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Diethyl Phthalate (DEP) in Leachate End-Processing Sites Worldwide and Remediation Technologies for The Treatment

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Abstract. Production of plastic is inseparable from the addition of additives in the form of plasticizer which adds the flexibility of the plastic in its final product form. One of said plasticizer is Phthalate Diester (PAEs). One PAE substance that has been declared as hazardous is Diethyl Phthalate (DEP). PAEs can easily migrate from the plastic polymer into the environment and endanger living beings. One potential site of DEP accumulation is final processing site due to it receiving huge amounts of plastic waste from domestic and industrial activities. Leachate is a transport medium for DEP substance from final processing site. Efforts must be done to reduce DEP toxicity in leachate to ensure safety of health and ecosystem. The objective of this paper is to describe the hazard of DEP towards health, correlation between landfill age and DEP concentration, and appropriate remediation technique for further treatment of DEP.

Keywords: *phthalate; Diethyl Phthalate (DEP); plasticizer, landfill; degradation.*

1 Introduction

Global generation of plastic waste in 2016 is expected to reach 322 million tons [1]. Impromper handling and management of plastic waste will cause environmental accumulation which poses severe risks towards living organisms and threatens the ecosystem [2]. When plastic waste enters the environment, the plastic will undergo fragmentation and degradation into smaller particles less than 5 mm in diameter which is known today as microplastics [3].

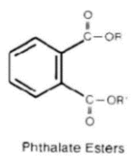

Figure 1 Chemical structure of PAEs

In the last decade, several researches have been done to observe microplastic, from its abundance to its effect on health. According to Ognowoski et al. [4], microplastic is shown to produce chronic effect in the form of increased rate of death and interbrood period, while also reducing D. Magna. Aside from the dangers of microplastic particles, additive substances could also separate and produce environmental health risks. Plastic production will always come with the addition of several additives to increase the plastic quality. The typicalgroups of additives for plastic are plasticizer, antioxidant, UV-stabilizer, lubricant, and coloring agent [5]. One of the common plasticizer additive used is Phthalate Diester (PAEs). PAEs are a group of synthetic chemical substances that is used as plasticizer to increase the usability of food packaging, toys, raincoat, personal care products, etc. [6,7]. The structure of PAEs is shown in **Error! Reference source not found.**. PAEs are only physically bound onto the polymer matrix of plastics, which could result in the migration of said substance from the product surface into the environment due to Van der Waals attraction force in ester functional groups [8,9,10]. Van der Waals force is a very weak force of attraction when compared to ionic and covalent bonds [11].

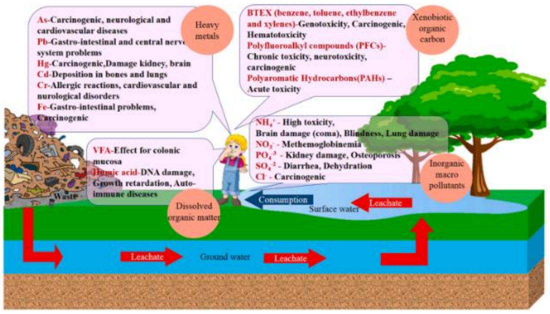

Figure 2 Illustration of hazardous substance exposure pathway from final processing sites to human [12]

The easily-removed nature of PAEs from plastic surface creates a situation where it is often leached in final processing site's leachate shown in **Error! Reference source not found.**, which makes PAEs to be found in high concentrations [13,14]. The concentration of PAEs in higher levels in final processing site leachate has also been identified by several researchers [14,15]. DEP is typically used in beauty, automotive, cleaning, dishwashing, and kids toys products [16]. Several researches have also shown that DEP usage poses risk for the nervous and reproductive systems on both male and female humans [17,18]. Due to this llooming risk, it is of essence to remediate this DEP from leachates in final processing sites, which reduces its toxicity and hazard levels for the environment and humans.

