



HAL
open science

Analysis of biokinetic parameters reveals patterns in mercury accumulation across aquatic species

Louise M Stevenson, Paul G Matson, Rachel M Pilla, Simon Pouil, Tom J Geeza, Amber Hills, Zapporah Ellis, Sydney Smith, Teresa J Mathews

► **To cite this version:**

Louise M Stevenson, Paul G Matson, Rachel M Pilla, Simon Pouil, Tom J Geeza, et al.. Analysis of biokinetic parameters reveals patterns in mercury accumulation across aquatic species. *Science of the Total Environment*, 2025, 959, pp.178129. <10.1016/j.scitotenv.2024.178129>. <hal-05106151>

HAL Id: hal-05106151

<https://hal.science/hal-05106151v1>

Submitted on 10 Jun 2025

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Copyright - All rights reserved

1 **Analysis of biokinetic parameters reveals patterns in mercury accumulation across**
2 **aquatic species**

3

4 Louise M. Stevenson^{a,*}, Paul G. Matson^a, Rachel M. Pilla^a, Simon Pouil^{a,b}, Tom J. Geeza^a,
5 Amber Hills^a, Zapporah Ellis^a, Sydney Smith^a, Teresa J. Mathews^a

6

7 ^a Oak Ridge National Laboratory, Environmental Sciences Division, Oak Ridge, TN, United
8 States of America

9 ^b Université Paris-Saclay, INRAE, AgroParisTech, GABI, 78350 Jouy-en-Josas, France

10

11 * Corresponding author: L.M. Stevenson

12 E-mail address: stevensonlm@ornl.gov

13 Abstract

14 Mercury (Hg) is a potent neurotoxicant and poses a risk to human health through the ingestion
15 of Hg-contaminated fish. Mercury, especially in its organic form methylmercury (MeHg),
16 biomagnifies up food chains such that even small aqueous concentrations of Hg can result in
17 significant concentrations of total Hg in fish. Understanding the ecological and human health
18 risks associated with Hg and MeHg exposure requires an understanding of the factors that
19 affect its bioaccumulation in aquatic species. We compiled estimates of three biokinetic
20 parameters: uptake rate (k_u), assimilation efficiency (AE), and efflux rate (k_e). These
21 parameters describe contaminant uptake from aqueous (k_u) and dietary (AE) exposure and the
22 rate of excretion (k_e). We found parameter values for 38 and 34 different species of fish and
23 aquatic invertebrates, respectively, and collected 502 parameter values in total. We used a
24 machine learning technique to establish the relationships between experimental and
25 physiological variables and these parameter values. We found differences in which variables
26 were associated with biokinetic parameter values for fish and aquatic invertebrates. The form
27 of Hg was the most impactful variable, influencing values of all parameters except k_u for
28 invertebrates, for which aqueous exposure time was the only significant predictor variable.
29 The parameter k_e were the only values significantly influenced by more than one variable,
30 with water type (freshwater, brackish, or marine), organism weight, and form of Hg
31 significantly impacting parameter values for fish and/or invertebrates. To our knowledge, this
32 study represents the most extensive review of biokinetic parameters of Hg and MeHg
33 accumulation in aquatic organisms. Environmental parameters found to significantly impact
34 Hg and MeHg bioaccumulation in past studies were not identified as important in our
35 analyses across aquatic ecosystems and species. Our dataset and analysis reveal novel patterns
36 that may help us better understand and manage Hg bioaccumulation.

37 Keywords: Mercury, Methylmercury, Bioaccumulation, Biokinetic modelling, Regression tree
38 analysis

39 1. Introduction

40 Contaminant bioaccumulation serves as a valuable indicator of the chemical health of an
41 ecosystem. Metal bioaccumulation is a particularly stable indicator as metals cannot be
42 metabolized into other forms, such as organic contaminants. However, metal accumulation, as
43 with other contaminants, is complex and impacted by abiotic and biotic variables depending
44 on the physicochemical characteristics of the focal element and its environment and the biotic
45 characteristics of the focal organism. Metal biomagnification across trophic levels can imperil
46 ecological and human health, turning low aqueous concentrations of some metals, including
47 mercury (Hg), into dangerous concentrations in higher-trophic-level organisms. Therefore,
48 there is great incentive to understand metal bioaccumulation rates and drivers across species.

49 Biokinetic or toxicokinetic models have been developed to understand and predict the internal
50 concentration of a chemical such as Hg in an organism (mass of contaminant per mass of
51 organism). Since responses to contaminant exposures can differ interspecifically and even
52 intraspecifically, biokinetic models can help unify assessments of ecological risk for a
53 chemical by calculating a common currency (internal body burden) for organisms in a shared
54 environment. Biokinetic models are also useful to help interpret field data and compare
55 laboratory results to other studies and, if successfully parameterized, can predict
56 concentrations of a contaminant in the future and allow for assessments of various
57 remediation schemes through simulations.

58 One of the most commonly used biokinetic models for metal bioaccumulation is known in the
59 literature as the dynamic multi-pathway bioaccumulation model (DYMBAM), also known as
60 a bioenergetic-based or kinetic bioaccumulation model (Luoma et al., 1992; Luoma and
61 Rainbow, 2005; Schlegel et al., 2002):

$$\frac{dC_i}{dt} = k_u C_W + AE * IR * C_F - C_i(k_e + g)$$

62 C_i is internal concentration of the contaminant, k_u is uptake rate, C_w is contaminant
63 concentration in water, AE is assimilation efficiency, IR is ingestion rate, C_F is contaminant
64 concentration in food, k_e is efflux rate, and g is growth rate. DYMBAM predicts the
65 concentration of a contaminant within an organism based on two modes of influx and two of
66 efflux. The internal concentration of the contaminant increases from aqueous exposure and
67 through ingestion of contaminated food. Water uptake occurs based on the contaminant
68 concentration in the water and the rate at which the organism takes up the contaminant (k_u).
69 The organism accumulates contaminant from food based on the concentration of that
70 compound in the food item and the proportion of that chemical the organism assimilates into
71 its own tissue (AE, assimilation efficiency). The internal contaminant concentration decreases
72 because of the rate of efflux (the organism excreting the contaminant in some way at a rate
73 k_e) and growth. Combined, these parameters estimate the bioaccumulation of metals of
74 concern in organisms and their magnification up the food web.

75 Mercury (Hg) poses a potent risk to ecological and human health across the globe. For
76 decades, researchers have studied the bioaccumulation of Hg and its organic form,
77 methylmercury (MeHg), in both field and laboratory settings to quantify this risk
78 (Environment, 2019). Hg bioaccumulation can vary widely between aquatic systems and
79 species; studies have identified abiotic factors and processes that significantly impact Hg
80 bioaccumulation such as alkalinity (Spry and Wiener, 1991), pH and dissolved organic carbon
81 (DOC; Watras et al., 1998), sediment resuspension (Kim et al., 2004), and solar radiation,
82 total suspended solids, and salinity (Clarke et al., 2023). The relative importance of these
83 influences on Hg fate and bioaccumulation differs between systems and between forms of Hg
84 (inorganic Hg vs MeHg; Watras et al. (1998). While many studies have identified important
85 drivers of Hg bioaccumulation within species or within a certain focal study site, these
86 patterns have not been investigated across a wide range of aquatic species for all relevant
87 toxicokinetic parameters.

88 To investigate patterns of Hg accumulation across aquatic species, model parameter values
89 were collected from the literature for three main parameters: uptake rate (k_u), assimilation
90 efficiency (AE), and efflux or elimination rate (k_e). Values for these parameters have been
91 measured for decades and there is extensive information in the literature. We created a
92 database to summarize these parameter values for fish and aquatic invertebrates and looked
93 for patterns across biotic (species, trophic level, phylogenetic association) and abiotic
94 (salinity, temperature) variables that may explain the variability across these parameter
95 values. By comparing parameter estimates to each other, comparisons of Hg and MeHg
96 biokinetics can be compared across species, taxa, empirical setups, time scales, and
97 environments. Further, this meta-analysis can help guide the development of more elaborate
98 biokinetic models specifically for Hg and MeHg by identifying which factors need to be
99 incorporated to successfully predict Hg/MeHg uptake, assimilation, and efflux in natural
100 systems. Recent studies (Madenjian et al., 2021; Yao and Drouillard, 2019) have identified
101 the need to predict Hg body burdens more accurately in fish by determining which factors
102 may be the most influential in the face of complex environmental challenges, such as
103 overfishing and climate change.

