

Phthalic Acid Esters Accumulation and Biodegradation in Mesozooplankton: A Comprehensive Review

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Abstract

Ocean is a place for various living organisms but the threat to marine life is increasing day by day due to various anthropogenic activities such as dredging, over exploration, dumping of waste, and land reclamation. Burning of fossil fuel causes an increase in CO₂ content that decreases the pH of the ocean. Zooplankton are organisms that drift along the currents, tide and waves of the ocean. They are distributed unevenly all over the ocean. Their diversity, abundance and distribution are influenced by various climatic and physico-chemical parameters. Their existence plays an important role and significance in biogeochemical cycles, vertical migration, deep scattering layer, bioluminescent plankton, discontinuous distribution, ooze formation and biological indicators. Wastes from municipal, industries and domestic are discharged into the marine ecosystem. Organic pollutants like polychlorinated biphenyls, DDT, Polyaromatic hydrocarbons have grabbed much attention. Phthalates are used and produced by various industries as plasticizers (plastic additives) for durability and flexibility of products. Thus, use of phthalates has increased and several toxic studies on it have grabbed much attention as marine persistent pollutants. The current chapter reveals the review of literature in relation to zooplankton and impact of pollution on zooplankton, concentration of phthalates and a group of persistent organic pollutants (POPs), its distribution and effects on zooplankton and biodegradation.

Keywords: Phthalic Acid Esters, Mesozooplankton, Bioaccumulation, Biodegradation

1. Introduction

Marine pollution and types of pollutants: POPs are Persistent pollutants that can have longer half-life in sediment and several days in atmosphere. [1]

Toxic chemicals have now become the greatest threat to the safety of water coming from the industries, metal plating, pesticides, and agricultural fields. Pollutants present in the industrial chemicals are chlorinated hydrocarbons and heavy metal includes nickel, zinc, iron and vanadium [2]. There are few persistent organic pollutants and inorganic pollutants that are becoming a major source of marine pollution, even leading to health hazards. POPs have been studied over the past three decades as many of them are studied to be persistent bio accumulative, toxic and are found to be present in every matrix on the planet [3]. These are man-made organic compounds which are now present on both the terrestrial and aquatic environments. These are highly toxic and have a wide range of toxic effects [4-5]. Organochlorine pollutants commonly can be referred to as POP, which are pesticides, chemically stable and hydrophobic. The organochlorine compound includes Dichlorodiphenyltrichloroethane (DDT), used in agriculture as an insecticide [3]. Other pesticides that are used in agriculture that are referred to as POPs are Aldrin, Chlordane, Heptachlor, Toxaphene (Stockholm Convention et al., 2008). Polychlorinated biphenyls (PCBs) are referred to as priority organic contaminants and consist of specific properties that point out the threat to human health [6]. Class of 209 compounds is constituted by PCBs called "Congeners"; they differ in biological activity and toxicity [7]. PCBs have been reached to the marine environment and to the terrestrial environment through leakage, disposal and evaporation. PCBs tend to accumulate in the sediment biota due to hydrophobic character, persistence and low water solubility [7]. Polycyclic aromatic hydrocarbons (PAHs) are chemicals that are formed due to incomplete burning of coal, gas, oil and other organic substances [8]. There are four ways of PAH input to the marine ecosystem such as production by organisms, derived from incineration processes, derived from fossil fuel and from transformation processes in soils and sediments [9-10]. PAHs are known to be a large group, ranging from 2-ring naphthalene and naphthalene derivatives to complex ring structures [9]. PAHs can also induce carcinogenic effects when in contact with biological systems [10].

Types of Phthalic acid esters (Phthalates): Phthalates can be classified into two classes according to their molecular weight.

High molecular weight phthalates include 7-13 carbon atoms in their chemical structure which allow them to have increased permanency and durability. The most common types of phthalates include diisononyl phthalate (DINP), diisodecyl phthalate (DIDP), di-(2-ethylhexyl) phthalates (DEHP) and dipropyl heptyl phthalate (DPHP). High phthalates are commonly used in PVC (Polyvinyl chloride) products for example, cable, flooring, wall covering, adhesive films, synthetic leather, coated fabrics, roofing, automobile application [11].