2 Methodology

This research is done through systematic review. Data and information obtained are from researches that correlate with Phthalate Diester (PAEs), specifically DEP. Literature review is done through collection of secondary data from books, scientific journals, and several past researches. The data is then summarized and presented as secondary data to answer the objectives of this paper.

3 Result and Discussion

3.1 Plasticizer and Characteristic of Diethyl Phthalate (DEP)

Plasticizer is an additive which is added to increase the flexibility of plastic polymers. Plasticizing agents are characterized as nonvolatile compounds with high boiling point, and contains material altering characteristics when added to another material [19]. Plasticizer oftentimes lack the covalent bond with the polymer, which makes them easily removed from a surface. Visual illustration of plasticizer usage is shown in **Error! Reference source not found.**.

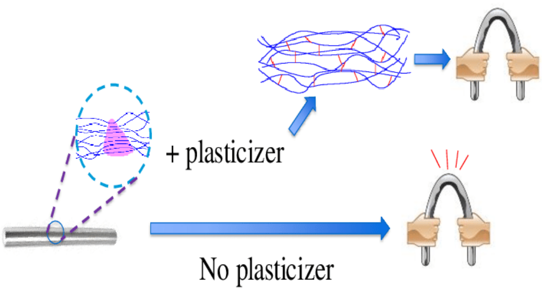

Figure 3 Plasticizer usage in a plastic matrix [20]

Diethyl Phthalate (DEP) is accumulated in the environment due to its plasticizing abilities, especially near the production and usage site [21]. Characteristics of DEP is shown in **Error! Reference source not found.**.

Tabel 1 DEP characteristics [22]

No	Characteristics	Value	Unit
1	Molecular weight	222.26	Gram/mol
2	Melting point	-40.5	ºC
3	Boiling point	295 – 302	ºC
4	Density	1.120	g/ml
5	Henry's law constant	7.8×10^{-7}	atm m^3/mol

DEP has poor solubility in water at 25ºC, which is 1.120 g/mol and tend to be more soluble in organic solvent such as alcohol, ether, acetone, and benzene [23]. This substance is colourless, coupled with slight aromatic scent and is denser than water [23]. Release of DEP into the environment must be halted due to its ability to penetrate soil, thus contaminating groundwater and nearby waterways [24]. According to Jonsson et al. [25], Dimethyl Phthalate and Diethyl Phtahalte are more hydrophilic and can easily migrate from plastic wastes into aqueous phase when compared to Butyl Benzyl Phthalate dan Di(2-ethylhexyl) phtahalate which are more hydrophobic in comparison.

The hydrophilic nature of DEP makes it easier to penetrate through soil, water, and even leachate media. When entering said media or human exposure pathway, DEP tends to be directly soluble in said media in a short period of time. This is directly correlated with the length of exposure and the concentration of DEP which will be discussed further in the following subchapter, specifically in final processing site leachate.

3.2 Landfill Leachates as A Source Diethyl Phthalate (DEP)

Leachate is the seepage of water that passes through wastes that has undergone aerobic and anaerobic processes [25, 26]. Leachate composition depends on the waste characteristics, operational time of final processing site, climate conditions, and surrounding soil conditions [26]. Leachate contains 4 main components, which are: nutrient (nitrogen), volatile organic compound, heavy metals, dan toxic organic substances [27,28,29]. Leachate component, when possessing higher levels of toxicity will cause further problem for the environment and health conditions. DEP is one of the toxic organic substances, in which US EPA has set a reference dose (RfD) for DEP exposure at 0.4mg/kg/day.

Table 1 Summary of several Diethyl Phthalate concentration in leachate

No	Location	DEP Concentration	Reference
1	Perungudi open dumpsite, Chennia, India.	17,2 mg/l	[31]
2	2 Landfill sites in Jepang	1,0 – 8,4 μg/l	[32]
3	6 Landfill sites in Denmark	<1 μg/l	[33]
4	2 Dumping Grounds,Chennia, India	56,80-495,20 μg/l	[34]
5	Landfill in Wuhan, China	N.D-43,27 μg/l	[14]
6	Landfill in Thailand	12,50 μg/l	[15]
7	Sarimukti Landfill	2,4 mg/l	(researcher's data)

*N.D = not detected

The continuously massive accumulation of plastic waste will further increase the concentration of Diethyl Phthalate in leachate. Primary data of researcher shows that DEP concentration in Sarimukti final processing is 2.4 mg/L. When compared to the other leachate from final processing sites, it is ranked as the 2nd highest in terms of DEP concentration in **Error! Reference source not found.**. This value is considered to be high concentration of DEP, due to it mostly being in the order of μg/l.