104 **2. Methods**

105 2.1. Literature review

106 We conducted two reviews of the peer-reviewed literature pertaining to biokinetic parameters
107 for mercury bioaccumulation. We started the first review in May 2020 and focused on fish
108 ('fish library'), while the second review was initiated in June 2021 and focused on
109 invertebrates ('aquatic invertebrate library'). We searched two online databases (Web of
110 Science and Google Scholar) for the keywords "fish" (or "invertebrate", respectively),
111 "mercury", "bioaccumulation", "assimilation efficiency", and "kinetics". We downloaded and
112 read articles matching the search criteria and identified additional literature by carefully

113 reviewing the articles and reference list of each article. For example, review papers such as
114 Reinfelder et al. (1998) and Trudel and Rasmussen (1997) are two primary literature sources
115 that yielded many relevant references but are not cited directly in our database. Multiple
116 researchers contributed to the compilation of literature, and we checked every database entry
117 on two separate occasions to ensure consistency and accuracy.

118 We extracted biokinetic parameters related to mercury bioaccumulation and associated
119 metadata from each article (Table S1). We included the following biokinetic parameters in the
120 database: 1) uptake rate, k_u ($L\ g^{-1}\ d^{-1}$), 2) assimilation efficiency AE, (%), and 3) efflux rate k_e
121 (d^{-1}). If these parameters were reported in different units, we expended all efforts to convert
122 them using information in the article. If unit conversion was not possible, we did not include
123 the parameter value in the database. We report all values on a wet weight basis because the
124 majority of papers reported parameter values in terms of wet weight. For the few reporting
125 values in dry weight, we converted either using data provided in the article or using
126 supplemental studies found in the literature that define relationships between wet and dry
127 weights of the focal organism. For fish, we identified parameter values as either measured in
128 the filet (muscle only) or whole-body, while all invertebrate values were measured whole-
129 body. In terms of fish, it is possible that whole body differs from filet accumulation rates such
130 that parameter estimations using filet concentrations would differ from estimates using whole-
131 body concentrations; however, there is no clear evidence that rates or AE should differ in this
132 regard (Van Walleggem et al., 2013).

133 We gave special consideration to values of k_e that were included in the database. Efflux rate
134 of Hg is generally acknowledged to be biphasic with the fast component being relatively short
135 (half-life of days to weeks) compared to the slower component, which can range from months
136 to years (Trudel and Rasmussen, 1997; Yao and Drouillard, 2019). Past studies have shown
137 that chronically exposed fish excrete persistent contaminants primarily from the slow
138 compartment and thus models incorporating the slow phase estimate for efflux are more likely

139 to predict Hg dynamics (Trudel and Rasmussen, 1997). As such, we used only efflux rate
140 estimates for the slow component of a biphasic efflux model, if it was presented in the article.
141 Most articles either presented one efflux rate (single compartment model of efflux as opposed
142 to two compartment or biphasic), explicitly reported efflux rates from both phases, or stated
143 that they were specifically only considering the slow phase of efflux and ignored fast, short-
144 term efflux. Many efflux rate estimates were reported as the “biological half-life” of mercury
145 ($t_{1/2}$), which was converted to efflux rates using the equation:

$$k_e = \frac{\ln(2)}{t_{1/2}}$$

146 which assumes a single phase of efflux likely more comparable to the longer, slow phase of a
147 biphasic model than the shorter, fast phase.

148 We extracted 51 variables that could be predictive of these parameter values from the
149 reviewed literature (Table S1). These were chosen for their potential relevance in predicting
150 Hg bioaccumulation based on past studies and expertise on Hg cycling. We will briefly
151 describe our reasoning for including each variable (column name denoted using “”) not
152 previously discussed in this section. We recorded the “salinity” reported in each study, both as
153 a general “Water type” (freshwater, brackish, or marine) and also as a continuous variable
154 (“‰/ppt”). Multiple studies have found that biokinetic parameter values vary across salinities,
155 either due to physiological differences between marine and freshwater organisms (Dutton and
156 Fisher, 2011; Pickhardt et al., 2006) or salinity itself (Denton and Burdon-Jones, 1981; Wang
157 and Wang, 2010). “Pathway”, how Hg was introduced, may also impact biokinetic parameter
158 values; for example, the AE of Hg from a prey item that a fish that was force-fed (gavaged)
159 may be lower than if the fish had been allowed to forage freely (Dutton and Fisher, 2012;
160 McCloskey et al., 1998). We also noted “How Hg was measured” (using radiolabeled Hg,
161 Atomic Absorption Fluorescence, etc.) to identify if there were temporal trends in
162 publications favoring any analytical technique. “Form of Hg” distinguished between studies

163 that explicitly measured MeHg (“MeHg” in the database) from those that measured total Hg
164 (“Hg”) in the database; papers that measured total Hg assumed that all of the Hg present was
165 inorganic Hg, however we do not specify these studies as measuring inorganic Hg since the
166 analysis did not distinguish between inorganic and other forms of Hg. Based on the results
167 reported in these papers and our results in this meta-analysis, it appears safe to assume that
168 these studies were measuring mostly inorganic Hg.

169 Exposure time, either through aqueous (“Aqueous exposure time (d)”) or dietary pathways
170 (“Dietary exposure time (d)”), and depuration time (“Depuration time (d)”) may impact
171 biokinetic parameter values; length of the experiment was found to impact Hg k_e values in
172 fish (Trudel and Rasmussen, 1997). If Hg is introduced via a dietary exposure route, the
173 choice of prey item (“Food item”) can impact biokinetic parameter values, especially AE, in
174 that artificial diets may be more or less digestible (leading to higher or lower AE estimates,
175 respectively) than natural prey items (Bowling et al., 2011). The exposure concentration of
176 Hg, either through water (“Aqueous [Hg]”) or food (“Food [Hg]”), can influence biokinetic
177 parameter values if any of these processes are not first-order. Organism size (“Organism
178 weight (g)” and “Organism length/size (cm)”) may impact biokinetic parameter values due to a
179 correlation between size and physiological rates, and multiple reviews of k_e values in fish
180 (Madenjian et al., 2021; Trudel and Rasmussen, 1997; Yao and Drouillard, 2019) also
181 identified the impact of organism size on parameter estimates. Temperature (“Temp (degC)”)
182 impacts physiological rates in complex ways, including altering biokinetic parameter values
183 intraspecifically (Denton and Burdon-Jones, 1981; Tsui and Wang, 2004). We also recorded if
184 the animals were acclimated and for how long (“Acclimation (yes/no)” and “Acclimation
185 duration”) along with the “Geographic Location” of where the organisms were collected or
186 sourced from, if reported.

187 2.2. Taxonomic matching and tree building

188 We compiled taxonomic information on organisms used in the reviewed articles containing
189 biokinetic data, with the level of identification differing across studies. We matched and
190 resolved scientific names to the Open Tree Taxonomy from the Open Tree of Life (Hinchliff
191 et al., 2015) using the R package “rotl” (Michonneau et al., 2016). Induced subtrees were
192 returned separately for the fish and invertebrate taxa to create phylogenetic trees with uniform
193 branch lengths. We visualized trees and their associated data using “ggtree” (Yu et al., 2017)
194 and “ggplot2” packages (Wickham, 2016).

195 2.3. Analysis of biokinetic parameters

196 For all analyses described in this manuscript, first we averaged values for each parameter
197 within the reporting article if different values were estimated based on factors/treatments that
198 were not selected for this study (Table S1). Our aim was to not bias the analyses with multiple
199 values from a single study. A good example of this is from Tsui and Wang (2005) which
200 tested the effect of Hg acclimation on the values of AE for multiple generations of *Daphnia*
201 *magna*. Since we did not distinguish generational exposure of Hg in this database, the 11
202 reported AE values in Tsui and Wang (2005) were combined into a single mean value for AE.
203 To compare biokinetic parameter values between Hg and MeHg within each database, we
204 conducted Wilcoxon rank sum tests in R version 4.3.1 (R Development Core Team, 2023).

205 We used regression tree analysis to assess the interacting and potentially non-linear variables
206 that were associated with the three biokinetic parameters of interest for fish and aquatic
207 invertebrates. This decision tree method is a type of machine learning technique that allows
208 for the analysis of variable importance and directional effects of many variables on a response
209 variable. Regression tree analysis is not constrained by the assumptions of a multiple linear
210 regression, as it can, for example, capture non-linear relationships and utilize both continuous
211 and discrete (i.e., categorical) predictor variables. We fitted a total of six regression trees, one
212 each for predicting AE, k_u , and k_e for fish and aquatic invertebrates separately. We included

213 14 potential predictor variables, which were selected based on established relevance for these
214 parameters as well as data availability across studies in the database: water type (categorical:
215 freshwater, brackish, or marine), type of study (categorical: field or lab), salinity (ppt),
216 pathway (categorical: water, diet, water+diet, injection, or oral), form of mercury (categorical:
217 Hg or MeHg), aqueous exposure time (days), dietary exposure time to free feed (days),
218 dietary exposure time after feeding (days), food Hg concentration ($\mu\text{g/g}$ wet weight prey),
219 aqueous Hg concentration ($\mu\text{g/L}$), depuration time (days), organism weight (g), organism
220 length (cm), and water temperature ($^{\circ}\text{C}$). Regression trees were pruned by selecting the model
221 with the lowest number of nodes that had a mean cross-validated error rate within ± 1 standard
222 error of the model with the overall lowest mean cross-validated error. All results presented
223 here reflect pruned regression trees. Regression tree analysis was conducted using the “rpart”
224 package (Therneau and Atkinson, 2022) and figures generated using the “ggplot2” package
225 (Wickham, 2016) in R version 4.3.0 (R Development Core Team, 2023).

