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Swedish Agency for Marine and Water Management

ARSENIC IN SEDIMENT

AN ENVIRONMENTAL QUALITY STANDARD OVERVIEW

Spring 2022

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This report was initiated and funded by the Swedish Agency for Marine and Water Management (SWaM).

Contents

Introduction

Arsenic exists in four valency states: -3, 0, +3 and +5 where the trivalent state, e.g. arsenite, dominates in reducing conditions and the pentavalent state, e.g. arsenate, is the most common state in aerobic conditions in both water and sediment (WHO, 2001). Arsenic can adsorb to clays, iron and aluminium hydroxides and organic material (WHO (2001) and references within) and settle in the sediment. The dominant accumulation mechanism of arsenic in oxygenated sediment is the co-precipitation of arsenic to hydrous iron oxides (Nicholas et al., 2003). Thus, in a shift to anoxic conditions where iron is reduced, the complexed arsenic, in the form of arsenate hydrous oxides, might be released to the water column in a similar matter as phosphates and further reduced to arsenite (Moore et al. (1988); WHO (2001) and references within). Inorganic arsenic is more toxic than organoarsenicals, i.e. arsenic bound to organic ligands (WHO, 2001).

Marine organism generally contains higher levels of arsenic than terrestrial organisms. This is partly attributed to the high arsenate-to-phosphate ratio where the ocean generally has very low phosphate concentrations (WHO, 2001). This was also confirmed by Van Ael et al. (2017) who investigated metal accumulation in biota and sediment along the Scheldt estuary, finding increased concentration of arsenic in shrimp muscle tissue and polychaeta moving from inland water towards the North Sea. This, despite the geographical decreasing trend of arsenic content in the sediment moving towards the more marine domain (Van Ael et al., 2017).

Arsenic can be responsible for both sublethal and acute toxicities towards benthic organisms (Martinez et al., 2006; Mahamoud Ahmed et al., 2018). Because of its chemical similarity to sulphur and phosphate, arsenic can induce toxic effects by replacing these elements in metabolic processes (WHO, 2001; Martinez et al., 2006 and references within) such as replacing phosphate in the energy carrier ATP (Mahamoud Ahmed et al., 2018). Also, acute toxicity effects on the photosynthetic efficiency of algae has been observed (Tuulaikhuu et al., 2015).

Major industrial processes that contribute to the arsenic input to the environment are mining, smelting of non-ferrous metals and burning of fossil fuels (WHO, 2001). What is less know is how arsenic affect marine and freshwater organisms in general and benthic communities in particular. However, in the European Union (EU), the Water Framework Directive (WFD, 2013) require member states to derive so called Environmental Quality Standards (EQS) for specific pollutants that are discharged in significant quantities into a water body, so-called river basin specific pollutants (RBSP). As of today, there is no Swedish environmental quality standard derived for arsenic in freshwater or marine sediment.

The derivation of EQS values shall be performed according to the protocol described in the Technical Guidance Document No. 27 (European Commission, 2018), hereafter TGD 27. The first version of TGD 27 was published in 2011 and an updated version was issued in 2018. Briefly, TGD 27 focus on the steps involved for deriving an EQS, e.g. types and quality of data required, extrapolation and choice of assessment factors and how to account for background concentrations and bioavailability. The TGD 27 does not consider the implementation phase (e.g. design of monitoring programs, sampling strategies and chemical analysis etc), this is instead described in more detail in the Technical Guidance for implementing Environmental Quality Standards (EQS) for metals No 38, hereafter TGD 38 (European Commission, 2019).

Despite the guidance in TGD 27, there is a huge variation in national EQS-values between EU member states, which for arsenic range from 3 to 400 µg/L in surface water, i.e. over three orders of magnitude (Irmer et al., 2014). No compilation has, to the best of the authors knowledge, been performed for arsenic EQS values in the sediment compartment.

The overall aim of the proposed report was to develop an Environmental Quality Standard overview for arsenic in marine and freshwater sediments, including to propose threshold values of arsenic in these two compartments. In addition, the proposed threshold values were compared to measured arsenic concentrations from Swedish monitoring data in freshwater and marine sediments.

