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1     Assessment of metal toxicity in marine ecosystems -  
2             Comparative Toxicity Potentials for nine cationic  
3                     metals in coastal seawater

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10    ABSTRACT:

11     This study is a first attempt to develop globally applicable and spatially differentiated marine  
12     Comparative Toxicity Potentials (CTPs) or ecotoxicity characterization factors for metals in coastal  
13     seawater for use in Life Cycle Assessment. The toxicity potentials are based exclusively on marine  
14     ecotoxicity data and take account of metal speciation and bioavailability. CTPs were developed for  
15     nine cationic metals (Cd, Cr(III), Co, Cu(II), Fe(III), Mn, Ni, Pb and Zn) in 64 Large Marine  
16     Ecosystems (LMEs) covering all coastal waters in the world. The results showed that the CTP of a  
17     specific metal varies 3-4 orders of magnitude across LMEs, largely due to different seawater  
18     residence time. Therefore the highest toxicity potential for metals was found in the LMEs with the

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19 longest seawater residence times. Across metals, the highest CTPs were observed for Cd, Pb and  
20 Zn. At the concentration levels occurring in coastal seawaters, Fe acts not as a toxic agent but an  
21 essential nutrient and thus has CTPs of zero.

## 22 **1 Introduction**

23 Life Cycle Assessment (LCA) “quantifies all relevant emissions and resources consumed”<sup>1</sup>  
24 associated with a good or service in a Life Cycle Inventory (LCI) and assesses “the related  
25 environment and health impacts and resource depletion issues”<sup>1</sup> by Life Cycle Impact Assessment  
26 (LCIA). LCA has been broadly used to support environmentally informed decisions in policy-  
27 making, product development and procurement, and consumer choices<sup>2</sup>. It is a valuable screening  
28 tool to facilitate identifying environmental hotspots<sup>2</sup>. The uncertainties associated with LCA results  
29 can be high due to data and simplified modelling<sup>2</sup>. This can be partially compensated by enhancing  
30 regional detailed modelling.

31 Metals are often ranked at the top of toxicity concerns in Life Cycle Assessment (LCA)<sup>3</sup>. Large  
32 quantities of metals are released from anthropogenic resources to the natural environment (up to  
33  $3 \times 10^5$  tons/year for selected metals, e.g. Mn)<sup>4</sup>. Waterborne emissions contribute 50-80%, and  
34 originate mainly in industrial sectors such as iron or steel production, thermal power stations,  
35 mineral oil and gas refineries etc.<sup>5</sup> Waterborne metal emissions typically reach freshwater first and  
36 move towards seawater through fluvial pathways, thus potentially causing ecotoxicity in both  
37 freshwater and marine compartments<sup>6</sup>. Hitherto, metal toxicity in the aquatic environment has been  
38 modelled in LCIA using models developed to simulate the behaviour of organic chemicals with  
39 poor representation of the speciation behaviour of metals and bioavailability (e.g. USES-LCA 2.0<sup>7</sup>  
40 used in ReCiPe, IMPACT 2002+<sup>8</sup>). Following the principles laid out in the Apeldoorn Declaration<sup>9</sup>  
41 and the Clearwater Consensus<sup>10</sup>, Gandhi et al.<sup>11,12</sup> developed a new method to calculate the toxicity

42 potential of six metals in freshwater ecosystems (expressed as a Comparative Toxicity Potential  
43 (CTP), also known as a Characterization Factor in LCIA), including fate, bioavailability and effect  
44 of metals. Their CTP was calculated for a number of archetypical freshwater chemistries. Dong et  
45 al.<sup>13</sup> further adapted the method, expanding its scope of metals and calculated freshwater CTP for  
46 14 metals. The results showed that for some metals (e.g. Al, Be, Cr(III), Cu and Fe(III)), freshwater  
47 CTP was highly dependent on the speciation of metal in a certain water chemistry, thus varying by  
48 2-6 orders of magnitude in different water archetypes. This reveals the importance of 1) including  
49 metal speciation and bioavailability in the modelling and 2) identifying spatially determined and  
50 differentiated water chemistries.

51 In comparison, marine CTP of metals has received less attention. Following the Apeldoorn  
52 Declaration<sup>9</sup>, “the oceans are deficient in essential metals, and the CTP for essential metals should  
53 be set at zero for toxicity in the oceans.”<sup>9</sup> In contrast, coastal seawater receives higher  
54 anthropogenic metal emissions not just through fluvial pathways<sup>6</sup>, but also from airborne emission  
55 and metals resuspended from the seabed<sup>14</sup>, leading to the observable metal concentrations in the  
56 coastal zones, and even reach the mmol/l level close to wastewater discharges<sup>15</sup>. This can lead to  
57 exceeding the levels where metal becomes toxic to organisms.

58 Not all metal forms are toxic. Only bioavailable forms, often within the truly dissolved forms, can  
59 access a sensitive receptor, the biotic-ligand, and become hazardous<sup>16,17</sup>. In addition to metal  
60 availability, also its residence time in the coastal seawater is essential for the exposure and hence its  
61 CTP. For most metals, a substantial removal happens after entering coastal zone, where complex  
62 binding to Suspended Particulate Matter (SPM) followed by removal through sedimentation is  
63 increased<sup>6</sup>. The fate of a metal in coastal seawater is thus strongly influenced by its tendency to  
64 adsorb to SPM, its solubility in seawater and its complexation affinity with particulate and  
65 dissolved organic matter<sup>18</sup>.

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66 Until now metal marine CTP in the previous LCIA models has either not been calculated (e.g.  
67 USEtox<sup>19</sup>, IMPACT 2002+<sup>8</sup>) or it has been derived neglecting speciation and bioavailability, and  
68 using freshwater toxicity data (e.g. USES-LCA<sup>7</sup>), with a questionable representativeness for  
69 saltwater organisms<sup>20</sup>. Moreover, as demonstrated by Gandhi and co-workers, metal freshwater  
70 CTP is highly sensitive to water chemistry<sup>21</sup>. While water chemistry parameters such as pH,  
71 Dissolved Organic Carbon (DOC), SPM and salinity affect the speciation of metals, different  
72 seawater residence times (SRT) in different coastal zones also play a large role when determining  
73 the fate of metal in coastal compartment<sup>22,23</sup>. So far, no study has given a coherent treatment of the  
74 global spatial variability of metal marine CTP, considering speciation and applying toxicity data for  
75 marine organisms. As a consequence, toxic impacts on the marine ecosystem were either not at all  
76 considered in LCA studies or they were assessed with methods of limited reliability based on  
77 questionable assumptions. These shortcomings and the strives for a coherent consideration of  
78 marine biodiversity in LCA studies set the objectives of this study.

