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Review

A critical review on the organo-metal(loid)s pollution in the environment: Distribution, remediation and risk assessment

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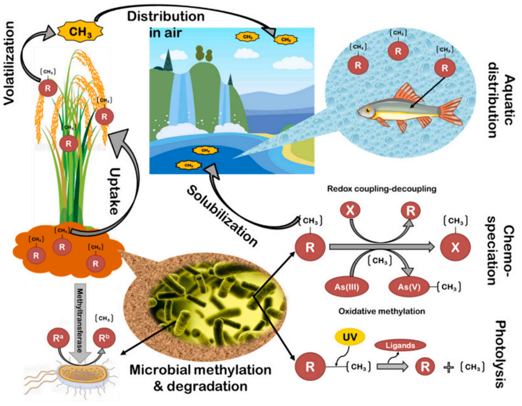
HIGHLIGHTS

- Bibliographic analysis has showed limited research focus on organometal(loid)s.
- Distribution of organo forms in soil-plant-aqueous environments is mostly unnoticed.
- Under changing natural conditions, inorganic forms get alkylated or dealkylated.
- Organometal(loid)s make greater health hazards than their inorganic forms.
- Both physico-chemical and biological remediations are possible with precautions.

GRAPHICAL ABSTRACT



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ABSTRACT

Toxic metal(loid)s, e.g., mercury, arsenic, lead, and cadmium are known for several environmental disturbances creating toxicity to humans if accumulated in high quantities. Although not discussed critically, the organo-forms

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of these inorganic metal(loid)s are considered a greater risk to humans than their elemental forms possibly due to physico-chemical modulation triggering redox alterations or by the involvement of biological metabolism. This extensive review describes the chemical and physical causes of organometals and organometal(loid)s distribution in the environment with ecotoxicity assessment and potential remediation strategies. Organo forms of various metal(loid)s, such as mercury (Hg), arsenic (As), lead (Pb), tin (Sn), antimony (Sb), selenium (Se), and cadmium (Cd) have been discussed in the context of their ecotoxicity. In addition, we elaborated on the transformation, speciation and transformation pathways of these toxic metal(loid)s in soil-water-plant-microbial systems. The present review has pointed out the status of toxic organometal(loid)s, which is required to make the scientific community aware of this pressing condition of organometal(loid)s distribution in the environment. The gradual disposal and piling of organometal(loid)s in the environment demand a thorough revision of the past-present status with possible remediation strategies prescribed as reflected in this review.

1. Introduction to organometal(loid)s

Heavy metal(loid)s, defined as metal(loid)s with high atomic weight and a density of 4 g/cm^3 or eight times more than that of water (Bánfalvi, 2011; Upadhyay and Majumdar, 2022), are usually present in trace quantities but can have extreme ecotoxicity (Egorova and Ananikov, 2017; Chowdhury et al., 2022). The formation of metal-organo compounds is primarily based on the alkyl bond rather than the metal-carbon π bond, which is limited in natural production. Alkylation of these heavy metals results in the formation of organometal(loid)s, which are more toxic and pose a severe threat to biotic components of the environment (Craig et al., 2000; Majumdar et al., 2022a). Under different environmental conditions, the chemical transformation of organometal(loid)s takes place via the formation or breakdown of metal-carbon bonds and transfer of organo groups to the metal(loid)s with frequent redox coupling and de-coupling reactions (Bolan et al., 2013; Frohne et al., 2011). In natural systems like soil, water or aerosols, this mobility depends on the nature of the ligand (organic or inorganic) (Shahid et al., 2017; Kumarathilaka et al., 2018) and any change in these attached ligands can cause alteration of adsorption or mobility of the compounds. Mobility and further adsorption of these organometal(loid)s to specific binding sites are also dependent on the number of organo groups attached to the metallic part. This elemental mineralization, transformation and volatilization are controlled by the physico-chemical parameters of the associated environment and biological, specifically microbial participation (Afsal et al., 2020; Majumdar et al., 2023a, 2023b, 2023c, 2024a).

Organometal(loid) exhibit some important characteristics that influence chemical speciation and availability to living organisms, resulting in toxicity. Among these properties, volatility remains a crucial factor. For instance, among other organic groups, methyl does not form any stable intermolecular bonding. Nitrogen, oxygen or halogen atoms attached to a metal moiety, replaced by an organic group, minimize the inter-molecular association and thus increase the volatility (De Jonge et al., 2012; Harlov and Aranovich, 2018). If all the available bonds of metal(loid) are attached to the methyl group, the chances of becoming volatile get triggered. These compounds are termed as 'permethyl', such as $(CH_3)_2Hg$, $(CH_3)_4Sn$, $(CH_3)_4Pb$ and $(CH_3)_3As$, and mainly these are gases or readily get volatilized from the liquid state at room temperature (Onuchak et al., 2019). Another fundamental property affecting the associated ligands is the water solubility of an organometal(loid) compound. The longer the alkyl chains are attached, the lower the water solubility of these aryl derivatives has been observed. Although methyl and ethyl groups sometimes show considerable solubility, permethyl compounds are almost insoluble in water. If this number of the alkyl group is low and the corresponding metal(loid) counterpart is one step lower in the periodic table, then the chances of solubility get increased to some extent; for example, among AHg^+, A_2Tl^+, A_3Pb^+ and A_4Sb^+, AHg^+ will be the best solute compared to the rest (Thayer, 2012).

In natural environments, organometal(loid)s show mobility from one point to another depending on these physico-chemical (abiotic) or microbial (biotic) activities. Being organo-groups associated, the metals and metalloids tend to dissolve more prominently in lipid (hydrocarbon) solutions, another important property, as this lipid solubility helps in organometal(loid) transportation carried out by several hydrocarbons in the natural water systems. Absorptivity of any metallic complex depends on the associated ligands, and external incorporation of such organo groups to the metal ions enables their adsorption onto free surfaces like suspended aerosols, plant leaf abaxial surfaces, and sediments. Such attachments reduce accessibility for compound dissolution and thereby alter availability. It is often observed that the organo forms of any element can increase the bioavailability. The redox transformation of any organic molecule can interact with the inorganic minerals to develop the organometal(loid)s complexes. The alkylation process of any inorganic metal(loid)s is significantly influenced by the intrinsic involvement of biological entities, whose metabolic activities affect the surrounding chemical speciation of elements of interest.

2. Bibliometrics analysis

2.1. Analysis protocol

Bibliometric analysis allows researchers to identify the most prolific authors, institutions and countries contributing to the body of knowledge on organo-metals and organo-metalloids pollution. This approach enables the exploration of the intellectual structure of a specific domain within the existing literature (Donthu et al., 2021a, 2021b, 2021c; Moulick et al., 2024b). We conducted a bibliometric study aiming to provide both qualitative and quantitative interpretations. This involved summarizing extensive bibliometric data extracted from the SCOPUS database, following the PRISMA protocol. The search string used to retrieve the data from the SCOPUS database was "TITLE-ABS-KEY (organo-metals OR organo-metalloids OR methylmercuric OR methylarsine) AND (pollution)) AND (LIMIT-TO (DOCTYPE, "ar")) AND (LIMIT-TO (LANGUAGE, "English")). Out of the initial 133 documents retrieved, 121 were included in the analysis following appropriate exclusion and inclusion criteria, as depicted in Supplementary Fig. 1. The analyzed bibliometric data based on different criteria, dataset was processed utilizing a variety of network-based visualisation software tools, including VOSviewer (Van Eck and Waltman, 2014), ScientoPy (Ruiz-Rosero et al., 2019), and the R Biblioshiny Package (Aria and Cuccurullo, 2017).

2.2. Global production trend

A total of 121 articles on organo-metals and organo-metalloids pollution in the environment were selected from the year 1971 to 2023. The first article was published in 1971 on 'Residues of total mercury and methyl mercuric salts in lake trout as a function of age'. Only one document was published in the first two years. Then, a gradual increment was observed till 1975, and a declining trend again was noticed (Fig. 1a). In 2015, maximum (12 publications) documents were published on this topic. Similarly, the mean total citations in the first year were 2.44 and the highest mean citation (7.19) was in 2018. Citation analysis is a fundamental tool in science mapping, based on the premise that citations indicate intellectual associations between publications, demonstrating when one publication refers to another (Appio et al., 2014).

Among the total 57 internet sources, the top 10 journals published almost 47 % of the total publications (57 documents) (Fig. 1b). The top five journals, including Environmental Toxicology and Chemistry, published a maximum number of publications (13 publications, 10 %), followed by Environmental Research (7 publications, 5.7 %), Archives of Environmental Contamination and Toxicology (6 publications, 4.94 %), Bulletin of Environmental Contamination and Toxicology (6 publications, 4.95 %), and Chemosphere (5 publications, 4.13 %). The contribution of the top ten countries towards research in organo-metals and organo-metalloids is depicted in Fig. 1c. The was most publications recorded from the USA, followed by China and India, respectively. The last ten years (2010 onwards) proved incredibly very important for China, Sweden, Italy, and India as 91 %, 84 %, 74 % and 70 % of their total documents, respectively, were published globally. A well-established collaboration map among the countries in the field of organo-metals and organo-metalloids pollution in the environment was demonstrated in Fig. 1d, where the USA contributed the most among the 52 countries.

The author's production over time is presented in Supplementary Fig. 2a, depicting the top ten authors and their production dynamics in organo-metals and organo-metalloids research. Article number and Total citation per year are represented by nodal size and darkness, respectively. The more the article number, the larger the nodal size and the darker the node, the larger the more significant the nodal size and the darker the node; the more significant the nodal size and the darker the node, the more the citation. Supplementary Fig. 2b presents a three-field plot diagram depicting the authors (AU) and respective countries (AU_CO) and used keyword (DE) in their research documents. The USA produced the most substantial highest number of researchers in the field of organo-metals and metalloids research.

2.3. *Association analysis based on the authors' used keywords*

A total of 814 keywords from the 121 papers selected for association analysis. A total of 8 clusters are formed in between 814 keywords and among which only 463 keywords made the largest cluster shown in Supplementary Fig. 3a. The keyword 'Perovskite solar cells' occurred 16 times with 59 total link strength followed by 'Organo-metal complexes' occurred 10 times with total link strength 50, 'Stability' occurred 7 times with total link strength 42, 'Heavy metal" occurred 6 times with total link 35 and 'Perovskites' occurred 6 times with total link strength 29. Supplementary Fig 3b, an overlay map is used, where several color nodes indicate the occurrences over time. In contrast, the light green colour indicates the distribution of keywords in recent years deep green colour reflects the occurrence of keywords earlier. In contrast, the light green colour indicates the distribution of keywords in recent years. The keywords 'perovskite solar cells', 'perovskite' and 'thin films' occurred during the year 2020; on the other hand, deep green colour keywords 'organo-metal complexes' happened during the year of 2010–2015. While blue colour keywords like 'soil' and 'soil organic matter' illustrate the occurrence during 2005. The top five keywords in organo-metals and organo-metalloids research based of based on occurrence in various documents are depicted in the Evolution of the author's keyword graph (Supplementary Fig. 3c). The most common keyword, "Methylmercury", was used before 1992, and it reached its peak in 2023.