Method selection and considerations

The derivation of a new EQS for arsenic in sediment was based on the guidelines published in the TGD 27 (European Commission, 2018). As the guidance document primarily focuses on the water column, expert judgement and previous experience from the derivation of an EQS for copper in marine sediment served as additional complement (see Lagerström et al. (2021)). An extensive literature study, to collect ecotoxicological data and provide supporting information, was conducted prior to the analysis (see section Data collection and selection below).

As suggested by the TGD 27 and supported by experts (workshop described in Lagerström et al. (2021)), the background concentrations of RBSP should be assessed during the implementation of the EQS rather than during the derivation process. In one of the tiered approaches, according to the TGD 38, it has been suggested that, as a second tier, the EQS should be recalculated with the added risk approach. However, as the TGD 38 also suggest deriving and implementing one specific EQS value and that this value should be of total concentration for tier 1 evaluations, the added approach might be used in the implementation phase. However, the implementation phase is out of the scope of this overview. Also, the organisms inhabiting the sediment have no ability to distinguish between arsenic of natural and anthropogenic origin, making the added risk approach ecologically inappropriate. Therefore, the total risk approach is the preferred approach and will be used to derive an EQS for arsenic in sediment.

Bioavailability

The bioavailability of arsenic in sediment is determined by several different factors, both due to arsenic's strong affinity of particles and the presence of other ligands such as organic compounds and iron oxides (WHO, 2001). Also, in environments where phosphate concentrations are high, the arsenate toxicity is generally reduced (WHO, 2001).

Due to the lack of detailed data, no corrections for bioavailability will be conducted within the scope of this overview.

Data collection and selection

There are only a few chronic ecotoxicological studies conducted on benthic species exposed to arsenic in the sediment (Table 1). In total, four species are represented by chronic studies; *Hyalella Azteca (crustacean)*, *Tubifex tubifex ("sludge worm")*, *Branchiura sowerbyi* ("red worm") and *Chironomus dilutus* (also known as *C. tentans; a midge larvae*). These four species represent different feeding and living conditions in the benthic environment but none of the chronic ecotoxicological studies represented marine organisms or marine test conditions.

All data that was included in the final assessment have been analysed with respect to reliability and relevance (Table 1), according to the CRED model (Moermond et al., 2016).

Table 1: An overview of the collected studies included in the derivation of an EQS for arsenic in marine and freshwater sediments. Reliability and relevance explanation (Moermond et al., 2016): C1 Relevant without restrictions: The study is relevant for the purpose for which it is evaluated. C2 Relevant with restrictions: The study has limited relevance for the purpose for which it is evaluated. R2 Reliable with restrictions: The study is generally well designed and performed, but some minor flaws in the documentation or setup may be present. R3 Not reliable: Not all critical reliability criteria for this study are fulfilled. The study has clear flaws in study design and/or how it was performed

** Chironomus dilutus is also known as Chironomus tentans*

Chronic studies						
Species	**End-point**	**Concentration (mg/kg dw)**	**Reference**	**SW/FW**	**Equilibration time after spiking**	**Relevance/reliability**
Hyalella azteca (crustacean)	Mortality (LC20)	45	Goulet and Thompson (2018)	FW	8 weeks	C1/R2
Hyalella azteca (crustacean)	Growth (EC20)	239	Goulet and Thompson (2018)	FW	8 weeks	C1/R2
Tubifex tubifex (annelida)	Mortality (LC10)	116	Lobo et al. (2016)	FW	1 week	C2/R2
Tubifex tubifex (annelida)	Autonomy (EC10) (swollen anterior body parts)	81	Lobo et al. (2016)	FW	1 week	C2/R2
Tubifex tubifex (annelida)	Mortality (LC10)	163.58	Lobo et al. (2021)	FW	9 days	C1/R2
Tubifex tubifex (annelida)	Reproduction (EC10)	96.1	Lobo et al. (2021)	FW	9 days	C1/R2
Branchiura sowerbyi (oligochaete)	Mortality (LC10)	86.54	Lobo et al. (2021)	FW	9 days	C1/R2
***Branchiura sowerbyi** (oligochaete)*	**Growth (EC10)**	**22.13**	**Lobo et al. (2021)**	**FW**	9 days	C1/R2
Chironomus dilutus (arthropoda)*	Development (NOEC)	30	Martinez et al. (2006)	FW	5-7 days	C2/R2
Acute studies/end-points						
Species	**End-point**	**Concentration (mg/kg dw)**	**Reference**	**SW/FW**	**Equilibration time after spiking**	**Relevance/reliability**
Americamysis bahia (crustacean - mysid)	Mortality (LC50)	88.8	Burgess et al. (2007)	SW	>96 h	C2/R3. Testing of resins (to fix As to sediment). Only nominal conc.
Ampelisca abdita	Mortality (LC50)	80.9	Burgess et al. (2007)	SW	>96 h	C2/R3. Testing of resins (to fix As to

