of total contamination, with particle size range between 0.39-9.36 mm. The average MPs abundance per kg salts reached 53.3 items (Fig. 4), dominated by red (48.27%) and blue (27.59%) colors, with line (93.1%) and fragment (6.9%) forms. Meanwhile, there was a trend of lowering MPs abundance in the order of water, sediments and salts (Fig. 5). Lastly, the FT-IR analysis came out with three different polymers, i.e. Polyvinyl acetate (abbreviated as PVAc) accounted for 41.7%, Polyethylene (abbreviated as PE: 33.3%) and Polystyrene or PS (25%) (Figs. 6, 7 and 8).

Yang *et al.* (2015) discovered MPs content of 550-681 items/kg sea salts sold for human consumption in China, while Cordova stated results of 10-20 MPs/kg sea salts sampled in several sea salt producing ponds in northern central Java (Nat. Geo. Indonesia, December 3, 2018). The most recent work examined salts from 21 countries by Kim *et al.* (2018) was also shown that sea salt from Madura, Indonesia as the dirtiest sea salt contained more than 660 MPs/kg. Consequences of microplastics contamination from three different kind of samples observed will definitely increase community concern on plastics pollution in the aquatic environment. Observation on water samples come out with fear-provoking level of contamination of 68.75% which pin-pointed on how high the level of MPs contamination, particularly when looking at location of sample

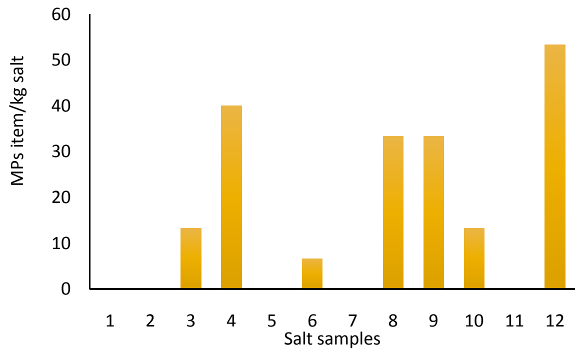

Fig. 4: MPs item abundance per- kg salt

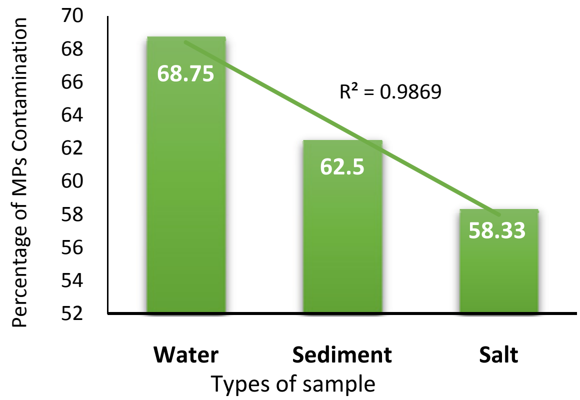

Fig. 5: Percentage of total MPs contamination level from different types of sample

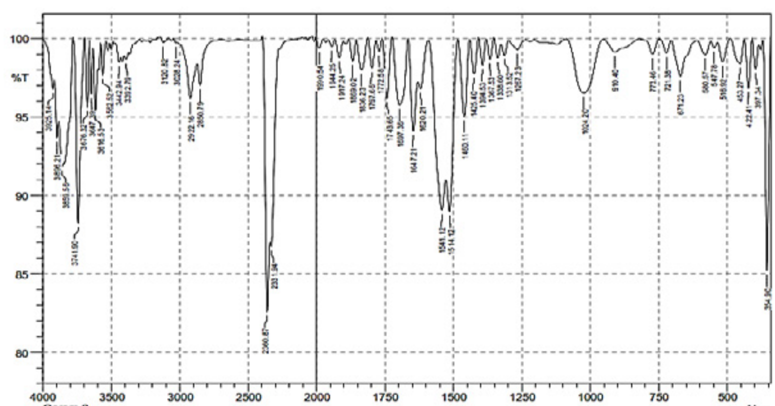

Fig. 6: Spectrum of polyvinyl acetate (PVAc) polymer from FT-IR Shimadzu reading (300 scans) of salt samples with wave numbers 350-4000/cm

