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ORIGINAL RESEARCH PAPER

Waste oil management: Analyses of waste oils from vehicle crankcases and gearboxes

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ABSTRACT: In accordance with waste strategy for Turkey, the study was carried out to analyses waste engine crankcase oils and waste gearbox oils generated from vehicle maintenance services in order to determine their suitability for recycling, recovery or final disposal based on regulation published by Turkish Ministry of Environment and Forestry on 21 January 2004. The regulation requires all waste oil neither abandoned nor released into the environment and all batches must be analyzed for arsenic, cadmium, chromium, lead, chlorine, total halogens, polychlorinated biphenyls, and flash points. The content analysis showed that the heavy metal concentrations in waste engine crankcase oils were varied considerably, between the metals analyzed; lead the highest is followed by chromium, arsenic and cadmium. In addition, higher amount of chlorine and total halogens, were detected in some samples, while polychlorinated biphenyls concentrations remained below regulatory limits for all samples. The analyses revealed that waste engine crankcase oils from fifteen to thirty five years old vehicles contained chromium, lead, chlorine and total halogens levels above legal limits set by Ministry of Environment and Forestry for recycling. Conversely, in comparison to the findings from the analyzed series of old vehicles, the waste engine crankcase oils samples from new vehicles and all waste gearbox oils are eligible for recycling.

KEYWORDS: *Conservation; Hazardous waste; Polychlorinated biphenyl (PCB); Recycling; Total halogens (TX); Waste engine crankcase oils (WCO); Waste gearbox oils (WGO); Waste management; Waste oils.*

INTRODUCTION

Lubricating oils are used in many types of machines to minimize friction, heating and wearing by preventing metal to metal contact between moving parts, especially in internal combustion engines and gears. Materials protect engines and gears in all operating conditions and minimize needed repairs and maintenance, extend equipment life and minimize adverse effects (e.g., dropout and corrosion). Engine and gear lubricants are highly complex mixture of base oil (mineral or synthetic) and additives mainly such as ash dispersants, detergents, antioxidants, metal deactivators, viscosity index improvers, pour point depressants. These chemicals are soluble in base oils and modify the properties with stable performance. Lubricants usually contain up to 30 volume percent additives in the finished lubricating oil (Speight and Exall, 2014) and provide maximum protection under extreme temperatures and operating conditions. Annually, the modern world's demand for lubricants is about 40 million metric tonnes (Fuentes *et al.*, 2007) and demand is expected to grow approximately 0,5% per year for

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Europe (Mandakovic and Novina, 2015). According to statistics, automotive and industrial sectors are the major end-use sectors and 50% is used in the automotive products (Fuchs, 2000). During service automotive lubricants lose lubricating capacity and become contaminated with water, metal particles, acids, or debris, need to retain. These oils must be changed and removed from vehicles at proper intervals, no later than the maximum number of driven miles, given by the car manufacturer. When the oil is not replaced, contaminants form sludge or the oil gels which provide the less protection from severe damages compared to operations with fresh lubricants. Several factors like equipment load type, mechanical condition and other variables such as severe environmental conditions give rise to the breakdown of lubricants during operation and determine the level of oil loss.

The waste oils are estimated worldwide that less than 45% available for collection and the remaining 55% are dominated by either misuse or improper set-up in the field (El-Fadel and Khoury, 2001). The amount of waste oils that is managed annually in Europe is very high approximately 3 million tonnes (EC, 2016). These wastes are considered as hazardous wastes (Rincon *et al.*, 2007) are not acceptable for landfills, thus should be collected separately to prevent pollution and enable reuse and recovery of materials. Mismanagement of waste oils can create not only environmental problems but also health hazards. The presence of waste oils in the environment requires continuous attention and management so. Because waste oils have a great tendency to bind to organic matter, clay particles (the soil adsorption complex) and are expected to migrate and accumulate in organisms (Makkar and Cameotra, 2002) that cause ecosystem pollution. On the other hand, drips of crankcase oils on the street surfaces are potentially sources of heavy metal contaminants in urban environments. These contaminants can travel great distances through surface waterways and estuarine environments (Brown and Peake, 2006) and pollute drinking water wells and other water resources. Metal contaminated sites represent hazardous conditions and threaten many species that live in narrow habitats. Although metals can be transformed from one to another by such situations as complexation, redox state etc., they are permanent and cannot be destroyed (Ghaemi *et al.*, 2015). Among the metal contaminants, lead is one of the most important environment pollutants and has gained increasing attention in recent years (Fontana *et al.*, 1996), with a soil retention time of 150 to 5.000 years (Kumar *et al.*, 1995).