(crustacean - amphipod)						sediment). Only nominal conc.
Hyalella azteca (crustacean)	Mortality (LC50)	134	Goulet and Thompson (2018)	FW	8 weeks	C1/R2. Juvenile.
Hyalella azteca (crustacean)	Mortality (LC50)	134	Goulet and Thompson (2018)	FW	8 weeks	C1/R2. Adult.
Hyalella azteca (crustacean)	Mortality (NOEC)	462	Liber et al. (2011)	FW	20 days	C1/R2
Hyalella azteca (crustacean)	Mortality (LC50)	532	Liber et al. (2011)	FW	20 days	C1/R2
Hyalella azteca (crustacean)	Growth (NOEC)	462	Liber et al. (2011)	FW	20 days	C1/R2
Chironomus dilutus (arthropoda)*	Mortality (NOEC)	39	Liber et al. (2011)	FW	20 days	C1/R2
Chironomus dilutus (arthropoda)*	Mortality (LC50)	642	Liber et al. (2011)	FW	20 days	C1/R2
Chironomus dilutus (arthropoda)*	Growth (NOEC)	<39	Liber et al. (2011)	FW	20 days	C1/R2
Tubifex tubifex (annelida)	Mortality (LC50)	251	Lobo et al. (2016)	FW	1 week	C2/R2
Tubifex tubifex (annelida)	Autonomy (EC50)	210	Lobo et al. (2016)	FW	1 week	C2/R2
Tubifex tubifex (annelida)	Mortality (LC50)	189.15	Lobo et al. (2021)	FW	9 days	C1/R2. Chronic study (28d), acute end-point.
Tubifex tubifex (annelida)	Reproduction (EC50)	254.52	Lobo et al. (2021)	FW	9 days	C1/R2. Chronic study (28d), acute end-point.
Branchiura sowerbyi (oligochaete)	Mortality (LC50)	102.87	Lobo et al. (2021)	FW	9 days	C1/R2. Chronic study (28d), acute end-point.
Branchiura sowerbyi *(oligochaete)*	**Growth (EC50)**	**36.61**	**Lobo et al. (2021)**	**FW**	9 days	C1/R2. **Chronic study (28d), acute end-point.**
Corophium volutator (crustacean)	Mortality (LC50)	45-52	Moriarty et al. (2014)	SW	No spiking but mixing, unclear of eq. time	C2/R2. Sediment not spiked but mixed sediment from unpolluted area (5.4 mg/kg dw) with high As area (628 mg/kg dw) to create exposure levels.

There were no chronic studies on marine species, but LC50 values derived from acute studies on three marine species (all crustaceans; *Americamysis bahia*, *Ampelisca abdita* and *Corophium volutato)* showed that the end-point concentrations of these are found at the lower end of the determined effect concentrations of both marine and freshwater species (Figure 1).