79 Aiming for consistency with the methodology developed for characterizing metal toxicity in  
80 freshwater<sup>11,13</sup> and applying marine ecotoxicity data availability in ECOTOX database<sup>24</sup>, the  
81 objective of this paper is to develop new, spatially differentiated and globally applicable marine  
82 CTPs for Cadmium (Cd), Cobalt (Co), Chromium(III) (Cr), Copper(II) (Cu), Iron(III) (Fe),  
83 Manganese(II) (Mn), Nickel (Ni), Lead (Pb) and Zinc (Zn), taking metal speciation and  
84 bioavailability into account, and investigating their variation over 64 Large Marine Ecosystems  
85 (LMEs) for emissions received in coastal seawater all over the world.

## 86 2 Methods

### 87 2.1 General framework

88 For metals,  $CTP_i$  for ecosystems is expressed as the Potentially Affected Fraction of species  
89 integrated over time and space  $[(PAF) \cdot \text{day} \cdot \text{m}^3/\text{kg}_{\text{emitted}}]$ , representing the ecotoxicity potential for  
90 the *total* metal in compartment  $i$ . It is calculated as the product of three factors: Fate Factor (FF),  
91 Bioavailability Factor (BF) and Effect Factor (EF) as presented in Eq. 1<sup>11</sup>.

$$92 \quad CTP_i = FF_i \cdot BF_i \cdot EF_i \quad (\text{Eq.1})$$

93 Where:

94  $FF_i$ : Fate Factor [day], representing the residence time of *total* metal in compartment  $i$ ,

95  $BF_i$ : Bioavailability Factor [dimensionless], representing the ratio between *truly dissolved*  
96 and *total* metal. Here truly dissolved metal contains metal free ion and inorganic metal  
97 complexes. Total metal includes truly dissolved metal, DOC complexed metal and  
98 SPM complexed metal.

99  $EF_i$ : Effect Factor  $[(PAF) \cdot \text{m}^3/\text{kg}]$ , representing the fraction of species potentially affected  
100 by the toxicity of the *truly dissolved* metal in compartment  $i$ .

101 This framework can be used for any single environmental compartment (e.g. freshwater, soil).  
102 When considering a multi-compartment system, the terms of eq.1 become matrices, which besides  
103 residence times also include inter-compartmental transfers<sup>25</sup>. In this paper we focus on metals  
104 received from adjacent environmental compartments or directly emitted into the coastal seawater  
105 compartment. Therefore, FF represents the persistence of the metal in coastal seawater, while BF  
106 and EF represent bioavailability and metal ecotoxicity effects in coastal seawater respectively. FF is  
107 modelled for the total metal rather than dissolved metal, due to the fact that this is the entity which  
108 is reported in LCI and that the metal in the water may re-partition between particulate and dissolved  
109 forms during transportation. Note that the partitioning pattern can vary over time and with local

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110 environmental conditions. This can have an impact on the FF of metals. For the purpose of LCA  
111 temporal variations need to be averaged over a year to be compatible to the information in the life  
112 cycle inventory.

## 113 2.2 Spatial differentiation of environmental conditions and parameters

114 To explore the spatial variability of CTP in coastal seawater, we worked with the LMEs  
115 following Cosme et al.<sup>26</sup>. The coastal compartment that is represented by a LME covers the marine  
116 area from the coastal line to the seaward boundary of the continental shelf and includes any  
117 estuaries. Thus defined, the coastal compartment with its adjacency to the continents receives  
118 emissions related to human activity through the influx of continental freshwater or direct discharges  
119 to the sea. 80%-90% of marine net primary production occurs in this compartment, which thus  
120 comprises the majority of species and biomass that potentially may be affected by metal  
121 emissions<sup>27</sup>. The global coastal seawater zone was divided into 64 LMEs according to “distinct  
122 bathymetry (seabed topography), hydrography, productivity and tropically dependent  
123 populations”<sup>28</sup>, where each LME represents a relatively independent coastal zone. Data on SRT,  
124 seawater surface area, temperature and water chemistry were collected for each LME from literature  
125 (Table S1 in Supporting Information (SI)). The values for these parameters show large variations  
126 across the 64 LMEs (Figure S1 in SI). SRT varies from 11 days-90 years, surface area from  
127  $1.5 \times 10^5$ - $5.7 \times 10^6$  km<sup>2</sup>, estuary discharge rate (water flow rate from freshwater to coastal seawater)  
128 from 0- $1.3 \times 10^5$  m<sup>3</sup>/s, temperature from -1°C-29°C, pH from 7.75-8.35, DOC from 0.6-6.5 mg/l,  
129 Particulate Organic Carbon (POC) from 31-802 ug/l, SPM from 0.2-2.9 mg/l and salinity from 6.2-  
130 40.3‰. For speciation modelling, salinity was translated into concentrations of the major ions (Na<sup>+</sup>,  
131 Mg<sup>2+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, and Cl<sup>-</sup>) by scaling from a standard salinity (35 ‰) and its corresponding  
132 major ion concentrations (Table S2 in SI), assuming a fixed relationship between the major ion

133 concentrations at different salinities<sup>29</sup>. For each LME, the relevant environmental parameter and  
134 water chemistry values were applied to derive a CTP value for each metal.

135 Note that also within one LME, environmental parameters such as pH, salinity, DOC, POC, and  
136 SPM show both spatial and temporal variation. The annual variation ranges are shown for pH,  
137 salinity and POC within each LME in Figure S2 in SI. Fe, Mn and Al oxides have been shown to be  
138 strong adsorbents for metal ions<sup>30-32</sup>, because of their large surface area. Due to lack of spatially  
139 differentiated concentration data for these oxides, fixed concentrations of 0.15, 0.02 and 0.4 µg/L  
140 for Fe, Mn, and Al oxides respectively had to be assumed across all LMEs<sup>33</sup>.