Newest research in 2019 in Chennia, India shows that DEP concentration in Perungdi Open Dumpsite India has reached 17.2 mg/l. This concentration level shows that DEP concentration increases along with the length of final processing site operation. The research of Wowkonowicz et al. [35] also shows the same result, in which the same sampling site will produce seasonal variability in 2015 and 2016. DEP concentration will again increase along with the time of operation of the final processing site. The sampling result of Wowkonowicz et al. is shwon in.

Landfill 2 is the landfill with the longest operation time when compared to other landfills. However, this landfill has some damaged sites due to earthquake and an in-progress remediation effort. This activity has caused several processes in the landfill to have reduced effectiveness compared to the start of operation. Landfill 5 shows significant hike in DEP concentration and has relatively higher concentration compared to other landfills. This is caused by roofed tank system landfilling, in which no dilution occur during the winter. Landfill 5 is also the only sitr that has the same operating condition as the beginning without any meaningful damage. This shows that changes in landfill operating condition might be related to the amount of DEP concentration accumulated in the landfill leachate.

Table 2 Concentration of DEP in Central Poland final processing site [35]

No	Location	DEP Concentration				Notes
		Summer 2015	**Autumn 2015**	**Winter 2015**	**Spring 2016**	
1	Landfill 1	<5.0	<1.0	<2.5	<0.6	First operated in 1978 and shut down in 2011, holding around 5.5 million m^3 of waste.
2	Landfill 2	<5.0	<1.0	<2.5	<0.6	First operated in 1965 and handles 2.5 million m^3 of waste. One block is damaged due to earthquake damages and is currently undergoing remediation.
3	Landfill 3	<5.0	<1.0	<0.6	<0.6	First operated in 1970 and handles 960 thousand m^3 of waste. Has several parts damaged due to earthquake. The landfill collection system is roofed tank.

4	Landfill 4	<2.5	<1.0	<2.5	<0.6	First operated in 1970 and handles 908 thousand m³ of waste. Landfill collection system is unroofed tank.
5	Landfill 5	<5.0	<0.6	4.3-4.72	<0.6	First operated in 1970 and handles 832 thousand m³ of waste. Landfill collection system is roofed tank.

Furthermore, the research of Jonsson, et al. [25], shows that the three sampling points in different year will produce different results. In 1995, DEP concentration in month-35 shows a significant increase with 220 μg/l observed. It is then slowly decreasing until it goes past the lower detection limit of <1 μg/l. Then in the year of 1997, DEP concentration is formed from the 10th month of sampling. Diethyl Phthatale on the same location with different age will create a different concentration value, too. In this case, the rate of release of DEP from plastic onto environment is shown to be faster in older landfills, which is within 10 months.

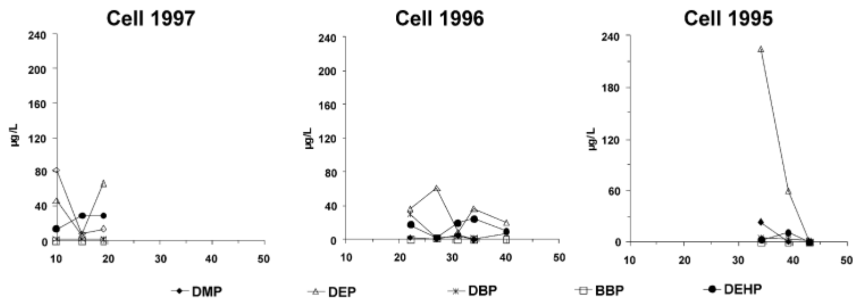

Figure 4 PAEs concentration trend in Sweden landfills in 1995-1997 [25]

Aside from the length of landfill operation, there are several factors affecting DEP concentration in leachate, which are seasonal variation, landfill size, and physical characteristics of leachate [35].