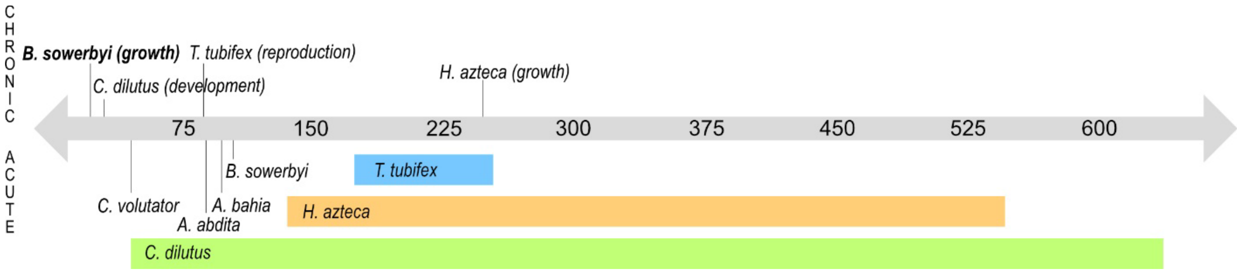

Figure 1: The range of the acute effect concentrations (mortality endpoint) compared to chronic (above arrow) end-points indicated in brackets from ecotoxicological studies of arsenic toxicity for the different species in table 1.

Selected approach for the derivation of environmental quality standard

Due to the scarcity of data, a deterministic approach was applied when deriving an EQS for arsenic in marine and freshwater sediment. The lowest NOEC/EC10, from chronic studies, thus represent the critical value which is then divided by an assessment factor to get the EQS.

The critical values, derived from the lowest NOEC/EC10 value for marine and freshwater tests respectively, was used for the EQS setting. In accordance with the TGD 27, the number of tests suggests an assessment factor of 10 for freshwater sediments (*Three long term tests (NOEC or EC10) with species representing different living and feeding conditions* in table 11 in TGD 27) and an assessment factor of 50 for marine sediments (*Three long term sediment tests with species representing different living and feeding conditions in table 13 in TGD 27)*.

The proposed EQS for arsenic in freshwater and marine sediments are:

$EQS_{sediment,fw}$: 2.2 mg/kg dw

$EQS_{sediment,sw}$: 0.4 mg/kg dw

Proposed environmental quality standard

Analysis

The lowest chronic effect concentration (EC10) of *B. sowerbyi ("red worm")* was based on the growth end-point and this was the lowest effect concentration within the entire dataset (EC10=22.13 mg/kg dw). This was used as the critical value for the deterministic approach of deriving an EQS for sediment in marine and freshwater sediments. As there were no marine long-term studies, the same critical value was used for both matrices.

From the study by Moriarty et al. (2014), where field sediments were mixed into different concentrations, it is shown that a significant mortality of the marine crustacean species *Corophium volutator* (>40%) occurs at concentrations just below 20 mg/kg dw. In a 10-day bioassay, the LC50 value of *C. volutator* was determined to be between 45-52 mg As/kg dw. Mamindy-Pajany et al. (2013) estimated that the EC50 of arsenic with respect to bioluminescence of *Vibrio fischeri* was somewhere between 22 (As(III)) to 121 (As(V)) mg/kg dw. Both these studies support a critical value around 20 mg/kg dw.

Uncertainties and determination of assessment factor

The TGD 27 clearly states the suggested assessment factors depending on the available data when applying a deterministic approach. As there were no chronic marine studies available but more than three long term tests for freshwater species, the same critical value was used in the derivation of both the marine and freshwater EQS. The assessment factor was set to 10 for freshwater sediments, fulfilling the criteria *Three long term tests (NOEC or EC10) with species representing different living and feeding conditions* in table 11 of the TGD 27, and 50 for marine sediments, fulfilling *Three long term sediment tests with species representing different living and feeding conditions in table 13 of the TGD 27.*

Background concentrations

Another aspect to account for when deriving an EQS, according to the TGD 27, is the natural background concentration. In the marine and coastal areas of Sweden, the Swedish EPA have compiled a status classification of sediments based on comparative values and the deviation from these values (Naturvårdsverket, 1999). The comparative value (Class I) is derived from the median reference value, calculated from sediments sampled at 55 cm below the water-sediment interface which should correspond to pre-industrial times. Arsenic concentrations above 10 mg/kg dw is classified as small to very large deviation. Other studies show that the mean sediment arsenic concentrations range from 5 to 3000 mg/kg, with the higher levels occurring in areas of contamination and cannot be considered as background levels.

Table 2: Deviation classification of metals in surface sediment based on the Swedish EPA report nr 4914. The concentration unit is mg/kg dw.