Low molecular weight phthalates include 3-6 carbon atoms in their chemical backbone. The common ones are diethyl phthalate (DEP), dimethyl phthalate (DMP) and di-butyl phthalate (DBP). Low phthalates are useful in medical devices, general purpose PVC, adhesive inks and cosmetics [11].

Impact of pollution on zooplankton: The pollutant can enter zooplankton's body into three ways viz from phytoplankton diet, as zooplankton feed on phytoplankton so any pollutant that is accumulated in phytoplankton can enter zooplankton's body, from assimilation of diet and direct uptake of sea water [12]. Increasing pollution in marine water causes loss of diversity, plankton patchiness or even increase in indicator species or invasive species. This study focuses on Mesozooplankton which is a sized based fraction of zooplankton community, this fraction includes taxa and stages of zooplankton with body length between 200 μ m-2000 μ m which include groups like Copepods, Siphonophores, Chaetognatha, Decapod larva, Cladocerans and many more. The hypoxic or anoxic condition of sea water caused by pollution can be responsible for the decrease in zooplankton population [13-14]. Domestic and industrial effluent makes the water turbid that can restrict the movement of zooplankton to the desired area [15]. Eutrophication caused by nutrient loading into the sea water causes an increase in the grazing activity and due to increased

phytoplankton growth causes shift in species composition [16]. Another problem faced by zooplankton communities are changes in physical flow, particularly longitudinal changes during downstream transport [17]. Zooplankton are bioindicators for water quality assessment, rotifers with their high population turnover rates are sensitive to water quality change [17]. Zooplankton without carbonate shells are impacted by decreased pH conditions- a) Climatic induced ocean acidification b) Carbon capture leaks [18]. Thus, zooplankton is affected by every kind of pollution: it is nutrient, heavy metal, chemical etc. Almeda and team studied the interaction between zooplankton pollutants, where they mentioned three types of interaction first; Pollutants can have direct toxic effects on zooplankton. Second, zooplankton can influence the characteristics of pollutants. Third, by playing an important role in biomagnification [19].

2. Literature Review

Sources of phthalates and their ingress into the aquatic Environment:

Phthalates are commonly used plasticizers that have been used to increase the elasticity and durability of the product. Phthalates are found mostly in daily care [20] products like cosmetics, perfumes, moisturizers, toys, baby care [21], flooring material [22], wastewater [23], industries producing plastic materials and phthalates. These phthalates enter marine via drainage of domestic waste, industrial waste and medicinal waste [24] causing change in water chemistry and entry into the food web. Thus, the entry of phthalates into aquatic ecosystems is mostly due to various anthropogenic activities [25]. In addition to this, a few species of algae are also known to produce phthalates naturally [26]. Once the marine environment is in contact with phthalate it gets distributed accordingly in water, in sediment, in zooplankton and other higher organisms like crustacean, mollusk, fishes, etc via food web. Studies by [27-28], have given the account of the presence of phthalate in the water column. Appearance of phthalate in biota samples enters through the food web it gets accumulated in zooplankton which is food for secondary consumer later this secondary consumer

is eaten up by tertiary and so on or it gets ingested by the organisms [29]. Distribution of organic pollutants between the environmental sample and biota sample are related to biochemical properties, as phthalates are lipophilic in nature thus get in contact with lipids present in the organism's body and accumulate [30]. Samples were collected from the Gulf of Mexico and North Atlantic and studied the presence of two ester phthalates DEHP and DBP, the samples also consisted of other pollutants like PCB and DDT, but the samples showed a higher concentration of DEHP than PCB and DDT in water [25].

Biodegradation of phthalates:

Biodegradation of phthalates is possible with the help of microbes and this has been analysed [31] in sediment [32] and water [33]. Biodegradation of phthalate is possible by both aerobic and anaerobic bacteria.