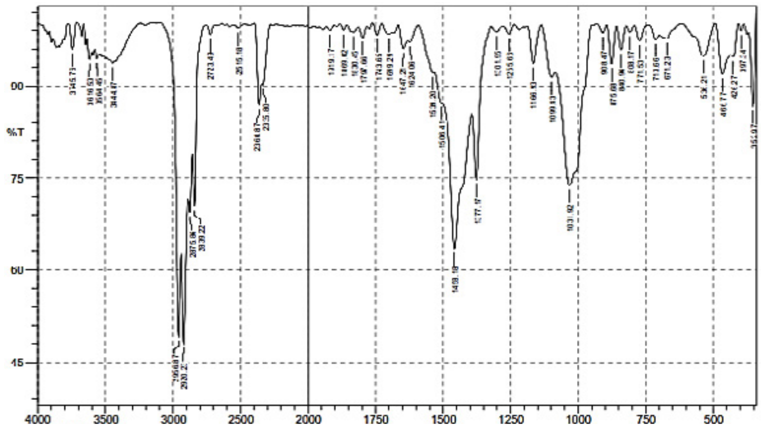

Fig. 7: Spectrum of polyethylene (PE) polymer from FT-IR Shimadzu reading (300 scans) of sediment samples with wave numbers 350-4000/cm

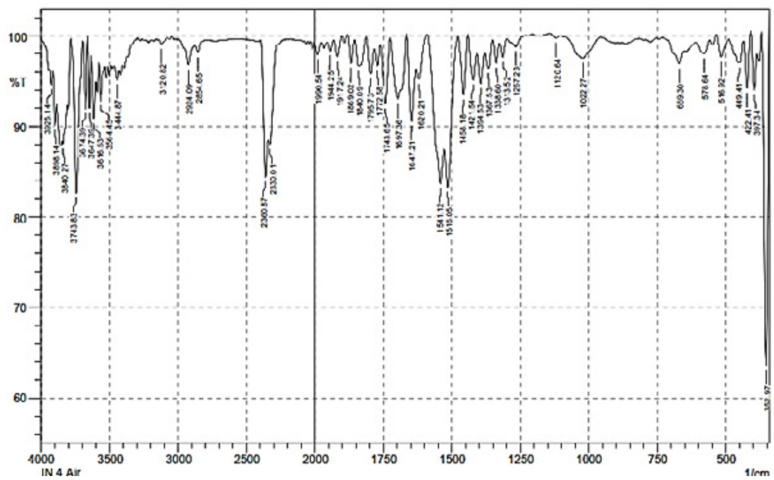

Fig. 8: Spectrum of polystyrene (PS) polymer from FT-IR Shimadzu reading (300 scans) of water samples with wave numbers 350-4000/cm

points at sedimentation/heating pools (HP1 and HP2), where highest numbers of MPs were observed. The same locations were also identified with highest numbers of MPs particles, although the correlation of coefficient is not high enough (r=0.46). In the marine environment, microplastics are typically found as fragments, lines, films and pellets (Fig. 9) and are composed of diverse polymers (Hidalgo-Ruz *et al.*, 2012).

Sources of microplastics pollution are diverse, making their types varies widely (Rochman *et al.*, 2019). During plastics production several distinct additives were used to provide several properties, including plasticizers, anti-microbial agents, UV-stabilizers, heat-stabilizers and flame retardants such as polybrominated biphenyl ethers (PBDEs) (Talsness *et al.*, 2009; Lithner, 2011). Chemical additives may cause toxic effects, and microplastics have ability to accumulate POPs and heavy metals which may therefore could transfer hazardous toxic substances from POPs to seafood and salts produced from sea water and subsequently to humans (Teuten *et al.*, 2009; Rochman, 2105; GESAMP, 2016; Lusher *et al.*, 2017). Moreover, since microplastics are ubiquitous in the marine environment, they are increasingly contaminating species and all kind of marine foodstuffs, including sea salts. Inevitably, humans are consuming microplastics at some level. Although