226 **3. Results/discussion**

227 3.1. Database size and characteristics

228 A summary of the data presented in the database is presented in Table 1 and the database
229 itself is publicly available at doi: <https://doi.org/10.5281/zenodo.14503336>. Table 1 reports
230 the median, sample size, and minimum and maximum values for the three focal parameters
231 for aquatic invertebrates and fish by the form of Hg. We calculated these summary statistics
232 on the database after the data were summarized by study (see details on this at the beginning
233 of Methods Section 2.1). The number of parameters values present in the database of fish and
234 aquatic invertebrate biokinetic parameters are reported in Table 2. The database contains
235 parameter values for organisms exposed to Hg or MeHg through dietary and aqueous
236 exposure routes at various temperatures, organism sizes, salinities, and experimental time
237 periods (Table 2). Some experiments were conducted in the field, while the majority were

238 conducted in the lab. The database is comprised of 84 publications (list in Table S2) with
239 publication dates ranging from 1970 to 2020. The database contains parameter values from 37
240 different species of bony fish and 34 different species of aquatic invertebrates which includes
241 insects, crustaceans, molluscs, and a segmented worm (list in Table S3). The study sites
242 represent 65 unique geographic locations; all from temperate regions in the northern
243 hemisphere. The majority occurred in the USA (26 sites), followed by China (11), Canada
244 (10), France (6), Finland (4), Denmark (3), England (2), Monaco (1), Norway (1), and
245 Portugal (1).

246 There are interesting trends in the rate of publication of the papers included in our database
247 (that include parameters we were searching for) – there was a regular pattern of a couple of
248 publications per year, especially for aquatic invertebrates, but studies of fish uptake and efflux
249 of Hg peaked in 1972, 1975, 2010, and 2012, and there was a break in relevant publications in
250 the early 1990s (Fig. S1). Most of the papers in the database used radiotracer techniques (65
251 %), which allows for non-destructive sampling of these organisms over time to measure Hg
252 uptake, assimilation, and efflux. The remaining 31 % (Fig. S2) of the studies use stable
253 isotopes of Hg which requires destructive sampling. The rise in papers from 2009 to 2012 is
254 mostly due to radiotracer experiments conducted in fish (Fig. S2).

255 3.2. Patterns in biokinetic parameter values

256 Fig. 1 illustrates that from the literature reviewed, k_u and AE are higher for MeHg than Hg for
257 both fish ($W = 34$, $p\text{-value} = 2.28e-07$ and $W = 511.5$, $p\text{-value} < 2.2e-16$ k_u and AE,
258 respectively) and aquatic invertebrates, although AE ($W = 20$, $p\text{-value} = 9.428e-08$) was
259 statistically significant and k_u was not ($W = 97$, $p\text{-value} = 0.12$). This pattern is consistent
260 with the literature (Bradley et al., 2017; Dutton and Fisher, 2010; Mason et al., 1996; Riisgård
261 and Hansen, 1990; Trudel and Rasmussen, 1997). Further, k_e was significantly slower for
262 MeHg than Hg in fish ($W = 3985$, $p\text{-value} = 8.80e-11$) and aquatic invertebrates ($W = 655$, $p\text{-}$

263 value = $2.85e-4$), also consistent with past reviews of efflux rates (Trudel and Rasmussen,
264 1997). This slower efflux for MeHg may be due to its affinity for sulfhydryl groups in
265 proteins (Houck and Cech Jr., 2004). Overall, this helps explain the higher accumulation of
266 MeHg in organisms, especially fish, than Hg (Trudel and Rasmussen, 1997).

267 3.3. Phylogenetic patterns

268 For fish, AE and k_e had the greatest sample size and taxonomic diversity, while k_u was less
269 available across taxa (Fig. 2). In addition, large differences were present between forms of Hg
270 in some but not all taxa, particularly in k_e (Fig. 2). The highest k_u values were found in the
271 grunt, *Plectorhinchus gibbosus*, and the western mosquitofish, *Gambusia affinis*, for MeHg
272 and Hg, respectively, while several species showed very low k_u for both forms of Hg (Fig. 2).
273 MeHg AE were generally high for most species, though much lower in the Atlantic salmon,
274 *Salmo salar*, while Hg AE were generally much lower than for MeHg (Fig. 2). The highest k_e
275 were found in the western mosquitofish, *Gambusia affinis*, and the goldfish, *Carassius*
276 *auratus*, for Hg and MeHg, respectively, while the lowest k_e were found in the lake trout,
277 *Salvelinus namaycush*, and the northern pike, *Esox lucius*, for MeHg and Hg, respectively
278 (Fig. 2).

279 Phylogenetic diversity was much higher for invertebrates in the database than the fish, which
280 offered a better opportunity for assessing variability in Hg biokinetic parameters.
281 Unfortunately, the total number of studies and biokinetic parameters available for each
282 species were lower than the fish, despite the greater invertebrate species diversity available
283 (i.e., many invertebrate species had only one parameter value from one study). Invertebrates
284 present in the database can be broken into six general taxonomic groups shown in Fig. 3: (A)
285 planktonic crustaceans (e.g., copepods and daphnids), (B) aquatic insects; (C) benthic
286 crustaceans (e.g., shrimps, crabs, and amphipods); (D) bivalve molluscs (e.g., oysters,
287 mussels, scallops, and clams); (E) cephalopod molluscs (e.g., cuttlefish); and (F) benthic

288 segmented worm (e.g., sandworm). The number of species present within each of these
289 groups ranged from a single species (F) to 13 species of bivalve molluscs (D). Some of the
290 highest uptake rates (k_u) were found in bivalve molluscs (D), particularly for MeHg, while
291 data was sparse (only a single study) for benthic crustaceans (C; Fig. 3). The lowest uptake
292 rates (k_u) were found in the daphnid, *Daphnia magna*, for both forms of Hg. Assimilation
293 efficiency (AE) was generally related to the form of Hg, with higher efficiencies shown for
294 MeHg, though efficiencies varied across taxa and no AE estimates were available for aquatic
295 insects (B; Fig. 3). The highest AE values were found in the planktonic crustaceans (A) and
296 the Pacific oyster, *Magallana gigas*, for MeHg and Hg, respectively, while the lowest AE
297 values for both forms of Hg were found in the blue mussel, *Mytilus edulis* (Fig. 3). The
298 highest efflux rates (k_e) were found in the aquatic insects (B), especially the chironomid
299 *Chironomus riparius*, while the highest MeHg k_e were found in the two daphnids, *Daphnia*
300 *magna* and *D. pulex* (Fig. 3). The lowest k_e were found in the benthic crustaceans (C) and
301 bivalve molluscs (D) for MeHg and Hg, respectively (Fig. 3). It should be noted that no
302 studies were found for AE and MeHg uptake and efflux in aquatic insects (B), no uptake
303 studies for most benthic crustaceans (C; the lone exception being the amphipod *Leptocheirus*
304 *plumulosus*), and no uptake or efflux studies were found for copepods.

305 Further phylogenetic analysis of this data set could identify nodes at which species diverge
306 from each other in terms of parameter values of Hg/MeHg biokinetic parameters, lending
307 more information on why Hg accumulates in certain species and their associated habitats.
308 This analysis could also potentially help identify key Hg bioaccumulating species that should
309 be identified as of particular human health risk if consumed and remediation techniques
310 resource managers could attempt.

311 3.4. Regression tree analysis

312 Results from all six regression trees are summarized in Table 3, including sample size,
313 explanatory power, and most important variables (listed in hierarchical order from most to
314 least important).

315 For three of the six regression trees, form of Hg was the sole important predictor for the
316 biokinetic parameter value (AE for fish and invertebrates and k_u for fish). For both aquatic
317 invertebrates and fish, MeHg was associated with a significantly higher average AE compared
318 to Hg (78 % versus 26 % in aquatic invertebrates and 71 % versus 27 % in fish; Fig. S4). For
319 fish k_u , form of Hg was also the sole important predictor variable (Fig. S3), with MeHg
320 leading to higher average uptake rates ($0.1 \text{ L g}^{-1} \text{ day}^{-1}$) compared to Hg ($0.01 \text{ L g}^{-1} \text{ day}^{-1}$).
321 Aquatic invertebrate k_u was also related to only one predictor variable, in this instance
322 aqueous exposure time. Here, aqueous exposure time < 0.67 days (i.e., 16 h) was associated
323 with very low mean k_u ($0.008 \text{ L g}^{-1} \text{ day}^{-1}$) compared to longer aqueous exposure times >0.67
324 days (mean $k_u = 0.9 \text{ L g}^{-1} \text{ day}^{-1}$), though notably this shorter exposure time was only ever
325 recorded in Anomopoda.

