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REVIEW

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Arsenic through aquatic trophic levels: effects, transformations and biomagnification—a concise review

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Abstract

Arsenic (As) contamination is a major global environmental concern with widespread effects on health of living organisms including humans. In this review, the occurrence (sources and forms) of As representing diverse aquatic habitats ranging from groundwater to marine environment has been detailed. We have provided a mechanistic synopsis on direct or indirect effects of As on different organismal groups spanning from bacteria, algae, phytoplankton, zooplankton and higher trophic levels based on a review of large number of available literature. In particular, special emphasis has been laid on finfishes and shellfishes which are routinely consumed by humans. As part of this review, we have also provided an overview of the broadly used methods that have been employed to detect As across ecosystems and organismal groups. We also report that the use of As metabolites as an index for tracking As_{tot} exposure in humans require more global attention. Besides, in this review we have also highlighted the need to integrate 'omics' based approaches, integration of third and fourth generation sequencing technologies for effective pan-geographical monitoring of human gut microbiome so as to understand effects and resulting consequences of As bioaccumulation.

Keywords: Aquatic food chain, Arsenosugar, Arsenolipid, Bioaccumulation, Biomagnification, Omics

Introduction

Arsenic (As) is a ubiquitous element present naturally in the earth's crust and one among the 20 most common elements (Nordstrom 2002). It was first isolated and purified in 1250 AD and since then it is used in agriculture, electronics, drugs and metallurgical applications (Nriagu and Azcue 1990). Though the origin and distribution of As is mainly geogenic, anthropogenic activities (including mining, fossil fuels burning and pesticide application) can also lead to As contamination across various environments (Bissen and Frimmel 2003). This has become a major environmental concern at a global scale (Bissen and Frimmel 2003; Ravenscroft et al. 2009; Chung et al. 2014; Upadhyay et al. 2018; McArthur 2019; review by Shaji et al. 2021). The metalloid As is found in two forms namely, organic arsenic and inorganic arsenic (iAs), while As can be further classified into four major oxidation states namely, arsenate [As(V)], arsenite [As(III)], elemental arsenic (As^0) and arsine (As^0). Additionally, there are few methylated derivatives termed as 'fish arsenic' (arsenobetaine—AsB and arsenocholine—AsC) and arseno-sugar compounds (Table 1) which have been reported from various environments (Fig. 1 for chemical structures) (Ng 2005). Other than that, arsenic is naturally found in more than 200 different mineral forms, of which ca. 60% are arsenates, 20% sulfides and sulfosalts and the rest are arsenides, arsenates, oxides, silicates and elemental arsenic (Mandal and Suzuki 2002).

Naturally iAs is easily dissolved and mobilized and hence is more toxic than organic As and reported in

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Table 1 Examples of major organoarsenical compounds (adopted from Sharma and Sohn 2009)

Organoarsenicals	Formula
Methylarsine	CH_3AsH_2
Dimethylarsine	$(CH_3)_2AsH$
Trimethylarsine	$(CH_3)_3As$
Monomethylarsonic acid	$CH_3AsO(OH)_2$, MMA^V
Monomethylarsenous acid	$CH_3As(OH)_2$, MMA^{III}
Dimethylarsinic acid	$(CH_3)_2AsO(OH)$, DMA^V
Dimethylarsenous acid	$(CH_3)_2AsOH$, DMA_{III}
Trimethylarsinic oxide	$(CH_3)_3AsO$, TMAO
Tetramethylarsonium ion	$(CH_3)_4As^+$, TMA^+
Arsenobetaine	$(CH_3)_3As^+CH_2COO^-$, AB
Arsenocholine	$(CH_3)As^+CH_2\ CH_2OH$, AC
Arsenosugars	**Structures**
Dimethylarsinoylribosides	Fig. 1 (a to k)
Triaklylarsonioribosides	Fig. 1, (l, m)
Dimethylarsonoulribtol sulphate	Fig. 1, (n)
Glycerophosphorarsenocholine	Fig. 1, (o)
Glycerophosphatidylarsenocholine	Fig. 1, (p)

varying concentrations from terrestrial and aquatic environments including associated biota (Shrivastava et al. 2015). Higher concentration of As is found in igneous/argillaceous sedimentary rocks (0–143 mg/kg) while marine sediment might contain up to 3000 mg/kg, which can co-precipitate with iron hydroxides and sulphides in sedimentary rocks (Boyle and Jonasson 1973). Besides iron deposits, sedimentary iron ores and manganese nodules can contain arsenic in high concentration (Mandal and Suzuki 2002). Similarly terrestrial soil can have varying concentration of As ranging between 0.1 and 50 mg/kg and can also vary based on geographical settings (Colbourn et al. 1975). Higher concentration of As in soil has been reported from China (mean: 11.2 mg/kg; Wei et al. 1991), India (mean: 14.8 mg/kg; Chakraborti et al. 2001) and Bangladesh (mean: 22.1 mg/kg; Nickson et al. 2000) compared to other countries. The As concentration also vary depending on the nature of soil and higher concentration has been reported from alluvial or organic soil compared to sandy soil (Kabata-Pendias and Pendias 1984; Punshon et al. 2017).

Over the last several decades, rapid industrialization has led to increased As emission in atmosphere (particularly those from geothermal plants and non-ferrous metal smelters) (Zhang et al. 2020a, b). According to Chilvers and Peterson (1987), emission in the atmosphere from natural sources is ca. 1.5 times higher than estimated emission from human activities. Nevertheless, due to continuous emission and accumulation, both organic and inorganic As have been reported in higher concentration from aquatic environments compared to terrestrial environments (IPCS 2001). Though geogenic in source, As contamination in aquatic biological communities occurs through accumulation from lower trophic levels to higher trophic levels (e.g. flora and fauna, particularly higher in members under the Class Pisces) (Jankong et al. 2007; Grotti et al. 2008; Taleshi et al. 2014; Srivastava and Sharma 2013; Williams et al. 2014; Oliveira et al. 2017; Han et al. 2019). Such accumulation of As in aquatic flora and fauna are typically based on geographical settings such as freshwater, estuarine, transitional and marine ecosystems while the degree of accumulation and biomagnification may also vary across species (Chen and Folt 2000; Oliveira et al. 2017), in addition to their trophic status within the food web, which strongly controls exposure and As uptake routes (McGeer et al. 2003; Schäfer et al. 2015) (Table 2).

In aquatic environments, small vertebrates (e.g. Pisces) play important role as an intermediate in energy transformation from lower trophic levels (e.g. phytoplankton, benthic microalgae, macroalgae and zooplankton) to higher trophic levels (e.g. large fishes, mammals and ultimately humans). Williams et al. (2014) reported higher concentration of As in small fishes compared to other aquatic flora and fauna. However, this may not be acceptable universally because varying concentration of different organoarsenical compounds have been reported from a variety of organisms (Schaeffer et al. 2006), thereby reflecting the complexity in overall comparison.

