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## 1 **Bioaccumulation of persistent organic pollutants in the deepest ocean fauna**

2 Alan J. Jamieson<sup>1\*</sup>, Tamas Malkocs<sup>2</sup>, Stuart B. Piertney<sup>2</sup>, Toyonobu Fujii<sup>1</sup>, Zulin Zhang<sup>3</sup>

3 <sup>1</sup>Oceanlab, Institute of Biological and Environmental Sciences, University of Aberdeen, Main Street,  
4 Newburgh, Aberdeenshire, AB41 6AA, UK.

5 <sup>2</sup>Institute of Biological and Environmental Sciences, University of Aberdeen, Zoology Building,  
6 Tillydrone Avenue, Aberdeen, AB24 2TZ, UK.

7 <sup>3</sup>The James Hutton Institute, Environmental and Biochemical Sciences, Craigiebuckler, Aberdeen,  
8 AB15 8QH, UK.

9 + Present address: School of Marine Science and Technology, Newcastle University, Ridley Building,  
10 Newcastle-Upon-Tyne, NE1 7RU, UK.

11 \*Corresponding Author: Jamieson, A.J. email: [alan.jamieson@ncl.ac.uk](mailto:alan.jamieson@ncl.ac.uk)

12 **The legacy and reach of anthropogenic influence is most clearly evidenced by its impact on the most**  
13 **remote and inaccessible habitats on Earth. Here we identify extraordinary levels of persistent**  
14 **organic pollutants in the endemic amphipod fauna from two of the deepest ocean trenches (>10,000**  
15 **metres). Contaminant levels were considerably higher than documented for nearby regions of**  
16 **heavy industrialisation, indicating bioaccumulation of anthropogenic contamination and inferring**  
17 **that these pollutants are pervasive across the World's oceans and to full ocean depth.**

18 The oceans comprise the largest biome on the planet with the deep ocean operating as a potential  
19 sink for the pollutants and litter that are discarded into the seas<sup>1</sup>. The spatial and bathymetric expanse  
20 of the deep sea infers that there are still large areas untouched by anthropogenic activity, although  
21 the intrinsic linkages between the deep sea and surface waters<sup>2</sup> would suggest this inference is ill-  
22 conceived<sup>3</sup>. The hadal zone (6000 to 11,000 m deep) is comprised of trenches formed at tectonic  
23 subduction zones and represents the least explored ecosystem on Earth and the last major marine  
24 ecological frontier<sup>4</sup>. Trenches have been considered both as pristine environments, but also given their  
25 locations and topography as likely sinks for contaminants that enter the marine environment<sup>2</sup>. Of  
26 particular concern are the persistent organic pollutants (POPs) that are highly detrimental to  
27 organismal health through their endocrine disrupting properties<sup>5</sup>. POPs possess an inherent  
28 hydrophobicity that confers a high binding affinity to organic or inorganic particles present in the  
29 water column that through vertical transport will collect in the deep ocean. They also have inherent  
30 lipophilicity, meaning these compounds readily bioaccumulate in organisms, with cumulative  
31 increases at each trophic level<sup>6</sup>.

32 POPs were released into the environment through industrial accidents and discharges, leakage from  
33 landfills or incomplete incineration<sup>7</sup>. Two key POPs are polychlorinated biphenyls (PCBs, used as  
34 dielectric fluid) and polybrominated diphenyl ethers (PBDEs, used as flame retardants). From the  
35 1930s to when PCB production ceased in the 1970s, the total global production was ~1.3 million  
36 tonnes<sup>8</sup>. Approximately 65% is thought to be contained in landfills or still within electrical equipment,  
37 with the other 35% residing in coastal sediments and open oceans<sup>9</sup>. These pollutants are invulnerable  
38 to natural degradation<sup>10</sup> and so persist in the environment for decades. Moreover they can spread  
39 great distances, including to seemingly isolated environments, such as polar regions and open ocean<sup>3</sup>.

