Article (refereed)

1 **Element patterns in albatrosses and petrels: influence of trophic**

2 **position, foraging range, and prey type**

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17 *Trophic position, foraging range, and prey type were found to influence element*

18 *compositions and concentrations in Procellariiformes from South Georgia.*

19

20 **Abstract**

21

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22        We investigated the concentrations of 22 essential and non-essential elements

23    among a community of Procellariiformes (and their prey) to identify the extent to which

24    trophic position and foraging range governed element accumulation. Stable isotope analysis

25    (SIA) was used to characterise trophic ($\delta^{15}$N) and spatial patterns ($\delta^{13}$C) among species.

26    Few consistent patterns were observed in element distributions among species and diet

27    appeared to be highly influential in some instances. Arsenic levels in seabird red blood cells

28    correlated with $\delta^{15}$N and $\delta^{13}$C, demonstrating the importance of trophic position and

29    foraging range for arsenic distribution. Arsenic concentrations in prey varied significantly

30    across taxa, and in the strength of association with $\delta^{15}$N values (trophic level). In most

31    instances, element patterns in Procellariiformes showed the clearest separation among

32    species, indicating that a combination of prey selection and other complex species-specific

33    characteristics (e.g. moult patterns) were generally more important determining factors than

34    trophic level *per se*.

35

36    *Key words*: Elements; Procellariiformes; Trophic Position; Diet; South Georgia

37

38    **1. Introduction**

39

40        Some metals and metalloids are essential elements (EE) involved in physiological

41    and biochemical processes (Abdulla and Chmielnicka, 1990), but can be toxic when

42    exposure and assimilation is excessive.  Others, such as lead (Pb), cadmium (Cd), arsenic

43    (As) and mercury (Hg), are non-essential elements (NEE) that are also potentially toxic.

44  Marine organisms can be exposed to elements from anthropogenic and natural sources, and

45  exposure is governed by various factors, including foraging location and trophic position.

46      Although the Southern Ocean is a remote environment, it is subject to

47  anthropogenic metal inputs due to global transport of elements in the atmosphere and

48  through oceanic circulation (Gaiero et al., 2003). Seabirds provide an ideal model with

49  which to investigate contaminant dynamics in marine food webs (see Gilbertson et al.,

50  1987; Furness, 1993; Furness and Camphuysen, 1997) and the Procellariiformes exploit a

51  wide range of ecological niches in the Southern Ocean. Many species occupy high trophic

52  positions and so can act as sentinels of exposure, highlighting potential threats to other apex

53  predators. Previous studies have examined mercury (Hg) and other elements in lower order

54  species in the Southern Ocean (e.g. Bargagli et al., 1998; Negri et al., 2006) but, apart from

55  Hg, concentrations of other elements in biota at higher trophic levels have received much

56  less attention (although see Lock et al., 1992; Stewart et al., 1999; González-Solís et al.,

57  2002; Bocher et al., 2003).

58      Conventional approaches for quantifying dietary composition and assessing metal

59  intake, such as analysis of gut content or regurgitate, have a number of biases (see Votier et

60  al., 2003) and samples are often difficult to obtain over long periods. In this study, we

61  analysed stable isotopes to elucidate broad-scale, inter- and intra-specific dietary patterns in

62  Procellariiformes (Hobson et al., 1994), and so determine whether differences in foraging

63  strategy explained variation in elemental uptake. Use of stable isotopes for this purpose is

64  based on the predictable increase in the ratio of $^{15}$N:$^{14}$N (by ca. 3.4‰) with each trophic

65  level,, and so $\delta^{15}$N can be used as a proxy for trophic position (e.g. DeNiro and Epstein,

66  1981; Hobson and Welch, 1992; Bearhop et al., 1999). $\delta^{13}$C can vary depending on whether

67 animals feed inshore or offshore, or on benthic or pelagic prey (e.g. Chisholm et al., 1982;

68 Hobson et al., 1994); $\delta^{13}C$ values also vary with latitude (Cherel and Hobson, 2007) and, as

69 such, provide a coarse-scale proxy for foraging location in wide-ranging organisms.