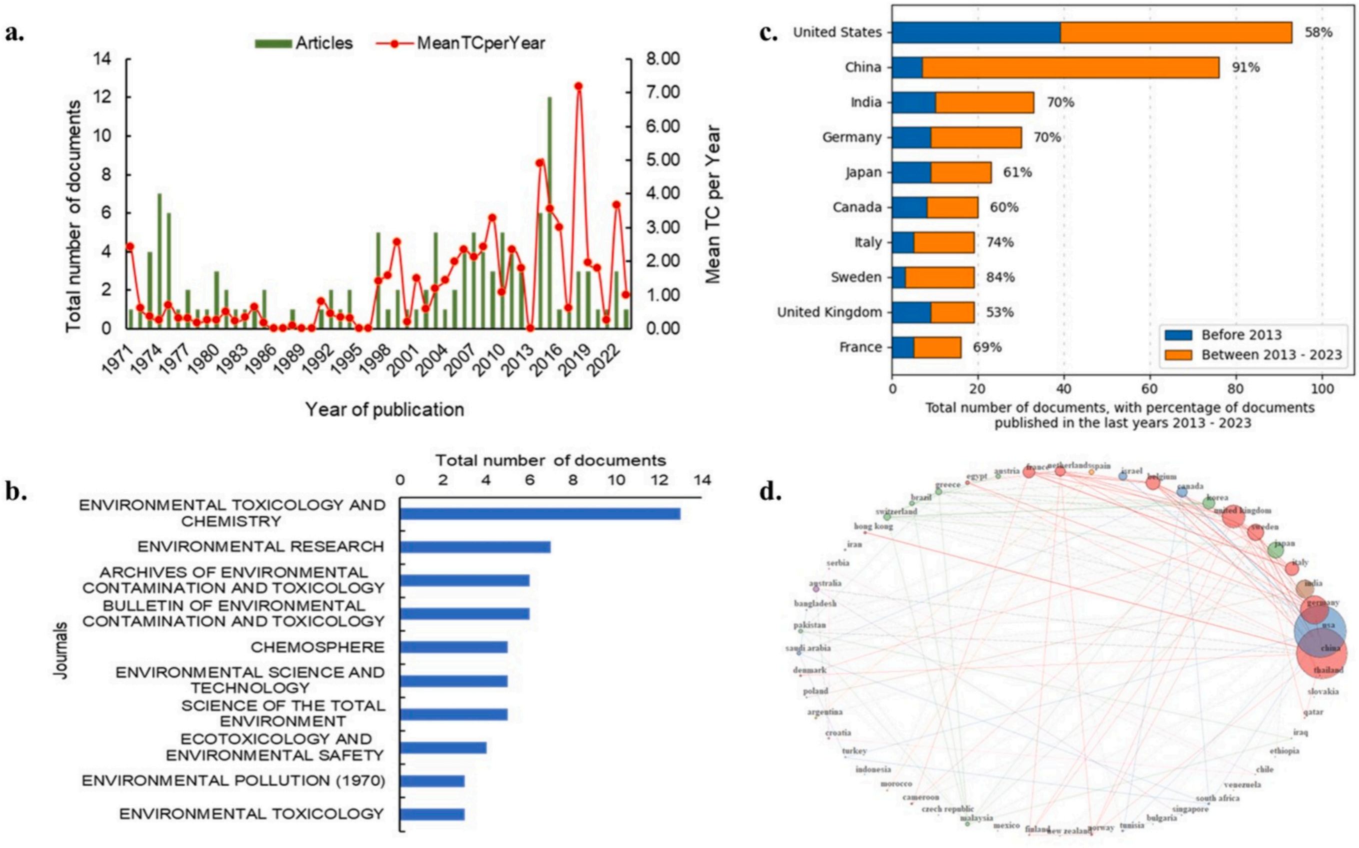

Fig. 1. (a) The global publication dynamics of the last fifty years (1971–2022), (b) The top Journals that published research related to Organo-metals and Organo-metalloids pollution in the environment. (Abbreviation: Mean TC per Year: Average Total Citation per year), (c) Total number of documents produced by top ten countries with percentage of documents published in the last decade, and (d) Country's collaboration in research on Organo-metals and Organo-metalloids pollution in the environment.

2.4. Thematic analysis of research trends

The assessment of various themes in organo-metals and organo-metalloids research is shown with the help of a thematic map (Supplementary Fig. 4). The thematic map is used to increase the precision and clarity of understanding various conventional and trending themes of the research areas. The thematic map is divided into four parts (quadrants) based on the centrality and density of the research themes. The tetra quadrants are as follows: a) Motor themes (positioned in the topmost right part); b) Niche themes (situated in the upper left part); c) Emerging/declining themes (located in the lower left part), and d) Basic themes (found in the lower right part). The quadrant of motor themes in research focusing on organo-metals contains two clusters. “Methylmercury”, “oxidative stress” and “eggs” form the primary cluster-driving themes. “Soils,” “acid mine drainage” and “jarosite” formed the niche theme cluster. The fundamental theme of the research includes “mercury”, immunotoxicity” and “metal toxicity”. “Organic matter” and “methyl mercury” are emerging and declining research themes in the field of organo-metals and metalloids.

In bibliometric studies, conceptual cluster analysis depends on the simultaneous occurrence of keywords. At the same time, several statistical methods are being used to break the complex keyword networks into more coherent and smaller groups (Ding, 2011). The multiple correspondence analysis (MCA) and hierarchical clustering analysis by using authors’ keywords have been utilized in this study. Initially, we find the keywords with the highest degree of similarity to compile into one larger cluster. Then a new cluster is merged with another large cluster in the conceptual map. This merging of keywords into a single cluster is repeated until all the individual keywords are clustered together. The author’s keywords are merged into three clusters positioned in the graph’s middle point resulted in greater attention in recent decades. Each cluster demonstrates a similar group of keywords in the specified domain of research (Supplementary Fig. 5). The keywords positioned in the graph’s middle point resulted in greater attention in recent decades. In contrast, the keywords located nearer graph’s edge gained the least attention. In this graph, Cluster I is situated in the lower right corner of the figure, and the keywords include ‘heavy metal,’ ‘lead,’ ‘zinc,’ and ‘copper,’ with 70.64 % variation. Therefore, the cluster’s location and variation percentage indicate an in-depth study is required in these specified keywords. Where cluster II (red cluster), located closer to the middle of the graph involves the keywords ‘metals,’ ‘organo-metals complex’ ‘organometallic compound,’ ‘chlorine compounds,’ ‘soil pollution,’ ‘soil organic matter’ and ‘carbon sequestration’. The result indicates a significant shift from nanostructured materials to organo-metals and organo-metalloids pollution in the environment causing soil pollution and the utmost requirement of soil carbon sequestration to mitigate the pollution and native effect on health. Cluster III, which is blue-colored and located at the edge of the graph involves the research of ‘electron transport properties,’ ‘nanostructured materials’, and ‘layered semiconductors and these clusters are mainly associated with the engineering and industrial properties of organo-metallic substances.

3. Sources of organometal(loid)s in the environment

3.1. Organometal(loid)s in the soil-sediment system

The transformation of several metallic and metalloid compounds to organo-compounds within a confined soil-sediment system is a crucial phase that we need to know more from environmental pollution, soil biota, crop quality and human exposure perspectives (Mondal et al., 2024). In the case of sedimentary depositions, factors like salinity also affect organo-transformation with both seasonal and inter-seasonal fluctuations (Chen et al., 2019; Panichev, 1997). Sedimentary depositions also showed that in the presence of a higher amount of chloride ions, the methylation increases while the particle size of sediments remained the same, helping in the adsorption of excessive organo-metals content (Frohne et al., 2012). Both biotic and abiotic factors in the soil can influence this methylation process. Transformation to the organo form and further volatilization were found to be increased under higher soil temperature whereas the volatilization was assessed to be inversely correlated to the soil water content or minerals like nitrate ion or glucose molecules (Feng et al., 2014; Zhang et al., 2018). The higher the alkalinity in soil, the greater organo-forms volatilize. A study showed Hg can be methylated in soil due to various soil categories and physico-chemical properties like pH, temperature, total organic matter content etc. along with types of inorganic mercuric compounds (Yin et al., 2018; Moulick et al., 2024b). Methylated Hg are also traced in the polar sediments within a range of 0.4–1.1 μg kg^{-1} and the in-situ alkylation accounts >80 % of the Hg distribution in the Arctic region (Tang et al., 2020). Due to the changing temperatures in the arctic region, chances of methylation also get increased due to the triggered activities of marine phytoplankton that release methyl groups to the soluble Hg that further settled down in the sediment (Oiffer and Siciliano, 2009). Reports also had revealed the influence of microbial bio-conversion of organo-mercurals in the soil at a 30 % rate in a 4-day period to elemental mercury. In activated sludge-treated soil, the methylation process was triggered as found similarly in agricultural soils, where labelled mercury was transformed into methylmercuric ions (Liu et al., 2021). For agricultural fields like paddy cultivation sites, the change of methylation is very high due to complex anaerobic microbial metabolic activities and methylation processes. Agricultural sites near industrial regions are prone to have Hg contaminations that results in the development of the methylated Hg production (Tang et al., 2019).

Report suggests that the organo forms of the As are more efficient towards temperature and osmotic stresses in soil-sediment phase, however, the rate of biomethylation is comparatively lower than the degradation of these methylated As species, as in primitive era, the organoarsenicals were much higher in concentration than this modern time (Chen and Rosen, 2020). Soil systems can also influence the speciation of arsenic to organo forms and back to inorganic complexes. In earlier days, the application of herbicides, having inorganic arsenicals, got transformed into arsine, methylarsine and dimethylarsine, under differential soil status, the physicochemical parameters including soil pH, moisture content, redox potentials, clay fractions, organic matter, nutrients and elemental hydroxide complexes are essential in the biomethylation process of As (Hemmat-Jou et al., 2024; Meharg and Meharg, 2021). The oxidation and reduction processes by microbial communities, largely depends upon the availability of these As fractions associated with the Fe and Mn oxides and layers in the soil-sediment phase (Kumarathilaka et al., 2018). In soil, pH and microbial arsM gene are positively correlated that also influence As methylation (Di et al., 2019). In natural soil, C^{14}-lebeled methylarsonic acid was found to be converted by 10 % as $^{14}CO_2$ within a span of three weeks, whereas that sterilized soil having all the parameters and conditions similar, produced only 0.7 % of $^{14}CO_2$, indicating the importance of soil biota in the elemental geocycling (Giovannoni et al., 2019; Majumdar et al., 2022a, 2022b, 2022c). Another report showed that the labelled cacodylic acid transformation under aerobic but moist soil accounted for around 80 % of the $^{14}CO_2$ within two months of the time, whereas the conversion under anaerobic waterlogged soil was different, producing much lesser carbon dioxide and higher methylarsonate (Thayer, 2012). Methylarsonate associated with disodium ions is adsorbed to the clay surfaces more readily than loamy soil with a greater tendency of methylation and demethylation compared to cacodylic acid.

Researchers have shown that the properties of sediments in natural water bodies, along with their microbiota, play a crucial role in influencing the formation of inorganic stannous compounds and their subsequent conversion into variable organotins. Both Sn(II) and Sn(IV) were considered in an experiment to demonstrate the methylation process using lake sediment. The result identified different methylated products with varying degrees of formation rate. Organotins are adsorbed more readily and firmly onto clay soil compared to other soil types (Fang et al., 2017). In the case of Pb species, the chemical associations and reactions with different elemental complexes following ionic exchange, adsorption, and precipitation vary greatly depending on the species of the Pb in the soil-sediment system, resulting in a variable bioavailability and chances of dealkylation process thereafter (Li et al., 2020a, 2020b).

3.2. *Organometal(loid)s stability in the aqueous system*

During hydrolysis of an organometallic compound, a nucleophilic attack occurs on the water oxygen atom's lone electron pair and is transferred to the empty metal orbital of the organometal. This hydro-instability is thus connected to the empty low-energy state orbitals of the metallic atoms and their capacity for increasing coordination number (Craig and Jenkins, 2004). It has been observed that the strong metal-carbon bonds ($M^{\delta-}C^{\delta-}$) are not stable enough as the hydrolysis rate is linked to the degree of polarity of metal-carbon bonds (Elschenbroich, 2016). Group I and group II organometals, along with Zn and Cd, show such examples of instability. With low polarity, alkylboron compounds were shown to have greater stability in the aqueous phase due to the restrictions on expanding the coordination number. With the increasing number of alkylation, the tendency of solubility also gets reduced like R_2Hg, R_4Sn and R_4Pb are generally hydrophobic with a greater tendency of being volatile if the R gets increased. On the other hand, CH_3HgX is comparatively more stable than R_2Hg and its solubility in the water depends on the associated X group. A study showed that $(CH_3)_nSn^{(4-n)+}$ is decently soluble in water where the methyltin part is stable and generates penta/hexavalent coordination with water/hydroxyl ions. In the case of $(CH_3)_2Pb^{2+}$, similar coordination with water molecules has been found that gradually results in the cleavage generating $(CH_3)_3Pb$ + and CH^{3+} with a concomitant loss of these methyllead ions from the water (Koo et al., 2016). Organoarsenicals like $(CH_3)_2As^+$ and $(CH_3)As^{2+}$ react differently in the aqueous phase, as the former one gets hydrolyzed to $(CH_3)_2AsOH$ and then to $[(CH_3)_2As]_2O$, whereas the second compound gets converted to $CH_3As(OH)_2$ and then to a more soluble $(CH_3AsO)_n$ form (Abbas et al., 2018; Hussain et al., 2021a, 2021b). Methylarsinic acid $[CH_3AsO(OH)_2]$ and dimethylarsonic acid $[(CH_3)_2AsO(OH)]$ are stable in oceanic aqueous phase with acidic pKa values between 3.6 and 6.2. However, methylarsine compounds are insoluble in water and readily defuse in the air (Mestrot et al., 2013). A generalized schematic of organo-metal(loid)s distribution in both the soil-aquatic media has been shown in Fig. 2, taking MeHg as a model element which represent other organo-elements as well.