Aerobic bacteria are well known for the degradation of phthalate esters; they are rod forms morphologically [34]. There is a progression of fungal species that is found to be beneficial in degrading phthalates [35]. Consumption of DEHP can be carried out by fungi- *Aspergillus parasiticus*, *Fusarium subglutinans*, and *Penicillium funiculosum* [36]. Marine microalgal species like *D. Salina*, *C. Meulleri* and *C. Closterium* is known to degrade phthalate [37]. A certain species of **Anaerobic bacteria** has been reported that are beneficial in degrading phthalates the species include *Clostridium sp.*, *Bacillus sp.*, *Pseudomonas sp.* and others [34-38]. Phthalate by anaerobic degradation is by decarboxylation to benzoate this benzoate is then cleaved via β -oxidation, to hydrogen, to carbon dioxide and acetate [39]. Bioaccumulation of phthalates in the marine ecosystem could be majorly due to the food web. Phthalates usually have smaller half-lives and start degrading within 28 days; prior degradation is by photodegradation [40]. High molecular weight phthalates get readily biotransformed and thus get excreted that gives lesser bioaccumulation; thus low molecular weight phthalates are more accumulative [41]. Bioaccumulation of phthalates in organisms is due to lipid content that has high affinity towards phthalates as they are lipophilic and in sediments it has high affinity towards the texture and organic carbon content

for accumulation. Bioaccumulation factor (BAF) can be calculated as the ratio of phthalates present in the biota sample to the total lipid content/ phthalates in sediments to the organic carbon in the sediments [30].

Impacts of phthalates on zooplankton:

Mesozooplankton often are known to ingest phthalates that are found in marine water due to increased plastic pollution. Phthalates get accumulated in the abdomen, alimentary canal, appendages and gills. They also obstruct the feeding, growth, respiration, locomotion and inhibit other metabolic activities [42]. Zooplankton being on the base of the food web for higher trophic level can cause accumulation in the organism's body. Once the phthalates enter into zooplankton's body they can have noxious effect on the organisms, even at deficient concentration as phthalates can express endocrine disrupting, mutagenic and teratogenic effects. Interaction between the zooplankton and phthalates have shown to affect the hormonal synthesis, alter in reproduction, increase in oxidative stress and immunotoxicity [43]. Accumulation of Dibutyl phthalate can lead to overproduction of reactive oxygen species. The quantity of phthalates in zooplankton and sediment samples was estimated, 80 % - 131 % of phthalates being present in zooplankton samples and 97% in water samples [44].

National scenario

Degradation of PAE in soil was studied by [45]. The study analyzed Di-n-methyl phthalate, Di-n-butyl phthalate, and Di-2-ethyl phthalate, finding higher degradation rates under aerobic conditions and anaerobic biodegradation. The study suggests that bacteria actively participate in degradation of DMP, DEP and DEHP.

Increasing pollution has brought several trends in zooplankton biogeochemical cycles and population. The presence of POP's and other contaminants gets accumulated in the body of zooplankton and transfers the pollutant to higher levels. The bioassay studies have been carried out by various species of zooplankton. The literature has mentioned various study areas along the sea (Arabian Sea, Bay of Bengal, North Sea, Sargasso Sea) and presence of pollutants and their concentration

levels present in the sea and biota samples [12]. A study investigated the biodegradation of polyethylene by marine bacteria found in pelagic water [46]. They isolated sixty bacterial colonies and found three positive isolates that could use polyethylene as a carbon source. These isolates showed 99% homology with *Kocuria palustris*, *Bacillus pumilus*, and *Bacillus subtilis*. The hydrophobicity of these isolates was confirmed using triphenyl tetrazolium chloride reduction tests, and the biodegradation was then confirmed by the increase in Keto Carbonyl Bond Index, Ester Carbonyl Bond Index, and Vinyl bond index.