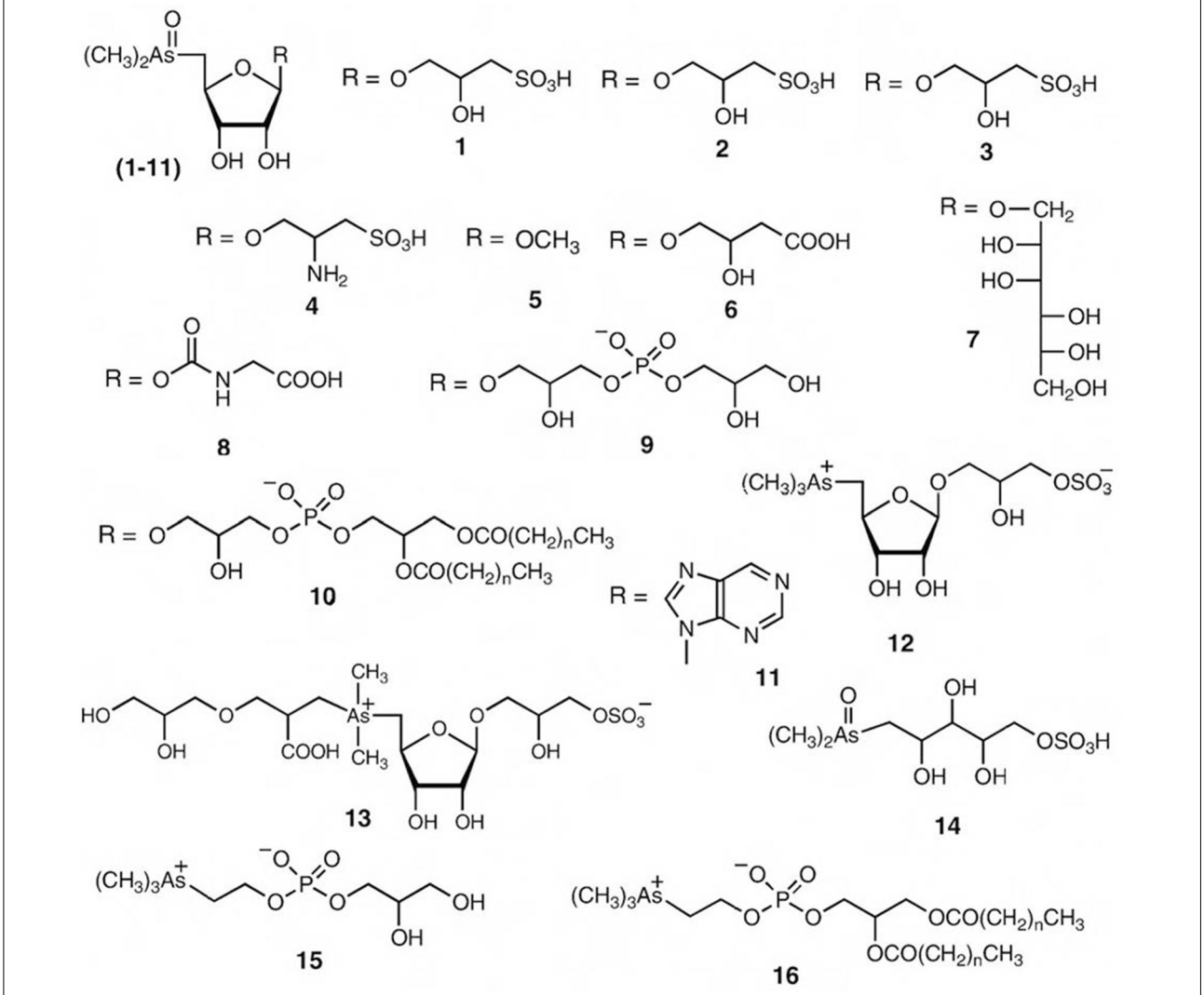

Fig. 1 Structures of arsenosugars, 1–16 correlated with Table 1 (modified from Sharma and Sohn 2009)

Table 2 Brief summary of various analytical methods for detecting various As species in different samples

Sample type/matrix	Species of arsenic	Sample pre-treatment (if any)	Separation and detection method
Blood	As(III), As(V), MMA, DMA, AB	Centrifugation/ dilution ($HgCl_2$) followed by ultrafiltration	LC-ICP-MS
Animal urine/ excreta		Dilution with deionized water and filtration	IC-ICP-MS HPLC-ICPMS HPLC-HG-ICPMS
Fish and oyster tissues		Lyophilization/microwave digestion	CE-ICP-MS
Fish		Ultrasonic extraction and 4 different experimental conditions	HG-AFS
Water Groundwater	As(III), As(V)	Dilution with deionized water and filtration	ICP-MS HPLC-ICPMS HPLC-HG-ICPMS
Sediment and fly ash Soil	Total As	Extraction with HNO_3, acetic acid, EDTA	HG-AAS

LC liquid chromatography, ICP-MS inductively couple mass spectrometry, IC ion chromatography, HPLC high-pressure liquid chromatography, CE capillary electrophoresis, HG-AFS hydride generation atomic fluorescence, HG-AAS hydride generation atomic absorption spectroscopy

Globally, numerous studies have looked into the available forms of As and their contamination in various environments (Smedley and Kinniburgh 2002; Bissen and Frimmel 2003; Borba et al. 2003; Ng 2005; Ghosh et al. 2015a, b; Farooq et al. 2019; review by Shaji et al. 2021) and also in organisms representing various habitats (Chen and Folt 2000; Jankong et al. 2007; Grotti et al. 2008; Taleshi et al. 2014; Williams et al. 2014). However, As contamination, effects, bioaccumulation and biomagnification are still not well established in terms of trophic levels. Based on this backdrop, the present review has two aims: (1) provide a brief overview of As distribution and contamination in different aquatic environments, and (2) discuss about the fate of arsenic at various trophic levels focusing on the marine environment.

Forms of arsenic in aquatic environment

As(III) and As(V) are major forms of As found in various types of aquatic environments and their fluxes as both inorganic [e.g. As(III) like $H_3AsO_3^0$, $H_2AsO_3^-$, $HAsO_3^{2-}$ and AsO_3^{3-}, and As(V) like $H_3AsO_4^0$, H_2AsO^{-4}, $HAsO_4^{2-}$ and AsO_4^{3-}] and organic forms have been found to be controlled by prevailing physicochemical parameters like pH, temperature and redox potential (Meng et al. 2000). Overall As fluxes across global aquatic environments have been reported to range from < 0.1 µg/l (unpolluted water bodies) to 5000 µg/l (areas of sulphide mineralization and mining) (Mandal and Suzuki 2002; Smedley and Kinniburgh 2002).

Groundwater

Arsenic contamination and their effects have been frequently noticed in groundwater aquifers across countries such as Canada, United States of America (USA), Mexico, Chile, Argentina, Spain, Italy, Hungary, Poland, Finland, India, Nepal, Bangladesh, Thailand, Burma, China, Vietnam, Cambodia, Taiwan, Japan, Australia and New Zealand (Valette-Silver et al. 1999; Argos et al. 2010; Shaji et al. 2021). There are three main sources of As contamination in groundwater: (1) As affected aquifers where geogenic As-bearing mineral dissolute into aquifer water; (2) geothermal water which contain high amount of As that seeps into aquifers and (3) mining-affected water which seep through and contaminate groundwater (Kossoff and Hudson-Edwards 2012; Morales-Simfor et al. 2020). The As concentration in groundwater vary globally due to various sources of arsenic. Higher As concentration (0–48 mg/l) has been recorded from Western USA because of prevailing geochemical environments (Welch et al. 1988), followed by India (0–23.08 mg/l) due to As rich sediment and pesticide production (Mandal et al. 1996; Ghosh et al. 2015a). Notably the Bengal Delta Plains (BDP) spanning across Eastern India and Bangladesh have arsenic-contaminated aquifers mainly due to geogenic activity (Smedley and Kinniburgh 2002) and also represent one of the worst affected regions globally (Mukherjee et al. 2009; Ghosh et al. 2014; Shrivastava et al. 2015; Ghosh et al. 2018). Moreover, 2% of the world's human population residing in the BDP region consume As-contaminated groundwater almost on a daily basis and this has become a serious public health issue over the last few decades (Ghosh et al. 2015a, b; McArthur 2019; Chikkanna et al. 2019).