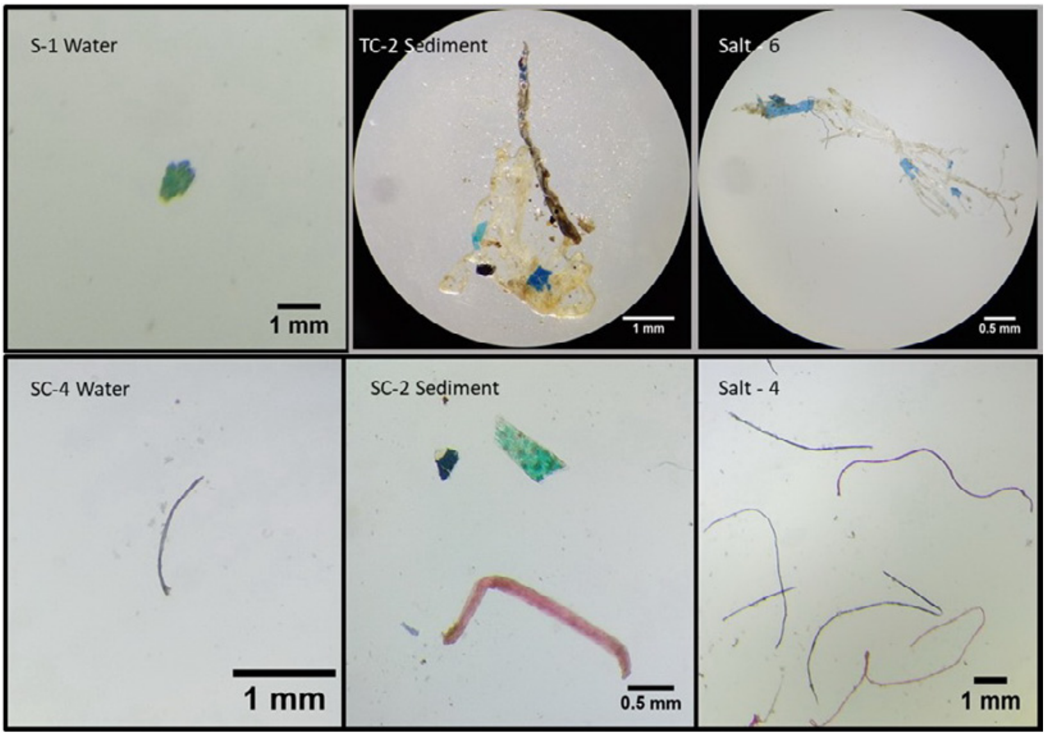

Fig. 9: Selected photographs of MPs ietmfrom water, sediment and salt samples observed with stereo microscope (magnification x45)

human body's excretory system is able to eliminate microplastics of >90% of ingested microplastics and nanoplastic via feces (Wright and Kelly, 2017), but retention and clearance rates are mainly affected by the size, shape, polymer type and additive chemicals of microplastics ingested by humans (Lusher *et al*., 2017), making the impact of MPs ingestion on human health remains unknown and is an issue of growing concern (Galloway, 2015). Wright and Kelly (2017) further predicted that ingested microplasticss may cause inflammation in tissue, cellular proliferation, and necrosis and may compromise immune cells. Direct exposures to POPs and other chemicals associated with microplasticss may affect biological systems and pose specific threats to juveniles of humans and animals, including at low doses (GESAMP, 2016; ATSDR, 2015).

Plastic polymers consist of repeating monomers, which form the backbone of the polymer. This backbone structure is the fundamental difference between polymer types, informing a plastic's physical and chemical properties. Polyvinyl acetate (PVAc) is a synthetic rubber polymer made from vinyl acetate (C4H6O2) monomer. The polymer is commonly used as mixture of anti-fouling paint to protect boats from barnacles and others. Pallengu village waters used to be one of prominent wooden boat/ship industry locations in south Sulawesi prior to 1990s, and is located at Mallosoro Bay with relatively little oceanography dynamics. It is then suspected that the high incidence of PVAc observed (41.7%) was mainly derived from paint residues of wooden boat/ship industry in the past. Polypropylene (PP) and polystyrene (PS) are two of the most produced and consumed polymers (PlasticsEurope, 2017). Polyprophylene lend themselves for application in packaging of consumer products, ropes, carpets and clothes. While polystyrene include polystyrene foam, regular polystyrene plastic, and polystyrene film. With such diverse appplications of the three polymers, microplastics both from primary and secondary entities could arrive in Mallosoro Bay, Jeneponto. Last but not least,with respect to a consistently decreasing MPs abundance in the order of water, sediment and salt samples (Fig. 5) with strong correlation value in recent finding, it is clear that there is a need to increase sea water management as raw material source for traditional sea salt production at Pallengu village, Jeneponto, Indonesia. First option would be to install micro-filter at the secondary channel as primer water-ways to each salt pond clusters. Second option would be a regular water siphoning (coud be executed during non-production time in rainy season) from sedimentation/heating pools where high MPs concentrations were detected.