## 141 **2.3 Model and parameter selection**

### 142 **2.3.1 Fate model**

143 With the intended use in LCA in mind, the multimedia fate model embedded in USEtox<sup>19</sup> was  
144 chosen for this study. USEtox is an LCIA model for assessing ecotoxicity and human toxicity  
145 impacts. It has been developed in a scientific consensus process involving LCIA and chemical fate  
146 modelling experts. It is the recommended characterization model for toxicity impacts in LCA<sup>34</sup>. In  
147 USEtox, the fate is calculated based on a steady-state mass balance. USEtox determines metal FF in  
148 the coastal seawater compartment by modelling of metal inflow, metal outflow and metal removal  
149 (including sedimentation and sediment burial/re-suspension). Metal inflow and outflow largely  
150 depend on the retention time of the coastal seawater. Thus the default SRT of seawater on  
151 continental scale in USEtox was replaced by the SRT representative for each LME. To be  
152 consistent, also the default surface area of continental seawater and the water flow rate from  
153 continental freshwater to continental seawater (estuary discharge rate) were replaced by the  
154 corresponding LME-specific data. Water flow from ocean to coastal seawater is then automatically  
155 calculated from parameters mentioned above. Details of LME-specific data and calculations are  
156 available in Table S1 in SI. Metal removal is simulated by metal sedimentation and diffusion of



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157 metal from seawater to sediment. The former process is modelled by metal complexation with SPM,  
158 followed by SPM sedimentation. The removal largely depends on the fraction of metal adsorbing to  
159 SPM, the concentration of SPM and the SPM sedimentation velocity. Metal diffusion into sediment  
160 is determined by the dissolved fraction of metal and the metal's mass transfer coefficient between  
161 sediment and water. The metal fraction adsorbed to SPM can be calculated from a spatially  
162 differentiated adsorption coefficient  $K_{pSS}$  (L/kg; the ratio of metal concentration between *SPM*  
163 *bound* metal and *truly dissolved* metal). The truly dissolved fraction of metals is calculated using  
164 both  $K_{pSS}$  and  $K_{DOC}$  (L/kg; the ratio of metal concentration between *DOC complex bound* metal and  
165 *truly dissolved* metal). All parameters mentioned above vary between different LMEs. Thus  $K_{pSS}$   
166 and  $K_{DOC}$  were recalculated in WHAM VII<sup>35</sup> for each metal in each LME respectively, to replace  
167 the default values in USEtox. WHAM<sup>35</sup> is a metal speciation modelling software. Based on the  
168 input of target metal concentration and relevant water chemistry, it can deliver the concentration of  
169 target metal in a specific form. In WHAM's calculation of  $K_{pSS}$  and  $K_{DOC}$  values, it is assumed that  
170 metals are in equilibrium with the discrete sites of DOC and the organic fraction of SPM. Here  
171 target metals have to compete with other cations (e.g.  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $K^+$  and  $Na^+$ ) to form complexes  
172 with SPM or DOC. The ratio between the concentration of metal that is truly dissolved in water and  
173 the concentration of metal forming complexes with SPM or DOC were calculated for each LME  
174 and each metal as the specific  $K_{pSS}$  and  $K_{DOC}$  value. Default DOC and SPM concentration in  
175 USEtox were also replaced by the corresponding specific parameter values for each LME. Other  
176 landscape parameters were kept unchanged. All parameters used in FF calculation are listed in  
177 Table S3 in SI. There were no substance parameter values for Mn and Fe in default USEtox  
178 inorganic database. They thus had to be collected from literatures. The retrieved values are  
179 presented together with substance parameter values for the other metals in Table S4 in SI.

### 180 **2.3.2 Bioavailability model**

181 BF,  $K_{pSS}$ , and  $K_{DOC}$  all represent ratios between different metal species in coastal seawaters. They  
182 are thus dependent on the metal speciation in each LME. In the modelling of this speciation  
183 behaviour, we assumed that metals remained at their background concentration (Table S4 in SI) in  
184 coastal seawater before the emission. BF,  $K_{pSS}$ , and  $K_{DOC}$  were then calculated for each LME with  
185 its specific water chemistry. This assumption is based on the fact that LCA assesses impacts caused  
186 by marginal changes. Nevertheless, a sensitivity analysis of the dependence of BF,  $K_{pSS}$ , and  $K_{DOC}$   
187 on background concentration change is performed in section 3.4.5.

188 WHAM VII<sup>35</sup> was used to calculate metal speciation in seawater. While originally developed for  
189 freshwater, its applicability for prediction of metal free ion activity in seawater has been validated<sup>36</sup>.  
190 Furthermore it contains data and has a good reputation for simulating metal binding to DOC, POC,  
191 Fe oxide and Mn oxide. These two criteria favoured the choice of WHAM VII over other speciation  
192 models (e.g. Visual Minteq<sup>37</sup>, MINEQL+<sup>38</sup>, PHREEQC<sup>39</sup>).

### 193 **2.3.3 Ecotoxicity model**

194 Currently there are two main ecotoxicity models to explain how cationic metals cause toxicity in  
195 organisms. The Free Ion Activity Model (FIAM) assumes that the toxic compound is free metal ion  
196 represented by its activity. The Biotic Ligand Model (BLM) further includes the competition  
197 between free metal ion and other cations (e.g.  $Ca^{2+}$ ,  $H^+$ ) for binding to biotic ligand – the receptor in  
198 the target organism where the metal binds to exert its uptake and/or toxicity. Due to lack of BLMs  
199 for metals in seawater, FIAM was chosen in this study. It has been validated to assess metal toxicity  
200 to marine organisms in saltwater<sup>40,41</sup>. As stated in Clearwater Consensus<sup>10</sup>, we calculated EF based  
201 on truly dissolved metal, assuming that free ion is a fraction of truly dissolved metal and is  
202 responsible for the toxicity. In risk assessment, Predicted No Effect Concentration (PNEC) is  
203 typically used as effect indicator to protect the sensitive species of the ecosystem. Compared to

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204 PNEC, the geometric mean  $HC_{50}$  calculated from  $EC_{50}$ , representing the Potentially Affected  
205 Fraction (PAF) of species exposed above chronic  $EC_{50}$  values, is more robust but less  
206 conservative<sup>42</sup>. The purpose of LCA is to compare alternatives, where robustness is highly required.  
207 Therefore  $HC_{50}$  values calculated from  $EC_{50}$  are normally applied in LCA. It can use all the  
208 available toxicity data for a metal and is a measure associated with less uncertainty than the  
209 PNEC<sup>43,44</sup>. Detailed descriptions of calculation methods for the PAF method and  $HC_{50}$  can be found  
210 in Larsen et al.<sup>44,45</sup>. EFs were calculated exclusively from data on chronic marine  $EC_{50}$  from  
211 literature. The availability of marine ecotoxicity data in the ECOTOX database<sup>24</sup> allowed us to  
212 apply our model to nine cationic metals, including Cd, Co, Cr, Cu, Fe(III), Mn, Ni, Pb, and Zn  
213 (Table S5 in SI). For metals where chronic marine ecotoxicity data were insufficient, extrapolation  
214 from acute marine ecotoxicity data was performed applying an Acute-to-Chronic Ratio (ACR)  
215 derived from the available toxicity data as described in Table S6 in SI. Total metal marine  $EC_{50}$   
216 reported in literature were translated into free ion  $EC_{50}$  using WHAM VII<sup>35</sup>, taking into account  
217 water chemistry of the test medium in which the reported  $EC_{50}$  was determined. This conversion  
218 reduced the standard deviation of the  $EC_{50}$  of each metal by at least an order of magnitude (Table  
219 S5), which also justifies the use of FIAM in EF calculation.