River water

As contamination in river water are generally inadequate and reported concentrations are known to vary between 0.13 and 2.1 µg/l in many regions (Kossoff and Hudson-Edwards 2012). However, the range of As is found to be higher in rivers with inputs from volcanic eruptions, weathering and leachate of bedrocks, and other contaminants arising from anthropogenic activities. For example, European rivers such as Tinto (Portugal) and Rios Odiel (Spain) draining through the Iberian pyrite belt contain higher As loads (441 and 1975 µg/l, respectively) due to weathering and dissolution of As-bearing sphalerite, chalcopyrite, tetrahedrite and arsenopyrite (Kuehnelt 2006; Sarmiento et al. 2009). Similarly, in North America, the Gibbon River which drains through the Norris Geyser Basin of Yellowstone National Park, USA contain up to 160 µg/l of As (McCleskey et al. 2010) and stream/river water and sediment of Alaska showed As concentration ranging from 5 to 4000 mg/kg which is usually dominated by metamorphic rocks with mineralized regions (Wilson and Hawkins 1978). However, studies on elevated As level in many global rivers including from India are limited to a large extent. In recent years, the Central Water Commission under directives from the Government of India, have started to monitor As in Indian rivers. Reports suggest that As ranges from 0.01 to 9.47 µg/l, which falls within the permissible limit set WHO. Studies undertaken in the Ganga River Basin system have shown that elevated As level pose serious threat to public health (review by Chakraborti et al. 2018). Studies have also shown that riverbed–aquifer interface constitutes a hotspot for elevated As concentration and sustained release of the same is facilitated by the river muds rich in labile forms of organic matter as well as reactive iron oxides (Wallis et al. 2020).

Marine water

The concentration of As in open ocean is generally low (ranges from 0.003 to 1.8 µg/l) (Maher 1985; Cullen and Reimer 1989; Santosa et al. 1994) compared to coastal water (1 to 4.3 µg/l) (Santosa et al. 1996) or other coastal ecosystems (0.14 to 147 µg/l) including estuaries, lagoons

and backwaters (Peterson and Carpenter 1983; Martin et al. 1993; Abdullah et al. 1995; Smedley and Kinniburgh 2002). The As concentration in the Indian Ocean has been reported to be even lower (0.03 to 0.766; Morrison et al. 1997) compared to Pacific (1 to 1.8; Santosa et al. 1996), Atlantic (1 to 1.58; Santosa et al. 1994) and Antarctic Oceans (0.003 to 1.078; Middelburg et al. 1988). The lower concentration of As in the Indian Ocean region could be due to huge fresh water discharged from major rivers and high tidal influx from sea that provides large mixing zone in the Northern part of the Indian Ocean. Cullen and Reimer (1989) suggested that due to this saltwater–freshwater interface, co-precipitation zone of iron (Fe) and As occur which results in floccules formation made up of poorly crystalline Fe oxides as well as oxyhydroxide and precipitates (Anninou and Cave 2009). It has been also reported that inorganic As is ubiquitous in modern ocean and ranges from 15 to 20 nmol/L in the open ocean (Ellwood and Maher 2002; Cutter and Cutter 2006).

Forms of arsenic in aquatic organisms: metabolism and toxicity

Terrestrial organisms such as plants are well known for absorption of iAs from air, water and also from soil. The amount of As in a plant mainly depends on its exposure to surrounding environment contaminated with As. Other than that, most of the terrestrial organisms absorb As residues through consumption of plants and contaminated soils (Mandal and Suzuki 2002). Even though few studies reported the presence of As in both organic and inorganic forms in aquatic biological communities including bacteria, phytoplankton, small and large fishes and mammals, concentrations and biotransformation mechanisms are not well documented. Similar to terrestrial organisms, aquatic organisms can absorb varying concentrations of As from surrounding ecosystems. Aquatic organisms are exposed to many different forms of inorganic and organic arsenic species (arsenicals) commonly through food, water and other environmental sources. Owing to a large variety of physicochemical properties and bioavailability of each form of As, arsenical metabolism is a complex phenomenon. This is further complexed by the influence of other metals and metalloids on arsenic metabolism within and between the species (Mandal and Suzuki 2002).

Organic forms of As have been widely observed in aquatic flora and fauna; among them arsenobetaine (AsB) which is the most commonly reported organoarsenical is virtually absent in most of the freshwater invertebrates and vertebrates (Schaeffer et al. 2006). At the same time, an opposite trend has been reported in marine organisms because of major difference in As speciation (Taylor et al. 2017). For example, S-adenosyl methionine (SAM) acts as a donor of methyl group and glutathione acts as a cofactor in the formation of methylated forms of As like monomethylarsonic acid (MMA^V) and dimethylarsonic acid (DMA^V) (Table 1). Nevertheless, organic As compounds have been also reported from fish lipid extracts (Schäfer et al. 2015; Taylor et al. 2017) and these are mostly arsenolipids that have been observed in higher volume in edible fishes (Arroyo-Abad et al. 2010; Taleshi

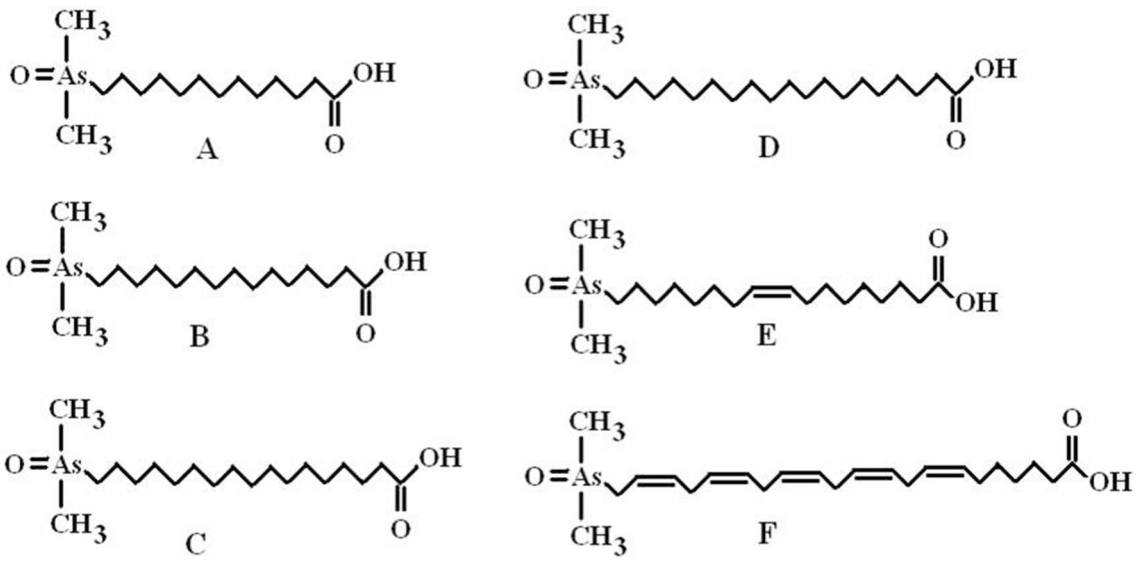

Fig. 2 Arsenolipids detected in fish cod oil. The molecular weight of A = 334, B = 362, C = 390, D = 418, E = 388, and F = 436 (adopted from Rumpler et al. 2008)

et al. 2010; Taylor 2017). In a recent study, considerable volume of non-phospholipid forms of As (Fig. 2 modified from Rumpler et al. 2008) has been reported which can be easily transported to upper trophic levels through food web including in humans (Amayo et al. 2011; Sele et al. 2012). Other than that, few of the arsenolipids have been characterized by gas chromatography–mass spectrometry (GC–MS) approach and their synthesis pathways have been elucidated by Taleshi et al. (2014). The ingestion pathway, including intake of food and water or through sediment exposure, controls the bioavailability of ingested inorganic arsenic. Besides, nutrients can also play a major role in iAs solubility. Arsenic distribution in aquatic organisms in terms of fate of ingested As in vivo depends on two factors: (1) oxidation and reduction reactions between iAs(V) and iAs(III) and (2) consecutive methylation reactions.