70       The overarching aim of the present study was to investigate the distributions and

71 dynamics of toxic heavy metals, essential metals and metalloids in Southern Ocean

72 seabirds, and assess the extent to which they may be bioaccumulated or biomagnified

73 through foodchains (see Bargagli et al., 1996; McIntyre and Beauchamp, 2007). We

74 analysed red blood cells (hereafter blood) and feathers collected during the breeding season

75 to relate element assimilation to trophic position. The residence times of dietary isotopic

76 signatures are dependent on the regenerative time of the tissue analysed (Hobson and

77 Clarke, 1992a, 1992b; Hobson 1999). Blood cells are short-term integrators of dietary

78 isotope signatures and are derived from prey consumed within the past 2-3 weeks. Blood

79 metal concentrations also reflect recent exposure, concentrations equilibrating between the

80 blood and the body organs (Burger and Gochfeld, 1997). Hence, the isotopic signature and

81 element concentrations in blood are temporally matched and reflect foraging in the

82 breeding season. In contrast, most Procellariiformes grow feathers during the non-breeding

83 period and so feathers provide a means for characterising diet and element burdens

84 acquired at this time. Feathers, once grown, are metabolically inert. They therefore

85 provide a dietary isotope and element exposure 'snap-shot' for the period of feather growth

86 (Mizutani et al., 1990, 1992, Bearhop et al., 2002). By randomly sampling body feathers,

87 the measured isotope signature and element concentration is averaged over the moult period

88 (Bearhop et al., 2000). The temporal matching of the feather isotope signature and element

89 concentrations is unlikely to be as good as that for blood because feather data are based on

90  averaged values obtained over a relatively long time period. Overall, however, it is possible

91  to gather information on trophic level and foraging range for the breeding (blood stable

92  isotopes) and non-breeding season (feather stable isotopes), although it is also important to

93  determine isotope values for likely prey items (as determined from previous conventional

94  dietary studies) so that the extent to which certain prey types may affect metal

95  accumulation in birds can also be evaluated.

96      In this study, our specific objectives were to investigate: (1) current levels of

97  exposure of Procellariiformes to a range of EE and NEE by measuring concentrations in

98  blood and feathers; (2) the extent to which assimilation varies between species; and, (3) the

99  extent to which foraging area, diet, and trophic level explains intra- and inter-individual

100  variation in metal accumulation in Procellariiformes.

101

102  **2. Materials and methods**

103

104  *2.1 Sample collection and preparation*

105

106  Fieldwork was undertaken on Bird Island, South Georgia (54º 00'S, 38º 03'W) from

107  December 2001 for a period of four months. Ten species were sampled in total: Antarctic

108  prion (*Pachyptila desolata*), black-browed albatross (*Thalassarche melanophrys*), blue

109  petrel (*Halobaena caerulea*), common diving petrel (*Pelecanoides urinatrix*), South

110  Georgian diving petrel (*Pelecanoides georgicus*), grey-headed albatross (*T. chrysostoma*),

111  northern giant petrel (*Macronectes halli*), southern giant petrel (*M. giganteus*), white-

112   chinned petrel (*Procellaria aequinoctialis*) and wandering albatross (*Diomedea exulans*).

113   Blood and feathers from adults of each species were taken from surface-nesting birds at the

114   end of the brood-guard period and from burrowing birds whilst mist-netting adjacent to

115   breeding colonies. Whole blood (0.2-1.0ml) (from the tarsal vein) was spun to separate

116   cells and plasma, immediately frozen, subsequently freeze-dried to a constant mass and

117   then again frozen until analysis. Only red blood cells were analysed for isotopes and trace

118   elements. Six to 8 feathers sampled at random were taken from the mantle region of each

119   bird. Feathers were stored in plastic bags at room temperature until analysed.  Fresh prey

120   samples (muscle tissue) were obtained from seabird regurgitates, or from fisheries vessels

121   operating in waters surrounding South Georgia, and frozen at -20°C after identification.

122   Cephalopods were identified from beaks according to Clarke (1986) and Rodhouse et al.

123   (1992), fish from external morphology according to Gon and Heemstra (1990), or from

124   otloliths according to Reid (1996), and crustaceans from Kirkwood (1984). Identifications

125   were checked against BAS reference collections. Prey were of unknown size and age.