220 The calculation of EF was based on the recommended principles for LCA<sup>43,45,46</sup>. For each metal at  
221 each trophic level (i.e. primary producers, primary and secondary consumers), a free ion activity  
222  $HC_{50-trophic}$  was calculated as the geometric mean of the corresponding free ion  $EC_{50}$  for all species  
223 with available data. The geometric mean of the resulting three  $HC_{50-trophic}$  represents the free ion  
224 activity  $HC_{50}$  in saltwater for that specific metal. Then, for each combination of metal and LME, a  
225 truly dissolved  $HC_{50}$  was calculated using WHAM VII, based on the free ion activity  $HC_{50}$  and  
226 corresponding LME water chemistry. Finally, EF was calculated as  $0.5/\text{truly dissolved } HC_{50}$ <sup>43</sup>.

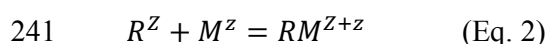
## 227 3 Results and Discussion

228 In this section the results for the spatially differentiated FF, BF, EF and CTP are discussed. The  
229 results are shown for all combinations of metals and LMEs in Table S7 in SI.

### 230 3.1 Fate Factors

231 Cr, Cu and Fe have the highest log  $K_{\text{DOC}}$  and log  $K_{\text{pSS}}$  among all metals, indicating their strong  
232 tendency of complexation with Organic Matter (OM, represented by DOC and the organic fraction  
233 of SPM (POC)) in seawater (Figure S3 in SI). This is in accordance with previous findings that Cr,  
234 Cu and Fe have high affinity for OM<sup>47</sup>. Compared with empirical values,  $K_{\text{pSS}}$  in this study were  
235 generally within an order of magnitude (Table 1).

236 Both log  $K_{\text{DOC}}$  and log  $K_{\text{pSS}}$  vary linearly with OM concentrations and salinity ( $0.31 < R^2 < 0.93$ ,  
237  $p < 0.001$ , Table S8) for all metals except Pb and Fe. OM in WHAM is considered as humic  
238 molecules, which are “rigid spheres, with proton-dissociating groups at the surface that can bind  
239 metal ions.”<sup>48</sup> Metal ion binding to a humic molecule can be simply expressed by the general  
240 reaction in Eq.2, which is described by the intrinsic association constant  $K_M$  (Eq.3)<sup>48</sup>.



$$242 \quad K_M = \frac{[RM^{Z+z}]}{[R^Z][M^z]} \quad (\text{Eq.3})$$

243 Here R is the humic molecule, M is metal and z is the net charge. Under similar conditions (e.g.  
244 pH value, temperature, etc.),  $K_M$  stays within a comparably narrow range. Therefore increasing OM  
245 concentration leads to a higher concentration of metal-OM complex, resulting in a higher log  $K_{\text{DOC}}$   
246 and log  $K_{\text{pSS}}$ . When salinity increases, the metal ions are in stronger competition with major cations  
247 in the seawater for the binding sites on OM, thus decreasing log  $K_{\text{DOC}}$  and log  $K_{\text{pSS}}$ <sup>49</sup>. The exception  
248 for Pb and Fe is due to the fact that the binding of Pb and Fe to DOC and particles is not only

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249 influenced by OM concentrations and salinity, but also by other parameters (e.g. temperature and  
 250 pH values).

251 **Table 1:** Log  $K_{pSS-D}$ <sup>a</sup> values (L/kg) in this study compared with empirical log  $K_{pSS-D}$ <sup>b</sup> values (L/kg)  
 252 developed in other studies.

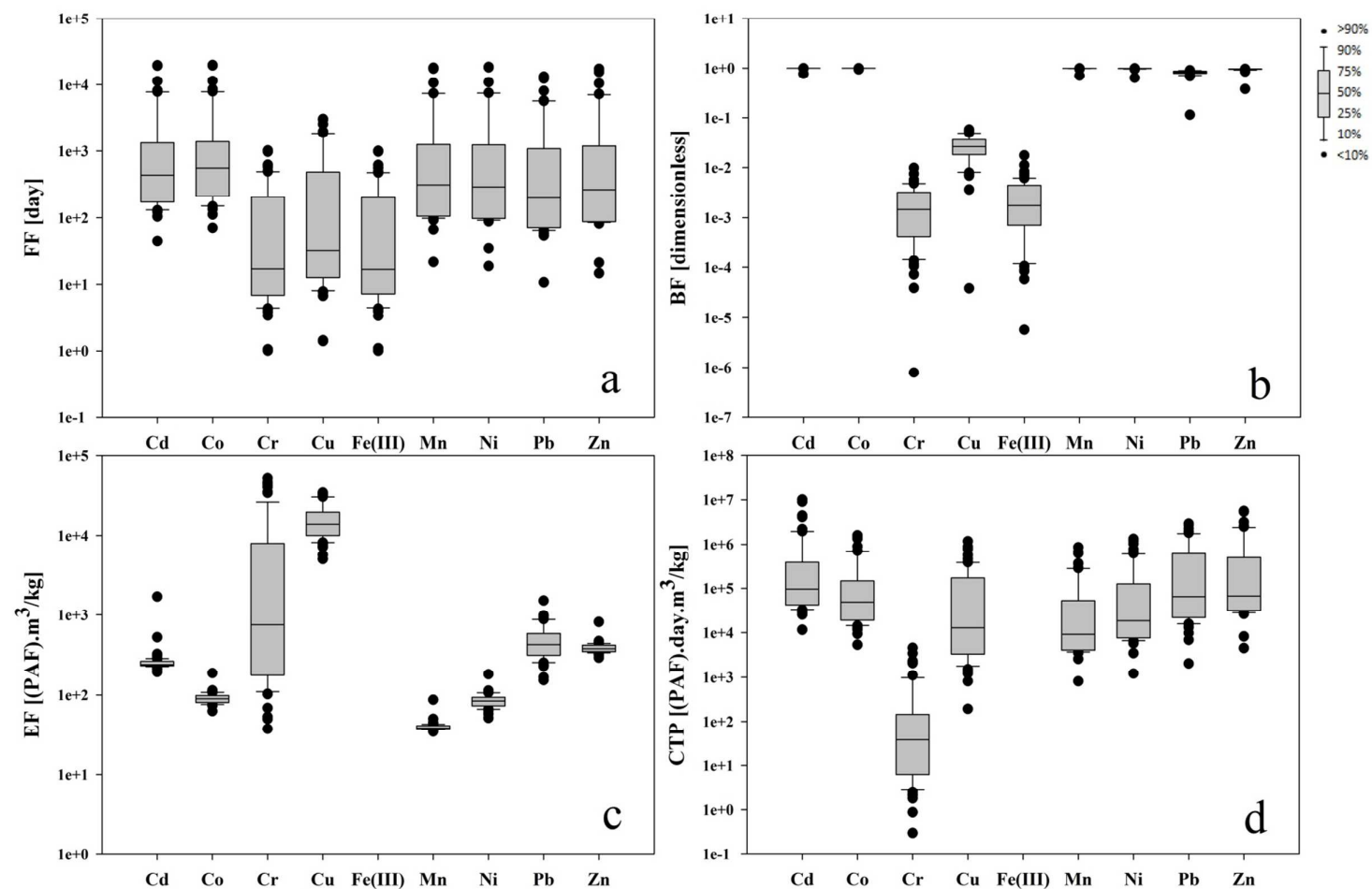
Metals	log $K_{pSS-D}$ in this study (mean±standard deviation)	log $K_{pSS-D}$ in other studies
<b>Cd</b>	2.9±0.3	4.0 <sup>50</sup> ; 4.3 <sup>51</sup> ; 3.5-5.5 <sup>52</sup>
<b>Co</b>	2.7±0.2	4.6 <sup>51</sup> ; 4.5-6.0 <sup>52</sup>
<b>Cr</b>	5.5±0.2	5.5 <sup>50</sup> ; 5.0-7.0 <sup>52</sup>
<b>Cu</b>	5.5±0.2	4.3 <sup>53</sup> ; 4.9 <sup>50</sup> ; 5.0 <sup>54</sup> ; 3.5-5.5 <sup>52</sup>
<b>Fe</b>	5.5±0.2	5.0-7.5 <sup>52</sup>
<b>Mn</b>	3.5±0.2	3.5 <sup>51</sup> ; 3.0-6.0 <sup>52</sup>
<b>Ni</b>	3.8±0.2	4.4 <sup>50</sup> ; 2.5-5.0 <sup>52</sup>
<b>Pb</b>	4.8±0.2	5.5 <sup>50</sup> ; 6.0 <sup>53</sup> ; 6.0 <sup>54</sup> ; 4.0-6.5 <sup>52</sup>
<b>Zn</b>	4.2±0.2	3.0 <sup>51</sup> ; 4.5 <sup>50</sup> ; 5.2 <sup>53</sup> ; 5.5 <sup>54</sup> ; 4.0-6.0 <sup>52</sup>