Class I	**Class II**	**Class III**	**Class IV**	**Class V**
Insignificant deviation	Small deviation	Apparent deviation	Large deviation	Very large deviation
<10	10-16	16-26	26-40	>40

Applying the EQS in Swedish waters

The derived critical value and the respective EQS value, when an assessment factor of 50 is applied, is compared to the marine monitoring data from 2014 (collected by SGU and published on ICES dome (ICES, 2020)). All sites will exceed the suggested EQS value (figure 2). The same is true for the freshwater surface sediment monitoring data from the Geological Survey of Sweden (SGU, 2022) where all monitoring sites exceeds the proposed EQS, when an assessment factor of 10 is applied (figure 3).

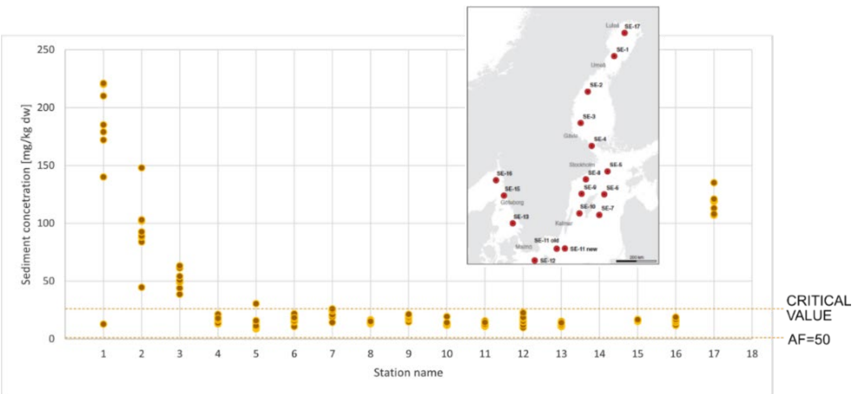

Figure 2: Surface sediment concentrations at Swedish marine stations (see map). The dashed lines show the critical value (i.e. EC10 Branchiura sowerbyi), determined from the deterministic approach, and the suggested EQS for marine sediments based on an assessment factor of 50.

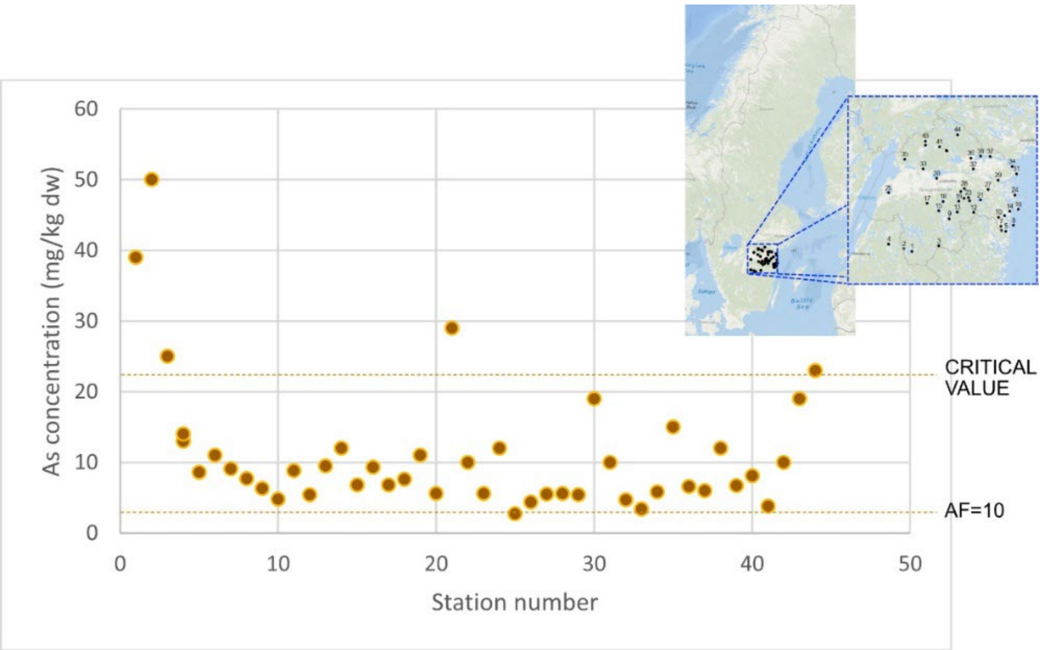

Figure 3: Surface sediment (0-2 cm) concentrations of arsenic from "background" environments according to monitoring data from 2019-2021 reported to and published in the SGU database of environmental pollutants. The dashed lines show the derived critical value and the threshold of the suggested EQS with an assessment factor of 10.