The best environmental options, for the management of waste oils follow the 'waste hierarchy' by recycling, recovering and then disposing. Waste oils can be used as an alternative fuel in a variety of engine configurations and other applications. Its gross calorific value is greater than 42,9 MJ/kg (Ketlogetswe, 1998). But this method represents another poor use of renewable sources and probably causes a disproportionate amount of emissions; contribute significantly to air pollution (Tiwari and Khatana, 2012). However significant technological advances and knowledge have accumulated in recent years and biomass resources can be used to generate fuel that does not require mining or drilling of hydrocarbon reservoirs (Golzary *et al.*, 2015). Thus, recycling of waste oils is the best option in financial and environmental terms. Pawlak *et al.* (2010) reported that 3,8 L of waste engine crankcase oil mainly may be regenerated into 2.3 kg material for manufacturing lubricating oils. And EPA (1996) also presented, while 67 L of crude oil is needed to extract 1 L of engine oil, only 1.6 L of waste oil is required to generate the same quantity of engine crankcase oils. For comparison, Lin and Mendelssohn (1998) indicated that a 150 kg barrel of crude oil typically contains only 1.9 kg of lubricant-quality based oil.

In Turkey, as in many developing countries, waste oils from vehicle engine crankcases and gearboxes had not adequately characterized, due to difficulties and lack off-site data. A limited volume of waste oil is recycled and turned back into its basic components, e.g. base oil. Thus, great interest must be given to the research of these waste oils. According to the data of the TurkStat (2014), the number of motor vehicles in Turkey reached 18 million in 2013 and it continues to grow. It is estimated by the application of waste factors, the total quantity of the waste engine crankcase oils (WCO) and waste gearbox oils (WGO) generated during vehicle maintenance and repair activities as 320.000 ton/year (Yılmaz and Yetis, 2008). Thus, the increasing generation of waste oils across the country has become a major

Table 1: Waste oil categories according to regulation (MoEF, 2008)

Pollutants (ppm)	Limit values		
	Category I	Category II	Category III
As	< 5	Max. 5	> 5
Cd	< 2	Max. 2	> 2
Cr	< 10	Max. 10	> 10
Cl^-	Max. 200	Max. 2000	> 2000
Pb	< 100	Max. 100	> 100
TX	Max. 200	Max. 2000	> 2000
PCBs	Max. 10	Max. 50	> 50
Flash point	Min. 38 °C	Min. 38 °C	-
APPLICATION	RECYCLING	RECOVERY	DISPOSAL

concern. In fact, these oils still contain lubricating fraction and therefore can be recycled to make new lubricants. Generally, WCO and WGO are listed as a hazardous pollutant as a consequence of their chemical composition which can cause adverse effects to human or the environment health. But when they are properly managed, they are excluded from hazardous wastes.

Since 2004, waste oil management systems are available in Turkey and regulated by the heavy metals, the halogens, polychlorinated biphenyl (PCB) content and flash point. The regulation on the control of waste oils is published on 21 January 2004 dated on the Official Gazette with No: 25353 which is in line with European Directive 75/439/EEC on the disposal of waste oil (MoEF, 2008). The regulation requires responsible management of waste oils through the entire recycling process and prohibits illegal disposal. The categories are based on whether or not the oil is contaminated with chemical impurities and arranged in a hierarchical structure. The regulation and the category limit values are given in Table 1.