This schematic representation of organomercurial compounds highlights the complex distribution pattern of organometal(loid)s in the environment. The transformation from inorganic species to organo-compounds is a multifaceted process that involves the environment at every step. These compounds further accumulate in plant tissues and are consumed by herbivores or aquatic animals, which act as the primary accumulator of organometal(loid)s. From this trophic level, human accumulation of organometal(loid)s occurs as a secondary accumulator, further complicating the distribution and transformation of these compounds. The role of the environment in this process is crucial and underscores the depth of the research.

4. Speciation mechanisms of organometal(loid)s

The formation of metal-organo compounds is mainly based on the alkyl bond rather metal-carbon π bond which is limited in natural production. Under different environmental conditions, the chemical transformation of organometal(loid)s takes place via the formation or breakdown of metal-carbon bonds, and transfer of organo groups to the metal(loid)s with frequent redox coupling and de-coupling reactions (Bolan et al., 2013; Frohne et al., 2011). In the natural environment, several pathways have been observed for the generation of organometal(loid)s with subsequent interactions that trigger the greater bioavailability or make them more immobile, confined to an enclosed system. Chemical and photo-catalytic speciation are primarily responsible for such alterations, although, biologically originated organometal(loid)s were also reported. Different forms of commonly found organometal(loid)s are listed in Table 1 with their common sources mentioned and the forms that are either stable or unstable at that environmental matrix.

4.1. *Chemical speciation*

A metal-carbon bond might get broken or reformed under environmental conditions without any biological intervention, although the appropriate occurrence of organometal generation and activity needs a favourable biological mediator. But the simple chemical change in the redox coupling-decoupling process can influence metal-carbon bonds like-

$$(CH_3)_aM^{(x+)} + (CH_3)_bN^{(y+)} \rightarrow (CH_3)_{a-1}M^{(x+1)+} + (CH_3)_{b+1}N^{(y-1)+}$$

where a and b are positive numbers, and x and y are redox states of

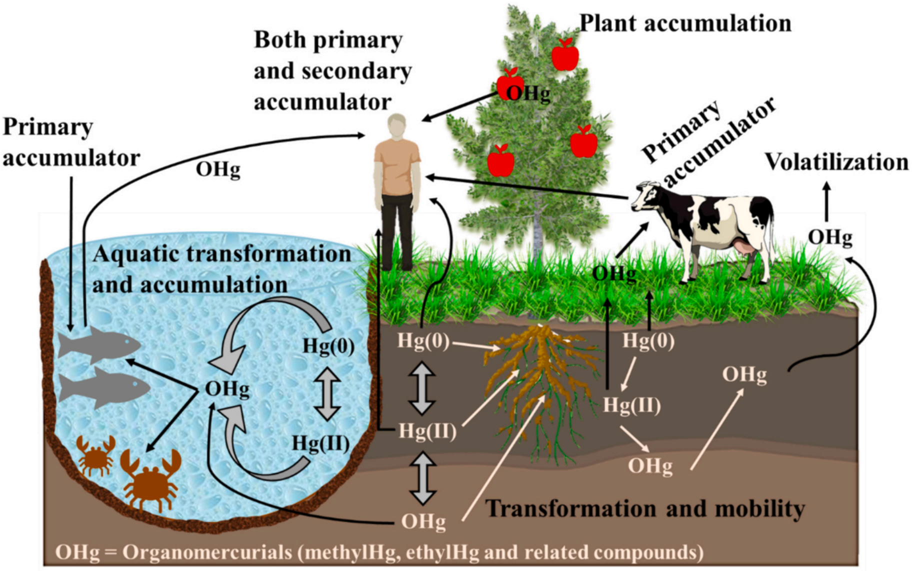

Fig. 2. Environmental distribution, transformation and speciation of organomercurial compounds as a representative of all organometal(loid)s.

Table 1
Some of the common organometal(loid)s with their stable and unstable forms in the environment.

Stable organometal (loid)s	Unstable organometal (loid)s	Commonly found and references
$(CH_3)_2Hg$, CH_3HgX (C_6H_5 and C_2H_5 also stable)	$C_6H_5HgSi(C_6H_5)_3$	Soil (Liu et al., 2019), freshwater (Zhang et al., 2019a, 2019b), seawater (Wang et al., 2018), stream sediment (Lino et al., 2019), estuaries (Chen et al., 2021), deep sea (Sun et al., 2020)
$(CH_3)_4Si$, $[(CH_3)_2SiO]_n$, $(CH_3)_nSi^{(4\text{-}n)+}$, $(CH_3)_6Si_2$	$(CH_3)4SiH_{4\text{-}n}$	Soil (Zhang et al., 2018), freshwater (Pelletier et al., 2022), sediment (Shen et al., 2018)
$(CH_3)_4Sn$, $(CH_3)_{4\text{-}n}SnX_n$	$(CH_3)_6Sn_2$ (At RT gives $[(CH_3)_3Sn]_2O$)	Lake water, river water, and seawater (Jiemin et al., 2003); soil (Noubissié et al., 2017); sediment (Chen et al., 2019); deep sea (Rüdel, 2003)
$(CH_3)_4Pb$, $(CH_3)_3PbX$, $(CH_3)_2PbX_2$	CH_3PbX_3, $(CH_3)_6$, Pb_2 (to methyl lead products)	Soil (Filella and Bonet, 2017); sediment (Escobar et al., 2013); fresh water (Mikac et al., 2001); seawater (Rao et al., 2005)
$\pi(CH_3C_5H_4Mn\ (CO)_3$, $CH_3Mn(CO)_4L$	$(\eta\text{-}\ C_5H_4Me)Mn\ (O_2CMe)$	Soil (Campillo et al., 2007); sediment (Feldmann, 2003); river water (Peñalver et al., 2011); seawater (Campillo et al., 2014)
$(CH_3)_2AsO(OH)$, $CH_3As(O)(OH)_2$	$(CH_3)_5As$, $(CH_3)_3As\mathit{e}$, $(CH_3)_2AsH$, CH_3AsX_2, $(CH_3)_3AsO$	Soil (Majumdar, 2021); sediments (Yang and He, 2015); seawater (Shimizu, 2010); river water (Rahman and Hasegawa, 2012)
$(CH_3)_2S$, $(CH_3)_3S^+$	Tetraphenyl sulfurane	Soil (Carrión et al., 2017); sediment (Deschaseaux et al., 2019); river water (Tan et al., 2017); seawater (Yang et al., 2012)
$(CH_3)_2Se$, $(CH_3)_3Se^+$	C_6H_5SeH	Soil (Favre-Bonté et al., 2006); sediment (Meseck and Cutter, 2012); river water (Kang et al., 2004); seawater (Simó et al., 2000); estuarine water (Tessier et al., 2002)
CH_3COB_{12} (solid state)	C6H5SeH	Soil (Hussain et al., 2021); sediment (Chemaly, 2002); seawater (Zhu et al., 2011); river water (Demuth and Heumann, 2001)
$(CH_3)_3SbO$, $(CH_3)_2SbO(OH)$, $CH_3SbO(OH)_2$	$(CH_3)_5Sb$, $(CH_3)_3Sb\mathit{e}$	Soil and sediment (Yang and He, 2015); river water (Craig et al., 2001); seawater (De Gregori et al., 2005)
$(CH_3)_2Ga^+$	$(CH_3)_3Ga$	Fresh water (Feldmann, 2003); seawater (Zheng et al., 2020)

positive or zero.

Mercury, tin, thallium, and lead have been reported to produce methylated derivatives following such reaction mechanisms; for example, methylcobalamin can be formed in this way from hydroxocobalamin (Ramezanpour Ahangar and Annamaraju, 2024). Another way of organo-transformation of metallic ions is to get oxidative methylation. Compounds of selenium (Deng et al., 2018; Duncan et al., 2017) and tellurium, $RX(CH_3)^{2+}$ where X = Se/Te, react with triphenylphosphine or triphenylarsine to generate methyltriphenylphosphonium or methyltriphenylarsonium salts. In the case of arsenic trioxide, it can be converted to the methylarsonate by transferring the methyl group from the trimethylsulfonium hexafluorophosphate. In natural water, a common organic compound, iodomethane, donates the methyl group most frequently to the other elemental minerals nearby. This compound was also reported to interact with either particulate or aqueous lead (Yang et al., 2020) along with tin compounds to generate methyllead or methyltin. Such diverse chemical interactions triggered the formation of organometals in the static environment. The redistribution tendency of heavy metals, specifically lead, has been observed. It was observed that the sulfide ion can redistribute some methylated metal compounds as per the given reaction mechanism-

$$\left[(CH_3)_{a-1}M\right]_2S \rightarrow (CH_3)_aM + 1\Big/x\left[(CH_3)_{a-2}MS\right]_x$$

where a = 2 for Hg and a = 4 for Sn or Pb. Due to this redistribution process, a release of volatile permethyl metal compounds occurs. Some examples are the formation of dimethylmercury from methylmercuric sulfide, tetramethyllead formation from bis(trimethyllead) etc. (Thayer, 2012).

4.2. Photolytic speciation

Some studies have shown that the development of metal-carbon bonds or breakdowns in the natural environment can be a result of photochemical reactions. The photolytic process of organo-metal and metalloids depends on the adsorption of electron-creating organic radicals. Such adsorption generates transitions in d-d electron pairings, in case of any transition element or charge transfer to/from metal orbitals (Craig and Jenkins, 2004; Horvat et al., 2003). Any charge transferred to the metal ion dissociates metal-organo ligand bonds, which in turn results in coordinative instability. Any charge transfer from the metallic ions causes a nucleophilic attack on the metal part. The development of methylmercuric ions was reported due to the photolysis of mercuric sulfide reacting with acetate ions (Luo et al., 2020) and similar products also generated during photolytic reactions on mercuric chloride in the presence of any α-amino acids. In natural temperatures, the generation of ethane is more predominant than methane, and with increasing temperature, methane production also increases, hence the methylmercuric ions predominate. In surface seawaters, irradiation of tetraethyllead resulted in the breakage of Pb—C bonds, triggering the formation of Pb(II) salts, hydrocarbon compounds and various alkylleads like *n*-butyltriethyllead (Chau and Wong, 2017). In the case of organomercurials and organotins, a similar cleavage process was reported in natural water bodies, the UV irradiation can break down methylmercuric chloride into several by-products and elemental Hg (DiMento and Mason, 2017). Photolysis of Sn—C bonds in soil systems has been observed where triphenyltin generates diphenyl and monophenyltins.

5. Accumulation of organometal(loid)s to the biological systems

Organometal and metalloids are distributed in all the natural forms possible in a soil-aqueous-plant triphasic system encompassing consumers with no chance of escape from getting affected by these compounds. Soil contamination with organometals led to the subsequent accumulation of these compounds within the edible plant tissues that in turn get entered the food chain, in all the trophic levels, from single to multicellular organisms. Bioaccumulation of these compounds varies with spatio-temporal influences of different ecosystems. Reports suggest that in flowing water with a low amount of methylmercury present in it, the aquatic plant *Elodea densa* can take up significantly (Gentès et al., 2021). In contrast, marine alga *Chaetoceros* sp. and *Phaeodactylum* sp. were reported to accumulate these methylmercuric compounds with increasing temperature (Deng et al., 2013; Yu et al., 2020). In confined and natural field soil systems, methylated arsenic compounds are reported to accumulate in different cereals, specifically rice (Majumdar and Bose, 2017; Hazra et al., 2023; Fasano et al., 2021). The translocation of organoarsenicals from soil to plant follows some transporter protein molecules that allow the passage of these compounds along with some other nutrients and essential element passage (Majumdar and

Bose, 2018; De et al., 2024). In a natural aquatic content range of MeAs, some common crayfish tended to accumulate this compound more than other available crustaceans nearby (Ruttens et al., 2012; Karaś et al., 2021). Supplementary Table 1 provides a detailed list of concentrations reported in different food items for different organometals and metalloids.