Bioaccumulation refers to the accumulation of any chemical component due to uptake by an organism from surrounding habitat (EPA 2003). It also depends on the type of organismal group (invertebrates, fishes), trophic status within the food chain and duration of exposure and routes of uptake (WHO 2001; Williams et al. 2006). In an aquatic environment, uptake of As by an organism can take place either directly (through ingestion, inhalation and absorption) or indirectly (through food web) (Moss 1998; Mandal and Suzuki 2002; Smedley and Kinniburgh 2002; Smith et al. 2002; Magellan et al. 2014). Smaller marine organisms such as bacteria, microalgae and macroalgae can take up dissolved arsenate from seawater via cellular phosphate transport system [e.g. As(V) is a structural analog of PO_4^{3-}]. Once inside the organism, the toxic As(V) form is converted into organic forms such as MMA and DMA. These organic As forms get biomagnified once they get accumulated in tissues of higher organisms (e.g. fish) in freshwater and marine environments and this could be possibly due to transformation or bioaccumulation from lower organisms through the food chain (Hellweger and Lall 2004; Rahman and Hasegawa 2012; Rahman et al. 2012; Hasegawa et al. 2019).

Arsenic bioaccumulation at lower trophic levels

Bacteria

Arsenic is toxic to most of the known bacterial groups, although some bacteria can metabolize as well as tolerate arsenic within its microenvironment. Members representing some of the bacterial phyla can use arsenic as an electron donor for autotrophic growth or as an electron acceptor for heterotrophic and anaerobic respiration (Oremland and Stolz 2003). Strains representing the bacterial genera *Agrobacterium* and *Rhizobium* utilize As(III) as a sole source of electrons (Páez-Espino et al. 2009) whereas members of Epsilonproteobacteria (e.g. *Sulfurospirillum arsenophilum* and *S. barnesii*) utilize arsenic as a respiratory oxidant by coupling along with oxidation of organic matter. Most aquatic bacterial groups metabolize As either through oxidation [As(III) to As(V)] and reduction [As(V) to As(III)], methylation [formation of monomethyl arsine (MMA) and dimethylarsenic acid (DMA)] and demethylation (Oremland and Stolz 2005). For example, strains of *Rhodopseudomonas palustris* have been reported to methylate arsenic forming mono-, di-, and/or tri-methyl derivatives such as trimethylarsine (Qin et al. 2006). Arsenic resistance in bacterial cell is mainly due to the presence of phosphate-specific transport systems or efficient efflux systems. Phosphate-specific transport system prevents the uptake of arsenic, whereas the plasmid or chromosomally encoded *ars* operon works to remove arsenic from cells. Of the three arsenate reductases reported in bacteria, ArsC has been widely studied in order to understand detoxification and resistance mechanism. It is a small molecular mass protein of 13–16 kDa which is located in the cytoplasm and plays a role by reducing As(V) to As(III). There are reports that in the bacterium *Rhodopseudomonas palustris* the expression of ars2 or ars3 operons showed an increase with increasing environmental As(III) concentrations (up to 1.0 mM) (Zhao et al. 2020). Macur et al. (2001) reported that *Caulobacter*-like, *Sphingomonas*-like and *Rhizobium*-like genera have metabolic ability to reduce arsenate rapidly. On the other hand, strains belonging to *Proteus*, *Escherichia*, *Flavobacterium*, *Corynebacterium* and *Pseudomonas* have been reported to transform As(V) into As(III) and other volatile methylarsines (Shariatpanahi et al. 1981). Thus the presence of highly efficient *ars* export system leads to low As levels in bacterial cells (review by Shi et al. 2020). There are also reports of parallel genetic pathways for organoarsenical detoxification by bacteria (review by Yang and Rosen 2016). Ghosh and Bhadury (2018) has shown that the stress from As can induce alteration of membrane phospholipid fatty acid (PLFA) in arsenite oxidizing bacterial members including *Hydrogenophaga bisanensis* strain BDP20 and *Acidovorax facilis* strain BDP24.

Algae

Algae are an important source of organoarsenic compounds in the marine environment. Organic forms of As are synthesized by algae and transferred through the food chain (Wrench et al. 1979). Micro- and macro-algae which form the basis of lower trophic levels may accumulate more As compared to members representing higher trophic levels (Garcı´a-Salgado et al. 2012).

In laboratory experiments it has been shown that euryhaline algae have ability to synthesize fat and

water-soluble arseno-organic compounds from iAs (Eisler 1988). Larsen et al. (1993) found that dissolved arsenate from seawater can be accumulated by marine micro- and macroalgae, and incorporated as arsenosugars (Fig. 1) after conversion into arsenobetaine (AsB) and arsenocholine (AsC). Other than MMA and DMA, several other As containing ribosides are also synthesized by marine algae (Francesconi et al. 1992), in addition to enzymatic methylation in the form of methylcobalamin and S-adenosylmethionine (Ridley et al. 1977). There are reports of the potential of algae to take up As(V) by more than one mechanism (Andreae et al. 1979). Several studies have also reported detoxification of As by algae and it is achieved by excreting out methylarsonic acid and DMA from inside the cell and DMA can also be reduced to dimethylarsinyl adenosine form in a reaction with adenosylmethionine. Dimethylarsinoriboses can be formed by glycosidation of this intermediate dimethylarsinyl adenosine which may reduce to trimethylarsonio ribosides. Other complex organoarsenical derivatives like arsenotaurine are also formed within marine algae by similar mechanism and intermediates (Ridley et al. 1977).

Microalgae such as *Chlorella* sp. and *Monoraphidium arcuatum* can uptake As(V) and reduce to As(III), while *M. arcuatum* excretes As(III) out of the cell (Levy et al. 2005). However it had been hypothesized that methylation of As(III) is not the sole detoxification method adopted by freshwater algae. Algal cells can also pump out As(III) from inside which can get oxidized to As(V) (Levy et al. 2005; Wang et al. 2013; Magellan et al. 2014). Other than that, algal cells can oxidize to form As(V). For example, macroalgae such as *Fucus spiralis* and *Ascophyllum nodosum* have been shown to accumulate four times more As(V) than As(III) from equivalent concentrations in seawater (Klumpp 1980a). Lai et al. (1998) reported that local climate conditions including seasonality can contribute to varying concentration of arsenosugars accumulation rate in the marine brown alga, *Fucus* sp. Similarly Francesconi and Edmonds (1993) reported that generally brown algae showed higher levels of total As (up to 230 µg/g dry weight) than red algae (up to 39 µg/g dry weight) and green algae (up to 23.3 µg/g dry weight) (Additional file 1: Table S1). In general it is known that detoxification of As by microalgae can be achieved through adsorption on cell surface (Wang et al. 2013; Dutta and Bhadury 2020; Zhang et al. 2020a, b), intracellular metabolism including As(III) oxidation and reduction of As(V), complexation with thiol compounds and sequestration into vacuoles (Olguı´n and Sa´nchez-Galva´n 2012), methylation and transformation into forms such as arsenosugars and arsenolipids and also by excretion (Levy et al. 2005; Jiang et al. 2011).