According to the Regulation, waste oil management incorporates recycling through regeneration for re-use, recovery as supplementary fuel in cement or lime production plants and disposal at hazardous waste disposal facilities in order to minimize the environmental impact. Category I waste oils are legally authorized in order to recycling without restriction and they include WCO and WGO. Therefore, the objective of the current research was: to determine of WCO and WGO categories within the framework of "The regulation on the control of waste oils" to assess their suitability prior to applying recycling, recovery or disposal options. This study had been carried out in many provinces (İstanbul, Kocaeli, Eskişehir and Ankara) of Turkey and had been performed in 2014, by Environment-Cleaner Production Institute and Chemical Technology Institute.

MATERIALS AND METHODS

Sample collection

Two types of waste oils have been studied. Two hundred WCO and twenty WGO samples from different regions of Turkey, randomly selected, were studied between January and October in 2014. In order to reflect the present state, samples were collected from the authorized and private service stations are located in four provinces (İstanbul, Kocaeli, Eskişehir and Ankara). That means that the samples of each waste type are drained from different sources. New and older diesel and gasoline powered vehicles covered a variety of models from major vehicle manufacturers. The vehicles were frequently run beyond recommended service intervals for oil change and under different driving conditions. The sample distribution is as given in Table 2.

Table 2: Sample distribution

Vehicle type	WCO	WGO	Model year interval
Passenger	112	-	1980-2014
Light commercial	37	-	1990-2008
Truck	8	3	1998-2010
Bus	43	17	1982-2010
TOTAL	200	20	1980-2014

During the sampling of the WCO, vehicles were operated for 15 minutes in order to take the representative sample. The first 0.5 liter of the oil sample was taken into a separate container and prevented from mixing into the sample. As distinct from engines, WGO were directly taken into the sample containers and sampled. The samples were kept in the sampling leak-proof and dark-colored glass containers. All analyses were completed by a particular operator within 30 days.

Characterization

Heavy metals

Waste oil samples were analyzed 'as received' in order to account the metals. The heavy metal (arsenic, cadmium, chromium and lead) contents of waste oils were analyzed by using Optical Spectrometer (Spectroil M/F-W) according to the ASTM D 6595 standard (ASTM, 2005). This method was chosen as the most appropriate experimental procedure since it does not require any dilution or preparation. Therefore, analyses were carried out faster and the contaminant losses caused by sample preparation were minimized. Before the analysis the waste oils were heated and stirred to ensure that it is homogeneous before use. Multi element calibration standard solutions of 5, 10, 30, 50, 100, 200, 500 and 900 µg/g were obtained from Spectro Laboratories Inc. and Conostan Oil Company Limited and used for the measurement of oil samples. The reported concentration could not deviate more than 5 percent from the standards.

Chlorine and total halogens

Total halogen content in waste oil was expressed as the sum of chlorine (Cl^-), fluorine (F^-) and bromine (Br^-) in ppm to demonstrate that waste oil has not been mixed with hazardous wastes. The halogen iodine (I) was excluded as it has no functional use in lubricants. The measurements of the samples were carried out by Ion Chromatograph (Dionex ICS 1000) according to Standard Method 4110 B (APHA, 1998). The method detection limits (MDL) for Cl, F and Br were 0.2, 0.02 and 0.1 ppm respectively. The chromatograph consisted of a Dionex DS6 conductivity detector, Dionex isocratic pump and Dionex AS9-HC 4 mm column.

PCBs

Existing regulation with regard to PCB concentrations in waste oil requires a total PCB content to be reported. PCB concentrations in waste oil samples were determined according to EN 12766-1 (2000) standard, which is recommended in European Union directives. During lubrication with chlorinated oils, high temperature and high pressure processes may lead to PCBs formation and these wastes are not easily disposed of without contamination. Such material needs to be treated as hazardous waste and is best destroyed by high temperature incineration. For PCBs analyses all samples were diluted and treated with solid phase extraction (SPE) in hexane solution. Seven chlorobiphenyls were used as primary standards in order to form the calibration curves. The hexane solvent and the PCB standards were obtained from Merck and Dr. Ehrenstorfer GmbH, respectively. The identification and quantification analyses were carried out by Gas Chromatograph (Agilent 6890) which consists of an electron capture detector (ECD) and a HT-8 column (25 m x 0.22 mm x 0.25). According to EN 12766-2 (2001), the concentrated extracts included internal standards (PCB 30 and 209) before analysis. All of the glassware for the preparation of samples and standard solutions were cleaned with deionized water, dried at 100°C and then baked at 550°C.