From such contaminated food consumption, like fish, studies showed direct accumulation of MeHg in humans irrespective of age or gender difference (Taylor et al., 2017; Upadhyay et al., 2019a; Majumdar, 2021). The Biomethylation process of arsenic in humans is reported well that controls the MeAs content in the body via the excretion process, although, some different organoarsenicals like arsenolipids or arsenobetaines are not toxic to humans and can be found in marine organisms in plenty (Luvonga et al., 2020). Some laboratory experiments found that European carps, goldfishes, and guppies can bioaccumulate and biomagnified MeHg several folds more than any natural water content (Alvarado et al., 2020; Kumari et al., 2017; Bossu et al., 2019; Meng et al., 2010). Rainbow trout can accumulate and excrete tetramethyllead more efficiently than other relative fishes. A group of researchers reported, by using a model system, that the primary producer (phytoplanktons) tends to generate lipid-soluble organoarsenicals, which gradually decreased with the higher trophic level of zooplanktons to large aquatic organisms (Caumette et al., 2014).

6. Distribution and toxicity of specific organometal(loid)s in the environment

6.1. Organomercurical compounds

6.1.1. Environmental distribution

Organomercuric compounds are the most predominant among the different organometals distributed in the environment. Some studies showed the modelling-based data of Hg reservoirs and distribution patterns, although, in the case of MeHg, such studies are not well established yet. The concentration of MeHg on the surface of freshwater bodies is greater than in the oceanic strata. In the sediment, the formation of MeHg is parameter-dependent, including the sulfate-reducing bacterial metabolism that stimulates the MeHg generation (Lu et al., 2016; Mehouel et al., 2019). However, at higher content of sulfate, the process of Hg methylation also gets inhibited due to complexation between Hg and Sulfide. Wetlands are reported to be a significant source of MeHg with intermediate anaerobic zones that triggers the methylmercurals due to biotic factors and finally releasing from the sediments (Lehnherr et al., 2012). Concentrations of MMHg in surface water of any temperate lake or river or stagnant water body range from 0.05 pM to 5 pM (clear water to eutrophic water) (Mason and Benoit, 2003). Reports suggest the more significant amount of dissolved organic carbon in surface waters encompasses the MeHg by complexation hence high DOC indicates higher MeHg content (Chandan et al., 2015; Li et al., 2008). Aquatic biota-microbiota also influence the DOC, pH, and other elemental content which in turn influences the distribution of MeHg. Compared to the aqueous content of the MeHg, its atmospheric dispersion is relatively low and accounts for a small percentage of the total Hg in the gas or particulate phase in the aerosol. It has been reported that the MMHg release from municipal sludge applied to the soil for different usage, can be a source of atmospheric MeHg (Tao et al., 2017; Li et al., 2014). In contrast, some other studies showed that gaseous discharge from sewage treatment plants can also increase the MeHg content in the air (Craig and Jenkins, 2004). But, overall, the atmospheric MeHg concentration over the terrestrial area lies within a range of 5–25 fmol m^{-3} and is mainly dependent on the dry and wet deposition of the atmospheric particulate matter (Mason and Benoit, 2003). So, the organomercural distribution in any matrix of the environment occurs due to either methylation or demethylation processes, which fall under two specific categories- one, where environmental factors control the matrix parameters itself to modulate the chemical forms of Hg triggering methylation/demethylation process and second, where these factors influence biota to modulate the rate of methylation/demethylation process. Temperature, pH of the concerned system, seasonal variations, and sulfate content in the aqueous phase controls both methylation/demethylation processes.

6.1.2. Formation and species development

In the terrestrial environment, monomethylmercury (MMHg) is dominant, whereas dimethylmercury (DMHg) is prevalent on ocean surfaces. Although Hg is exposed in the environment as divalent salts of Hg (II) or elemental Hg (0), the formation of DMHg is still unknown to scientists. The probable mechanisms for the formation of DMHg can involve the methylation of Hg (0) by methylating bacteria or any other specific methylating enzymes. Still, the dominance of MMHg in the aquatic environment over DMHg is pretended due to the photochemical degradation of DMHg, which is however debatable. However, in 2022, S. Jonnson et al. have experimentally proven that DMHg is readily converted to MMHg by undergoing photochemical degradation using sunlight in a range of natural waters, including the water source from the Arctic Ocean, Baltic Sea and DOC-rich stream (Fig. 3) (West et al., 2022).

6.1.3. Effects and toxicity

Toxicity of organo-mercury towards organisms at different trophic levels or in various matrices are directly linked via the ability to get bioaccumulated and the highest transfer of MeHg in its bioavailable form has been found from water to the phytoplanktons (Li et al., 2013; Meng et al., 2014). A gradual transfer and biomagnification of MeHg from phytoplanktons to zooplanktons to small fishes to large predatory fishes to tertiary consumers/humans is an obvious route of accumulated toxicity of organomercurals. The percentage of MeHg of the total Hg bioaccumulated in the tissues of a different group of organisms clarifies the scenario of organomercural distribution. In the report by Mason and Benoit (2003), they showed around 10 % of MeHg is bioaccumulated in phytoplanktons or macroalgae followed by other deposit feeders like polychaetes (18–25 %), bivalves (20 %), echinoderms (25–50 %), amphipods (30–40 %) with greater bioaccumulated MeHg. As primary consumers or herbivores, zooplanktons (20–35 %), cockles (30–80 %) and some stonefly-mayfly larvae can accumulate 5–40 % of the MeHg intracellularly. Among suspension feeders and their predators also retain a high amount of MeHg as in mussels (15 %), clams (5–25 %), large bivalves (1–35 %), crayfish (50–75 %), different crustaceans (65–90 %), crabs (70–90 %) was accounted. Apart from these aquatic contaminations and further accumulation in variable organisms, organomercurals were also observed among terrestrial animals and humans. Birds were found to be susceptible to the MeHg concentrations and the process of demethylation was also reported as a means of detoxification (Craig and Jenkins, 2004). In poultry animals, MeHg concentrations of >1500 ppb of MeHg in eggs were reported to be toxic. In mammals, accumulation of organomercury can get demethylated in the brain within a span of 2 months, depending on the type of mammal (Takanezawa et al., 2019). The passage of MeHg towards a fetus mammal's brain via the placental route. In humans, the toxicity of MeHg comes from fish consumption and other aquatic food sources. Being a neurotoxic chemical, MeHg can specifically damage brain activity due to smooth passage through the blood-brain barrier (Craig and Jenkins, 2004; Gimenes et al., 2021). In the USA, >40 states have their rules on fish consumption having high MeHg content. No-Observed-Effects-Level (NOEL) is a limit that determines the number of fish and other sea-foods to be consumed with estimated MeHg intake per day, whereas Lowest-Observed-Effect-Level (LOEL) was calculated to be more than NOEL and numerically 0.07 μg kg-1 d-1 to develop any health issues (Suomi et al., 2017). In two studies, marine mammals (whale meat) and marine fishes were considered to assess the MeHg effects on isolated native populations in Faroe and Seychelles islands. The Faroe human population showed a neurological malfunction, but the other group did not

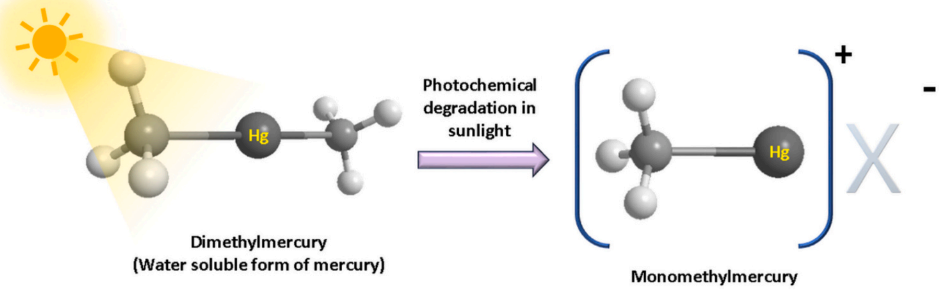

Fig. 3. Schematic representation of photochemical degradation of dimethylmercury to monomethylmercury (X denotes the halides) in the natural environment.

develop any such signs (Mason and Benoit, 2003). So, the distribution of MeHg is evident in all forms of life, but mostly, those are associated with aquatic relevance.

6.2. Organoarsenical compounds

6.2.1. Environmental distribution

Among any stable element that has organo forms, arsenic is different due to having a clear difference between marine and terrestrial organoarsenicals (Majumdar et al., 2020a, 2018; Shrivastava et al., 2020). These compounds are either toxic, like dimethylarsinic acid (DMA), monomethylarsonic acid (MMA), trimethylarsine oxide (TMAO) or arsine (AsH3) and mainly get released from the soil-plant tissues or aquatic surfaces or nontoxic like arsenolipids and arsenobetaine (Gupta et al., 2022). Compared to the terrestrial system, marine or aquatic arsenic compounds have been studied more intensively for both inorganic and organic forms. Marin algae are the primary source of organoarsenicals having simple arsenosugars, dimethylarsinoylribosides, and trimethylarsonioribosides (Craig and Jenkins, 2004; Majumdar et al., 2019a, 2024b). However, arsenobetaine (AB), another major form of organoarsenic in marine animals, has not been found in marine algae. Besides the oceanic distribution, organoarsenicals are also present in freshwater sources, coming either from the natural geogenic dissolution of As compounds or anthropogenic interventions. Reports showed the presence of DMA and MMA in river water (48 % DMA, 14 % MMA), pond and lake waters (7.7 % DMA and 2.6 % MMA), underground well water (9 % DMA and 3.1 % MMA; as a fraction of the total As present (Rahman and Hasegawa, 2012; Goessler and Kuehnelt, 2003; Suzuki et al., 2022). Studies have shown that a varied range of Arsenobetaine has been found in different marine animals like Elasmobranchs (2.4–38.1 mg kg^{-1}), teleosts (0.04–147 mg kg^{-1}), shrimps (2.1–18.3 mg kg^{-1}), lobsters (3.1–24.7 mg kg^{-1}), bivalves (0.31–1.9 mg kg^{-1}), gastropods (1.5–98.4 mg kg^{-1}), cephalopods (28–39 mg kg^{-1}). This distribution of organoarsenic in different freshwater systems has been reported from all around the globe-USA, Japan, Spain, Indonesia, and Austria. This diversified distribution has been researched and correlated to the active participation of phytoplankton metabolism, bacterial decomposition of organic matter bound As, microbial redox coupling-decoupling of Fe—Mn oxyhydroxides depending on seasonal variation or soil physico-chemical properties (Majumdar and Bose, 2018; Majumdar et al., 2019, 2020b, 2021).

6.2.2. Formation and species development

The biotransformation of inorganic As by microbes resulted from the generation of the MMA, followed by DMA and TMAO (Fig. 4). The biomethylation process involving different biological pathways are controversial due to inadequate experimental support. In a report by Challenger (1945), S-adenosylmethionine (AdoMet) acts as a methyl donor for such a methylation process. Later, in 2017, R. Bernier-Latmani et al. experimentally proven that sulfate-reducing bacteria (SRB) is primarily responsible for the sequential arsenic methylation in the paddy soil, which converts the arsenious acid (water-soluble salt) to mono, di, and trimethylated salts of As (Reid et al., 2017). They have also envisaged that the arsenite S-adenosylmethionine methyltransferase (ArsM) enzymes present in the amino acid fermenting enzymes are most effectual in the arsenic methylation processes (Qin et al., 2006).

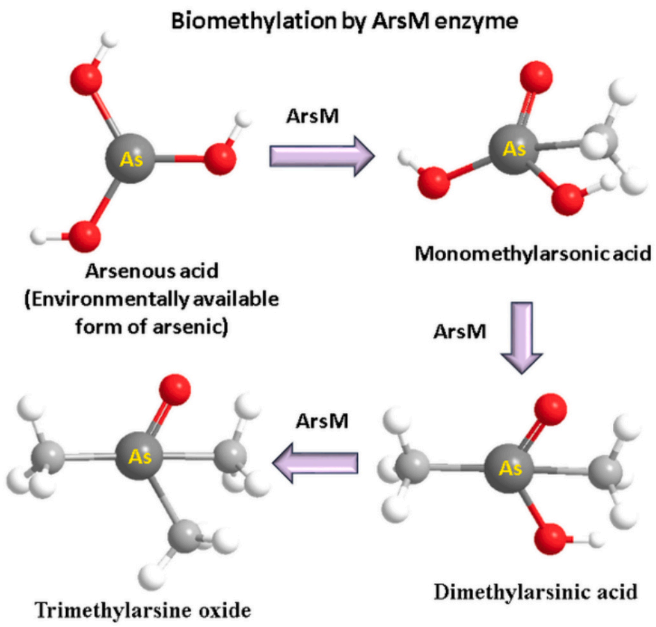

Fig. 4. Schematic representation for the step-wise biomethylation of aqueous arsenous acid-to-trimethylarsine oxide with the help of ArsM.