253 a. Log  $K_{pSS}$  (L/kg) in this study represents the calculated partitioning coefficient between metal  
 254 bound to SPM and truly dissolved metal. Log  $K_{pSS-D}$  (L/kg) in other studies represents the  
 255 partitioning coefficient between metal bound to SPM and total dissolved metal. To make the  
 256 values comparable, we calculated log  $K_{pSS-D}$  values from the log  $K_{pSS}$  that we determined in  
 257 this study.

258 b. Log  $K_{pSS-D}$  values in literature were presented as a function of other water chemistry  
259 parameters (e.g. salinity and SPM). Here we took approximate values derived from water  
260 chemistry similar to this study (e.g.  $SPM \approx 1 \text{ mg/L}$ ,  $Salinity \approx 30\text{‰}-35\text{‰}$ , etc.)

261 FF is largely influenced by log  $K_{pSS}$  and log  $K_{DOC}$ . Metals with high log  $K_{pSS}$  and log  $K_{DOC}$  (e.g.  
262 Cr, Cu and Fe) have an efficient removal, due to complex formation with OM followed by  
263 sedimentation. Therefore they have the lowest FF in all LMEs (Figure 1a). In contrast, FFs of Cd  
264 and Co are the highest across all metals, due to their low log  $K_{pSS}$  and log  $K_{DOC}$ . For a given metal,  
265 FF increases with SRT across LMEs (Figure S4 in SI). For Cd, Co, Mn, Ni, Pb and Zn, FF and SRT  
266 are linearly correlated with SRT ( $R^2 > 0.97$ ,  $p < 0.001$ , Table S8 in SI). It means that FF variation  
267 mainly depends on SRT and metal removal processes play a minor role. For the metals with high  
268 log  $K_{pSS}$  and log  $K_{DOC}$  (e.g. Cr, Cu and Fe), metal removal processes show a stronger influence on  
269 FF. Thus FFs for these three metals are less strongly correlated to SRT, but rather determined by the  
270 variation of SRT, log  $K_{pSS}$ , and log  $K_{DOC}$  together. Note that the metals with lower  $K_{pSS}$  and log  
271  $K_{DOC}$  (Cd, Co, Mn, Ni and Zn) can have a FF that is higher than SRT in some LMEs. The reason is  
272 that for these combinations of metal and ecosystem, the removed fraction is insignificant compared  
273 to the total input. A non-negligible fraction of the metal flows out to the ocean, from where some of  
274 it eventually recirculates back to the coastal seawater system after reaching steady state that USEtox  
275 calculates. This results in a longer FF than the water that originally carried them out. The effect is  
276 most pronounced in the LMEs with short SRTs because the inflow from the ocean is more  
277 important compared to the volume and the freshwater input for these LMEs.

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278

279 **Figure 1.** Variation of Fate Factor (FF, 1a), Bioavailability Factor (BF, 1b), Effect Factor (EF, 1c) and Comparative Toxicity Potential

280 (CTP, 1d) for nine metals determined across the 64 LMEs in this study.

281 FF varies 2-3 orders of magnitude across LMEs for each metal. Within one LME, FF variation  
282 between different metals is within two orders of magnitude (Figure S5a in SI). It indicates that FF is  
283 slightly more sensitive to environmental parameters than to properties of metal.

284 We compared our FF with data from other studies. The age of water constituent models the  
285 residence time of seawater constituents in particle forms in seawater by simulating particle cycling<sup>55</sup>  
286 and is similar to the concept of FF in this study. The constituent age of Baltic Seawater varies  
287 between a few days and up to 40 years<sup>56</sup>, which is similar to the range of metal FF in the Baltic Sea  
288 (LME 23) in this study (3-21years). The constituent age of Kara Seawater is 1-2 years<sup>57</sup>, which is  
289 within the range of the metal FF in the Kara Sea (LME 58) in this study (1-4 years). The constituent  
290 age of Norwegian Seawater and North Seawater combined together is 5-8 years<sup>58</sup>, which is slightly  
291 larger than the sum of metal FF ranges in the Norwegian Sea (LME 21) and the North Sea (LME  
292 22) in this study ( 1-5 years).