40 Pollutants entering the deep sea are deposited in sediments and can readily accumulate into the food-  
41 chain<sup>11</sup>. Studies on deep-sea organisms have reported higher concentrations than in nearby surface  
42 water species<sup>11,12</sup>. However, although these studies are described as 'deep sea' they rarely extend  
43 beyond the continental shelf (<2000m), so contamination at greater distances from shore and the  
44 extreme depths is hitherto unknown.

45 We measured the concentrations of key PCBs and PBDEs in multiple endemic and ecologically  
46 equivalent Lysianassoid amphipod Crustacea from across two of the deepest hadal trenches - the  
47 oligotrophic Mariana Trench in the North Pacific, and the more eutrophic Kermadec in the South  
48 Pacific. Two endemic amphipods (*Hirondellea dubia* and *Bathycallisoma schellenbergi*) were sampled  
49 from the Kermadec between 7227 and 10,000 m, and one (*Hirondellea gigas*) from the Mariana  
50 between 7841 and 10,250 m. Samples were obtained using traps deployed on deep-sea landers<sup>13</sup>. The  
51 concentrations of seven PCB congeners identified by ICES<sup>14</sup> for marine pollution assessment and seven  
52 PBDE congeners were measured both in sample dry weight (dw) and lipid weight (lw).

53 The salient finding was that PCBs and PBDEs were present in all samples across all species at all depths  
54 in both trenches. The  $\Sigma$ PCB7 concentrations ranged from 147.3-905 ng g<sup>-1</sup> dw in the Mariana and  
55 18.03-42.85 ng g<sup>-1</sup> dw in the Kermadec, with mean values of 382.28 ng g<sup>-1</sup> dw  $\pm$ 281.6 S.D and 25.24 ng  
56 g<sup>-1</sup> dw  $\pm$  9.1 S.D respectively. Across individual PCB congeners, PCB 153 was detected in the highest  
57 concentration (mean 64.45; range 5.03–373.63 ng g<sup>-1</sup> dw). Congeners PCB 138 and 153 alone  
58 accounted for 65% of the total PCB concentration, which suggested the heavier congeners were more  
59 recalcitrant towards degradation.

60  $\Sigma$ PBDE7 concentrations ranged from 5.82-28.93 ng g<sup>-1</sup> dw in the Mariana Trench and 13.75-31.02 ng  
61 g<sup>-1</sup> dw in the Kermadec. Concentrations of congeners PBDE 153 and 154 were below limit of detection  
62 (LOD) in some samples, while PBDE 183 was not detected. Congener PBDE 47 and 99 were found the  
63 higher concentrations (mean: 8.81, range 2.55–21.36 ng g<sup>-1</sup> dw; mean 3.31, range 0.78-8.38 ng g<sup>-1</sup> dw  
64 respectively) and accounted for 71% of the total PBDE concentration.

65 For both  $\Sigma$ PCB7 and  $\Sigma$ PBDE7 there were significant correlations between dry and lipid weights  
66 (Pearson's  $r = 0.98$   $p = 8 \times 10^{-9}$  and  $r = 0.70$ ,  $p = 0.012$ , respectively). There was no statistically significant  
67 relationship between concentration and depth within either trench (see supplementary information).  
68 In both trenches the highest values were found in the upper trench; 7841 m in the Mariana and 7227  
69 m in the Kermadec.

70 These data clearly indicate that potent anthropogenic contamination and bioaccumulation has  
71 occurred in a dominant macrofaunal group inhabiting the complete depth range of two of the deepest  
72 marine trenches. Placing the levels of contamination into a broader comparative context with values  
73 published from the western Pacific is complicated by variations in POP congeners assayed across  
74 different studies and level of replication. Notwithstanding, some insightful comparisons are possible.  
75 World baseline levels for  $\Sigma$ PCBs arising from atmospheric transport found in clean coastal sediments  
76 are  $<1 \text{ ng g}^{-1} \text{ dw}^{15}$ . In grossly polluted areas, levels can be far higher<sup>16</sup>, reaching up to  $314 \text{ ng g}^{-1} \text{ dw}$  in  
77 in Guam,  $240 \text{ ng g}^{-1} \text{ dw}$  in Japan and  $160 \text{ ng g}^{-1} \text{ dw}$  in Australia. Indeed, in the Mariana, the highest  
78 level of PCBs were fifty times more contaminated than crabs from paddy fields fed by the Liaohe River,  
79 one of the most polluted rivers in China<sup>18</sup>. The only NW Pacific location with values comparable to the  
80 Mariana Trench is Suruga Bay (Japan), a highly industrialised area with historically heavy usage of  
81 organochlorine chemicals<sup>19</sup>.