126

127   *2.2  Stable isotope analyses*

128

129        All blood and feather samples were homogenised in a freezer mill prior to analysis

130   and oven-dried for >24hrs at 50ºC to a constant mass. In the case of prey samples, lipids

131   were extracted prior to SIA over a 4 hour period using Soxhlet apparatus containing 1:1

132   methanol to chloroform solvent mixture. Stable carbon and nitrogen isotope analyses were

133   performed by continuous flow isotope ratio mass spectrometry (CF-IRMS) on 0.7mg sub-

134   samples of material loaded into tin cups and combusted in a Costech ECS 4010 elemental

135 analyser coupled to a Thermo Finnigan Delta Plus XP mass spectrometer. Every 8-10

136 samples were followed by two laboratory standards allowing correction for drift. Isotope

137 ratios are expressed in standard δ notation against international reference standards, vPDB

138 (virtual Pee Dee Belemnite) for $\delta^{13}C$ and atmospheric nitrogen for $\delta^{15}N$ according to the

139 equation:

140

141 $$\delta X = [(R_{sample}/R_{standard}) - 1] \times 1000$$

142

143 where $X$ is $^{15}N$ or $^{13}C$ and R is the corresponding ratio $^{15}N:^{14}N$ or $^{13}C:^{12}C$. Precision for

144 both $\delta^{13}C$ and $\delta^{15}N$ was routinely estimated to be $\leq 0.2‰$.

145

146 *2.3 Element analyses*

147

148    Samples were oven-dried for 24hrs at 50ºC to a constant mass. Between 0.05 and

149 0.1g dry weight of sample (mean weights for each tissue type; 0.6g for prey, 0.09g for

150 blood, and 0.05g for feathers) was weighed accurately and added to 2ml nitric acid and left

151 to cold digest for 24hrs. Samples were then hot digested for 50 minutes at 120ºC, after

152 which 0.5ml of hydrogen peroxide was added and left for a final 15 minutes. The samples

153 were made up to 5ml with deionised water. Total absolute concentrations for 22 essential

154 and non-essential elements were measured using an ELAN 6100 dynamic reaction cell

155 inductively coupled plasma mass spectrometer (DRC ICP-MS) (PerkinElmer, Connecticut,

156 US) with a reference material and internal standard run with every 10 unknowns. Spiked

157 samples and blanks were also run. Limits of detection (LoD) for particular elements are in

158 Table 1. Precision and accuracy (Table 2) were measured using replicate samples and

159 certified reference material (TORT-2 lobster hepatopancreas, NRCC, Canada). Element

160 concentrations are expressed throughout as mean $\pm$ SD in $ng.g^{-1}$ on a dry weight basis,

161 unless otherwise specified.

162

163 *2.4 Data analyses*

164

165      Elements with mean concentrations below LoD (beryllium (Be) for prey; aluminium

166 (Al), scandium (Sc), cobalt (Co), zinc (Zn), antimony (Sb), tungsten (W), and Be for blood;

167 and Sc, caesium (Cs), lithium (Li), Pb, and Be for feathers) are reported in the summary

168 statistics but are excluded from further consideration or statistical analyses. Among the

169 remaining elements, concentrations in some samples in some species were below the LoD

170 (maximum 13% of samples for rubidium (Rb) in feathers, 1-3% of samples for vanadium

171 (V), iron (Fe), molybdenum (Mo), nickel (Ni), Cs, Hg, manganese (Mn), and selenium (Se)

172 in various tissue types) and were assigned a value equal to half the LoD for the particular

173 tissue. Where element concentrations were below the LoD in >30% of samples overall for a

174 particular tissue type (V, barium (Ba), uranium (U), Li, and Pb for blood; V, Sb, and W for

175 feathers), those elements were also included in summary statistics but excluded from

176 subsequent statistical analyses, as in other studies (Borgå et al., 2006). Fe was excluded

177 from all statistical analyses of blood data because of its physiological function in this tissue.

178      For elements that were included in the statistical analysis, we first used principal

179 component analyses (PCA) in Canoco 4.5 for Windows[®] to identify co-variance among

180 elements, and between elements and isotope ratios (the latter entered as passive variables,

181    following Borgå et al., 2006) in seabird tissues. PCA produces ordination axes that reduce

182    the total residual sum of squares among all the variables included (i.e. all elements, $\delta^{15}$N,

183    and $\delta^{13}$C). The primary axis (PC1) represents the greatest proportion of component

184    variation among the samples. The secondary axis (PC2) explains the greatest proportion of

185    any remaining variation that is uncorrelated with PC1. Variables are shown as lines, with

186    the length of each line demonstrating relative contribution of a particular variable to the

187    separation of samples in ordination space. Angles between lines represent a greater or lesser