326 For both aquatic invertebrates and fish, k_e was predicted by a combination of two predictor
327 variables: water type followed by form of Hg in aquatic invertebrates (Fig. 4a) and organism
328 weight followed by form of Hg in fish (Fig. 4b). For aquatic invertebrates, brackish and
329 freshwater environments resulted in the highest mean k_e (0.07 day^{-1}), regardless of form of
330 Hg. However, in marine environments, form of Hg further distinguished grouping of k_e , with
331 MeHg in marine environments leading to higher average k_e (0.004 day^{-1}) compared to Hg in
332 marine environments (0.03 day^{-1}). In fish, heavy organisms ≥ 95 g had the lowest mean k_e
333 (0.001 day^{-1}) regardless of form of Hg. However, in smaller organisms < 95 g, form of Hg
334 influenced k_e . In these smaller organisms, if the form was Hg, average k_e was 0.04 day^{-1} . On
335 the other hand, if the form was MeHg in these smaller fish, then weight further influenced
336 their k_e . Very small fish < 11 g exposed to MeHg had k_e averaging 0.01 day^{-1} , but when
337 weight is between 11 and 95 g under MeHg exposure, average k_e was 0.005 day^{-1} .

338 3.5. Comparison to the literature

339 Form of Hg was the most important variable across all parameters and species; this variable
340 was selected by the regression tree analysis for all parameters except for k_u for invertebrates.
341 The importance of the form of Hg emphasizes a distinction already acknowledged in the field:
342 inorganic and organic Hg pose different bioaccumulation risks to aquatic organisms. The lack
343 of form of Hg as the most important predictor variable is more distinct: we were surprised that
344 form of Hg was not an important predictor for one parameter (k_u for invertebrates) and not the
345 most important variable for k_e in invertebrates and fish.

346 Further, the limited number of important variables in the regression trees was surprising, since
347 we chose the explanatory variables for the database because these variables had been either
348 empirically shown to influence or hypothesized to influence the focal biokinetic parameters.
349 However, by bringing together data from a wide variety of species in terms of their life
350 history traits, thermal regimes, feeding strategies, etc., we found that the only variables that
351 significantly predicted the values of the focal biokinetic parameters were form of Hg, aqueous
352 exposure time, water type, and organism weight (Table 3). This differs from the literature in
353 quite a few ways. Salinity, temperature, and organism size were the most frequently reported
354 variables that influence uptake rate, assimilation efficiency, and/or efflux rate during our
355 literature review, and organismal weight and salinity (broadly defined) were the only
356 significant predictor variables of that group (for k_e for fish and aquatic invertebrates,
357 respectively). It is also worth noting that discrete categories of salinity coded as “freshwater”
358 (0 ppt), “brackish” (0.5–30 ppt), and “marine” (>30 ppt) impacted invertebrate k_e , but
359 continuous salinity values did not. This is particularly interesting as there was only one study
360 for which we could not find a relevant salinity value either in the paper itself or in the
361 literature, so this is not due to the data set being larger for the coded salinity column than the
362 continuous numeric value salinity column. This result indicates that a simpler discrete
363 classification of salinity (freshwater, brackish, and marine) gives equal or possibly even better

364 predictive power than the continuous salinity variable. The regression trees indicate that we
365 can successfully predict k_e knowing the salinity category and the added data resolution of the
366 continuous salinity variable did not add predictive power.

367 Numerous studies have shown that biokinetic parameter values of Hg and MeHg are
368 influenced by salinity, including increased uptake of Hg at lower salinities in fish (Wang and
369 Wang, 2010) and oysters (Denton and Burdon-Jones, 1981). The same studies did not find an
370 effect of salinity on Hg or MeHg AE or k_e in fish (Wang and Wang, 2010) but did find an
371 impact on k_e in oysters (Denton and Burdon-Jones, 1981), as we found in aquatic
372 invertebrates collectively. Studies have hypothesized that marine and freshwater fish may
373 accumulate and efflux metals differently due to osmoregulatory differences (Pickhardt et al.,
374 2006). Marine fish may take up metals more quickly than freshwater as marine fish may have
375 adapted more efficient or rapid ion uptake (Dutton and Fisher, 2011; Wang and Wang, 2010).
376 Similarly, marine fish must compensate for water loss from their tissues and active salt
377 elimination, so metal efflux values may be larger for marine compared to freshwater fish
378 (Dutton and Fisher, 2011; Pickhardt et al., 2006). However, salinity was not an important
379 predictor for any of the biokinetic parameters for fish in our analysis (Table 3). Further, we
380 found the opposite pattern for aquatic invertebrates – marine invertebrates had lower k_e values
381 than freshwater or brackish studies.

382 Organism size has also been found to influence biokinetic parameters of Hg and MeHg,
383 commonly due to the correlation between size and physiological rates (metabolic rate,
384 ingestion rate, etc.). Uptake rate of Hg has been proposed to increase with decreasing size due
385 to smaller organisms having higher metabolic and ingestion rates per unit biomass, leading to
386 higher rates of digestion and thus Hg accumulation in fish (Dang and Wang, 2012; Leaner and
387 Mason, 2004) and mussels (Riisgård and Hansen, 1990). Similarly, k_e can vary with organism
388 size: reviews of mercury mass balance models have found that body size is an important
389 predictor for k_e estimates (Madenjian et al., 2021; Trudel and Rasmussen, 1997; Yao and

390 Drouillard, 2019). Our analysis supports these findings, showing that organism weight was
391 the most influential predictor variable for fish k_e , even more important than the form of Hg,
392 and smaller body size was associated with higher k_e . However, no organism size variables
393 (length, weight) were important predictors for any other fish parameter nor for any parameters
394 for aquatic invertebrates.

395 Temperature did not impact the values of AE, k_e , or k_u for aquatic invertebrates nor fish in the
396 regression tree analyses. This is despite numerous studies finding temperature having a
397 significant impact on Hg k_u (Denton and Burdon-Jones, 1981; Tsui and Wang, 2004) and k_e
398 (Fowler et al., 1978; Madenjian et al., 2021; Trudel and Rasmussen, 1997; Yao and
399 Drouillard, 2019), though one study found AE specifically was not impacted by temperature
400 (Tsui and Wang, 2004). Temperature is hypothesized to affect these rates because it increases
401 metabolic rates within an organism's optimal thermal window; there is likely a non-linear
402 relationship between temperature and mercury uptake that tracks the organisms' metabolic
403 rate (Denton and Burdon-Jones, 1981; Yao and Drouillard, 2019). Metabolic rate will increase
404 with temperature up to the organism's optimal temperature at which its metabolic rate is
405 highest; metal uptake rate may track metabolic rate up to that optimum, especially since other
406 physiological processes such as feeding/ filtering, oxygen consumption, and other
407 physiological rates increase with temperature and similarly increase uptake. Above this
408 optimum, metabolic rate and thus uptake rate decline (Denton and Burdon-Jones, 1981; Yao
409 and Drouillard, 2019). Interestingly, a study comparing uptake and efflux rates of Hg,
410 cadmium (Cd), and lead (Pb) in oysters found that temperature impacted Hg and Cd
411 accumulation more than for Pb, implying that Hg uptake may be more affected by
412 temperature than other metals (Denton and Burdon-Jones, 1981). Similarly, a study that found
413 increasing temperature increased uptake of Hg and MeHg in fish intestines hypothesized that
414 this result implies that the mechanisms driving uptake or assimilation of mercury may require
415 metabolic energy (Leaner and Mason, 2002). In this large review across many species, we did

416 not find that temperature was an important predictor of these biokinetic parameters, which
417 may be obscured by species-specific thermal tolerances. Explicit phylogenetic analysis
418 (statistically controlling for species in some way) may reveal an influence of temperature on
419 these parameters, however that analysis is outside the scope of the present study.

420 Temperature has also been identified as impacting Hg biomagnification across trophic levels
421 in contaminated systems globally. A meta-analysis of Trophic Magnification Slopes (TMS, a
422 marker of biomagnification potential) for Hg and MeHg found that the mean TMS value for
423 MeHg was 1.5 times greater than Hg. This finding is in agreement with our analysis that
424 MeHg biokinetic parameter values lead to higher MeHg bioaccumulation compared to Hg
425 (higher k_u and AE and lower k_e ; Fig. 1) and that the form of Hg was the most important
426 variable for most biokinetic parameter values (Table 3). The authors also found that both Hg
427 and MeHg TMS values showed a significant positive correlation with latitude, higher in polar
428 and temperate sites and lower in tropical sites (Lavoie et al., 2013). The authors propose that
429 this could be due to temperature, with higher temperatures decreasing Hg body burdens by
430 increasing growth and colder temperatures decreasing efflux rates. However, our analysis
431 would indicate that their temperature hypothesis is unlikely, as we did not see an impact of
432 temperature on any biokinetic parameter value, and instead the correlation is more likely due
433 to the authors' third hypothesis, that more complex food webs at lower latitudes compared to
434 higher reduces Hg trophic transfer efficiency (Lavoie et al., 2013).