Zooplankton

Apart from micro- and macro-algae, drifting organisms such as phytoplankton (horizontal drifter) and zooplankton (vertical drifter) can also accumulate As. Aquatic primary production undertaken mainly by phytoplankton, which can uptake As(V) from the surrounding water and reduce it to As(III) (Sanders et al.1989) and can readily incorporate large quantities of As within their cellular components (Sanders and Cibilt 1985; Blanch and Wangberg 1988). Similarly, Froelich et al. (1985) reported that phytoplankton can also uptake and biomethylate As(III) followed by excreting outside the cell into the environment in forms such as MMA and DMA. These methylated arsenicals and organoarsenic compounds can be transferred to higher trophic levels through accumulation processes (Sanders 1985; Irgolic et al. 1977). A study undertaken by Šlejkovec et al. (2014) has shown that phytoplankton can also accumulate large quantities of iAs and/or can convert into other organic forms like arsenobetaine.

Similarly, traces of AsB have been reported in herbivorous zooplankton while other major As forms have been reported in carnivorous zooplankton (Šlejkovec et al. 2014) and these can ultimately get biomagnified through the marine food chain. Chen and Folt (2000) examined the accumulation and fate of As in different sized zooplankton and reported As bioaccumulation factor (BAF) in small zooplankton to be significantly higher (0.026 to 1.98 mg/kg) compared to larger zooplankton (0.022–0.598 mg/kg). Similarly, they reported a temporal increase in As concentration in zooplankton indicating potentially greater effect through food and their uptake (Chen and Folt 2000). In a very recent study, Hasegawa et al. (2019) showed that some cultured strains of freshwater phytoplankton can rapidly release inorganic and methyl forms of arsenic out of their cells during logarithmic growth phase while organic forms of As remain inside the cells. These findings have huge implications in terms of long-term bioaccumulation of forms of arsenic within zooplankton as part of freshwater trophic levels and beyond.

Arsenic bioaccumulation at higher trophic levels

Like lower trophic level organisms (e.g. microbes, algae, and phyto-zooplankton), higher trophic level organisms (e.g. fishes, crabs, prawns, and shrimps) play a vital role in As speciation within aquatic environments. Varying concentration of total As (As_{tot}) has been reported in various group of Pisces, while iAs levels were found to be uniformly low in most cases (Rahman and Hasegawa 2012). Accumulation of As_{tot} was mainly determined by various primary factors: (1) foraging habits, (2) metabolism and individual variation of As owing to their age, (3) regional

differences (farmed *vs.* wild caught), (4) differences in preparation and sample handling methods, and (5) differences in analytical methods in aquatic foods (Schoof and Yager 2007). Many studies revealed variations of As_{tot} in both marine and freshwater fishes which are important components of the aquatic food chain and directly consumed by higher tropic levels including humans (Schoof and Yager 2007; Rahman and Hasegawa 2012). Bioaccumulation of As in Pisces mainly depends on their habitat and feeding behaviour which may lead to resulting presence of inorganic, methylated and other organoarsenic compounds in their body (Rahman and Hasegawa 2012).

Bioaccumulation and biomagnification of As in Fin fishes

Arsenic can be oxidized or reduced or methylated and subsequently metabolized in all living tissues leading to recurrent bioaccumulation and biomagnification. However, biomagnification is seldom observed (Henry 2003). The As_{tot} concentrations are lower in freshwater fishes and typically have higher iAs:As_{tot} ratios as compared to anadromous, estuarine and marine fishes (EPRI 2003). Trimethyl arsenic have been detected at higher levels than that of dimethyl arsenic compounds in several freshwater fish species such as *Plecoglossus altivelis*, *Oncorhynchus masou*, *Rhinogobius* sp., *Sicyopterus japonicus, Phoxinus steindachneri* and *Abramis brama danubii*. These species are representatives of various global geographical locations (Kaise et al. 1997; Šlejkovec et al. 2004).

Many studies observed fish gills, skin and digestive tract as potential sites of absorption of water-soluble forms of As. Although skin may act as an important As absorbing site in small fishes due to their high surface area-to-volume ratio (Rahman and Hasegawa 2012; Magellan et al. 2014), As bioaccumulation and chemical speciation varies greatly in body tissues representing different species (Kar et al. 2011). The organoarsenical form (accumulation of AsB) has been widely reported in freshwater fishes (Jabeen and Javed 2011) and accumulation can vary in different organs (e.g. gills, liver, kidney, intestine, reproductive organs, skin, muscle, fins, scales, bones and adipose tissue) of different species (0.33 ± 0.01 to 1.42 ± 0.04 µg/g; Jabeen and Javed 2011). A diverse group of freshwater fish species such as salmonids (*Salmo marmoratus*, *Salmo trutta fario* and *Oncorhynchus mykiss*), common nase (*Chondrostoma nasus*), common barbel (*Barbus barbus*), Danube roach (*Rutilus pigus*), burbot (*Lota lota*) and Wels catfish (*Silurus glanis*) had been seen to accumulate higher As concentrations in muscle tissues compared to other organs (Šlejkovec et al. 2004).

Based on trophic status, bottom feeder fishes can be exposed to greater quantities of As in tissues due to their proximity to contaminated sediments whereas predatory fishes can bioaccumulate As either from surrounding water or from feeding on other fishes. It has been found that juvenile fish have higher concentration of As accumulation compared to adult forms (Schmitt and Brumbaugh 1990). A preliminary study conducted on bluegill fishes in 1960s showed the presence of arsenic residues within the entire body of 16-week-old immature fishes whereas, in mature fishes, arsenic residues were recovered only from their muscle tissues (Gilderhus 1966). Another such study conducted in late 1990s on mullet fishes showed similar concentrations of As in liver and muscle tissue of both juvenile and mature stages (Suner et al. 1999). To measure the difference in As bioaccumulation in different foraging fishes such as *Alosa pseudoharengus* (alewife), *Pomoxis nigromaculatus* (black crappie), *Lepomis macrochirus* (bluegill sunfish), *Kryptolebias marmoratus* (mangrove killifish) and *Perca flavescens* (Yellow perch) caught from Upper Mystic Lake, Massachusetts it was found that As burdens of all fishes were 30 to 100 times lower than its burdens in zooplankton. This further reinstates the position within the food chain to be a crucial factor regulating As bioaccumulation and biomagnification (Chen and Folt 2000; Jayaprakash et al., 2015). A recent study has also shown that bioaccumulation of As_{tot} in fishes were significantly correlated with As_{tot} level of pond water ($n = 10$; $R^2 = 0.80$; $p < 0.05$; Kar et al. 2011). Thus bioaccumulation of As is a well debated issue in freshwater fishes and alike mercury (Hg) it can get bioaccumulated, transported and biomagnified across higher trophic levels.

The magnitude of accumulation of any element depends widely on its physicochemical properties, fluxes/abundance and hydrophobicity. Inherent different of biological systems could result in high chemical concentrations of As in fin and shellfishes (several orders higher) than in the surrounding aquatic system. Thus, marine fishes which have limited ability to methylate arsenic, accumulate less As(V) from the surrounding in comparison to the fresh water fishes from lower trophic levels (Guven et al. 1999; Neff 2002).