3.1 Route Exposure and Health Impact of Diethyl Phthalate (DEP)

3.1.1 Routes of Exposure of Diethyl Phthalate (DEP)

Diethyl Phthalate released into the environment could come from manufacturing process, disposal in landfills,or by leaching from products DEP-containing products [36]. Human DEP exposure has several route, which are skin/dermal, oral, inhalation, and intravein injection. Potential route of exposure phthalate in human is shown in **Error! Reference source not found.**.

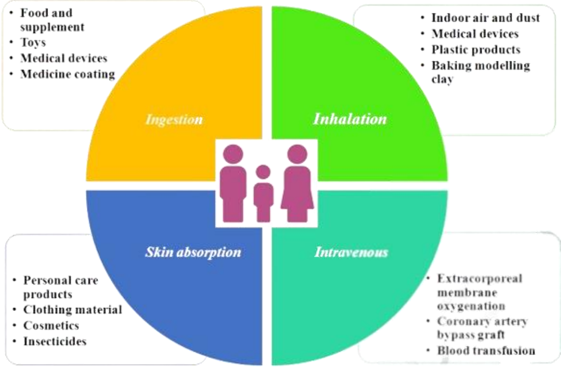

Figure 5 Potential routes of exposure to phthalates in human [13]

Several studies show that food intake is an important PAEs exposure pathway, followed by inhalation [37–39]. The following is a summary of different route of human and animal DEP exposure based on past researches shown in **Error! R eference source not found.**. DEP impact has been analysed often on dermal exposure route.

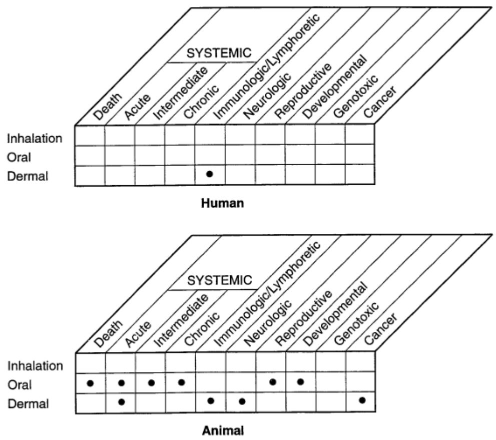

Figure 6 Summary of DEP exposure route on human and animals based on current researches [36]

Dermal exposure can be controlled and prevented by substituting chemicals used, which is practically impossible, so that another control procedure needs to be done through proper usage of personal protective equipment (PPE) [40]. PPE that works against dermal exposure of DEP includes working clothes, gloves, and safety boots [41]. Aside from PPE usage, there also needs to be hygienic practices so that dermal exposure of DEP can be minimized. One of which is through proper and often handwashing using appropriate amount soap.

3.1.2 Impact of Diethyl Phthalate (DEP) Exposure on Human Health

Several studies have proven that Diethyl Phthalate (DEP) exposure onto human via inhalation, dermal, or oral pathways causes negative impact towards human health. The following **Error! Reference source not found.** shows several summaries regarding the correlation between concentration of DEP versus its impact on human and animal health.