Arsenosugars from either algal cells or as dissolved natural compounds undergo biotransformation by microbes under an anaerobic environment in sedimentary systems where these arsenosugars get converted to dimethylarsinoylethanol which is the precursor of arsenobetaine (Edmonds and Francesconi, 2003). Like in aqueous systems, soil organoarsenic are also dependent on microbial activities and a study reported the difference between using sterilized and non-sterilized soils for the generation of these organoarsenicals where the sterilized soil did not produce any such compounds (Hare et al., 2020). Along with soil and sediments, in a study, DMA and MMA were found in sedimentary rock samples, indicating a geogenic production of these compounds underground for a longer period (Maguffin et al., 2015; Nawrocka et al., 2022). Temperature, pH, moisture content, and types of microbial community prevalent, are determining factors for the methylation or demethylation process. Several organoarsines were reported to be generated from the soil under the influence of different fungal species like *Scopulariopsis* sp., *Candida* sp., *Aspergillus* sp., *Penicillium* sp., and *Gliocladium* sp. (Wang et al., 2014; Afsal et al., 2020). In earlier days, when arsenic was used in many herbicides profoundly, soil microbes getting exposed to these herbicides, generated volatile arsines like $MeAsH_2$, Me_2AsH and Me_3As (Craig and Jenkins, 2004; Khalid et al., 2017). Not only aerobic microbes, under anoxic conditions, but researchers have also shown that different anaerobic oligotrophic microbes, Fe-Mn-S reducers and nitrate dissimilators can generate organoarsines. There, DMA, MMA and TMAO were traced from the instrumental headspace of the culture set of oligotrophs and sulfur reducers whereas only MMA and TMAO were found from the Fe—Mn reducers (Goessler and Kuehnelt, 2003). Along with the bacterial conversion of organoarsenicals, soil fungi are also well documented for their containment of such compounds. Different genera of ascomycetes and basidiomycetes, like *Sarcosphaera* sp.*, Tarzetta* sp.*, Agaricales* sp.*, Boletales* sp.*, Gomphales* sp.*, Lycoperdales* sp.*,* and *Poriales* sp. were reported to have MMA, DMA, TMAO, AC and AB (Zhang et al., 2020: Braeuer and Goessler, 2019). Another study reported that the low concentration of inorganic As in the system can minimize the formation and accumulation of organoAs in edible fungal (oyster mushrooms) tissues, although, the fruiting bodies stored some amount of MMA after a longer period of incubation (Quarcoo and Adotey, 2013; Hackethal et al., 2021). Terrestrial plants contain differential enzyme and stress-responsive mechanisms which they use for bypassing the abiotic stresses generated due to the deposition of inorganic As (Moulick et al., 2024a). The further conversion of the As molecules to their organo forms takes place readily. Studies showed that the generation and further release by volatilization of organoAs are common in wetland-associated plants like Typha, Lemna, Hordeum, Agrostis, Carex and rice (Lin et al., 2021; Kumar et al., 2019; Sarkar et al., 2017; Majumdar and Bose, 2017; Upadhyay et al., 2020). Some forms of arsenoribose compounds were found along with the other common organoAs in Lemna, Potomogetan and Myriophyllum (Pell et al., 2013).

6.2.3. *Effects and toxicity*

Studies have also shown arsenobetaine can also get degraded to other organo forms like arsenocholine (AC), TMAO, DMA etc. in different marine organisms. In marine organisms, the replacement of choline moiety in phosphatidylcholine, a major cellular biomolecule, by arsenocholine has been reported (Braeuer et al., 2018) which remains bound to these phospholipid molecules. Studies suggested the potential adversity of inorganic As to human health at very low concentrations, coming from sea-food consumption issues although organoarsenicals did not show such health issues. As a primary source, organoAs come from the marine or freshwater algae and phytoplanktons which are inevitably grazed by zooplanktons and other aquatic organisms and further bioaccumulation at the higher trophic level occurs. Arsenobetaine, if not a toxic form, but DMA and MMA (Fig. 4) are also accumulated in shrimps and prawns for human consumption and might pose a threat. Any inorganic forms of As or even organoarsenical like MMA, AB, AC or arsenosugars from seafood sources and other edible algae, get converted to the DMA inside the human system due to metabolic activities and excreted through urination (Francesconi, 2010). Analysis of human urine samples for arsenic speciation showed the presence of 10–15 % of inorganic As, 10–15 % of MMA and 60–80 % of DMA, indicating the transformation of organoAs inside the human system and non-absorbance nature to the tissues resulting in excretion and detoxification (Goessler and Kuehnelt, 2003). Other mammals like mice, rats, and rabbits were tested to understand the mechanism and found to be similar to humans. Thus, toxicity and health might develop in humans due to organoAs if the bioconversion process becomes malfunctions.

6.3. *Organolead compounds*

6.3.1. *Environmental distribution*

Elemental Pb usage and its pollution have a history of thousands of years, although the organoPb came into play around the 19th century when tetraethyllead (TAL) compounds were used to infuse with gasoline, triggering lead contamination around the globe. Among these TALs, tetraethyllead (TEL) and tetramethyllead (TML) are the most used compounds (Chau and Wong, 2020). Distribution of these compounds to the environment resulted from mishandling, lack of infrastructures, and controlling guidelines in the industrial areas and petroleum refineries (Abadin and Pohl, 2010). The U.S. Environmental Protection Agency (EPA) banned these compounds after analyzing the adversity caused to the atmosphere due to the release of the organoPb compounds and accounted that around 78 % of the atmospheric pollution was coming from industrial sources, 12 % from transportation and 10 % due to traffic fuel combustion (Michaels, 2020). OrganoPbs, dispersed in the atmosphere, finally, settle down to the land due to wet or dry deposition, and from that point, they might take entry into the aquatic environment or can infiltrate the soil strata. Depending upon the soil characteristics and nature, like pH, soil particle sizes, sand/silt/loam/clay types, the content of soil organic matter (SOM), involvement of different inorganic colloidal particles and degree of cation exchange capacity, the fate of the organoPb alter (Sigel and Sigel, 2019). In soil, the chelating effect of humic or fulvic acids, or surface adsorption activity of clay particles or immobilization by exchanging ions with oxyhydroxides determines the fate of solubilization and mobility of organoPb in soil (Rigoletto et al., 2020). A study showed that with a greater content of SOM and pH $\geq$5, the chance of retention of organoPb to SOM was increased and due to such inaccessibility and insolubility, TAL compounds do not leach out in the soil horizons. However, trialkylleads, which are partially soluble in the water, were found to be dealkylated and finally leached out to the groundwater system (Yoshinaga, 2003), although TELs were reported to be percolating through the soil column when infused with types of gasoline. Sometimes, the application of chelating agents like sodium DDTC or EDTA to mask the inorganic Pb might be useful to extract the trialkyllead compounds as a measure of removal from the soil, although for dialylleads, such extractions are unsuitable well due to the stronger affinity of dialkyllead compounds towards the soil particles (Abadin and Pohl, 2010).

6.3.2. *Formation and species development*

The environmental fate of alkyllead is a complex process which does not occur directly from the combustion of Pb-infused types of gasoline. Instead, the Pb halides are produced first (mostly in the form of PbBrCl) and some double-salt compounds with ammonium halides like 2PbBrCl·NH4Cl, $Pb_3(PO_4)_2$, and $PbSO_4$ (Horvath et al., 2021). The study also reported that around 75 % of the bromine and 40 % of the chlorine gets dissipated generating Pb-carbonates or oxides or oxycarbonates. The TEL and TML have been traced in the vapour phase in the air with a chance of rapid disintegration into tri- and dialkyllead compounds. These compounds are more stable and take time to decompose due to photolysis or reacting with hydroxyl radicals in the aerosols and ozone (Abadin and Pohl, 2010). Contamination of organoPb in the soil-aquatic systems confers a toxicological effect on the associated organisms. The gasoline spillage is one of the primary sources of organoPb derivatives in water because of the presence of organoPb derivatives (e.g., TEL, Fig. 5) (Landmeyer et al., 2003).

Although most of the organoPb compounds are not soluble in water due to their high lipophilicity, the consecutive dealkylation reactions of the organoPb derivatives ultimately make them water-soluble. The consecutive dealkylation processes involve the degradation of TEL to triethyllead ($(CH_3CH_2)_3Pb^+$) and then diethyllead ($(CH_3CH_2)_2Pb^{2+}$) which finally converted to the stable inorganic lead (Pb^{2+}) via formation of very unstable monoethyllead ($(CH_3CH_2)_1Pb^{3+}$) (Gallert and Winter, 2002; Ouyang et al., 1996).

6.3.3. *Effects and toxicity*

Among aquatic animals, fish, shrimps and crustaceans accumulate organoPb in a considerable amount which might create a health issue for consumers. In a study, different fish genera were observed- Siganus, Terapon, Gerres, Oblada, Liza, and Balistes having respectively 114, 77, 3, 76, 140 and 14 μg kg^{-1} of tetraethyllead while methylated lead

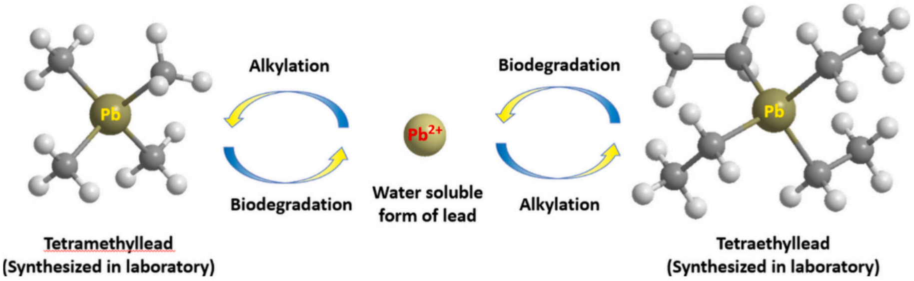

Fig. 5. Schematic representation for alkylation of Pb^{2+}-salt via biodegradation of TML and TEL to the Pb^{2+} form by environmental factors.

compounds were found almost 10-fold lesser in concentration (Shreadah et al., 2011). In another study, cod, shrimps and scallops were analyzed for the methylated-Pb and around 60–95 % of the total Pb content was found to be MePb for codes, around 55–90 % for shrimps and 70–95 % for scallops were MePb. For ethylated Pb, the concentrations were found within 85–95 % of the total Pb content (Forsyth and Iyengar, 1989). From different food sources, organoPb intake can occur in humans, and it has been established that the toxicity of tetraalkyllead is more than trialkyllead, and ethylated Pb compounds are more hazardous than methylated Pb. Both inorganic and organoPb toxicity effects are similar in pattern affecting almost every organ in the human body but predominantly the neurotoxic effect. Inside human tissues, Pb shows a structural mimic of calcium, causing physiological imbalances, whereas organoPb interferes with the heme synthesis, causing physiological imbalances, whereas organoPb interferes with the heme synthesis, resulting in haematological, renal, and hepatic and neurological disturbances (Brightman, 2017; Sigel et al., 2010). Some of the common symptoms of alkyllead intoxication include insomnia, anorexia, weakness, tremors, fatigue, vomiting, nausea, and impairment of memory. Prolonged and untreated conditions might lead to convulsions, coma and death (Gunatilake et al., 2014). Due to adverse effects on the neurological system, neuron death and brain oedema in the cerebellar and cerebral cortex, basal ganglia dysfunction and reticular formations are frequently reported with a coarse muscular tremor as a physiological indication (Abadin and Pohl, 2010). In another long-term study, persons with OrganoPb exposure in their work areas were tested for cognitive responses and memory testing with a conclusion of decreased cognitive ability along with higher blood and bone Pb content due to such long-term organoPb exposures (Mason et al., 2014). Such health complication arises due to the nature of alkyllead compounds to be less chelated and excreted from the body as compared to the inorganic forms.

6.4. Organotin compounds

6.4.1. Environmental distribution

Organotin compounds are primarily associated with anthropogenic activities resulting in terrestrial, sedimental and marine contamination (Dubalska et al., 2013). Depending on the solubility, adsorption-desorption of the organotin ions to the colloidal particles in water, participation of the microbial bioconversion of the organo-part of the tin compounds, the distribution of organotins in the environment. Several agricultural sprays, volatile biocidal compounds, incineration of treated or untreated wastes, and glass sprays allow the organotins to become a part of the aerosols. Residuals from the agricultural spraying and landfill of organotin compounds to the soil create a contamination profile which further transmits to the water bodies. Runoff from field soils, industrial discharges, leach out from shallow burials can contaminate water bodies with organotins (Cima et al., 2003).