Flash points

The flash point of waste oils is an indication of the possible presence of volatile products such as light fuels (Abro *et al.*, 2013). The flash points of the samples were measured by Miniflash FLP according to ASTM D 6450 (2010). Flash point is the temperature at which the vapors of a fuel ignite when a test flame is applied. This test is used to detect contamination from fuel oil. The samples were mixed and homogenized for 10 minutes prior to entering the device and results were determined in Celsius.

RESULTS AND DISCUSSION

Waste engine crankcase oils (WCO)

To achieve both light weight and maximum durability, the engine block is made of aluminum or of alloy that contains aluminum as its principal content, iron and lead (Udonne, 2011). During combustion of fuel in the engine block, vehicle engines generate a high power output, fine wear and larger particles as the wear becomes. Changes in WCO during

Table 3: Average concentration of heavy metals, halogens and PCBs in waste oils

Contaminants (ppm)	Meinz *et al.*, 2004	Vermont Agency, 1996		Lulek, 1998	Brinkman and Dickson, 1995	Results of WCO/WGO analysis
		Gasoline	Diesel			
As	0 – 0.45	0	0	-	NG*	1.0-2.0/0.9-1.0
Cd	0 – 0.86	0 – 3.3	0.8 – 6.6	-	0 – 5	0.1-1.0/0.7-1.0
Cr	2.0 – 17,6	0 – 4.2	6.9	-	0 – 233	1.0-90.4/1.0-5.4
Pb	0.2 – 66.1	0 – 104	23.6 – 146	-	0 – 265	1.0-577/1.0-10.2
Halogens	NG	< 350	< 234	-	NG	52-270/<100-157.2
PCBs	NG	< 5.0	< 5.0	2.88–53.42	NG	< 0.1/<0.1
Sample size	9.0	17	4.0	13	96	200

*NG: concentrations not given

operation are dependent on the original base oil, mechanical conditions, type of engine, retention time and type of fuel combusted (Mazur *et al.*, 2004). The most common source of contamination problems in crankcase engine oils are degraded additives, engine blow-by, burnt oil, dirt, dust, wear metals from engine components and incomplete combustion of fuels such as gasoline. Wear metals are important waste oil components and heavy metal particles are introduced into the oil in the crankcase of the engine as a result of wear, corrosion or external particles from the surrounding environment. Several authors have reported elevated metal concentrations (primarily chromium and lead) in WCO. A survey of the literature and results of WCO analysis was tabulated in Table 3 according to the contaminant of interest. The literature indicated that very little information is available on the presence of hazardous constituents that occur in waste oils, because of only a few studies have focused on characterization and management needs related to regulation policies.

Brinkman and Dickson (1995) investigated waste oil samples from gasoline engine crankcase, quick lube oil, waste oil tankers and industrial oil, and measured metals, chlorinated solvents and polycyclic aromatic hydrocarbons (PAHs). The study showed that the values ranging from 0 to 5 ppm, 0 to 233 ppm and 0 to 265 ppm for cadmium, chromium and lead respectively. The highest mean concentration obtained in the study is lead, thus showing affinity to lead accumulation. The profile of the metal content of waste oils in the study were found in the order Pb > Cr > Cd. Other studies, likewise, reported higher concentrations of chromium and lead than arsenic and cadmium in waste oils. Vermont Agency (1996) also found very high concentrations of lead in WCO, but concentrations of heavy metals were high only in diesel WCO, and arsenic did not found in waste oil samples. This is also similar to Meinz *et al.* (2004) that lead accumulates in WCO.

The halogens, being essential for additives, due its chemical property were also accumulated in significant. Furthermore, the Vermont Agency revealed comparatively low levels of PCBs in waste oils. Also, comparable to other study the highest concentration was 53.42 ppm reported by Lulek (1998). Levels of PCBs obtained in this study were higher than 5 ppm reported by Vermont Agency (1996). In comparison to standard limits, highest concentration of PCBs in WCO revealed value upper the Stockholm Convention limits.