435 Aqueous exposure time explained values of k_u for aquatic invertebrates, and interestingly this
436 is the only biokinetic parameter which was not influenced at all by the form of Hg (Table 3).
437 This is likely due to the influence of one study that measured the effect of temperature on
438 accumulation and elimination of Hg and MeHg in *Daphnia magna* (Tsui and Wang, 2004).
439 This study was the only study of *Daphnia* k_u , and reported eight k_u values after short-term
440 exposure (8 h), while the average aqueous exposure time for all k_u estimates for aquatic
441 invertebrates in the database was 5.83 days. Hence, it is unclear if our analysis highlights

442 aqueous exposure time specifically or the uniqueness of *Daphnia* as the primary driver of k_u in
443 invertebrates.

444 Species-specific characteristics have been shown to impact biokinetic parameter estimates for
445 Hg and MeHg. Characteristics like growth rate (Baumann et al., 2017; Peng et al., 2016), in
446 which higher growth rates lead to higher uptake due to increased ingestion or lower Hg
447 accumulation due to dilution due to growth. Similarly, differences in growth efficiencies or
448 food conversion efficiencies, which can vary inter- or intra-specifically, have also been shown
449 to impact mercury accumulation (Baumann et al., 2017). Trophic strategy may also impact
450 mercury accumulation – one study proposed that herbivorous fish may have higher k_e values
451 for Hg, leading to lower tissue concentrations (Peng et al., 2016). This may be due to
452 differences in gut retention time and/or gut passage times, allowing for higher or lower Hg
453 and MeHg assimilation from food (Dutton and Fisher, 2010; Wang, 2012). Gut passage time
454 can also vary with prey item (Bradley et al., 2017), and numerous studies hypothesized that
455 the amount of Hg assimilated from a prey item depends on how easily digested the prey is:
456 AE in fish can be related to the concentration of Hg associated with the soft body of the prey
457 (Dutton and Fisher, 2011) and organisms fed artificial diets that are less digestible could
458 underrepresent AE values from prey in natural systems (Bowling et al., 2011).

459 Other variables that have altered biokinetic parameter estimates in other studies include the
460 exposure concentration of Hg/MeHg and pathway or method of introduction of Hg. Most
461 studies found that the concentration of Hg/MeHg did not impact k_e , lending evidence to the
462 claim that Hg elimination is a first-order process (Li et al., 2015; Trudel and Rasmussen,
463 1997). AE was sometimes found to be correlated to Hg concentration (Zhong and Wang,
464 2006) and sometimes not (Wang, 2012). Previous studies hypothesized that force-feeding or
465 gavage/intubating fish lead to decreased AEs (Dutton and Fisher, 2012; McCloskey et al.,
466 1998), however neither exposure concentration nor pathway of introduction were significant
467 predictors of biokinetic parameters (Table 3). Some factors that we did not explicitly consider

468 that have been shown to impact biokinetic parameter estimates include sex (Madenjian et al.,
469 2014; Madenjian et al., 2021), or other factors influencing Hg bioavailability such as DOC
470 (Wang and Wang, 2010) and co-exposure with other contaminants such as selenium (Wang et
471 al., 2004). Indeed, some of these variables, such as DOC, and others not considered among
472 the papers reviewed, such as phosphorus concentrations, were shown to significantly impact
473 TMS values of Hg in freshwater locations (Lavoie et al., 2013).

474 Analysis of our database identified many variables traditionally found to influence
475 intraspecies Hg/MeHg bioaccumulation as not significantly impacting interspecific variation
476 in Hg/MeHg biokinetic parameter estimates across fish and aquatic invertebrates. This may
477 indicate that Hg/MeHg bioaccumulation is not influenced directly by environmental and
478 physiological factors previously studied, but rather these factors drive mercury methylation,
479 which explains how Hg so readily bioaccumulates in food chains and warrants further
480 analysis. An additional avenue for future work would involve more in-depth phylogenetic
481 analysis to identify key Hg bioaccumulating species or investigate how Hg/MeHg biokinetic
482 parameters have diverged evolutionarily.

483 **4. Conclusion**

484 Collection of bioaccumulation parameter values enabled the analysis of patterns of Hg and
485 MeHg uptake, assimilation, and efflux across aquatic species to identify important variables
486 driving Hg accumulation. Overall, we found that most of the variables that have traditionally
487 been studied as significantly impacting Hg and MeHg accumulation do not have a significant
488 influence on these parameters across a wider taxonomic scale (e.g. across fish species and
489 across aquatic invertebrates). For most parameters, only the distinction between Hg and
490 MeHg was a significant predictor of the parameter value. Interestingly, organism weight was
491 a significant predictor variable for fish efflux rates, and aqueous exposure time and water
492 salinity type were significant predictor variables for aquatic invertebrate uptake and efflux

493 rates, respectively. Overall, this analysis can help develop more accurate biodynamic or
494 bioenergetic models of Hg dynamics in aquatic systems by identifying which factors are
495 important to incorporate into these predictive models.

496 **Acknowledgments**

497 This work was funded by the US Department of Energy's Oak Ridge Office of Environmental
498 Management (ORO DOE) and URS CH2M Oak Ridge LLC (UCOR) and is a product of
499 ORNL's Mercury Remediation Technology Development Program. We would like to thank
500 Sujithkumar Surendrannair for his comments on the manuscript and Adam Malin (Oak Ridge
501 National Lab, Creative Services) and Delaney Bellis (Oak Ridge National Lab) for help with
502 the graphical abstract. This manuscript has been authored by UT-Battelle, LLC, under
503 contract DE-AC05-00OR22725 with the US Department of Energy (DOE). The US
504 government retains and the publisher, by accepting the work for publication, acknowledges
505 that the US government retains a non-exclusive, paid-up, irrevocable, world-wide license to
506 publish or reproduce the submitted manuscript version of this work, or allow others to do so,
507 for US government purposes. DOE will provide public access to these results of federally
508 sponsored research in accordance with the DOE Public Access Plan (<https://energy.gov/doi-public-access-plan>).
509

510 **References**

- 511 Baumann, Z., Mason, R.P., Conover, D.O., Balcom, P., Chen, C.Y., Buckman, K.L., et al.,
512 2017. Mercury bioaccumulation increases with latitude in a coastal marine fish (Atlantic
513 silverside, *Menidia menidia*). *Can. J. Fish. Aquat. Sci.* 74, 1009–1015.
- 514 Bowling, A.M., Hammerschmidt, C.R., Oris, J.T., 2011. Necrophagy by a benthic omnivore
515 influences biomagnification of methylmercury in fish. *Aquat. Toxicol.* 102, 134–141.
- 516 Bradley, M.A., Barst, B.D., Basu, N., 2017. A review of mercury bioavailability in humans
517 and fish. *Int. J. Environ. Res. Public Health* 14.

518 Clarke, R.G., Klapstein, S.J., Keenan, R., O'Driscoll, N.J., 2023. Salinity and total suspended
519 solids control mercury speciation in a tidal river: comparisons with a photochemical
520 mercury model. *Chemosphere* 344, 140313.

521 Dang, F., Wang, W.X., 2012. Why mercury concentration increases with fish size? Biokinetic
522 explanation. *Environ Pollut* 163, 192–198.

523 Denton, Burdon-Jones, 1981. Influence of Temperature and Salinity on the Uptake,
524 Distribution and Deputation of Mercury, Cadmium and Lead by the Black-lip Oyster
525 *Saccostrea echinata*.

526 Dutton, J., Fisher, N.S., 2010. Intraspecific comparisons of metal bioaccumulation in the
527 juvenile Atlantic silverside. *Aquat. Biol.* 10, 211–226.

528 Dutton, J., Fisher, N.S., 2011. Bioaccumulation of As, Cd, Cr, Hg(II), and MeHg in killifish
529 (*Fundulus heteroclitus*) from amphipod and worm prey. *Sci. Total Environ.* 409, 3438–
530 3447.

531 Dutton, J., Fisher, N.S., 2012. Bioavailability of sediment-bound and algal metals to killifish.
532 *Aquat. Biol.* 16, 85–96.