82 Contamination by PBDEs did not reach the magnitude of PCB levels. The closest data for PBDE levels  
83 to the Kermadec Trench are estuarine sediments from New Zealand's North Island ( $0.55$  to  $573 \text{ ng g}^{-1}$   
84  $\text{dw}^{23}$ ). These results are however highly variable with some locations, such as Puketutu Island, being  
85 exceptionally high ( $>500 \text{ ng g}^{-1} \text{ dw}$ ). However, the median value of  $10.3 \text{ ng g}^{-1} \text{ dw}$  is considerably lower  
86 than the Kermadec Trench ( $18.4 \text{ ng g}^{-1} \text{ dw}$ ) and close to that in the Mariana Trench ( $10.44 \text{ ng g}^{-1} \text{ dw}$ ).  
87 The levels of PBDEs from New Zealand estuaries were not considered excessively high<sup>23</sup>.  
88 Notwithstanding, the salient finding is that PBDEs are present in the hadal samples at comparable or  
89 higher levels than in coastal waters.

90 PCB concentrations were notably higher in the Mariana than in the Kermadec Trench. There are  
91 several possible explanations that are not mutually exclusive and at this juncture speculative. First,  
92 the high levels of the Mariana PCBs may originate from proximity to the industrialised regions in the  
93 NW Pacific<sup>21</sup> and the North Pacific Subtropical Gyre, famed for its reputation as the 'Great Pacific  
94 Garbage Patch'<sup>22</sup>. As such, it is located beneath a mass accumulation of trapped plastic debris which  
95 ultimately sinks as the plastics degrade and fragment, transporting POPs to depth. Second, the  
96 amphipods may be accumulating POPs through surface derived carrion-falls that have experienced  
97 contrasting levels of contamination in the surface. This however appears unlikely given the consistent  
98 patterns of contamination at different depths and the oligotrophic nature of the Pacific around the

99 Mariana. Third, it may also be likely that the different species bioaccumulate these contaminants  
100 differently through physiological differences, lipid turnover rates, metabolic capacity, and ingestion  
101 throughput. Again, this is somewhat less likely given the ecological equivalence of the species though  
102 there is a lack of information on their respective physiologies.

103 The most parsimonious explanation for PCB and PBDE accumulate in these remote and deep trenches  
104 is via long range oceanic and atmospheric transport and association with particulate matter and  
105 carrion-falls sinking through the water column. The potentially rapid rate of vertical transportation of  
106 surface derived material was clearly demonstrated following the 2011 Fukushima Dai-ichi nuclear  
107 disaster where the detection of  $^{134}\text{Cs}$  radiation at 7553 m deep in the Japan Trench suggested a sinking  
108 rate of 64 to 78 m day<sup>-1</sup><sup>24</sup>. Therefore the travel time between the surface and seafloor at full ocean  
109 depth is between ~110 and 170 days, and likely shorter for carrion-falls. Given that  $370 \times 10^3$  t of PCBs  
110 are estimated to now reside in the oceans<sup>9</sup>, a rapid influx of surface material suggests little  
111 bathymetric variation of seafloor concentrations. Indeed, topographically closed trench morphologies  
112 may hamper contaminant dispersal.