Figure 7 Several researches on DEP exposure impact on humans and animals

No	Location	DEP Concentration	Results	Reference
1	Puerto Rico	8-37 μg/l	Continuous exposure is predicted to cause endocrine system disruption in humans Case : premature growth of breasts in female adolescents.	[17]
2	China	0-7 ng/ml	Continuous exposure of DEP will affect absorption onto skin. Researchers concluded that skin exposure is more dangerous compared to oral exposure.	[42]
3	India	-	Testing on swiss albino mice creates a deformity in the histoarchitecture of the testes causing azoospermia and thinning of basal membrane.	[43]
4	South Korea	11 mg/l – 22 mg/l	DEP exposure has a strong proaterogenic effect towards the human macrophage and dermal through heavy modifications of lipoprotein and disfunction in HDL production Embryo and adult zebrafish exposure causes damages in nutritional hemostasis, growth, and development, due to hypolipidemia (abnormal increase in lipid or lipoprotein in blood).	[44]
5	India	10 mg/l – 50 mg/l	Results on mice testing shows DEP consumption will increase hepatic	[45]

		enzyme causing metabolic damages in glicogen, cholesterol, triglyceride, and changes in liver histology.

Due to DEP's health risks onto human mentioned above, mitigation efforts need to be done to prevent said negative impacts. Therefore, effective techniques must be implemented to repair degraded land and sites contaminated by DEP through remediation [46].

4 Remediation Strategies

One promising pathway to control plastic and microplastic pollution to minimize PAEs contamination, aside from source reduction strategies, is developing a remediation technology to cleanup contaminated sites [47]. Several PAEs remediation strategy from a certain wastewater or site is shown in pada **Error! Reference source not found.**.

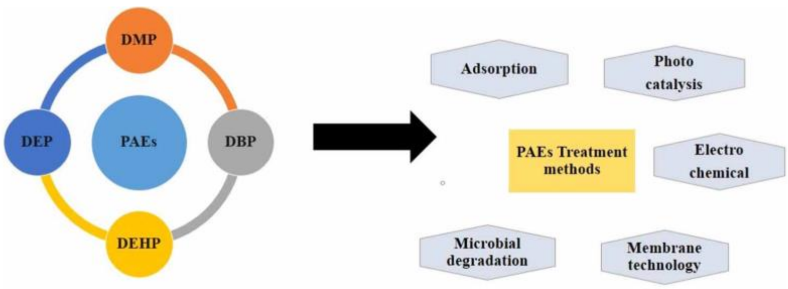

Figure 8 PAEs remediation strategies

4.1 Adsorption

Adsorption is an exothermic reaction in which a gas molecule or chemical substance will be accumulated on the surface of an adsorbent material [48]. This technique has several advantages which are easily applicable, low costs, low energy requirements, and high effectiveness and recovery of adsorbent [49]. According to Venkata Mohan, et al. [50], which removes DEP from aqueous phase using activated charcoal, shows that efficiency of adsorption ranges from 50 – 80% at 200 minutes contact time with DEP concentration of 0.5 mg/L to 3 mg/L.

4.2 Photocatalysis

Photocatalysis is a catalytic conversion process using light-induction as an oxidating agent to produce chemical species used to degrade pollutants. Photocatalysis is an emerging method in development and is aimed to have an important role in PAEs decomposition through several innate advantages it has, which are very simple in operation, eco-friendly, low cost, gentle operation method, and has the ability to remove several other pollutants in a complex water system at once [51]. According to Xu, et al. [52], which researches the efficiency of photocatalytic chemical reaction to remove DEP using UV-radiation self-photolysis, dark H_2O_2 oxidation, and a combination of both pathway shows that DEP removal is gauged at 98.6% with less than 60-minutes reaction time in combined UV and H_2O_2 photocatalysis pathway.

4.3 Electrochemical

Electrochemical method is an identified viable process option for PAEs removal due to its environmentally friendly, strong catalytic ability, no secondary pollutant production, light operating conditions, and easy operational characteristics [48]. A research conducted by Sun et al. [53] shows that catalytic electrochemical removal of PAEs using Nano-Fe_2O_3 catalyst produces promising result in which a 3-hour removal period is able to reduce more than 50% of the initial concentration using Fe-Mt medium for Fe_2O_3 planting, which is better than the simple addition of Fe_2O_3 catalyst.

4.4 Membrane Techonology

The fundamental mechanism of membrane technology is shown in **Error! Reference source not found.**. In the illustration, the red dots are soluble particles and the yellow dots are the solvent. The principle of this technology is similar to filtration in which soluble particles are filtered by the membrane surface and pure solvent will pass through the membrane [48].