Phthalate concentration in the atmosphere was studied. The study area was southern India. The sampling was done using a passive air sampler. Spatio-temporal trends of phthalates were studied. The 6 major phthalates chosen for the study were DEHP, DBP, DMP, DEP, BBP, DnOP. Phthalates were found in all samples, with highest concentrations in pre-monsoon, summer, and monsoon. They contributed 11-31% and 59-68%, respectively, mostly in urban areas. Exposure level for humans was found to be normal based on reference dose and tolerable daily intake (TDI) [47].

Global scenario

The effect of dredging in polluted estuaries on the larval zooplankton was evaluated. Water samples were collected from three dredging locations in Charleston harbor and its tributaries for the study. For location I the zooplankton used was *Daphnia*, the salinity was zero, for location II *Paleomonetes* with moderate salinity and for high salinity *Polydora*. The most toxic water was from the disposed area, the dredging samples were found to be least toxic [48].

The toxicity of phthalates to *Gymnodinium breve* dinoflagellates was investigated. Sea water was used to cultivate *Gymnodinium breve*. For the first assay, the median tolerance limits for the four phthalates they examined—DBP, DPP, DEP, and DMP—were 0.6, 6.5, 33.0, and 125.0, respectively, and 0.02, 1.3, 23.5, and 185.0. For the first assay, the median growth limit for the four phthalates—DBP, DPP, DEP, and DMP—was 0.0034, 0.9, 3.0, and 54.0, respectively. They also

experimented with the highest concentration of DEHP but their culture population did not reduce significantly [49].

Studies showed the factors that influence the adsorption of phthalic acid esters. It was seen that Di-n-butyl phthalate and Bis (2-ethylhexyl) phthalate dissolved in seawater, absorbed and adsorbed from three clay minerals, calcite, a sediment sample. The adsorption of phthalate was seen to be inversely proportional to aqueous solubility of phthalates. It was also seen that the lipophilic nature of the adsorbent/salinity was directly increasing phthalate bound. The probable binding mechanisms include Van der Waals and hydrophobic interactions [50].

A study describes the environmental fate of phthalates. The literature represents the environmental relationship of phthalates and abundant data on physicochemical properties of phthalates, partitioning behavior, abiotic and biotic transformation and bioaccumulation process of phthalates. Phthalates exhibit a four order of magnitude decrease in vapor pressure and an eight order of magnitude increase in the octanol-water coefficient (K_{ow}). Water solubility measurements of phthalates are also included in the literature. It also discusses photodegradation caused by free radical attack from phthalates, which is anticipated to be the main degradation pathway in the atmosphere with a half-life of about one day. Phthalates are biodegraded by a variety of bacteria and actinomycetes under both anaerobic and aerobic environments. The primary degradation in half-life in soils ranges from more than one week to several months, whereas in surface and marine water it is more than one day to two weeks. In cold or anaerobic oligotrophic environments, phthalates have prolonged half-lives [40].

A comparative study between the abundance of zooplankton in presence of neustonic plastic in southern California's coastal water. The sampling was carried out after an extended dry period and second after a storm when runoff was extensive. The neustonic sample was collected from 5 stations along a transect parallel to shore using a manta trawl of mesh size 333μ . The study concluded that the presence of plastic mass per cubic

meter was more in after storm samples whereas before storm shows 8 pieces per cubic meter [51].

A given literature about bioaccumulation of phthalate esters in aquatic food webs. High molecular weight phthalate esters (DEHP, DnOP and DnNP) show evidence of trophic dilution in aquatic food webs. A significant relationship has been observed between bioaccumulation patterns of DBP, DiBP and BBP and trophic position consistent with lipid-water partitioning model. Studies of laboratory and field have shown that low molecular weight phthalate esters (DMP and DEP) have bioaccumulation factors that are greater than predicted lipid-water partitioning models. Species-specific differences in metabolic transformation can show effect on observed bioaccumulation [41].