113 The extreme hydrostatic pressures that characterise the hadal zone require non-trivial evolutionary  
114 adaptations for survival, and present major engineering challenges in accessing full ocean depth. Such  
115 statements underpin the perspective that the hadal zone is remote and inaccessible, with popularist  
116 analogies generally reinforcing this view, such that if Mount Everest was placed into the Mariana  
117 Trench its summit remains a mile below the surface. However, the distance from the surface to full  
118 ocean depth is actually only equal to the widest point of the Mississippi River, and half the length of  
119 Manhattan Island. These alternative views emphasize that our proximity to these extreme locations is  
120 far from remote, which is why even the deepest chasms of the ocean are no longer pristine. The  
121 challenge moving forward is to determine the physiological consequences of such contamination and  
122 understand knock-on effects on ecosystem function.

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129 **Author contributions.** A.J.J. conceived the experiment, designed the sampling equipment and was  
130 awarded the analytical costs. A.J.J. and S.B.P. performed the sampling at sea. S.B.P identified species,

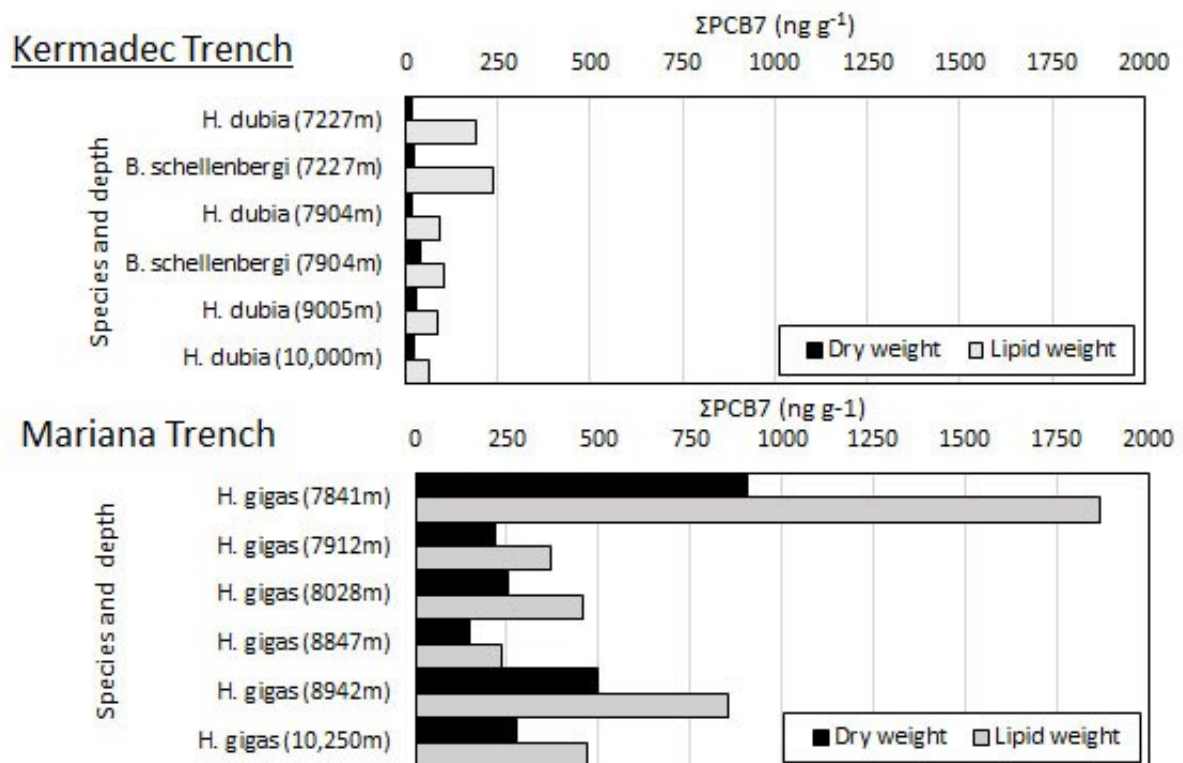
131 and T.M. performed the laboratory analysis under the supervision of Z.Z. and A.J.J. T.M. and T.F.  
132 performed the statistical analyses and the manuscript was written by A.J.J., T.M., S.B.P. and Z.Z.