533 Environment U, 2019. In: Branch UEPCaH (Ed.), *Global Mercury Assessment 2018*. Geneva,
534 Switzerland.

535 Fowler, S.W., Heyraud, M., Larosa, J., 1978. Factors affecting methyl and inorganic mercury
536 dynamics in mussels and shrimp. *Mar. Biol.* 46, 267–276.

537 Hinchliff, C.E., Smith, S.A., Allman, J.F., Burleigh, J.G., Chaudhary, R., Coghill, L.M., et al.,
538 2015. Synthesis of phylogeny and taxonomy into a comprehensive tree of life. *Proc. Natl.*
539 *Acad. Sci. U. S. A.* 112, 12764–12769.

540 Houck, A., Cech Jr., J.J., 2004. Effects of dietary methylmercury on juvenile Sacramento
541 blackfish bioenergetics. *Aquat. Toxicol.* 69, 107–123.

542 Kim, E.-H., Mason, R.P., Porter, E.T., Soulen, H.L., 2004. The effect of resuspension on the
543 fate of total mercury and methyl mercury in a shallow estuarine ecosystem: a mesocosm
544 study. *Mar. Chem.* 86, 121–137.

545 Lavoie, R.A., Jardine, T.D., Chumchal, M.M., Kidd, K.A., Campbell, L.M., 2013.
546 Biomagnification of mercury in aquatic food webs: a worldwide meta-analysis. *Environ.*
547 *Sci. Technol.* 47 (23), 13385–13394.

548 Leaner, J.J., Mason, R.P., 2002. Factors controlling the bioavailability of ingested
549 methylmercury to channel catfish and Atlantic sturgeon. *Environ. Sci. Technol.* 36, 5124–
550 5129.

551 Leaner, J.J., Mason, R.P., 2004. Methylmercury uptake and distribution kinetics in
552 sheepshead minnows, *Cyprinodon variegatus*, after exposure to CH_3Hg -spiked food.
553 *Environ. Toxicol. Chem.* 23, 2138–2146.

554 Li, J., Drouillard, K.G., Branfireun, B., Haffner, G.D., 2015. Comparison of the toxicokinetics
555 and bioaccumulation potential of mercury and polychlorinated biphenyls in goldfish
556 (*Carassius auratus*). *Environ. Sci. Technol.* 49, 11019–11027.

557 Luoma, S.N., Rainbow, P.S., 2005. Why is metal bioaccumulation so variable? Biodynamics
558 as a unifying concept. *Environ. Sci. Technol.* 39, 1921–1931.

559 Luoma, S.N., Johns, C., Fisher, N.S., Steinberg, N.A., Oremland, R.S., Reinfelder, J.R., 1992.
560 Determination of selenium bioavailability to a benthic bivalve from particulate and solute
561 pathways. *Environ. Sci. Technol.* 26 (3), 485–491.

562 Madenjian, C.P., Blanchfield, P.J., Hrenchuk, L.E., Van Walleghem, J.L., 2014. Mercury
563 elimination rates for adult northern pike *Esox lucius*: evidence for a sex effect. *Bull.*
564 *Environ. Contam. Toxicol.* 93, 144–148.

565 Madenjian, C.P., Chipps, S.R., Blanchfield, P.J., 2021. Time to refine mercury mass balance
566 models for fish. *Facets* 6, 272–286.

567 Mason, R.P., Reinfelder, J.R., Morel, F.M.M., 1996. Uptake, toxicity, and trophic transfer of
568 mercury in a coastal diatom. *Environ. Sci. Technol.* 30, 1835–1845.

569 McCloskey, J.T., Schultz, I.R., Newman, M.C., 1998. Estimating the oral bioavailability of
570 methylmercury to channel catfish (*Ictalurus punctatus*). *Environmental Toxicology and*
571 *Chemistry: An International Journal* 17, 1524–1529.

572 Michonneau, F., Brown, J.W., Winter, D.J., 2016. rotl: an R package to interact with the Open
573 Tree of Life data. *Methods Ecol. Evol.* 7, 1476–1481.

574 Peng, X., Liu, F., Wang, W.X., 2016. Organ-specific accumulation, transportation, and
575 elimination of methylmercury and inorganic mercury in a low Hg accumulating fish.
576 *Environ. Toxicol. Chem.* 35, 2074–2083.

577 Pickhardt, P.C., Stepanova, M., Fisher, N.S., 2006. Contrasting uptake routes and tissue
578 distributions of inorganic and methylmercury in mosquitofish (*Gambusia affinis*) and
579 redear sunfish (*Lepomis microlophus*). *Environmental Toxicology and Chemistry: An*
580 *International Journal* 25, 2132–2142.

581 R Development Core Team, 2023. R: A Language and Environment for Statistical
582 Computing. R Foundation for Statistical Computing, Vienna, Austria.

583 Reinfelder, J.R., Fisher, N.S., Luoma, S.N., Nichols, J.W., Wang, W.-X., 1998. Trace element
584 trophic transfer in aquatic organisms: a critique of the kinetic model approach. *Sci. Total*
585 *Environ.* 219, 117–135.

586 Riisgård, H.U., Hansen, S., 1990. Biomagnification of mercury in a marine grazing food-
587 chain: algal cells *Phaeodactylum tricornutum*, mussels *Mytilus edulis* and flounders
588 *Platichthys flesus* studied by means of a stepwise-reduction-CVAA method. *Mar.*

589 Ecol. Prog. Ser. 259–270.

590 Schlekat, C., Lee, B., Luoma, S., 2002. Dietary Metals Exposure and Toxicity to Aquatic
591 Organisms: Implications for Ecological Risk Assessment. CRC Press, Boca Raton, FL,
592 pp. 151–188.

593 Spry, D.J., Wiener, J.G., 1991. Metal bioavailability and toxicity to fish in low-alkalinity
594 lakes: a critical review. Environ. Pollut. 71, 243–304.

595 Therneau, T., Atkinson, B., 2022. rpart: Recursive Partitioning and Regression Trees. R
596 package version 4.1.19.

597 Trudel, M., Rasmussen, J.B., 1997. Modeling the elimination of mercury by fish. Environ.
598 Sci. Technol. 31, 1716–1722.

599 Tsui, M.T., Wang, W.X., 2004. Temperature influences on the accumulation and elimination
600 of mercury in a freshwater cladoceran, *Daphnia magna*. Aquat Toxicol 70, 245–256.

601 Tsui, M.T., Wang, W.X., 2005. Multigenerational acclimation of *Daphnia magna* to mercury:
602 relationships between biokinetics and toxicity. Environ. Toxicol. Chem. 24, 2927–2933.

603 Van Walleggem, J.L., Blanchfield, P.J., Hrenchuk, L.E., Hintelmann, H., 2013. Mercury
604 elimination by a top predator, *Esox lucius*. Environmental Science & Technology 47,
605 4147–4154.

606 Wang, W.-X., 2012. Biodynamic Understanding of Mercury Bioaccumulation in Freshwater
607 Tilapia (*Oreochromis niloticus*).

608 Wang, R., Wang, W.-X., 2010. Importance of speciation in understanding mercury
609 bioaccumulation in tilapia controlled by salinity and dissolved organic matter. Environ.
610 Sci. Technol. 44, 7964–7969.

611 Wang, W.X., Wong, R.S., Wang, J., Yen, Y.F., 2004. Influences of different selenium species
612 on the uptake and assimilation of Hg(II) and methylmercury by diatoms and green mussels.
613 *Aquat. Toxicol.* 68, 39–50.

614 Watras, C.J., Back, R.C., Halvorsen, S., Hudson, R.J.M., Morrison, K.A., Wente, S.P., 1998.
615 Bioaccumulation of mercury in pelagic freshwater food webs. *Sci. Total Environ.* 219,
616 183–208.

617 Wickham, H., 2016. *ggplot2: Elegant Graphics for Data Analysis*. Springer-Verlag, New
618 York.

619 Yao, S., Drouillard, K.G., 2019. Prediction of mercury elimination rate coefficients of fish is
620 improved by incorporating fish temperature classification into models. *Bull. Environ.*
621 *Contam. Toxicol.* 103, 657–662.

622 Yu, G., Smith, D.K., Zhu, H., Guan, Y., Lam, T.T.Y., 2017. *ggtree*: an R package for
623 visualization and annotation of phylogenetic trees with their covariates and other
624 associated data. *Methods in Ecology and Evolution* 8, 28–36.

625 Zhong, H., Wang, W.-X., 2006. Metal–solid interactions controlling the bioavailability of
626 mercury from sediments to clams and sipunculans. *Environ. Sci. Technol.* 40, 3794–3799.