### 293 **3.2 Bioavailability Factors**

294 Representing the fraction of total metal in coastal water that is truly dissolved, BF of Cd, Co, Mn,  
295 Ni, Pb and Zn varies less than a factor of eight across LMEs (Figure 1b). For Cr, Cu and Fe the  
296 variations in BF are much larger with 3-4 orders of magnitude, due to their large variations in log  
297  $K_{\text{DOC}}$  and log  $K_{\text{pSS}}$  across LMEs (Figure 1b). For all metals, clear correlations were observed  
298 between BF and log  $K_{\text{DOC}}$  or log  $K_{\text{pSS}}$  (Figure S6). This implies that BF is largely determined by  
299 metal binding to DOC (log  $K_{\text{DOC}}$ ) and SPM (log  $K_{\text{pSS}}$ ). Co has the highest BF in all LMEs across  
300 metals, due to its low log  $K_{\text{DOC}}$  and log  $K_{\text{pSS}}$ . Similarly, Cr, Cu and Fe have the lowest BF across all  
301 LMEs, due to their high log  $K_{\text{DOC}}$  and log  $K_{\text{pSS}}$  values (Figure 1b and Figure S6 in SI).



### 302 3.3 Effect Factors

303 Some nutrient metals are essential for biota growth (e.g. Co, Cu, Fe, Mn, Ni and Zn)<sup>59,60</sup>.  
304 However, some of them may not reach the essential concentration to support biota growth under  
305 normal conditions in seawater, due to their low concentrations (at nmol level, Table S4 in SI).  
306 Under such circumstances, instead of being a toxic pollutant, a metal emission is more likely to  
307 facilitate biota growth. It is meaningless to talk about contribution to ecotoxicity under these  
308 circumstances. Therefore a true zero value of coastal CTP is given for those metals, in agreement  
309 with the recommendation in the Apeldoorn declaration<sup>7</sup>. For the metals covered in this study the  
310 essentiality condition appears to be relevant only for Fe, where the essential concentration range lies  
311 above its background concentration in coastal waters. This is caused by efficient removal of Fe in  
312 the estuary (ca. 90%) via precipitation, flocculation, and sedimentation<sup>6</sup>. Meanwhile, fluvial  
313 pathways contribute 75% of Fe inputs to seawater<sup>18</sup>, which leads to a low concentration of dissolved  
314 Fe in seawater. Morel et al.<sup>59</sup> reviewed the essential concentration of metals in seawater and found  
315 that for the metals Co, Cu, Mn, Ni and Zn, the background concentration in seawater is sufficient to  
316 support biota growth. This is in accordance with other studies showing that iron is the only limiting  
317 nutrient metal for algae growth in seawater<sup>61-63</sup>. Therefore, a true zero was given to the EF of Fe in  
318 all LMEs, which were excluded from the discussions in the rest of this section.

319 EFs show a modest variation, staying within one order of magnitude difference across all LMEs  
320 except for Cr, which shows a larger variation of three orders of magnitude (Figure 1c). Cu has the  
321 highest EFs in 90% of the LMEs, while Mn has the lowest EFs in all LMEs.

322 EF is influenced by temperature, pH, salinity and OM through their impacts on the speciation (the  
323 fraction of free ion activity within truly dissolved metal). In general, with increasing pH, the metal  
324 may form hydroxide or carbonate complexes, decreasing the metal free ion concentration in  
325 solution, which leads to a lower EF<sup>64</sup>. Increases in salinity leads to a higher ionic strength, which

326 results in lower free ion activity for a given free ion concentration, and thus a lower  $EF^{65}$ . When  
327 OM decreases, a fraction of metal may be released into truly dissolved forms, which leads to a  
328 higher truly dissolved  $HC_{50}$ , thus lower EF.

### 329 **3.4 Comparative Toxicity Potentials**

330 The comparative toxicity potentials are calculated as the product of FF, BF and EF following  
331 Eq.1. Results are shown in Figure 1d. Due to its background concentration below essentiality levels  
332 in coastal seawater ecosystems, the effect factor of Fe was set to zero and as a consequence its CTP  
333 also becomes zero.

#### 334 **3.4.1 Spatial variability of Comparative Toxicity Potentials**

335 Cr and Cu show the largest variation in CTP across LMEs with four orders of magnitude (Figure  
336 1d). For Cr the variation is mainly driven by the variation in EF ( $R^2=0.60$ ,  $p<0.001$ , Figure S7 in  
337 SI), and less influenced by variation of FF and BF ( $R^2<0.15$ ). For Cu no single individual parameter  
338 shows a significant correlation with CTP.

339 CTPs of Cd, Co, Mn, Ni, Pb and Zn vary by three orders of magnitude across LMEs (Figure 1d).  
340 These metals have rather stable BF and EF, which vary less than one order of magnitude across  
341 LMEs. Thus CTP variations are largely caused by FF. As FF of these metals is linearly correlated  
342 with SRT, CTP is overall strongly driven by the variation in SRT ( $0.64<R^2<0.96$ , Figure S8b in SI),  
343 with higher CTP for longer SRT.

#### 344 **3.4.2 Ranking of Comparative Toxicity Potentials**

345 Among all metals, Cd has the highest CTP in 45% of the LMEs (Figure S5d in SI), followed by  
346 Zn (31%) and Pb (24%). These three metals have high FF, BF and middle to high EF. They are  
347 ranked among the top four CTPs in all LMEs. In contrast, Cr has the lowest CTP in all LMEs (apart  
348 from Fe, for which CTP is zero). Although its EF is in the middle range compared to the other

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349 metals, its BF and FF are constantly low in all LMEs, due to its high log  $K_{\text{DOC}}$  and log  $K_{\text{pss}}$ . Also  
350 Mn and Ni are consistently in the lower ranking of CTP in all LMEs (5<sup>th</sup>-7<sup>th</sup>), due to their low EFs.

351 For Cd, Co, Mn, Ni, Pb and Zn, variation in CTP is significantly driven by SRT. Thus, the highest  
352 CTPs for these metals are observed in LME 5 (Gulf of Mexico), LME 26 (Mediterranean), and  
353 LME 62 (Black Sea), which have the longest residence time across LMEs (90 years). In contrast,  
354 the lowest CTP is observed in LME 35 (Gulf of Thailand), which has the 2<sup>nd</sup> shortest SRT among  
355 all LMEs (15 days).

356 CTPs ranking for Cr and Cu across LMEs are largely determined by SRT and by temperature  
357 through its influence on speciation. The highest CTPs are found in LME 64 (Antarctic), where the  
358 2<sup>nd</sup> lowest temperature (-1.20 °C) and long SRT (11 years) are observed. In contrast, they have the  
359 lowest CTP value in LME 35 (Gulf of Thailand), which has the 2<sup>nd</sup> highest temperature and 2<sup>nd</sup>  
360 shortest SRT.