133 **Author Information.** The authors declare no competing financial interests. Correspondence and  
134 requests for materials should be addressed to A.J.J. (alan.jamieson@ncl.ac.uk), S.B.P.  
135 (s.piertney@abdn.ac.uk) or Z.Z. (Zulin.Zhang@hutton.ac.uk).

136 **Data availability.** The data that support the findings of this study are available from the  
137 corresponding author upon reasonable request.

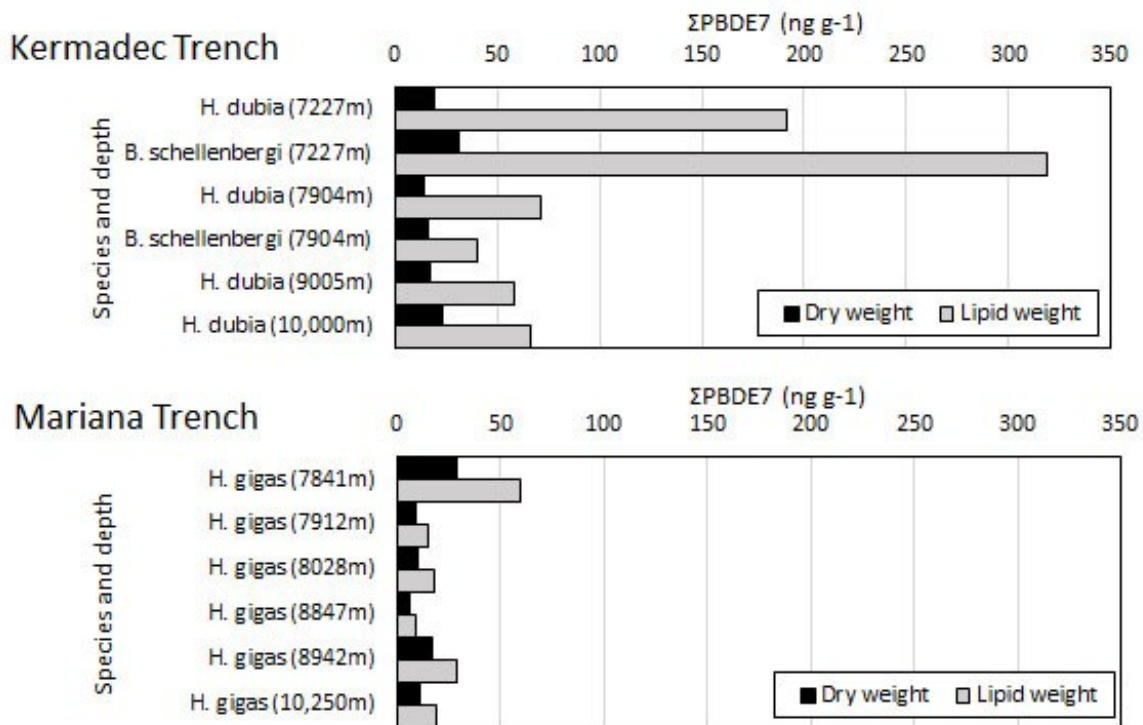
138

139 **Figures**



140

141 **Figure 1.** Polychlorinated biphenyl (PCB) concentration (ng g<sup>-1</sup>) for both dry and lipid weight found in  
142 endemic hadal amphipods across the bathymetric ranges of the Kermadec and Mariana trenches.



143

144 **Figure 2.** Polybrominated diphenyl ether (PBDE) concentration (ng g<sup>-1</sup>) for both dry and lipid weight  
 145 found in endemic hadal amphipods across the bathymetric ranges of the Kermadec and Mariana  
 146 trenches.