6.4.2. Formation and species development

Most organotin derivatives were synthesized by the nucleophilic substitution reaction between alkyl halides and Sn followed by oxidation for forming bis(tributyltin) oxide (Barug, 1981). In the nature, organotins (e.g., triphenyltin chloride, Fig. 6) are probably undergo degradation to the low-molecular derivatives utilizing UV light present in the sunlight.

The other organotins e.g., Bis(tributyltin) oxide, follow biochemical degradation pathways to form dibutyltin derivatives and release carbon dioxide in the air. Some possible routes are reported through which the organotin derivatives can enter the air, soil or water systems. The R and X groups attached to the stannous part determine the compound's solubility with a general conformation of ($[R_{4n}SnX_n]$, $n = 0–3$). In the aqueous phase, the natural pH range of 5–7 with a temperature of low to moderate (10–25 °C and variable salt concentrations, the solubility of triorganotin concentrations falls within 50–70 mg/l whereas the di- and monomethyltins shows a greater solubility of 104–105 mg/l (Rumengan and Ohji, 2012; Wang et al., 2019). The adsorption of organotins ions to other sea sedimentary particles depends upon the degree of cation exchange capacity of other negatively charged metal oxides and minerals. Such adsorption and desorption of organotins can be reversible, although tributyl and triphenyltin derivatives (Fig. 6) can remain settled for a long time (Fang et al., 2017). The Sn-carbon bond in an aqueous system can sustain a high temperature. Still, the UV radiation, the effect of strong acid, and the electrophilic attack might break this bond, influencing the solubility and availability of the organotins. In the aqueous phase, the generation of the methyltins is thought to be due to the methylation processes by microbes, and such production of monobutyl (MBT) and dibutyl (DBT) derivatives takes place due to the degradation of tributyl (TBT) compounds rather than a direct input of these compounds. Reports suggest that the decaying of TBT occurs slowly in sediments but rapidly gets converted in the water system. The hydroxylation of the butyl group by microbial or chemical attack releases butene or butanol (Cima et al., 2003). Reports demonstrated that the half-life time for these organotins in aqueous media varies between days to a couple of weeks whereas in shallow freshwater sediments, the half-life is approximately a few months and in anoxic soils, it can reach up to years.

6.4.3. Effects and toxicity

Organotins are naturally toxic compounds which are divided into a few categories of low-molecular-weight trialkyltins like (e.g., trimethyl and trimethyltin, butyltins like (dibutyl and tributyltins), triphenyltins like (triphenyltin acetate and triphenyltin chloride), phenylalkyltins like (phenbutatin oxide), cycloalkyltins like (tricyclohexyltin and azocyclotin) (Cima et al., 2003; Ebah et al., 2016). The risk of organotins becomes prominent due to the consecutive bioaccumulation of those compounds in aquatic animals and algae. In fishes, it has been observed that the accumulation of butyltins and triphenyltins is through the dietary sources and branchial uptake while the excretion of these organocompounds is very low inside the fish tissues (Lee et al., 2016). In vertebrates, some organs like the liver, kidney, lymphatic tissues and brain can accumulate such organotins at the acute level. Hepatic microsomal systems can utilize and metabolize the tributyltins to dibutyl form via dealkylation, which results in metabolite deposition accumulation in several tissues (Xu et al., 2019). Marine trophic predators like cetaceans, tuna, and sharks have been reported to accumulate a considerably high

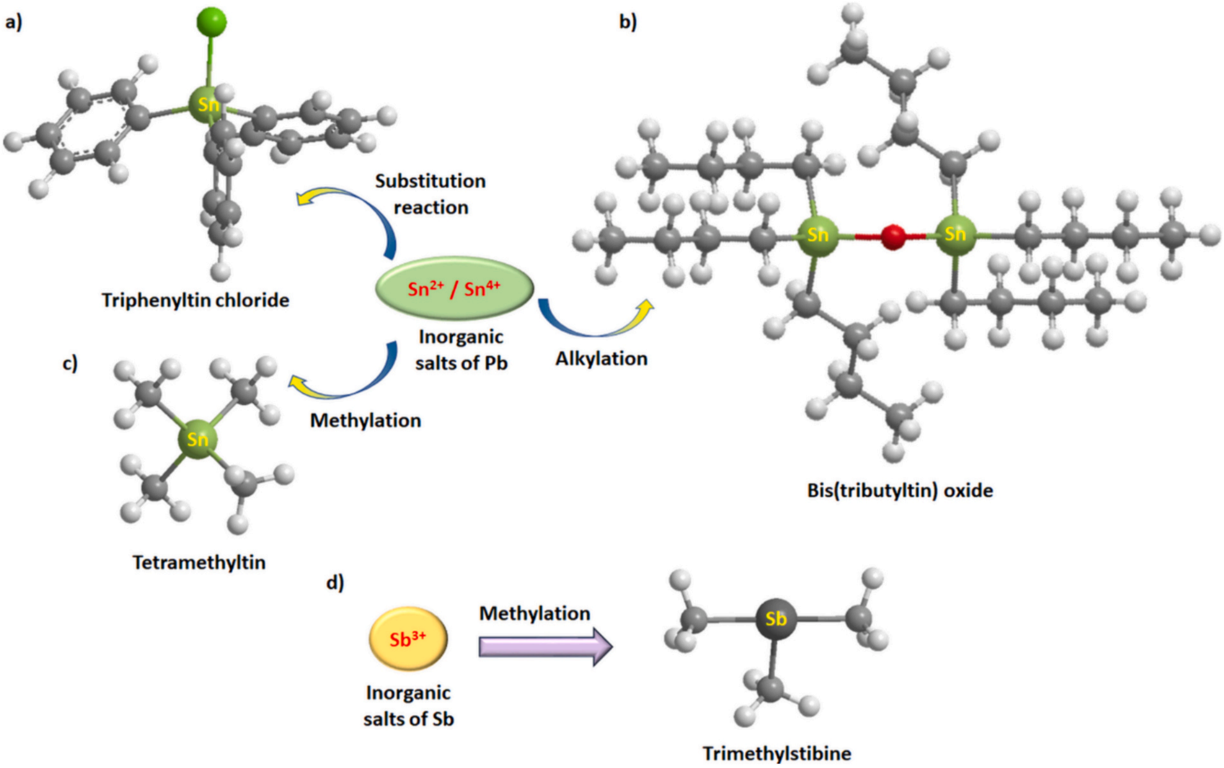

Fig. 6. Schematic representation of the formation of a) triphenyltin chloride, b) bis(tributyltin) oxide, and c) tetramethyltin, d) Schematic representation of the formation of trimethylstibine.

content of butyltins. These TBT are prominently stored inside the blubber of dolphins, kidneys of sharks and muscles of tuna, whereas DBT specifically gets accumulated in the liver and tissues of dolphins, tunas and sharks respectively (Fortibuoni et al., 2013; Chamorro-García et al., 2018). Bivalves and molluscs retain the organotins in their gills and tissues to a much greater extent compared to their body weight ratio while this accumulation is somewhat dependent on the seasonal variation and the fact that these animals do not have the required cellular machinery to detoxify the organotin derivatives from their bodies (Quintas et al., 2017). Due to this cumulative bioaccumulation in aquatic animals, a potential human health risk is associated with consuming organotin-contaminated foods. The acute toxicity of organotins can result from neurological imbalances by disrupting neurons within the hippocampus pyramidal band and fascia dentata area (by TMT) or by creating brain-spine oedema, demyelination and GABAergic neuron malfunctions (by TET) (Yu et al., 2015; Ferraz da Silva et al., 2018). TBT and DBT are reported to be hepatotoxic, causing inflammation of the bile duct, hepatomegaly, lesions and necrosis (Zhou et al., 2016). Most organotins are known for hemolytic activities due to the chemical nature of lipophilicity and amphipathic disruption of membranes in the order of TBT > TPT > TET > DBT > TMT > MBT. Not only these cellular and neural disturbances, but organotins are also potent for causing immunological interferences by generating atrophy of the thymic cortex and apoptosis of lymphoid tissues, resulting in a severe depletion of T-lymphocytes in the peripheral blood (Cima et al., 2003). Also, the natural killer cell activities like tumour suppression are retarded by TPT and TBT, causing a risk of cancer development.

6.5. *Organoantimony compounds*

6.5.1. *Environmental distribution*

The occurrence of organometallics in the environment results from either geogenic distributions or anthropogenic introduction of these compounds. In the case of antimony (Sb), the organo forms are not well applicable for larger aspects (Islam et al., 2016). The environmental distributions are mostly assumed to be natural via the biomethylation process. In the case of Sb, only methylated species are essential from the environmental aspects. Volatile forms of organoSb have been detected in diverse environments. Oxide deposition areas of antimony ores showed the presence of volatile Sb compounds around sewage and landfill sites releasing gases. Trimethylstibine has been identified from the leaking gases of sewage sites by using GC-ICP-MS from Germany, and Canada (Andrewes and Cullen, 2003). A similar mono/di/trimethylantimony release was detected from landfill sites' gas-collection pipelines where a methanogenic micro-environment has been generated. Such a methanogenic environment triggers the growth of a diverse microbial community, and there, both the di and trimethylstibine compounds were detected produced by algal biofilms (Li et al., 2016). Soil and sediment containment of these organoSb compounds are poorly reported due to poor distribution and fewer available study reports. Being less available in the soil, organoSb accumulation in plants and animals is also very rare, and only a few studies reported the presence of mono and dimethylSb species in plants like *Funaria* sp. and *Typha* sp. after a complex chemical extraction process and analysis using HG-GC-AAS (Ji et al., 2018).

6.5.2. *Formation and species development*

Till date, the mechanism of trimethylstibine formation in nature is still unknown to the researchers. Still, from the similarity of chemical properties between arsenic and antimony it has been believed that the formation of trimethylstibine was evolved from biomethylation of antimony in the nature (Parris and Brinckman, 1975). Monomethylantimony species is the only type detected in the river water whereas, in the marine water, mono and dimethylSb were found except the trimethyl form. In other study reports, methylSb was detected in water columns on the surface to deep layers of the Baltic Sea and the Gulf of Finland. monomethylSb were found with variable concentrations with a greater content at the near surface but the dimethylSb were detected occasionally (Filella, 2010). The generation of trimethylSb, a rare form of SB in natural water, along with the other two methylated forms, was reported in the hydrothermal vent water of New Zealand (Andrewes and Cullen, 2003). The primary generation of organoantimony results due to the biomethylation process by diverse microbes and one of the first trimethylstibine reports was documented using an anaerobic mix culture. From Sb-infused soil samples and anaerobic systems, microbes like *Methanobacterium* sp., *Clostridium* sp., and *Desulfovibrio* sp. generated a variable and minor quantity of trimethylstibine. Under aerobic conditions, microbes like *Scopulariopsis* sp. and *Phaelus* sp. can perform the Sb biomethylation where the production of mono/di/trimethylstibine depends on the microbial species involved (Caplette et al., 2021) although Sb(V) compounds are biomethylated in a much slower rate than Sb(III) compounds.

6.6. Organosilicon compounds

6.6.1. Environmental distribution

Silicon (Si) is one of the main components of the earth's crust. Still, organosilicons have only been reported in the last couple of decades to be involved in environmental distribution and toxicology. Anthropogenically produced organoSi has a Si-O-Si bond, also termed as siloxanes, and primarily, there are three major siloxanes predominate-polydimethylsiloxane (PDMS), volatile methylsiloxane (VMS) and polyethermethylsiloxane (PEMS). Due to its high molecular weight, PDMS is refrained from being bioaccumulated. Due to other nutritive enrichment prospects, sludge usage in the agricultural soils also allows PDMS distribution, as subsequent irrigation flushed out some fraction of PDMS out of the field. Unscreened fractions of PDMS in wastewater discharge allow it to settle down to the bottom of the sediment (Hirner et al., 2003). Volatile methyloxysilones (VMS) are generally cyclic or linear and play a crucial role in organoSi distribution in the environment. In a study, it was shown that sewage sludge application can certainly introduce VMS to the environment, and a concomitant dispersion into the soil and uptake by the plant is possible where it might cause further problems to the residents in the long span (Fernandes et al., 2022). Having low molecular weight and high vapour pressure, VMS has several industrial applications, so the chance of consumer encounter is also high. VMS molecules can be small enough to pass through the biological cell membranes and hence show a very low biodegradation rate while having a very high bioaccumulation factor (Powell et al., 2018). Octamethylcyclotetrasiloxane or D4 can get volatilized easily with a residual half-life of 3 h to 6 days in aquatic systems (Flassbeck and Gruemping, 2003), which can be traced by using aquatic microflora with radio-labelled carbon. When monomeric organosilicons contain at least one hydroxyl group, it is known as organosilanols and these molecules are highly soluble in the water system with very low volatilization properties.