In the present study, the WCO survey included analysis of two hundred samples from various services. The focus in the survey is placed on passenger and light commercial vehicles, nevertheless the fuel-type-drive type. The results indicated that analyzed samples were contaminated by arsenic and cadmium in low concentration, below 1 ppm for most analyses. Generally, heavy metal levels ranged from 1 to 2 ppm arsenic, 0.1 to 1 ppm cadmium, 1 to 90.4 ppm Cr and from 1 to 577 ppm lead. In the case of arsenic and cadmium, the accumulation in WCO never exceeded the limit values. Because the accumulation behavior of chromium and lead was different (significantly higher) than of arsenic and cadmium (significantly low). The total concentrations of lead and copper are higher in WCO, where there is also high variability in the concentrations. This is consistent with the findings of Vermont Agency (1996), Meinz *et al.* (2004) and Brinkman and Dickson (1995). These metals are believed to be largely associated with engine block wear and are also found in the fresh lubricating oil to enhance the performance of the formulation. The concentration of lead increased with increased usage, possibly indicating engine bearing damages over time and use of galvanized containers as long term storage vessels (Hamawand *et al*., 2013). The heavy metals that gave rise to the utilization of WCO, other than recycling, are chromium and lead. These contaminants are detected over the limit values given in the regulation, especially in passenger and light commercial vehicles which are over fifteen years of age. Fifty seven samples of WCO were found to contain high concentration of chromium (av. 16.5 ppm) and lead (av. 228 ppm). Different results were obtained with WCO samples. The difference in the accumulation behaviour chromium and lead may be due to solubility properties and to the chemical structures. Although lead-free gasoline is used today, lead continued to be present in WCO probably due to lead deposited from fuel combustion in old car engines. Thus, WCO become enriched with lead during the operation in old vehicles. Because, from the early 1920s lead has been blended with gasoline primarily to improve combustion (Durrani, 2013). And also extremely high content of these elements in WCO mainly depends on lubricating conditions inside in the crankcases. Fig. 1 shows the chromium and lead content of WCO.

Apart from halogen content in waste oils, the determination of PCBs is required. Among the persistent organic chemicals, PCBs have been used in many different products and applications as coolants and insulating fluids for machining operation. Thus they are the most concern as environmental contaminants worldwide. But, Tanabe (1988) concluded that PCBs never used as oil additives in the automotive industry. This is because of the dioxins can build up from PCBs and may result in increased emission level (Hewstone, 1994). The most significant finding, related to the content of PCBs, is that the analytical results verified that no PCBs were detected at a detection limit of 0.1 ppm. In WCO, total PCB concentrations fall within the recycling limits. The presence of heavy metals in WCO due to wear of metallic engine