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179 **METHODS**

180 **Sampling** - Samples were collected using a full ocean depth rated lander vehicle<sup>25</sup> deployed to the  
181 seafloor by free-fall and acoustically recalled by jettisoning ballast weights. The lander typically  
182 remained on the seafloor for 8 to 12 h. Coupled to the lander footpads were a cluster of three small  
183 funnel traps (30 cm length by 6 cm diameter with trap openings of approximately 2.5 cm diameter).  
184 The traps were baited with ~100 g of mackerel that was enclosed in a mesh bag to allow the  
185 development of an odour plume, but prevent the amphipods consuming any of the bait that might  
186 otherwise affect POP levels in downstream assays. Once the samples were landed on deck they were  
187 not exposed to any plastics during preparation or preservation. The samples were transferred to a  
188 cold room (4 °C) on glass plates and sorted using metal implements before being transferred into pre-  
189 muffled (450 °C) foil wrapped tightly in several layers and frozen at -80 °C in natural fibre pouches.

190 **Study sites and species** – Samples of Lysianassoid amphipods were obtained from across two of the  
191 deepest hadal trenches - the Mariana Trench that underlies the oligotrophic surface waters of the  
192 North Pacific, and the Kermadec Trench situated under moderate primary productivity off New  
193 Zealand in the South Pacific. Two endemic species of amphipod (*Hirondellea dubia* and *Bathycallisoma*  
194 *schellenbergi*) were sampled from the Kermadec (32° S 177° W), at depths of 7227, 7904, 9005 and  
195 10,000 m. One species of endemic amphipod (*Hirondellea gigas*) was sampled from the Mariana (12°  
196 N 145° E), at depths of 7841, 7912, 8028, 8847, 8942 and 10,250 m. In all cases, samples were obtained  
197 using traps deployed on autonomous deep-sea lander vehicles<sup>25</sup>.

198 **Contaminant analyses.** The concentrations of seven PCB congeners (28, 52, 101, 118, 138, 153 and  
199 180) identified by ICES<sup>14</sup> for marine pollution assessment and seven PBDE congeners (28, 47, 99, 100,  
200 153, 154 and 183) with a wide range of bromination. Concentrations of PCBs and PBDEs in amphipods  
201 were determined using gas chromatography/mass spectrometry (GCMS), following extraction and  
202 sample clean-up, using method described previously<sup>26</sup>. Procedural blanks were run in parallel with  
203 each batch of samples and the results were corrected accordingly. The mean recoveries of the method  
204 were 40-77% for PCBs and 71-93% for PBDEs, respectively. Limits of detection (LOD) were 0.02 ng g<sup>-1</sup>  
205 for all PCBs and PBDE congeners 28, 47, 99 and 100, and 0.50 ng g<sup>-1</sup> for PBDE congeners 153, 154 and  
206 183.

207 **Lipid content.** Total lipid content was assessed using a single step extraction method<sup>27</sup>. 90 mg of  
208 sample were extracted with 2:1 chloroform-methanol (v/v) mixture in a glass centrifuge tube. After  
209 adding NaCl water solution, the phase separation was facilitated by centrifugation to recover the  
210 chloroform phase into a clean, weighed scintillation vials. Chloroform was evaporated and the vials  
211 were re-weighed to calculate total lipid content (%) for each sample.

212 **Statistical Methods.** Simple linear regression was performed to examine the possible effect of depth  
213 on the measurements of each pollutant (i.e. PCB7 dw, PCB7 lw, PBDE7 dw and PBDE7 lw) in both  
214 trenches (supplementary information Table 3). The pollutant measurements were highly skewed and  
215 therefore natural-log transformation was applied to normalise and stabilise the variance. Estimated  
216 regression model residuals were examined for statistical adequacy using standard graphical tools to  
217 verify the assumption of normality and homogeneity. In addition, Welch's t-test was performed to  
218 test for the difference in the respective pollutant levels between the two trenches (supplementary  
219 information Table 4). For the results of both linear regression analysis and Welch's t-test, adjusted  
220 significance levels ( $\alpha = 0.05$  divided by  $n=8$  and  $4$ , respectively) were applied using the Bonferroni  
221 inequality correction. All analyses were performed using R v3.2.3 package (R Development Core  
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