6.6.2. Formation and species development

PDMS is an organoSi compound generally synthesized in the lab following different synthetic pathways. In 2022, a study reported a convenient path for the synthesis of PDMS in which dichlorodimethylsilane was hydrolysed in an alkaline condition to form a monomeric unit of dimethylsiloxane which was further self-polymerized and formed PDMS (Supplementary Fig. 6) (Fauziah et al., 2022). PDMS is primarily found in wastewater discharge. Still, due to the physicochemical properties, PDMS can be removed from the sewage water to the sludge particles. Therefore, the sludge processing further determines this PDMS's fate (Surita and Tansel, 2014). Complete pyrolysis can convert these PDMS into carbon dioxide, water, and silica, whereas, in the soil system, PDMS is also converted to these residual forms after a series of soil-driven catalysis and degradation. Organosilanols can get transformed into organosiloxanes under acidic or alkaline environments, so when PDMS gets hydrolyzed, the generation of organosilanols was reported to produce dimethylsilanediol and monomethylsilanetriol in dry soil (Scholey et al., 2018; Xu and Knoerr, 2020). The hydrolysis was subjected to the production of CO_2, which was further detected by the gas chromatography-mass spectrometer (Supplementary Fig. 6) (Sabourin et al., 1996).

Moist soils were tested for the PDMS conversion to silanols to be around 3 % after 6 months of incubation, while dry soil triggered the PDMS hydrolysis and about 50 % silanol generation was recorded. These organosilanols are very water soluble and hard to separate from the water. Due to low-moderate vapour pressure, these substances can remain in both the aqueous and aerosol states. Reports showed evidence that silanols can interact with humic substances and hence can be associated with the soil-sediment phases (Kulikova et al., 2021). Hydroxyl radicals can react with these organosilanols, and trimethylsilanol was found to be more reactive than methylsiloxanes in the atmospheric environment. Demethylation of organosilanols due to biodegradation and photolysis can produce subsequent silica, carbon dioxide and water. Biodegradation can be via enzymatic hydrolysis generating the C-O-Si, N—Si or Si—H bonds when co-cultured with *Pseudomonas* sp., *Proteus* sp. (Flassbeck and Gruemping, 2003). The toxic effects of organosilicons were tested on various organisms, although no severe toxicity was found.

Other organometal compounds are also mentioned related to their availability and distribution in the environment, briefly in the Supplementary section.

7. Health risk assessments of predominant organometal(loid)s

Distribution of organometal(loid)s in terrestrial or aquatic environments can have direct or indirect health issues and related consequences that are necessary to discuss for a precautionary measurement (Majumdar, 2021; Majumder et al., 2023). Globally, inorganic As and its organic species have resulted in the largest human health calamity, and several researchers have shown the route and pattern of this toxicity depending on the sources of As, age groups of the native people, types of food consumed and prolonged time of exposures (Upadhyay et al., 2019b; Khan and Muhammad, 2020). While considering the risk assessment of such environmental toxicants, a couple of parameters are generally analyzed, including risk quotient I (RQ I), the average daily dose (ADD), hazard quotient (HQ) and cancer risk (CR). RQ I is generally considered when a preliminary understanding of the toxicants is necessary to know the degree of pollution it is causing (Dai et al., 2022). An RQ I value >1.0 indicates a high ecological risk is associated with the organometal(loid)s. In contrast, a value within 0.1 to 1.0 is moderately risky and needs attention for control measurements. In contrast, a value between 0.1 and 1.0 is moderately risky, and a value <0.1 is a low-risk indicator. The following formula is used to calculate the RQ I of an organometal(loid)-

$$\text{RQ I} = MEC_{95} / PNEC_{fw}$$

where MEC_{95} indicates the 95th percentile of the environmental organometal(loid) measured and $PNEC_{fw}$ indicates the lowest value of predicted no-effect concentration in the freshwater of the concerned organometal(loid) (Dai et al., 2022). Further assessment suggests the ingestion of the concerned organometal(loid)s via water or food items using ADD, indicating the quantitative amount of the organometal(loid) is getting inside the consumer. For organic species of As, primary toxicity develops due to DMA and MMA rather than arsenobetaine or arsenocholine and these DMMA or MMA mainly get transferred from contaminated crops (Chen et al., 2020). In the study by Khan and Muhammad (2020), DMA and MMA were reported in the native people of Pakistan and deposited in their hair, nails, urines, and blood samples. Another report by Wei et al. (2017) showed the deposition of these organoAs species in the nails and skin tissues of the native population of China. These studies have shown a greater toxicity rate and As accumulation in children or young people who had a higher ADD value than older people. In this respect, the ADD can be calculated as-

$$\mathrm{ADD} = (\mathrm{C} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED})/(\mathrm{BW} \times \mathrm{AT})$$

where C is the concentration of organoAs species (mg kg^{-1}), IR is the ingestion rate or consumed volume of water (avg. 3 lit day^{-1}), EF represents the exposure frequency (365 days $year^{-1}$), ED is the exposure duration during which time the organoAs got accumulated to the consumers, BW is the average body weights of the residents (average BW will be different for adults and children), and AT is the average time or the total time in ED. The risk assessment of organometal(loid)s considering RQ I and ADD can be used for calculating HQ or hazard quotient, which finalises if the concentration of the concerned organometal(loid)s is creating a health hazard to the consumers (Sarwar et al., 2021). This HQ can be calculated as-

$$\mathrm{HQ} = \mathrm{ADD}/\mathrm{R_fD}$$

RfD is the reference or background value of the organometal(loid)s and is considered a threat to the consumers if the HQ value is >1 (Adimalla, 2019). All these risk assessments parameters are finally linked to the CR which suggests the acute toxicity level of the pollutant and can be calculated as-

$$\mathrm{CR} = \mathrm{ADD} \times \mathrm{SF}$$

where SF, the cancer slope factor, differs depending on the involvement of organometal(loid)s (Khan and Muhammad, 2020). Here, HQ values play a crucial role along with the duration of exposure to the concerned organometal(loid)s.

A few recent studies have been reviewed in this section, where researchers have demonstrated human health risk assessments for specific organometal(loid)s. Salazar-Camacho et al. (2022) have pointed out the effect of organomercurials on the native population of adults and childbearing women in Columbia. They used another equation for the assessment of estimated weekly intake (EWI) of organoHg in the population as-

$$\mathrm{EWI} = (\mathrm{IR} \times \mathrm{C})/\mathrm{B_w}$$

where C is the median concentration of MeHg and IR is the weekly food intake rate. Another study by Uc-Peraza et al. (2022) indicated the combined and species-specific effect of methyl/butyl/phenyltins on the coastal population of Mexico where a rise in health risk is found. A similar study was conducted by You et al. (2022) on three rivers in China where aquatic animals are found to contain butyl and phenyltin compounds, which increases health risks to the native population, especially children. The report has used another equation to assess the trophic level (TL) of organotin compounds using radio-labelled isotopes. The equation is-

$$\mathrm{TL} = \left[\left(\delta^{15}N_{consumer} - \delta^{15}N_{baseline}\right)/\Delta\delta^{15}\mathrm{N}\right] + 2$$

where $N_{consumer}$ and $N_{baseline}$ are the organotin concentrations in the contaminated site and background (uncontaminated site), respectively, both these terms should be defined by the stable isotope assessment. Guo et al. (2022) conducted a study where young college students from China were tested for methylsiloxanes under different situations indoors, outdoors and in personal care products. The result suggests a greater tendency of organosilicon risk development in girls than boys, mainly from personal care products. Another study by Guo et al. (2020) shows the involvement of this methylsiloxanes risk possibility in the blood plasma of volunteers from different sites like industry, universities, and household colonies where the presence of organosilicons has been highly associated with the peoples of industries and surrounding areas. OrganoAs species are reported by Chen and Liu (2023) in mushrooms in China. Related health risk possibilities are also presented, which suggests the involvement of organometal(loid)s in foodstuff must be analyzed regularly to identify the status of these toxicants and follow the possible remediations.

8. Remediation strategies for organometal(loid)s toxicity

Organo-metals and metalloids distribution and contamination are crucial aspects of environmental quality maintenance, and they need tactful attention so that the production of these compounds is not hampered for mass utilisation while the toxicity is also under control. Environmentalists can provide a clear idea of the degree of contamination and how those problems should be addressed. For practical remedial purposes, removing organo-elements or biotransformation is necessary. Depending on the type of organoelement, bioremediation or physico-chemical remediation approaches can be opted.

8.1. Bioremediation of organometal(loid)s contamination

The term bioremediation indicates involving biological organisms to minimize environmental hazards to restore balance. When organo groups are bound to the metallic ions, the degree of toxicity varies with the concerned element and dispersed medium. As a primary mode of remediation, bio-agents cleave the metal(loid)s-carbon bond and release the inorganic form of that element (Thayer, 2012; Klein and Thayer, 2017). On the other hand, these bioremediatory agents can also opt for the sequestration of metal(loid)s to chelate down the contaminants. Bioremediation can be performed by choosing one of the two primary sources- plants (phytoremediation) or microbes (microbial remediation).

8.1.1. Phytoremediation approach for organometal(loid)s removal

Depending on the environment, different plants can be used for the removal of contaminants from soil or aqueous systems, for example, water hyacinths (*Eichornia crassipes*) is well documented for taking up such aquatic contamination (Majumdar et al., 2018) or ornamental plants for the urban soil clean-up (Majumdar et al., 2022a, 2022b, 2022c) or cattails (*Typha latifolia*) for the marsh wetland pollutant removal (Sarkar et al., 2017; Majumdar et al., 2023a, 2023b, 2023c). Studies showed that the bioaccumulation of methylated As and inorganic As in duckweed (*Spirodela polyrhiza*) followed different pathways while the fern *Pteris vittate* and *Boehmeria nivea* showed higher tolerance towards methylAs with lower accumulation rate (Jang et al., 2016). For organoHg removal, plants are usually genetically engineered using *mer A* and *mer B* genes, which encode mercuric ion reductase and organomercurial lyase enzymes, respectively (Li et al., 2020a, 2020b). Rice, tobacco, cottonwood, and arabidopsis were used for enhanced tolerance and uptake of organoHg compounds by roots with subsequent translocation to the shoot and leaves. Some plants like willow, lettuce, barley, brassica, and sorghum, were also tested for removing organotins (mainly TBT) with further degradation to DBT and MBT (BD, 2014). Some halophytic plants, *Halimione portulacoides, Sarcocornia fruticose,* and *Spartina maritima*, were tested for TBT uptake and removal from sediments efficiently (Finnegan et al., 2018). In the case of organoSe, plants use a combination of both intracellular sequestration and volatilization. Transgenic hydrilla, ryegrass, and Indian mustard can take up organoSe greater than the wild-type species.

8.1.2. Microbial remediation approach for organometal(loid)s removal

Microbes are widely distributed in all possible environmental systems and can potentially transform metal(loid)s during their metabolism and tolerance (Eswayah et al., 2016; Zoni et al., 2012), as shown in Fig. 7 (a–b).

Here, microbial tolerance and remediation of organomercurials and organolead are shown in Fig. 7a, and organoarsenic compounds in Fig. 7b. According to some reports, the microbial tolerance of organoHg and organoPb are linked and mostly mediated by biochemical reactivity and enzymatic actions. Tolerance towards organoAs develops due to a combined action of transporter proteins and enzymatic activities.