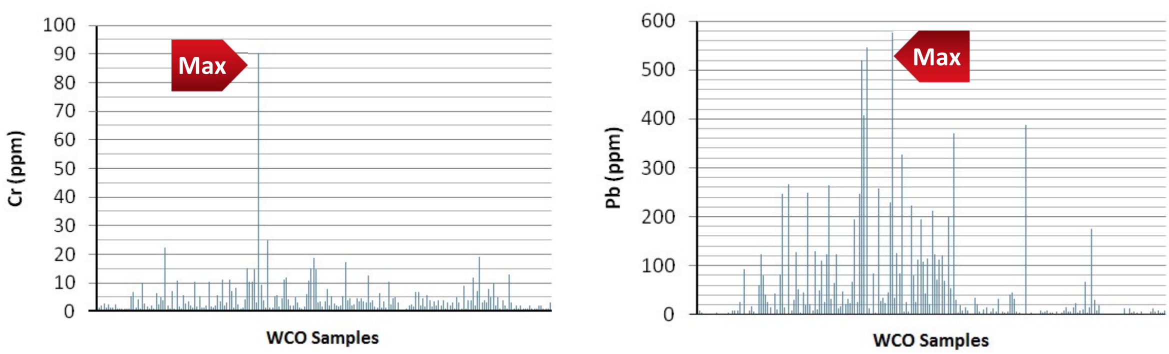


Fig. 1: Chromium and lead content in examined WCO

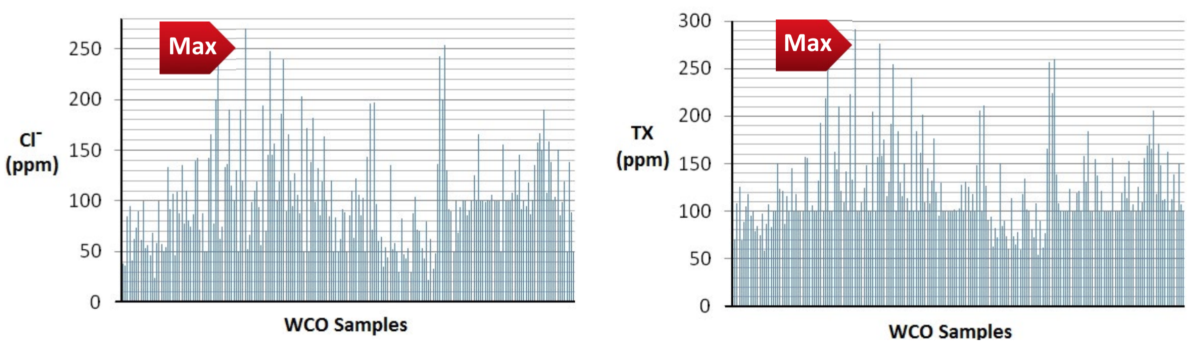
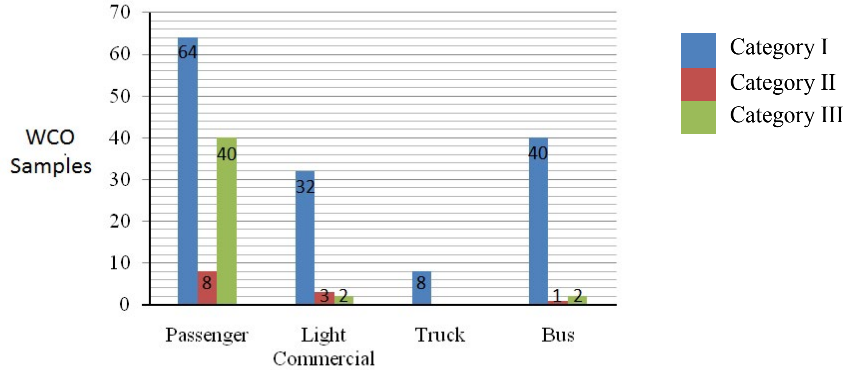
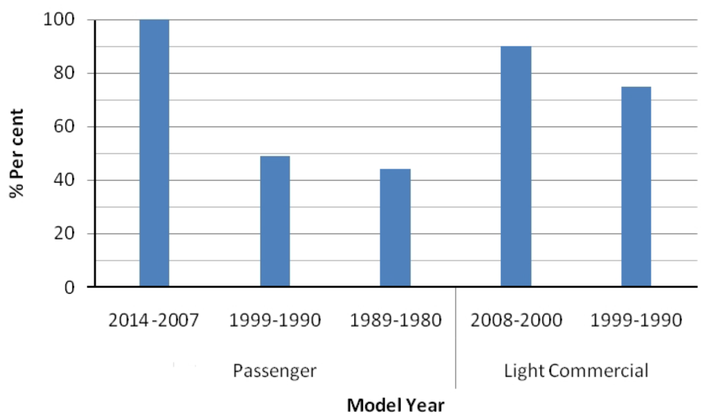


Fig. 2: Concentration of Cl- and TX in WCO



	Passenger	Light commercial	Truck	Bus
Category I	64	32	8	40
Category II	8	3	0	1
Category III	40	2	0	2

Fig. 3: Distribution of the WCO samples according to the categories



Model Year	% Passenger		Model Year	% Light Commercial
2014-2007	100		2008-2000	90
1999-1990	49		1999-1990	72
1989-1980	44			

Fig. 4: The suitability of the passenger and light commercial vehicles for recycling according to the model year intervals.