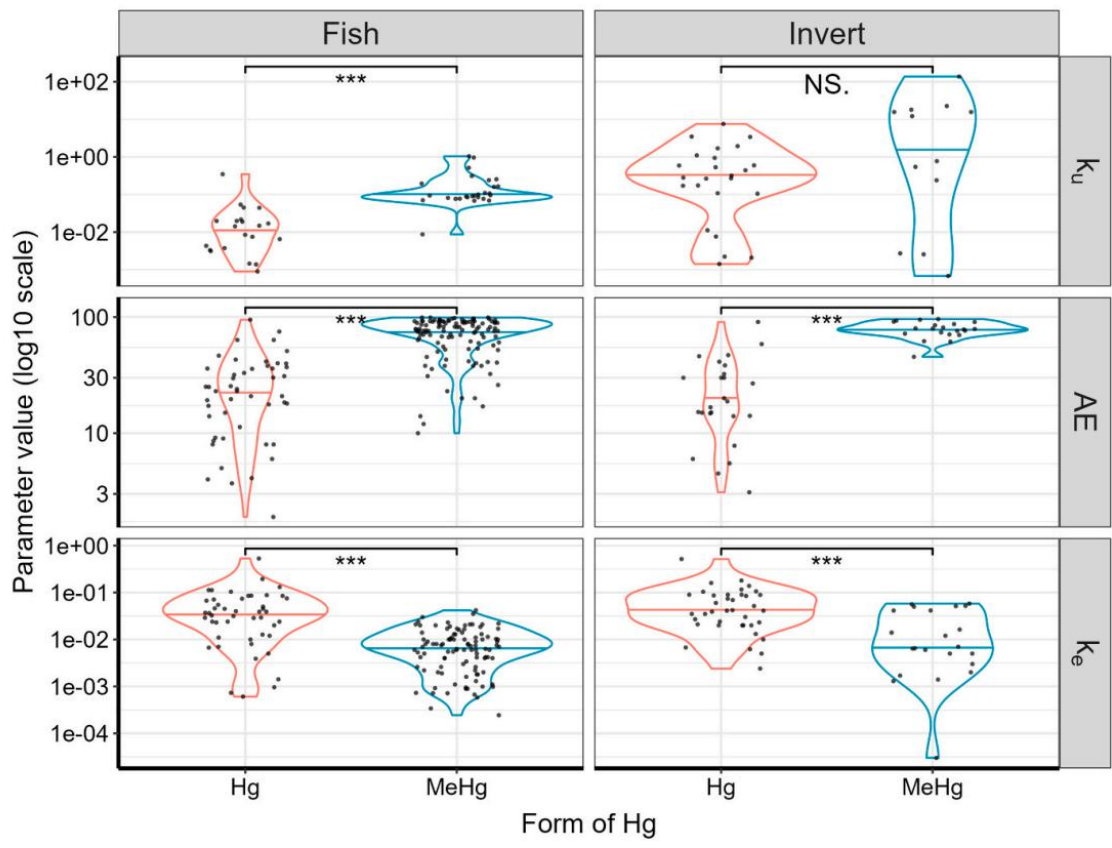
627 **Captions to figures**

628 Figure 1. All parameter values in database for fish (left panel) and aquatic invertebrates (right
629 panel). Note the y-axis is log-transformed. The data points represent every value in the
630 database and the violin plot denotes the distribution of the data with the horizontal line
631 representing the median value. We denote parameter values with statistically significant
632 values comparing Hg to MeHg using asterisks (***) indicates p-value < 1e-3 and “NS”
633 indicates no statistical difference).

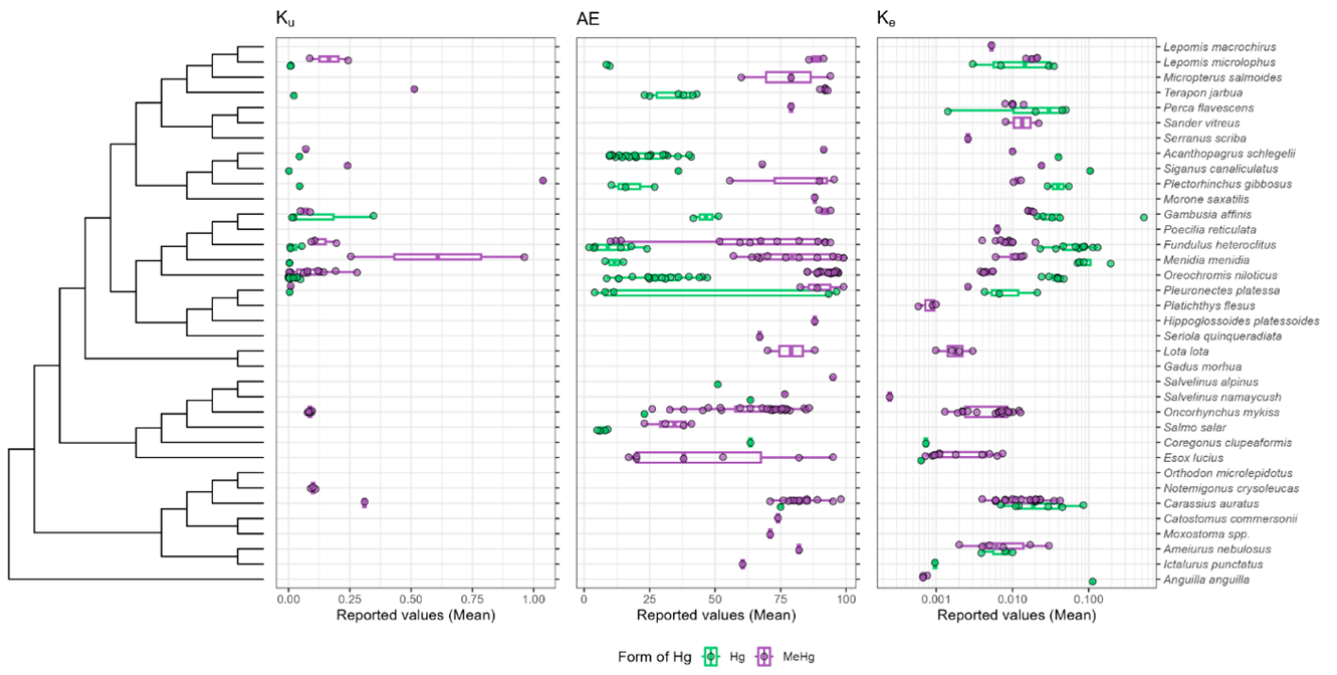
634 Figure 2. Uniform branch length phylogenetic tree for fish included in the database with
635 parameter values for Hg uptake (k_u), assimilation efficiency (AE), and efflux (k_e) from whole-
636 body samples. Fill and boxplot colors indicate the form of Hg (green, Hg; purple, MeHg).
637 Each point represents value for a single study. Note that values for k_e are presented in log
638 scale while the others are not.

639 Figure 3. Uniform branch length phylogenetic tree for invertebrates included in the database
640 with parameter values for Hg uptake (k_u), assimilation efficiency (AE), and efflux (k_e). Fill
641 and boxplot colors indicate the form of Hg (green, Hg; purple, MeHg). Each point represents
642 value for a single study. Letters indicate clades of similar taxa; (A) planktonic crustaceans
643 (e.g., copepods and daphnids), (B) aquatic insects; (C) benthic crustaceans (e.g., shrimps,
644 crabs, and amphipods); (D) bivalve molluscs (e.g., oysters, mussels, scallops, and clams); (E)
645 cephalopod molluscs (e.g., cuttlefish); and (F) benthic segmented worm (e.g., sandworm).
646 Note that values for k_e are presented in log scale while the others are not.