Sedimentary bacteria were able to convert organoHg into inorganic elemental Hg by breaking the methyl-Hg bond, producing Hg^{2+} ion and

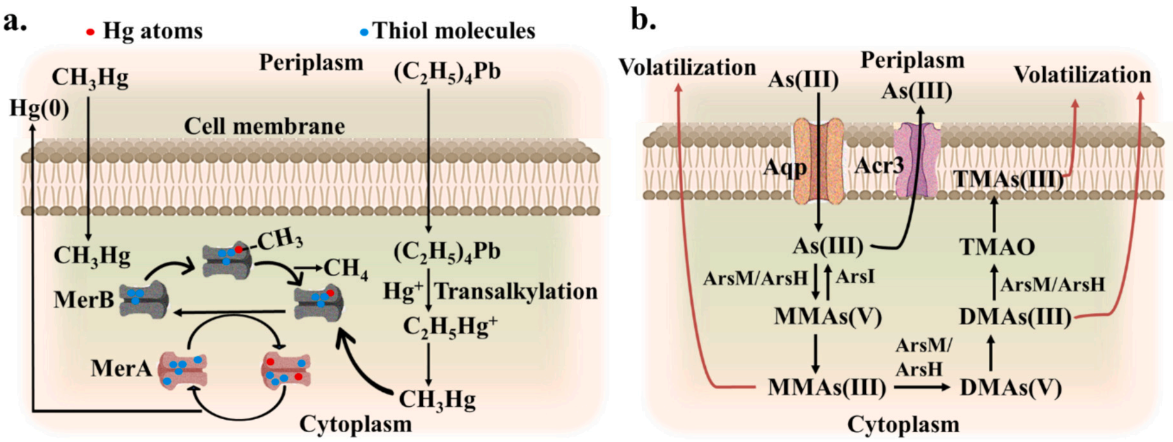

Fig. 7. Schematic representation of microbial remediation mechanisms towards organomercurial, organolead and organoarsenic compounds.

further to Hg^0. These conversions involve enzymes *mer A* and *mer B* from *mer* operon, identified by Hg-resistant bacteria (Christakis et al., 2021). In another co-metabolic pathway of methylotrophic metabolism of anaerobic microbes (methanogens and sulfate-reducing bacteria), via oxidative demethylation and methylHg degradation follows carbon dioxide production, a small amount of methane and elemental Hg (Zhou et al., 2020). Reports suggest that the reductive Hg demethylation via *mer* activity prevails in the aerobic condition at high Hg concentrations. In contrast, in the case of lower Hg content, under anaerobic conditions, oxidative demethylation predominates (Barkay et al., 2003). For organoAs, the distribution is mainly associated with plant accumulation and thus microbial participation is primarily associated with biotransformation and cellular sequestration at a lower extent. Bacteria like *Pseudomonas* sp.*, Arthrobacter* sp.*, Bacillus* sp.*,* and *Stenotrophomonas* sp.*,* were tested for the degradation of various soil contaminating organoAs compounds like di/triphenylarsine, triphenylarsinesulfide, methyldiphenylarsine, diphenylchloroarsine etc. slowly depending on the soil physico-chemical parameters and moisture contents (Köhler et al., 2001). The *As*-methyl bond cleavage by *Mycobacterium neoaurum,* and *Pseudomonas putida* generates both As(III) and As(V) from the methylated As species. In organic soil, microbial degradation of arsenobetaine and DMA was reported in the order of arsenobetaine to dimethylarsenoylacetate to DMA to MMA to arsenate (Xue et al., 2021). Triphenylarsine degradation by soil microbes suggested a gradual conversion to phenylarsonic acid and finally to inorganic arsenic. The degradation of organolead compounds is mainly triggered by environmental factors followed by some additive microbial dealkylation of Pb. In a study, tri- and dimethyllead chloride was degraded by inoculated microbes while adding nutrients was used compared to the sterilized soil/aqueous media where no microbes were present. This indicates the enzymatic co-metabolic activity of microbes, allowing alkylated Pb degradation while modulating other nutrients (Mousavi et al., 2021). The bacteria *Arthrobacter* sp. and fungus *Phaeolus schweinitzii* were observed to decompose these organoPb compounds when changed from minimal culture media to a complex enriched culture media. Under an aqueous system, tetraethyllead degrades due to microbial intervention in order of tetraethyllead to triethyllead to diethyllead to elemental lead. During the incubation with microbial cultures, tetraethylPb was found to be broken down with a half-life of 2 h, while without any microbial participation, the time was increased up to five times (Teeling and Cypionka, 1997). Several microbial genera, including Bacillus, Pseudomonas, Klebsiella, Aeromonas, Alcaligenes, Citrobacter, Enterobacter, etc., are reported to be resistant towards tributyltin compounds and degrade under differential environmental conditions (Cruz et al., 2015). In a study, the measurement of TBT was carried out using GC–MS to trace the residual concentration after microbial degradation after an incubation period of 120 days. It was found to be reduced by half of the primary TBT content (Finnegan et al., 2018). In some other studies, it was shown that bacterial secondary metabolite secretions can break the Sn—C bon and degrade TBT as in *Pseudomonas chlororaphis*, producing pyoverdins, and *P. aeruginosa*, producing pyochelin (Sampath et al., 2012). Microbial community in activated sludge can also participate in organotin decomposition, although it was observed that the rate of dibutyltin breakage was faster than the tributyltin, although triphenyl and monobutyltins were degraded slowly (Khanolkar et al., 2015).

8.2. Physico-chemical remediation of organometal(loid)s contamination

Apart from the conventional bioremediation approaches, some other physico-chemical means can be taken into consideration for remedial measurements including electrolysis and electrocoagulation of organometallics and photolysis or irradiation lysis of organo-complexes. In environmental aspects, applications of these methods are new and case-specific; hence, they need more thorough research and investigation. Sorption of organometal(loid)s onto suitable natural or engineered substances is a way of physical mode of removal based on chemical associations. Reports are evident that methylHg can be sorbed onto the natural clay and sediment depositions as well as activated charcoals and manganese dioxides (Murphy et al., 2021; Chowardhara et al., 2024; Schwartz et al., 2019; Desauziers et al., 1997). However, natural substances like clay or soil get some additional effect from the DOM which can alter the sorption rate and kinetics of this methylHg group (Hsu-Kim et al., 2013; Mridha et al., 2024). Organoarsenicals are more prominently researched for their sorption degree onto various chemical substances, including iron oxides, aluminium oxides, and ferralsol (Yang and He, 2015; Shimizu et al., 2010; Lafferty and Loeppert, 2005). Under a natural environment, organic matter has a sorption effect on the organoAs (Ou et al., 2017), whereas iron compounds like goethite, ferrihydrite, hematite and magnetite have variable sorption efficacy for the organoAs depending on the surrounding pH between 3 and 7 (Campbell and Nordstrom, 2014). However, the general trend of sorption is not very high for these organo forms of As compared to their inorganic forms. The report has suggested the sorption ability of tributyltin on sediment or particulate matters depends on the sorption coefficient, which in turn depends on the grain size of the sediment or sorption substance; hence, a finer particulate matter will have a higher sorption capacity (Ohtsubo, 1999). Also, these organotins are prone to alter their cationic form to hydroxides with increasing pH, which makes this sorption ability more prominent. In another study, it has been reported that natural organic matter, which has fulvic and alginic acids, can act as a sorption media for the trimethyltin within a natural pH range of 5–8 (Giacalone et al., 2006). Cationic exchange reactions with organolead make it difficult to adsorb on a soil-sediment system. However, a study successfully demonstrated the application of a solution mixture of calcium chloride, tropolone, and glacial acetic acid can efficiently adsorb organoPb on the media with >70 % recovery rate (Huang et al., 2003; Huang et al., 2018). To understand the proper desorption mechanism, Cukrowska et al. (2010) found that the nonpolar species of organolead like tetraethyl and tetramethyllead are more easily desorbed from the soil-sediment than other ionic organometals like butyltins. These organoPbs are strongly adsorbed at the pH 7–8, however, the redox fluctuations and pH change causes the release of these compounds from the soil.

In the electrolytic process, the metal-carbon bond is broken, and the metallic part is separated from the organo-complex in subsequent steps. In the electrolysis process, the electrocoagulation technique is an integral part where a simple electrochemical unit cell with a single cathode and anode is used and the anode part gets oxidized due to applied potential (Ali et al., 2011). Photolysis, on the other hand, via UV irradiation, is the common form of organometal breakage option, which can be either natural or as a treatment. In natural water, demethylation of organoHg has been reported in several studies due to photolysis (Lehnherr and Louis, 2009). The primary mechanism as suggested involves singlet oxygen generated from the sunlight or UV radiation in the natural environment and reactive oxygen species trigger the decomposition of the organo group from the metallic counterpart. While associated with a ligand like glutathione or EDTA, the organoHg breakage rate was reported to be enhanced (Zhang and Hsu-Kim, 2010). UV-lysis of methylated As species was reported also with demethylation of MMA and DMA to the inorganic As(V), and a merged UV/$S_2O_8^{2-}$ was proved to be a better oxidizer than UV/H_2O_2 or UV/TiO_2 combinations (Ryu et al., 2013). Reports showed that the photolysis due to 200 to 254 nm wavelengths is naturally present and able to break the methyl Hg and methyltin compounds even if these compounds are present in the atmosphere in a volatile form (Craig et al., 2000). Other than the photolysis, physical breakage of organometal(loid)s by hydroxyl radicals or ozone is also possible. Energy associated with the breakage of these metal-carbon bonds has been recorded to be 88–141 kJ/mol for organoHg, 218 kJ/mol for organotin, 155 kJ/mol for organoPb and the corresponding energy produced by the photolysis or hydroxyl radicals reaches up to 350 kJ/mol which is sufficient to break any such atmospheric metal-carbon bonds prevail (Craig et al., 2000; Himeno et al., 2022).

9. Suggestions and future perspectives

Growing industrialization and pollution load created havoc on the environmental stability of different naturally toxic elements that becomes more toxic when get associated with organo compounds. Many of these organo groups are generated in the environment under an undisturbed zone due to the redox status change of the inorganic elements (Majumdar et al., 2023a, 2023b, 2023c). Higher mobility and bioavailability are associated with these organometal(loid)s, and uncontrolled anthropogenic activities can increase the chance of exposure to these highly toxic organometal(loid)s, resulting in health issues. This review analyzes the involvement of various organometal(loid)s in the environment and how they chemically behave and interact with environmental and biological beings. It becomes evident that the necessity of regular research on these organometal(loid)s is critical and we must prioritize organometal(loid)s and their effect on environmental sustainability. United Nations has declared the motto of sustainable development goals (SDG) where environmental pollution removal and clean-up strategies are focused along with the well-being of the world's population (SDG 2, 3, 13, 14, and 15) and dealing with organometal (loid)s contamination can correlate all these points (Lal et al., 2021). At present, the interest in organometal(loid)s activity within the regional area is scattered with a focus which is concentric more on organoAs, organoHg, organoSn and organoPb. Continued monitoring of these organo-compounds should be practiced. So, researchers from different academic expertise should focus together to deal with such complex bio-environmental chemistry of organometal(loid)s and their remediation aspects.

10. Conclusion

To date, researchers are involved in the diversified assessment of organometal(loid)s from every possible aspect to gain the lacking knowledge of toxicity and the effect of the compounds on our environment with possible mitigation strategies. This extensive review tried to elucidate the chemical and environmental perspectives of these organo-compounds and as discussed, mainly mercury, lead, tin, arsenic and antimony have some strong pieces of evidence of generation, and distribution in the environment with further ways of degradation by either microbes and plants or by physico-chemical interventions. Environmental release of these elements follows two ways- either via biological alkylation or via redox reactions in the environment. Whereas the degradation of such organo-metallics is also possible due to biological interferences, especially by microbes. But, as this review has found, many aspects are still unclear and need a series of experiments both at the molecular level and ecological ground. With gradually increasing pollution to the environment, it is important to use the microbes at their fullest capacity to degrade organo-pollutants minimizing health risks at the higher trophic level. Trace elements like bismuth, selenium, cobalt, and tellurium are also known for producing organo-complexes, although very few studies are available. Overall, more rigorous studies are necessary for a better understanding of these organo-elemental distributions in the environment prior to any proposal for mitigation strategies and environmental protection laws.

CRediT authorship contribution statement

Arnab Majumdar: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Munish Kumar Upadhyay:** Writing – original draft, Visualization, Validation, Resources, Formal analysis. **Megha Ojha:** Writing – original draft, Visualization, Resources, Data curation. **Rakesh Biswas:** Writing – original draft, Validation, Software, Methodology, Formal analysis. **Saikat Dey:** Visualization, Validation, Methodology, Data curation. **Sukamal Sarkar:** Visualization, Validation, Software, Methodology, Formal analysis. **Debojyoti Moulick:** Writing – original draft, Validation, Software, Formal analysis, Data curation. **Nabeel Khan Niazi:** Writing – review & editing, Writing – original draft, Validation, Data curation. **Jörg Rinklebe:** Writing – review & editing, Writing – original draft, Validation, Resources, Data curation. **Jen-How Huang:** Writing – review & editing, Writing – original draft, Validation, Resources, Data curation. **Tarit Roychowdhury:** Writing – review & editing, Writing – original draft, Supervision, Resources, Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.175531.

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