Conclusions

Based on the current data availability of ecotoxicological studies with arsenic exposure in sediments, a deterministic approach was applied to determine a critical value (=22 mg/kg dw). The assessment factors were determined based on the guidance in the TGD 27 where the number of studies supported an assessment factor of 10 for freshwater sediments and an assessment factor of 50 for marine sediments, as no chronic studies were available.

Even though the proposed EQSs of 2.2 and 0.4 mg/kg dw in freshwater and marine sediments are low relative to previous classifications (Appendix 1) and measured concentrations at monitoring stations (figure 2-3), they have been based on reliable and relevant ecotoxicological studies following the framework of the TGD 27 and should be accepted until more data becomes available.

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Appendix 1 - previously determined threshold values and background concentrations

Threshold value (mg/kg dw)	Method	Remark	Matrix	Country	Year	Reference
5.9	TRA	ISQG	FW sediment	Canada		Canadian Sediment Quality Guidelines for the Protection of Aquatic Life
17	TRA	PEL	FW sediment	Canada		Canadian Sediment Quality Guidelines for the Protection of Aquatic Life
7.24	TRA	ISQG	SW sediment	Canada		Canadian Sediment Quality Guidelines for the Protection of Aquatic Life
41.6	TRA	PEL	SW sediment	Canada		Canadian Sediment Quality Guidelines for the Protection of Aquatic Life
6	TRA	Lowest Effects Level (LEL)	FW sediment	US	1990	US-EPA: https://www.nj.gov/dep/srp/guidance/ecoscreening/esc_table.pdf
8.2	TRA	ER-L Effect range Low	SW sediment	US	1990	US-EPA: https://www.nj.gov/dep/srp/guidance/ecoscreening/esc_table.pdf
17	TRA	PEL (=Probable Effect Level)	FW sediment	US	1990	US-EPA: https://www.nj.gov/dep/srp/guidance/ecoscreening/esc_table.pdf
25	Background	Background value	SW sediment	OSPAR	2009	The Background Assessment Concentration (BAC) is normalised to 5% aluminium in all subregions except the Iberian Sea and Gulf of Cadiz, where BACs are not normalised.
20	TRA	GV	Guideline value	AUS		ANZEC/ ARMCANZ interim sediment quality guideline
70	TRA	GV-high	Guideline value	AUS		ANZEC/ ARMCANZ interim sediment quality guideline
<10	Klass I Ingen avvikelse	Comparison value (=jämförvärde) totalanalys	SW sediment	Sweden	1999	Naturvårdsverket (1999) rapport 4914
10-16	Klass II Liten avvikelse	Comparison value (=jämförvärde) totalanalys	SW sediment	Sweden	1999	Naturvårdsverket (1999) rapport 4914

16-26	Klass III Tydlig avvikelse	Comparison value (=jämförvärde) totalanalys	SW sediment	Sweden	1999	Naturvårdsverket (1999) rapport 4914
26-40	Klass IV Stor avvikelse	Comparison value (=jämförvärde) totalanalys	SW sediment	Sweden	1999	Naturvårdsverket (1999) rapport 4914
>40	Klass V Mycket stor avvikelse	Comparison value (=jämförvärde) totalanalys	SW sediment	Sweden	1999	Naturvårdsverket (1999) rapport 4914
25	N1	French sediment quality guideline for dredging (Below the level N1, the ecological impact is view as negligible. Between N1 and N2, chemical analyses must be supplemented with toxicity tests.)	SW sediment	France	2001	Mamindy-Pajany et al. (2013)
50	N2	French sediment quality guideline for dredging (Below the level N1, the ecological impact is view as negligible. Between N1 and N2, chemical analyses must be supplemented with toxicity tests.)	SW sediment	France	2001	Mamindy-Pajany et al. (2013)