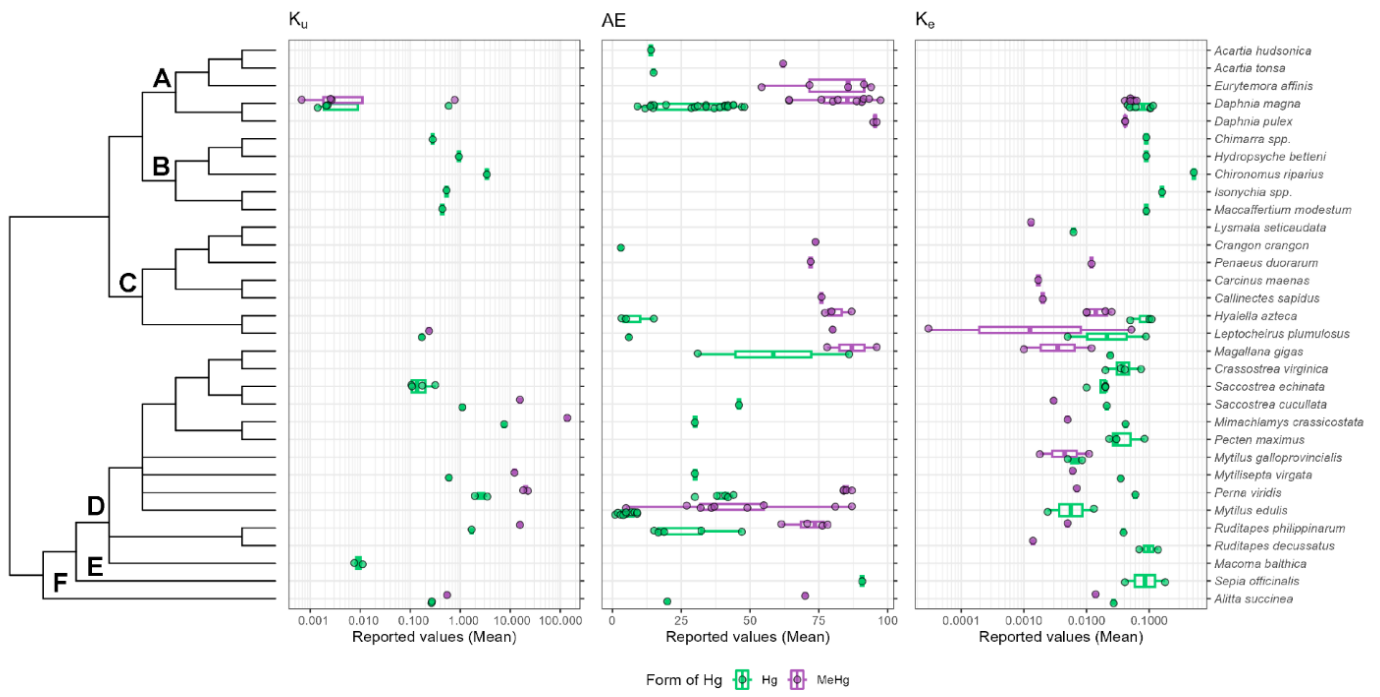
647 Figure 4. Pruned regression tress for k_e in (a) aquatic invertebrates and (b) fish as predicted by
648 multiple hierarchical variables. Terminal nodes list the mean k_e within each group, and the
649 percentage indicates the proportion of the data within each node. Violin plots and histograms
650 underneath each node display the spread of the data within each group and the taxonomic
651 groups within each.



652 Figure 1

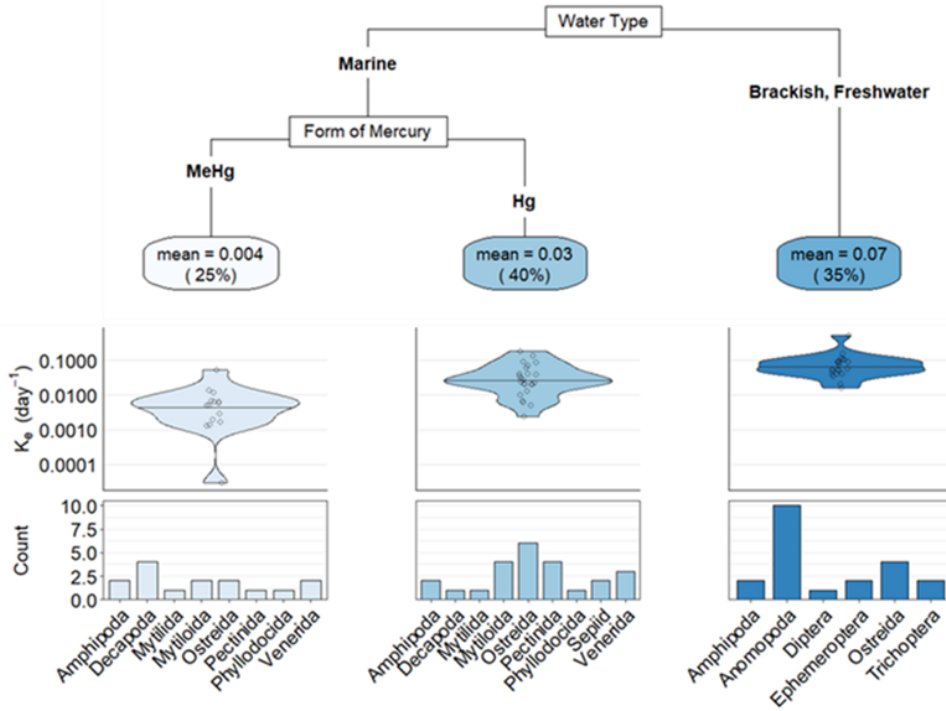


653 Figure 2

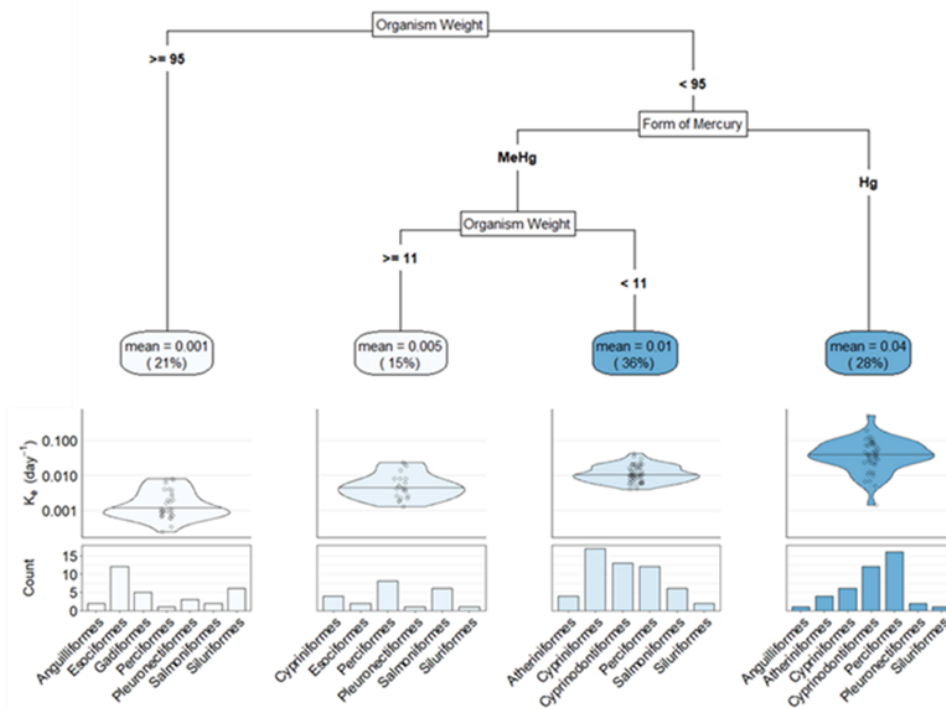


654 Figure 3

a) k_e Aquatic Invertebrate Pruned Regression Tree



b) k_e Fish Pruned Regression Tree



655 Figure 4

656 *Table 1. Summary of database, analyzed by study (see Methods). The data are reported as the*
 657 *median (sample size) with the (min-max) values of the parameter range reported below.*

Parameter	Aquatic invertebrates		Fish	
	Hg	MeHg	Hg	MeHg
k_u (L g ⁻¹ d ⁻¹)	2.98e-1 (24) (1.41e-3 – 7.54)	6.46 (12) (6.79e-4 – 1.37e2)	1.42e-2 (21) (6.96e-5 – 3.47e-1)	9.35e-2 (27) (2.21e-3 – 1.04)
AE (%)	18.8 (25) (1.0 – 90.7)	78.9 (21) (5.0 – 97.4)	23 (49) (1.9 - 96.3)	77.3 (114) (10 - 99)
k_e (d ⁻¹)	4.10e-2 (38) (2.40e-3 – 5.17e-1)	6.75e-3 (22) (3.00e-5 – 6.30e-2)	3.64e-2 (47) (5.81e-4 – 5.30e-1)	6.97e-3 (102) (2.44e-4 – 4.20e-2)

658

659 Table 2. Number of parameter values in databases of biokinetic parameters of Hg in fish and
 660 aquatic invertebrates. Note that this is the total number of parameter values; for the analysis,
 661 some were averaged if they differed in terms of variables that were not taken into account in
 662 our analysis (see details in Materials and Methods).

Parameter	Aquatic invertebrates			Fish		
	Hg	MeHg	Both	Hg	MeHg	Both
k_u	21	9	30	32	36	68
AE	40	54	94	77	150	227
k_e	28	24	52	59	164	223
Total	89	87	176	168	350	518

663

664 Table 3. Summary of pruned regression tree analysis for three biokinetic parameters for
 665 aquatic invertebrates and fish. Most important predictor variables are listed in descending
 666 order of importance.

	Bioaccumulation parameter	Sample size	Explanatory power (pseudo- R^2)	Most important predictor variables
Aquatic invertebrates	k_u	36	0.42	Aqueous exposure time
	AE	46	0.71	Form of mercury
	k_e	60	0.50	Water type, form of mercury
Fish	k_u	48	0.53	Form of mercury
	AE	163	0.47	Form of mercury
	k_e	149	0.66	Organism weight, form of mercury

667