A study was done highlighting the effects of pollution on zooplankton in relation to their abundance and distribution. Two estuaries of Basque coast were selected as the study area. Zooplankton abundance and distribution was studied in relation to pollution. The hydrological parameters measured were salinity, temperature, dissolved oxygen saturation, chlorophyll-a and particulate matter. Mesozooplankton groups were more abundant in Bilbao at higher salinities and in Urdabai more abundance was seen at lower salinities. The study reflects that in Bilbao the zooplankton abundance was much more due to pollution [14].

Pollutants have sorption behavior so that they can accumulate, and a similar study done on sorptive behavior of benzyl butyl phthalate (plasticizer) that is known to have endocrine disrupting ability. The study states that the sorption of Benzyl butyl phthalate on sediment was a rapid process that can reach equilibrium in 6h. The relationship between BBP sorption, temperature and salinity is seen, BBP on sediments increased with increasing salinity and decreased temperature. The study suggests that such findings can be used for the assessment of pollutants and their fate [52].

A study showed the degradation of four phthalates in the marine environment. The breakdown of two oxo-biodegradable plastics, compostable plastic and

standard polyethylene in the environment. It vanished from their test rig for between 16 and 24 weeks whereas approximately 98% of the other remained after 40 weeks. Their data concluded that plastics can degrade quickly compared to oxo-biodegradable conventional plastics [53].

A study recorded the abundance of neustonic microplastic and zooplankton. The study area was the Northwestern Mediterranean Sea: Microplastics were present in 90% out of 40 stations. Microplastic was present in the form of filaments, polystyrene, and thin plastic films. 30% of the samples contained more than 0.1 particles/m². Total 4,371 microplastic particles were observed with a total dry weight of 7.9g. Average weight of zooplankton was found to be 0.480mg/m², and it was found to be similar in both western and eastern parts. Between microplastics and mesozooplankton the average ratio was 0.5 [54].

The effect of sewage pollution on the mesozooplankton community in a shallow estuary was studied. The effluent discharge zone resulted in maximum values of particulate organic matter, nutrients mainly phosphate. Their statistical calculation for taxa richness, Shannon diversity values resulted in showing lowest values in the effluent discharge area when compared to other regions of estuary. *Eurytemora Americana* and *Acartia tonsa* and larvae of *Balanus glandula*, *neohelic granulata* were present in lower densities in the discharge area. Thus, the study confirms the effect of sewage on the mesozooplankton community [55].

The toxic effect of plasticizers on species of calanoid copepod was analysed. The study assessed the effect of pollutants (Polyethylene terephthalate and Di-2-ethylhexyl phthalate). On mortality, productivity, population size and gene expression. The organisms were exposed to pollutants for studying the toxicity test results showing nauplii being sensitive to even small concentrations up to 5120 µg L⁻¹. Adults, when exposed to sub-lethal concentration of pollutants showed reduction in egg production. 24 days exposure to microplastics when given to the copepod population the population was seen to be decreasing, whereas 6 days

exposure showed less depletion in population. No recovery in the population of copepods was observed, when exposed to DEHP and it yielded the same average population at the termination of the experiment. Thus, the study suggests that DEHP can produce reproductive disorders and can be inherited by generations [56].

A study showed the presence of organophosphorus and phthalates in coastal beaches of north China. The sample was collected from 28 coastal beaches of the Bohai and Yellow Sea in China. Investigation of microplastics showed the presence of polyethylene flakes and fragments and polystyrene foams. The study resulted in Tris (2-chloroethyl) phosphate (TCEP), Tris (1-Chloro-2-propyl) phosphate and DEHP where the most abundant organophosphorus was also found in higher concentration. Polypropylene flakes and Polystyrene foams showed the presence of much more additives than polyethylene pellets which contained the lowest. The presence of such compounds suggests that they can be a hazard to organisms. The additives from microplastics were found to be from different origins which suggested spatial and compositional variation and thus this information can be useful for tracing routes of microplastics in the environment [29].

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