Most of the marine fishes are planktivores and feeds directly phyto/zooplankton as a major food and thus can accumulate various forms of As through plankton (Peshut et al. 2008). On the contrary digestive tissues of estuarine mullets and luderick have shown to accumulate iAs in their tissues (Peshut et al. 2008). Other than iAs, marine fishes also accumulate organic As mainly in the form of arsenobetaine (AsB) which is a major water-soluble compound. It constitutes more than 95% of overall As compounds found in marine fishes (Kirby and Maher 2002). The biomagnification of arsenobetaine has been detected at different trophic positions of marine fishes such as planktivores, herbivores, detritivores and carnivores.

However, studies have shown pelagic carnivores to accumulate higher amount of AsB owing to their larger sizes (Kirby and Maher 2002). However, other organic arsenicals such as arsenocholine, tetramethylarsonium ion and arsenosugars have been also found in very low concentrations in marine fishes (Molin et al. 2012; Rahman et al. 2012). Moreover, varying concentrations of As_{tot} have been detected in different tissues of marine fishes irrespective of their trophic position (Langston 1984).

Bioaccumulation and biomagnification of As in shellfish

The amount of As accumulation and biomagnification in shellfishes can also vary based on their feeding preferences as well as on habitats. Arsenic speciation and their amounts fluctuate among marine shellfishes and also with respect to prevailing environmental factors such as temperature, salinity, pH and Eh. Shellfishes accumulate both organic and inorganic forms of As (Taylor et al. 2017), while ca. 90% are organic forms and found in edible portions. Similar to other aquatic finfishes, shellfishes also convert iAs to organic As like arsenobetaine (AsB) and arsenocholine (AsC) and dimethylarsinic acid (DMA), and bioaccumulate in their body parts (Lawrence et al. 1986). Among the shellfishes, crustaceans accumulate higher As_{tot} (170 mg/kg) (Calabrese et al. 1985) compared to others (Penrose et al. 1977). For example, common littoral crab (*Carcinus maenas*) accumulates both organic and iAs through ingestion; however, they cannot inter-convert these forms in their tissues and rapidly excrete out the inorganic forms compared to organic forms (Andersen and Depledge 1993). Thus, shellfishes have limited ability to accumulate As and there is strong support towards the fact that these levels have increased in the current era (Rodney et al. 2007).

Similarly in marine bivalves, AsB is the prevalent form absorbed in their body muscles; it can come directly via ingestion of phyto-zooplankton, detritus and sediment particles while carbonate shells accumulate from surrounding water column (Shibata and Morita 1992). On the other hand, freshwater clams are known to accumulate higher tetramethylarsonium ion (Shiomi et al. 1987). Lai et al. (1998) reported higher levels of As in the form of arsenosugars in scallops particularly muscles and gonads whereas Rodney et al. (2007) studied As accumulation in an estuarine oyster species (*Crassostrea virginica*) and showed increasing concentration in muscle tissues with bio-concentration factor (BCF). Nonetheless, depth-wise distribution of oyster shell exhibit marked variation in organic As bioaccumulation (top: 0.213 ± 0.037 µg/g, bottom: 0.092 ± 0.018 µg/g) whereas iAs accumulation is not distinctly detected (Sanders et al. 1989). The higher concentration of As in oyster shell can be found based on studied geographical regions such as the West coast of Florida, USA which receives more phosphates from mineral deposits. Other studies involving deposit-feeding bivalve *Scrobicularia plana* and filter-feeding bivalves *Cerastoderma edule* and *Mytilus edulis* are shown to accumulate As from sediment particles through ingestion whereas grazing gastropod such as *Littorina* sp. which fed on various macroalgae can also accumulate As (Klumpp 1980b). Similarly, As in soft tissues of the bivalve *Modiolus capax* (commonly known as fathorse mussel) has been detected and ranges from 6.62 to 44.7 µg/g of dry weight (Gutierrez-Galindo et al. 1994). In bivalves such as the blue mussels (*Mytilus edulis*), high concentrations of iAs ranging between 0.001 and 4.5 mg As/kg has been reported (Sloth and Julshamn 2008). However, As speciation and accumulation and magnification rates are not very well documented in most of the edible shellfishes including shrimps, prawns and molluscs.

Effect of arsenic on aquatic biota

Aquatic environments are exposed to arsenic through atmospheric deposition of combustion products and run-off from fly-ash storage areas. Arsenical compounds have been detected in aquatic biota and can go up to 48 µg/l in water, 120 mg/kg in diets and 5 mg/kg (fresh weight) in tissues (Eisler 1988). Effects of arsenicals in aquatic organisms including toxicity are significantly modified by number of biotic and abiotic factors.

Freshwater biota

Only few studies have been undertaken to investigate effect of As on freshwater algae. Thegrowth rate experiments conducted on three different freshwater algal species namely *Ankistrodesmus falcatus*, *Scenedesmus obliquus* and *Selenastrum capricornutum* indicated a decrease in growth at 0.075 mg/l of As(V) concentration (EPA 1985). Whereas, freshwater invertebrates species like *Bosmina longirostris, Daphnia magna, Daphnia pulex* and *Simocephalus serrulatus* when treated with As(III) concentration of 1.5 mg/l for 96 h resulted in 50% immobilization and followed by three weeks exhibited impaired reproduction (Lima et al. 1984; Passino and Novak 1984). Spehar et al. (1980) conducted 28 days (LC-10) experiment using As(III) and As(V) on *Helisoma campanulata* and showed that As(III) had more lethal effect than As(V). In another study on *Pteronarcys californica* and *Pteronarcys dorsata* showed a LC-50 of 96 h (3.8 mg/kg) and 28 h (0.96 mg/kg) of As concentration ultimately resulting in mortality (Johnson and Finley 1980). But, it should be noted that LC-50 values are markedly affected by surrounding water temperature, pH, Eh, organic load and phosphate concentrations, suspended solids and presence of other substances and

toxicants (Kumari et al., 2017). Besides, As speciation and duration of exposure can also affect these values (Eisler 1988).

Among freshwater vertebrates, marbled salamander (*Ambystoma opacum*) and eastern narrow-mouthed toad (*Gastrophryne carolinensis*) showed death or malformations in developing embryos when exposed to As at a concentration of 0.04 mg/l. The juvenile of bluegill fish (*Lepomis macrochirus*) showed reduced survival rate and histopathological changes when treated with 0.69 mg/kg of As for 16 weeks (EPA 1985) while goldfish (*Carassius auratus*) shows 15% behaviour impairment in 24 h and nearly two fold in 48 h when exposed to As. In another study, the flagfish (*Jordanella floridae*) showed mortality when exposed to As with and without food resulting in 96 h LC_{50} estimate of 14,400 µg/l (Lima et al. 1984). Another experiment carried out using 96 h exposure to As (based on LD_{50} values) in spottail shiner (*Notropis hudsonius*), chum salmon (*Oncorhynchus keta*), common minnow (*Phoxinus phoxinus*) and fathead minnow (*Pimephales promelas*) showed fins were more affected than edible muscle tissues (Lima et al. 1984; EPA 1985). Similarly, the embryo and adults of rainbow trout (*Oncorhynchus mykiss*) showed a depression in growth with avoidance of food and impaired feeding efficiency at different concentrations of As (Johnson and Finley 1980; Cockell and Hilton 1985).