### 361 3.4.3 Comparison between freshwater and coastal CTPs

362 Cd, Co, Cr, Mn, Ni and Zn marine CTPs show similar ranges to freshwater CTP determined by  
363 Dong et al.<sup>13</sup> using a parallel approach (Figure 1d). These similarities hide remarkable differences in  
364 fate and effect behaviour in freshwater and coastal waters, which tend to neutralize each other in the  
365 calculation of the CTPs. For these metals, EFs are thus up to two orders of magnitude lower in  
366 seawater due to higher free ion activity  $\text{HC}_{50}$  in seawater (Table S9 in SI). This is in accordance  
367 with previous research that freshwater species are more sensitive to metals than marine species<sup>66</sup>. In  
368 contrast, FFs are up to two orders of magnitude higher in seawater due to longer water residence  
369 times in many LMEs (the residence time of freshwater is 143 days at maximum in USEtox<sup>13</sup>). For  
370 the metals Cd, Co, Cr, Mn, Ni and Zn, BF in freshwater and seawater are rather similar. Cd, Co,  
371 Mn, Ni, and Zn were insensitive to variations in water chemistry in freshwater<sup>13</sup>. Thus it may be  
372 reasonable to expect similar BF in freshwater and seawater for these metals. BF of Cr is correlated

373 to log  $K_{\text{DOC}}$  and log  $K_{\text{pSS}}$ . These two values are negatively correlated with both SPM and salinity in  
374 estuaries<sup>49</sup>. From the freshwater end to seawater end, salinity increases and SPM decreases, which  
375 in combination leads to similar ranges of log  $K_{\text{DOC}}$  and log  $K_{\text{pSS}}$ , and thus similar BF ranges in  
376 seawater and freshwater for Cr. In summary, a combination of similar BF in sea- and freshwater,  
377 lower EF in seawater, and higher FF in seawater results in a similar range of CTP in seawater and  
378 freshwater for Cd, Co, Cr, Mn, Ni, and Zn (Figure S9 in SI).

379 Cu has up to two orders of magnitude higher FF in freshwater. It has a similar BF in freshwater  
380 and seawater, for similar reasons as Cr. But its EF is 2-4 orders of magnitude lower in seawater,  
381 which results in a slightly lower CTP in seawater (Figure S9 in SI).

382 Pb has a FF up to three orders of magnitude higher and a slightly lower EF in seawater than in  
383 freshwater. At the same time its BF is 1-2 orders of magnitude higher in seawater, possibly due to  
384 lower SPM and OM concentrations in seawater. This results in 1-4 orders of magnitude higher CTP  
385 in coastal seawater than in freshwater (Figure S9 in SI).

386 CTP is expressed in potentially affected fraction of species integrated over time and space.  
387 However, the species density varies considerably depending on location - from  $7 \times 10^{-12}$  to  $5 \times 10^{-4}$   
388 species/ $\text{m}^3$  in different freshwater ecosystems at various locations<sup>67</sup>. Thus, even if two different  
389 archetypes have the same CTP, the number of affected species can in extreme cases differ up to  
390 eight orders of magnitude in freshwater. Variation would also be expected for species density in  
391 coastal marine ecosystems. Moreover, species density in freshwater is generally about three orders  
392 of magnitude higher than in seawater<sup>68</sup>, which should be taken into account when comparing CTP  
393 values in freshwater and seawater.

#### 394 **3.4.4 Comparison of Fate Factors and Bioavailability Factors from USEtox**

395 The current version of USEtox does not provide marine CTP and only has seawater as a fate  
396 compartment supporting FF and the eco-exposure factor (XF) calculation for seven of the metals

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397 covered in this study (Cd, Co, Cr, Cu, Ni, Pb and Zn). USEtox operates with a default SRT of one  
398 year, which is at the middle range of SRTs applied for the LMEs in this study. The default USEtox  
399 FF thus falls within the range of the new FF in this study for all the metals (Figure S9 in SI). BF in  
400 this study is similar to the concept of eco-exposure factor (XF) in USEtox. The default XF in  
401 USEtox falls within or close to the range of BF found in this study for most metals. The only  
402 exceptions are Cr and Cu, for which the USEtox XF is 1-6 orders of magnitude higher (Figure S9 in  
403 SI). This is because the default  $K_{\text{DOC}}$  and  $K_{\text{pSS}}$  values in USEtox were taken from literature<sup>69</sup>, where  
404 it was defined as the ratio between absorbed metal and total dissolved metal. Recall that  $K_{\text{DOC}}$  and  
405  $K_{\text{pSS}}$  calculated in this study represent the ratios between absorbed metal and truly dissolved metal.  
406 This results in a lower  $K_{\text{DOC}}$  and  $K_{\text{pSS}}$  in USEtox, which leads to a higher XF.

#### 407 3.4.5 Sensitivity analysis

408 Several water chemistry parameters (DOC, POC, SPM, pH, salinity, metal background  
409 concentration and concentrations of Fe oxides, Mn oxides and Al oxides) and environmental  
410 parameters (SRT, surface area, freshwater inflow and temperature) are involved in the calculation  
411 of CTP in this study. In the following section, we will test the sensitivity of CTP to these  
412 parameters.

413 Salinity and pH values were extracted from a complete datasets<sup>70</sup>. Surface area and freshwater  
414 inflow were measured data taken from a global database<sup>71</sup>. They are well established values and  
415 their uncertainty are only caused by measurement error. Thus the uncertainty is hence judged to be  
416 low (e.g. uncertainty of pH meter measurement accuracy  $<0.1$ <sup>72</sup>, salinity probe  $<3\%$ <sup>73</sup>).

417 LME-specific land surface areas were applied in USEtox to calculate CTPs for metals in this  
418 study. Compared to the CTPs calculated by applying default land surface area in USEtox, the  
419 differences are less than 2%, caused by slightly different air deposition (which is also proportional  
420 to the land area).

421 The importance of the uncertainty accompanying the Fe, Mn and Al oxide concentrations was  
422 tested by changing them by a factor of 10. As a result CTPs varied less than 10% for all metals  
423 except Pb, for which the variation amounted to 1-35% across all LMEs.

424 DOC, POC and SPM affect metal partitioning in water, and thus the CTP. These three parameters  
425 show a significant positive correlation in natural waters (Figure S10 in SI). The parameter values  
426 are accompanied by variation among different seawater and locations, along with the transition  
427 from fresh to marine waters. We therefore tested the sensitivity of CTP to these three parameters, by  
428 varying them all together by a factor of 0.1-10. These variations can cover DOC, POC, and SPM  
429 concentrations ranging from conditions in freshwater to the open ocean. For these variations, CTP  
430 of Cr and Cu show the highest sensitivity, varying between a factor of 0.004 and 168. The other  
431 metals show very modest sensitivity, varying between a factor of 0.2 and 2.2 (Figure S11 in SI).  
432 This indicates that CTP is sensitive to DOC, POC and SPM concentrations for Cr and Cu, but less  
433 sensitive for the other metals. Note that within each LME, DOC, POC and SPM vary across  
434 locations and time. The average value of these parameters in a specific LME was applied in our  
435 study to calculate the corresponding CTP in that LME. Considering the large water volume and  
436 surface area in each LME, and the comparatively constant pH, salinity and POC values (Figure S2  
437 in SI), the average value of DOC, POC and SPM, thus CTPs are not likely to change dramatically  
438 within one LME. However, the uncertainty associated with CTPs of Cr and Cu is still comparably  
439 larger than the other metals. This needs to be noted when comparing CTPs across metals.