CONCLUSION

The presence of microplastics in salt is obviously distressing, particularly with the fact that the sea salts produced in Jeneponto were chiefly used as cooking constituent in most community members' kitchens. Recent findings on the occurrence of microplastics in traditional sea salt producing pond components: water, sediment and freshly harvested salts, have clearly shown the presence of solid microplastics pollutant which may have absorbed a wide range of pollutants, and the possibility of toxic effects associated to direct consumption by human do exist. However, current knowledge on toxic effects of microplastics to human is mostly based on research conducted in last 5 years making the collective understanding is limited regarding the sources, fate, modes of exposure, bioavailability, and toxicity of microplastics and their associated chemicals in the marine environment. Lastly, if this situation continued without any intervention from relevant institutions aiming for microplastics reduction in sea water as raw material in salt producing ponds, high risk for consumer health and community business in sea salt production might be hindered which will eventually loosing their livelihoods.

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CONFLICT OF INTEREST

The author declares that there is no conflict of interests regarding the publication of this manuscript. 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 observed by the authors.

ABBREVIATIONS

ASTDR	Agency for Toxic Substances and Disease Registry
BPS	Badan Pusat Statistik (Indonesian Government Agency for Statistics)
cm	Centimeter a metric unit of length equal to 1/100 m
cm^2	centimeter square is a unit area with 1 centimeter on each side
^oC	temperature in Celsius
FT-IR	Fourier transformed infrared spectroscopy
GESAMP	Group of Experts for Scientific Aspects of Marine Protection
g/cm^2	unit of concentration where a mass unit (g) mixed and/or dissolved in a centimeter square of unit area
HDPE	High density polyethylene (kind of plastic polymer)
HP	heating pool is a unit in salt producing pond where sea water was heated for 24 hours prior to be drained into production pool
kg	kilogram (basic unit of mass in the metric system, 1 kg is equal to exactly 1000 cubic cm of water or equal to 2.2 ponds or 1000 g
KOH	Potassium hydroxide is an inorganic compound and is commonly called caustic potash
L	liter is a metric unit of capacity equal to 1,000 cubic centimeters
min	one of the 60 parts that an hour is divided into, consisting of 60 seconds
MPs	Microplastics is term given to plastic sized 1 µm - 5 mm
MT	metric ton is a unit of weight equal to 1000 kg
mm	Millimeter is unit of length is metric system and equal to 1/10 cm
PBDEs	Polybrominated biphenyl ethers are organobromine compounds that are commonly used as flame retardant
PC	Primary channel is the first place where sea water flow into and used as source of raw water in salt producing ponds clusters
PE	Polyethylene is a kind of plastic polymer mainly used for plastic bags, food containers, and other packaging

POPs	persistent organic pollutants are organic compounds that are resistant to environmental degradation through chemical, biological, and photolytic processes
ppt	part per-thousand is quantity of 1 g of salts in 1000 ml sea water
PS	Polystyrene (a kind of plastic polymer) is a synthetic aromatic hydrocarbon polymer made from the monomer styrene, which can be solid or foamed in application.
PVAc	Polyvinyl acetate (a kind of plastic polymer) is a type of thermoplastic made from aliphatic rubbery synthetic polymer.
%	percentage (part per-hundred)
r	is a correlation coefficient measures the strength and direction of a linear relationship between two variables
r^2	is a statistical measure that represents the proportion of the variance for a dependent variable that's explained by an independent variable(s) in a regression model
rpm	rotate per-minute, a measure of rotation frequency in a minute
S	sea
SC	secondary channel is part of sea salt producing pond surrounding a cluster of producing system where sea water later drained into the tertiary channel.
TC	tertiary channel is part of sea salt producing pond which later to be pumped into sedimentation/heating pool at each cluster of producing system.
UNEP	United Nations Environmental Program
ZnBr	Zinc bromide
$ZnCl_2$	Zinc chloride

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