Marine biota

Marine bacteria and algae are thought to preferentially utilize As(III) form of arsenic in their environment (Johnson 1972; Johnson and Burke 1978). It is also believed that As(V) form has more profound effect on growth and morphology of marine algae compared to As(III). An experiment involving incubation of marine algae in media containing various concentrations of As(V) and As(III) have shown arsenic incorporation and release from algal cells thereby indicating that the differences between uptake and release rate have hinted towards chemical changes of As(V) upon incorporation inside algal cells (Bottino et al. 1978). There are numerous reports of changes at the morphological and physiological levels in marine algae due to exposure from arsenic. For example, reduced sexual reproduction and sporulation have been reported in red algae (*Champia parvula* and *Plumaria elegans*) following exposure up to 0.6 mg/l of As (Thursby and Steele 1984; Sanders 1986). Similarly, inhibition to growth, reduction of Chlorophyll-*a,* and overall biomass reduction has been observed in *Skeletonema costatum* and *Thalassiosira aestivalis* when exposed to arsenic (EPA 1985).

Among marine invertebrates, amphipods (*Gammarus pseudolimnaeus*) showed 50% immobilization in 96 h at 0.87 mg/l of As in water (Lima et al. 1984). Ricevuto et al. (2016) conducted an *in-situ* experiment (30 days) on polychaete (*Sabella spallanzanii*) near the CO_2 vent and reported elevated concentration of arsenic in their gills, particularly dimethylarsinic acid (DMA), stored as an anti-predatory compound. This study also concluded low pH and high pCO_2 may have detrimental effects on arsenic metabolism and oxidative status of this polychaete species provide an insight to understanding of species-specific vulnerability to ocean acidification. Similarly, very low As concentration were found in the copepods (e.g. *Acartia clausi*) and Dungeness crabs (*Cancer magister*) when exposed to As treated water (LD_{50} at 96 h) (EPA 1985) while reduced survival rate has been observed in juvenile and adult stages of calanoid copepods (*Eurytemora affinis*) when exposed to arsenic (arsenate form-100 µg/l) (Sanders 1986). The gastropod shells (*Nassarius obsoletus*) showed decreased consumption of food whereas bivalve shells (*Mytilus edulis*) showed mortality within 3 to 16 days of As treated water (NAS 1977). In higher tropic level, fishes like thicklip grey mullet (*Chelon labrosus*) and common dab (*Limanda limanda*) reported discoloration of skin and respiratory problems, respectively, in environments with high As concentration (Taylor et al. 1985; Scott and Sloman 2004).

Biomagnification in aquatic ecosystems and trophic levels

The effects of As on aquatic ecosystems and beyond including on human health are influenced by the nature of persistence as well as ability to increase toxicity potential from lower to higher trophic levels (e.g. microbes to fish to humans). This process is known as biomagnification. As discussed, forms of arsenic can disrupt the growth of various groups of organisms in aquatic ecosystems including plankton, molluscs and crustaceans as well as can adversely affect photosynthetic process with cascading impacts on productivity of these ecosystems (Newman 2015; review by Córdoba-Tovar et al. 2022). The adverse effects of As biomagnification and consequences such as decreased reproductive capacity, changes in embryo viability, teratogenesis as well as changes in enzymatic processes have been documented in many higher trophic levels inhabiting aquatic ecosystems (Zheng et al. 2019). Many studies have highlighted that biomagnification in aquatic ecosystems represents a combination of several factors including environmental, ecological and biological factors (Zhang et al. 2012; Huang 2016). It seems that bioaccumulation and biomagnification of As in aquatic ecosystems are not very consistent when demonstrating increase in concentrations of forms of arsenic such as from algae to zooplankton or from zooplankton to fish (Majer et al. 2014; Kato et al.

2020). Studies have shown that the concentration level of forms of As in freshwater systems is dependent on the rate of biodilution compared to marine systems where it is more linked to enrichment of organic form (arsenobetaine). As a result, there is degradation of food with high arseno-sugar contents (Caumette et al. 2012; Huang 2016). There are reports that have shown biomagnification of forms of As can be favoured by a combination of factors including benthic habitat and environmental factors in marine ecosystems (Du et al. 2021). In general, due to the effect of biodilution of As along food webs, negative effects may be prominent in organismal groups representing low trophic levels in aquatic ecosystems (Trevizani et al. 2018). Studies have also shown a temporal trend of biomagnification in higher trophic levels across aquatic ecosystems with higher concentrations reported in benthic fish during winter while pelagic fish had lower concentration in summer (Du et al. 2021). There are reports of a rise in lipid-soluble As concentrations in pelagic organismal groups from marine environment with concentrations higher in crustaceans compared to bottom dwelling fish (Hayase et al. 2010; Córdoba-Tovar et al. 2022). Some studies also indicate that biomagnification of As in higher trophic levels can be linked to age and accumulation tends to be species-specific (Agusa et al. 2008). It has been also observed that biomagnification of As can vary with respect to type and complexity of food web (Dovick et al. 2015; Huang 2016). Based on a number of studies it has been also proposed that retention capacity, assimilation, metabolization and exposure time can be crucial factors that can strongly influence biomagnification of forms of As across trophic levels and linked feeding habitats (Maher et al. 2011). While numerous studies have shown some indication of the steps of biomagnification of As in aquatic ecosystems; however, answers pertaining to trophodynamics and link with biomagnification are yet to be fully resolved.

Arsenic metabolism and toxicity in humans

Food is considered to be the primary source of As intake in human besides exposure from occupation or from drinking water. In humans, As(V) is rapidly reduced to As(III) and partly methylated in vivo. DMA is an important metabolite reported in most of the animals whereas 20% inorganic arsenic, 20% MMA and 60% DMA have been found in human urine under normal conditions (Mandal and Suzuki 2002). Under in vivo condition this iAs gets methylated to MMA and DMA and absorbed MMA is further methylated to DMA and ultimately excreted mainly in unchanged forms (Buchet et al. 1981, 1996a) while AsB is absorbed and excreted as unchanged form (Brown et al. 1990). Mandal et al. (2001) found that MMAV can be reduced to their trivalent analogues, monomethylarsonous acid (MMAIII) and DMAV as dimethylarsinous acid (DMAIII) based on the detection in human urine.

An experimental observation showed 33% of As excreted in urine within 48 h and remaining 45% within 96 h when using 500 g of As(III) and similarly radioactively labelled (^{74}As) showed 38% excretion in 48 h and 58% in 120 h (Tam et al. 1979; Buchet et al. 1981). However, As excretion can also happen through other routes (e.g. sweating) and it can also accumulate in keratin-containing tissues (e.g. skin, hair and nails) and mother's milk whilst these latter routes of excretion are not that frequent (Grandjean et al. 1995; WHO 2001). Although these keratin-containing tissues are used as indicators for identifying As exposure to humans but blood is also used to detect recent As poisoning or in terms of chronic stable exposure (Ellenhorn 1997). Other than that, consumption of aquatic products particularly seafood (e.g. shrimp, marine fishes, other crustaceans, bivalves and seaweeds) and other products (e.g. freshwater fishes, prawns and clams) by human is increasing day-by-day which can ultimately lead to As toxicity provided that the source of food have significantly higher amount of As metabolites (e.g. MMA, DMA and arsenosugars; Kumari et al., 2017). Therefore, As metabolites (iAs + MMA + DMA) are used as an index to elucidate As_{tot} exposure in human urine to correctly estimate iAs exposure in human population (WHO 2001). However, the As metabolites index is not well recognized globally. In particular, across parts of South and South-east Asia there is a need to integrate and rapidly use As metabolites index so as to understand the scale and magnitude of As bioaccumulation in human populations. This is particularly relevant given changing groundwater scenarios in many parts of Asia induced by anthropogenic climate change (Shah 2019) which can increase exposure to As toxicity across local, regional and transboundary scales (e.g. India and Bangladesh). There is an urgent need to develop cost-effective methods of estimation iAs exposure in order to effectively reach out to many of the developing and least-developing countries facing As exposure issues across pan continents. The Figure 3 provides a representation of translocation of arsenic from lower to higher trophic levels.