440 SRT has a strong influence on FF for all metals and hence also on the CTP. We varied SRT by  
441 two orders of magnitude (0.1X-10X of original values) resulting in a variation in CTP by a factor of  
442 0.05-21(Figure S12 in SI). The variations of CTP and SRT show a similar trend, indicating that  
443 CTP positively covariates with SRT. Therefore, SRT is an important parameter determining CTP

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444 when comparing metal CTP across LMEs, but it is less relevant for comparing CTP across metals  
445 within the same LME.

446 Temperature has influence on metal speciation, thus potentially influencing FF, BF, and EF. We  
447 calculated CTP by changing temperatures to 10°C lower or 10°C higher than the original values.  
448 This variation range covers the surface seawater temperature for the whole year, judging from data  
449 in the MODIS database<sup>74</sup>. We found that CTP only varies within a factor of 0.4-2.8 (Figure S13 in  
450 SI) for all metals. For Cr, BF and EF vary up to one order of magnitude. However, BF and EF have  
451 positive and negative correlation respectively with temperature and hence partly compensate each  
452 other, which results in a moderate change of CTP. It can be concluded here that CTP is not very  
453 sensitive to temperatures.

454 BF,  $K_{pSS}$ , and  $K_{DOC}$  were calculated from metal background concentration in generic seawater,  
455 which may differ in different locations. Therefore we tested the dependence of BF,  $K_{pSS}$ , and  $K_{DOC}$   
456 on metal background concentration, by varying background concentration by a factor of 10 (0.1X-  
457 10X of original value). For the metals with higher  $K_{pSS}$  and  $K_{DOC}$  values (e.g., Cr, Cu and Fe), BF  
458 can vary up to one order of magnitude and  $K_{pSS}$ , and  $K_{DOC}$  can vary up to two orders of magnitude.  
459 The variation is largely caused by metal binding with OMs. For the other metals, the variations of  
460 BF,  $K_{pSS}$ , and  $K_{DOC}$  are less than 2X. This result is similar to the observation in Gandhi et al.<sup>21</sup>. It  
461 shows that in the systems with higher background concentrations, BFs thus CTPs of metals with  
462 higher  $K_{pSS}$ , and  $K_{DOC}$  values may be underestimated. However, this might be offset by the  
463 adaptation of aquatic biota in those systems, which is not considered in the current effect  
464 modelling<sup>21</sup>.

### 465 3.5 Practical implications

466 This study is the first attempt to derive marine CTP considering speciation, bioavailability,  
467 seawater specific toxicity, and spatial differentiation. The results show that CTP for one metal can

468 vary 3-4 orders of magnitude across LMEs, except for Fe, for which CTP is zero due to its low  
469 background concentration and essentiality to marine biota. It was clearly demonstrated that it is of  
470 great importance to apply spatially differentiated CTP for metals in coastal seawater, as shown for  
471 all metals covered by this study except Fe. This raises the requirement for LCA practitioners to  
472 consider the emission location in the inventory. The variation of CTPs is primarily driven by SRT  
473 for most metals except Cr and Cu. If there is any updates on SRT in future research, it is strongly  
474 recommended to recalculate metal CTPs correspondingly. Due to limited ecotoxicity data for  
475 marine species and the metal coverage of the speciation model WHAM VII, it is difficult to derive  
476 marine CTP for additional metals at this point. It is recommended to look into methods to estimate  
477 marine ecotoxicity data by extrapolation from freshwater ecotoxicity data, or from known metal  
478 properties. This can potentially provide ecotoxicity data for more metals and thus allow calculation  
479 of additional marine CTPs. Where measured chronic data was missing, acute toxicity data was  
480 extrapolated to chronic EC<sub>50s</sub> for the EF calculation of some metals (e.g. Co, Cr, Mn, Ni, Pb and  
481 Zn, Table S5 in SI). It is recommended to revise these data when chronic data is available. The  
482 speciation model WHAM VII cannot simulate metal redox reactions and precipitation except Al  
483 and Fe hydroxide. Due to the fact that the CTP developed in this study is for metal in coastal  
484 seawater where water column depth is modest and presence of oxides are limited, the occurrence of  
485 extreme redox conditions will be rare in most LMEs. E.g., When Cr(III) is emitted to coastal  
486 seawater, its oxidation to Cr(IV) is limited and slow, unless abundant Mn dioxide and hydroxides  
487 exist<sup>75</sup>. However, the lack of precipitation modelling in WHAM can cause some uncertainties,  
488 especially for metals which may form insoluble compounds with major anions in seawater.  
489 Therefore, it is recommended to explore the possibility of applying other metal speciation models to  
490 complement WHAM VII (e.g. MINEQL<sup>38</sup>, Visual Minteq<sup>37</sup>, CHEAQS Pro<sup>76</sup> or PHREEQC<sup>39</sup>)  
491 covering other metals and supporting the modelling of precipitation and redox reactions where



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492 needed. Literature reported that eutrophication can increase metal bioavailability up to an order of  
493 magnitude<sup>77,78</sup>. However, this may be offset by decreasing EF due to organism adaptation, which is  
494 not considered in this study. Comparing to 3-4 orders of magnitude variation in CTPs, the  
495 uncertainty introduced by differences in eutrophication across LMEs will not have significant  
496 influences on the result. FIAM was used to assess EF in this study. However, unlike BLM it does  
497 not include competition between free metal ion and other cations for binding to biotic ligands. Thus  
498 it is recommended to estimate EFs with marine BLM when available. This study only developed  
499 CTP for metals in the water column of the seawater compartment. Ecotoxicity potentials in  
500 sediments were not considered here. In LCIA this is typically considered as a separate compartment  
501 (if at all) and would require a separate study.

502

503 ASSOCIATED CONTENT

#### 504 **Supporting Information**

505 9 tables and 13 figures addressing additional data were presented in supporting information. This  
506 material is available free of charge via the internet at <http://pubs.acs.org>.

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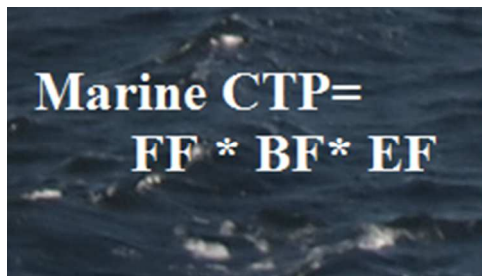
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