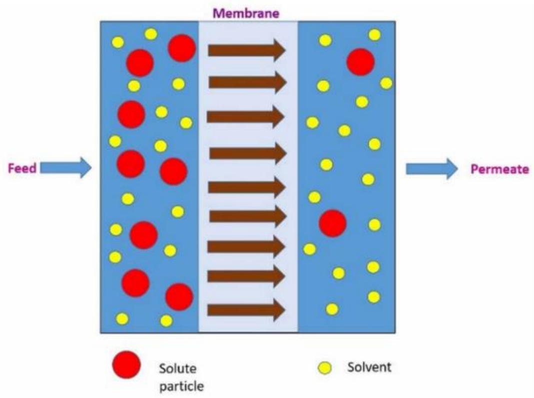

Figure 9 Illustration of membrane technology

Due to its high efficiency and ease of operation, PAEs removal is often done using this removal pathway [54]. Degradation of DEP and DEHP are observed using three membrane alternatives, which are reverse osmosis (RO) membrane RO-DS3SE, nano-filtration membrane NF-DS5DK, and ulltrafiltration membrane UF-DSGM. Each membrane used shows promising results with 97.6 – 99.9% efficiency. This technology however, requires high investments for membrane purchase, with similarly high operating costs for membrane maintenance to maintain the target efficiency.

4.5 Microbial Degradation

Several researches show that biological treatment using microorganism is able to take the main stage in degradation of PAEs substance [9]. Removal using free living microorganism is known to be beneficial and highly efficient due to their ability to adapt to their environment [47]. According to Chiellini et al. [55], degradation of plastic polymer is able to be done using fungi, bacteria, predating organisms, and other higher-order organism. Degradation of PAEs is very dependent on the surface area of bacterial colony and other nutrient requirements [36].

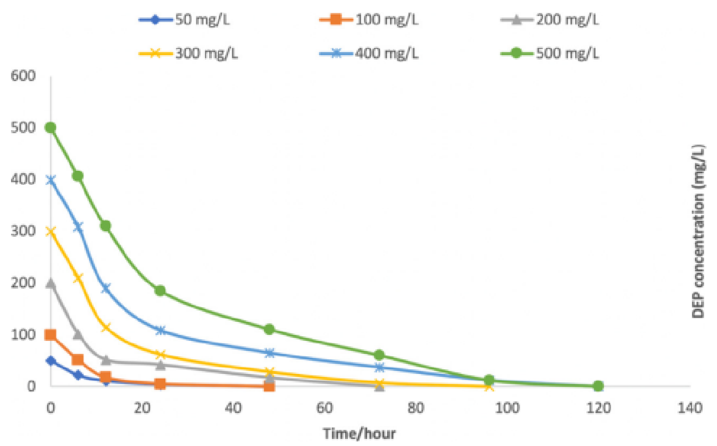

Figure 10 Result of DEP degradation using bacterial isolate [21]

In the research of Khadka et al.[21], it is shown that continuously plastic-exposed environments of mangrove and river ecosystems will provide an environment for strong PAEs-removing bacteria. Three bacterial isolates from Pseudomonas genus are observed to remove close to 100% of the DEP within 2 hours.

5 Conclusions

Based on the review done within this paper, it is known that the accumulation of DEP substance in leachate is tightly connected with how many years has passed since the Final Processing Site's first operation, in which the longer the operation, the more DEP will be accumulated. Many studies have shown that DEP produces negative impact towards living organisms, including human, animal, and the surrounding environment. The exposure pathway on human is typically oral exposure so that proper personal protection equiment (PPE) is needed, along with the habit of washing hands. Control of DEP is done through monitoring and treatment to ensure that the DEP-containing leachate is safe to dispose of into the environment. One of the most promising way to deal with DEP is through remediation. Remediation conducted using microbial metabolism and membrane technology are shown to reach efficiencies nearing 100%, which can be utilized and studied further for DEP degradation in leachate.

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