Conclusions and way forward

Arsenic poses serious health risk as it enters the human body through drinking water and contaminated food sources such as rice grains. However, the entry of arsenic in human body through consumption of freshwater and marine fish or shellfish has not been thoroughly investigated across large geographical scales, in particular from many countries which are reeling from

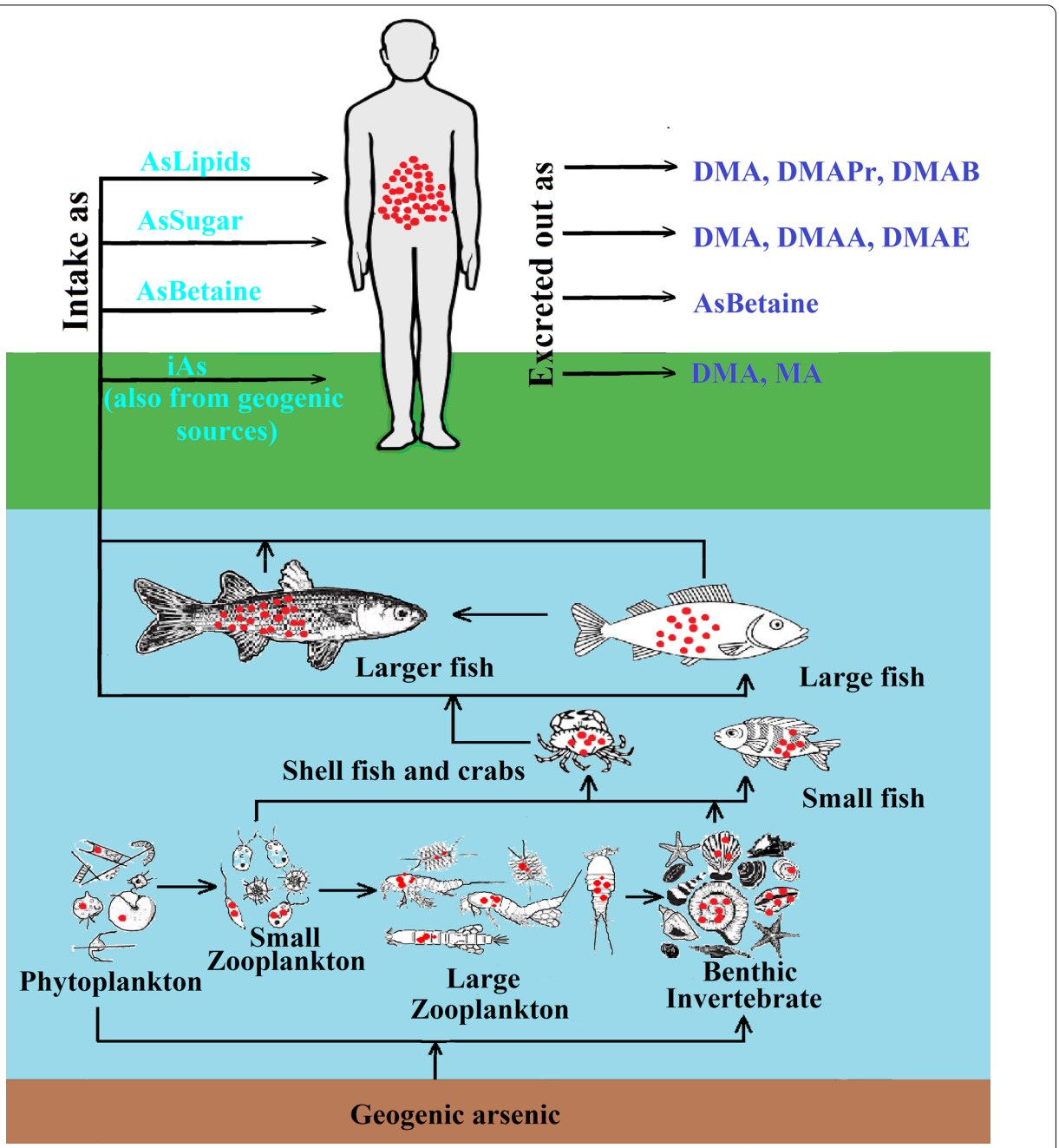

Fig. 3 Schematic representation of translocation of arsenic from lower to higher trophic levels (the red dots represent arsenic bioaccumulation and biomagnifications; see Additional file 1: Table S1 for more details; partly adopted from Taylor et al. 2017)

geogenic arsenic issues. A large number of investigations have established the presence of high concentration of As in aquatic environments including aquatic biota through routes of bioaccumulation. Therefore, As bioaccumulation in inorganic forms or organic forms such as momomethylarsenic acid (MMA) and dimethylarsenic acid (DMA) and arsenobetaine (AsB) can result in biomagnification at higher trophic level (including human) and ultimately lead to serious public health issues. However, only a limited number of investigations have looked into As bioaccumulation and their contamination on fresh and marine water including associated biodiversity which warrants further scientific investigations. This review provides a

much-needed understanding of As bioaccumulation and biomagnification through various trophic levels including fin fishes and shellfishes which are consumed by humans and are known to be potential health hazard.

In the era of 'omics' there is a need to integrate genomics, proteomics and metabolomics approaches in order to better understand the effect of bioaccumulation and biomagnification particularly in aquatic biota by investigating influx and efflux of inorganic arsenic species. For example, it is now becoming increasingly clearer that As(III) can enter living system through the involvement of other transport systems such as aquaporins (Thomas 2007). It is essential to characterize the genes that code for aquaporins in different organismal groups including in fishes. Other pathways such as the hexose permease transporter (HXT) can modulate uptake of As (III) with a higher efficiency compared to aquaporins. However, the distribution patterns of HXT needs to be more thoroughly investigated using 'omics' based approach in different organisms which have shown tendency to bioaccumulate As. Similarly, mitogen-activated protein kinases (MAPKs) are important regulatory proteins through which extracellular signals are transduced into intracellular events. It seems that arsenic can affect the MAPK pathway and therefore this pathway has to be thoroughly investigated in higher trophic levels.

Given the ability of microbiome in human gut to biochemically transform arsenicals (Coryell et al. 2019), there requires an international effort to elucidate the responses of human gut microbiome to As toxicity spanning geographical scales and representing diversity of populations. Such effort can be crucial towards linking As induced toxicity and manifestations in terms of other pathological ailments including cancer. The availability of third and fourth generation sequencing technologies (e.g. Illumina and Oxford Nanopore) (review by Bhadury and Ghosh 2022) along with robust computational biology tools provide the right time and opportunity to undertake human gut microbiome studies involving As affected populations of South Asia and beyond. The possibility of initiating long-term monitoring programmes of deep and shallow aquifers in countries such as India and Bangladesh through pan collaborative network can be immensely helpful towards understanding As bioaccumulation in the era of 'Anthropocene'. Overall, in future, understanding the consequences of bioaccumulation particularly in aquatic biota using 'omics' tools can ultimately pave the way for As metalloid-free environment and food resources.

Supplementary Information

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Additional file 1: Table S1. Arsenic concentration observed in various organismal groups.

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