components did not lead to enhanced PCBs levels in WCO. Our own data are in agreement with the findings of Vermont Agency (1996) on WCO. WCO were contaminated not only with chromium and lead but also with Cl^- and total halogens (TX). Samples were significantly contaminated with Cl^- and TX and a good correlation between measured Cl^- and TX was obtained. The examination of individual results further revealed that six from two hundred samples exceeded the Cl^- maximum allowable limit of 200 ppm for recycling. Their results indicated that chlorine contamination seems to be primarily associated with the fuel additives, base oil contaminants, lubricant additives, salt drying of fuels and salt from roads (Tanabe, 1988). As expected, TX was higher in WCO than Cl^-. A total of twenty from two hundred samples also exceed the limit of 200 ppm. It should also be pointed out that the presence of Cl^- did not enhance PCB content in samples. Fig. 2 shows the data obtained. The distribution of the WCO samples according to the classification is given in Fig. 3. Arsenic can be seen, most of the WCO from passenger cars and all truck are inventoried as Category I.

The suitability of the waste oils from passenger and light commercial vehicles for recycling according to the model year intervals is given in Fig. 4. The vehicles' age seriously affect oil composition to a significant degree, resulting in inconsistent lubrication. Thus, lubricants were contaminated with wear debris generated by engine pistons during extended usage. Arsenic shown in Fig. 4, WCO from new vehicles are suitable for recycling depending on whether there is a collection mechanism in place within the services. Further the flash point ranges for WCO lay between 190 and 210 °C. Thus, the present flash points for this study are agreement with the limit value set by the regulation.

Waste gear oils (WGO)

The gear oils are special oils that have the chemical stability to prevent the formation of substances which would cause thickening of the oil and corrosion on the surfaces of the sensitively processed gears. Being different from the crankcase oils, gear oils are not exposed to the effects of the combustion process during their use and have greater load-carrying capacity. During the study twenty WGO samples were analyzed for heavy metals, Cl^-, TX, PCBs and flash point. The experimental results showed that heavy metals in WGO are in the order: Pb > Cr > As > Cd. In the analyzed WGO Pb was noted in twelve samples. The highest concentration for this element was 10.2 ppm and the lowest 0.8 ppm. The variation between the minimum and maximum value was 9.4 ppm. Similar to lead, chromium was found in the analyzed samples, ranging from 0.5 to 5.4 ppm. In half of the analyzed samples the content of Cr did not exceed 1 ppm. In the analyzed samples the average content of Cl^- and TX in the samples amounted to 108 and 111 ppm, respectively. In half of the analyzed samples the content of Cl^- and TX was above 100 ppm. It can be seen that there are significant correlations between Cl^- and TX. The flash point of WGO lay between 178-216 °C. The values obtained in WGO are higher than WCO. None of the analyzed samples of WGO showed the presence of PCBs. It may be concluded that the risk posed by PCBs is minimal.

In general, total concentration of heavy metals, Cl^- and TX was much lower than WCO meaning that the WGO are good alternative sources in an environmental point of view. This was very valuable information on WGO, which were less contaminated than WCO, which were collected from services. The analyses conducted on the WGO showed that contaminants below regulatory limits and these waste oils are recyclable through refining and regeneration. During the collection of WGO without being mixed with the WCO is highly important for protection of their chemical composition which is required for recycling.

CONCLUSION

Large quantities of engine crankcase and gear oils are consumed every year in Turkey and further contributing to waste oil's generation originated from maintenance services. The waste oils, if not carefully disposed of, will contaminate environment and cause serious damage to human and environmental health. Proper management will minimize problems, and help conserve our valuable natural resources. WCO and WGO are valuable types of wastes with hydrocarbon content and they have potential for material recovery via refining processes. The collection and storage of WCO and WGO separately based on their categories are the most important conditions for recycling. Waste gear oils are already available for recycling without any need for analyzing. But, the chromium, lead, chlorine and total halogen content of WCO is a problem. In consequence, the age of the vehicle is important with regards to the recycling of the WCO that are generated from the passenger and light commercial vehicles. Overall, the results indicate that WCO from new vehicles and all waste gear oils obtained from services can be used as a feedstock for the production of the base or new oils.

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CONFLICT OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

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