188    degree of co-variance between elements; with acute angles representing the greatest degree

189    of co-variance, a 180º angle demonstrating a negative correlation, and a 90º angle represent

190    elements that are completely un-correlated. All PCA were performed on standardised

191    values, so that those elements with vastly different ranges could be analysed together. We

192    then used univariate general linear models (GLMs) to test the significance of relationships

193    between species, $\delta^{15}$N, $\delta^{13}$C, and elements of high factor loadings that were identified

194    through PCA, (elements with $\geq$ 0.50 factor loadings for PC1 and/or PC2). These analyses

195    were applied to both blood and feather data separately and the most parsimonious

196    univariate models were determined using Akaike Information Criterion (AIC) (Akaike

197    1973). Absolute element concentrations were logarithmically transformed ($\log_{10}$) in some

198    instances to reduce skewness and heterogeneity.  Model residuals were also examined by

199    way of Q-Q plots to assess goodness of fit. Statistical tests were performed in statistical

200    packages R ver. 2.6.2 and SPSS$^{®}$ ver. 14.

201

202    **3.  Results**

203

204          In total, we analysed samples for 22 essential and non-essential elements. Mercury

205   was detected in all three tissue types but, because of the particular biomagnification

206   properties and risks associated with this element, the results have been examined in greater

207   detail and reported elsewhere; essentially, mercury levels were found to increase with

208   trophic level, indicating strong biomagnification among Procellariiformes (Anderson et al.

209   2009). Fourteen elements were quantifiable in blood (Table 3) and seventeen in feathers

210   (Table 4). Species varied in the extent to which they accumulated different elements and no

211   single species accumulated the highest concentrations of all elements (Tables 3-5). Of all

212   the elements, Fe and Se had the highest concentrations in blood (Table 3), and Fe and Zn

213   the highest concentrations in feathers (Table 4).

214          In prey samples, 21 elements were detected, only Be was below detection limits.

215   Iron and Zn had the highest concentrations of all elements tested for (Table 5). Aluminium

216   concentrations were generally an order of magnitude higher in crustaceans than in fish and

217   cephalopods, and Cd concentrations were likewise some 10-fold higher in several squid

218   species and in *Themisto gaudichaudii*, compared with other species (Table 5). Benthic-

219   feeding fish, such as Patagonian toothfish (*Dissostichus eleginoides*) (García de la Rosa et

220   al., 1997), often had the highest As concentrations. Isotopic analyses indicated a high

221   degree of variation in trophic position ($\delta^{15}$N) and foraging range ($\delta^{13}$C) between seabird

222   species (Fig. 1), and considerable trophic overlap between fish and cephalopod prey.  The

223   greatest degree of spatial variation (largest range in $\delta^{13}$C ratios) in prey was between fish

224   species (Fig. 1).

225    Initial PCA analyses identified strong associations between As concentrations and

226    both $\delta^{15}N$ and $\delta^{13}C$ in blood (Fig. 2a).   There was a similar but weaker association for

227    caesium (Cs), as indicated by the shorter line on the PCA plot (Fig. 2a), but no similar

228    associations for other elements in blood. The distribution of As in ordination space was

229    predominantly governed by white-chinned petrel, suggesting that blood As concentrations

230    were elevated in this species during the breeding season; this association did not occur in

231    feathers (Fig. 2b). There were other similar associations between specific elements and

232    seabird species.  The positions of Rb and Cd in ordination space were largely governed by

233    grey-headed albatross, indicating that blood concentrations of these elements were highest

234    in this species during the breeding season (Fig. 2a); again there were no such associations

235    for feathers (Fig. 2b). The ordinal positions of white-chinned petrel and wandering

236    albatross were influenced by $\delta^{15}N$ and $\delta^{13}C$ in both blood and feathers. Overall, As and Cd

237    (NEE) contributed most to PC1 in blood, while Se (EE), Rb (NEE), and Mo (EE)

238    contributed most to PC2 (Table 2, supplementary material). In contrast, Mn (EE), Fe (EE),

239    Co (EE), As (NEE), and Mo (EE) contributed most to PC1 in feathers, while Se and U

240    (NEE) contributed most to PC2.

241    With the exception of blood As, all elements contributing $\geq 0.50$ to factor loadings

242    in the PCA were found to differ significantly between seabird species (Table 6). Univariate

243    GLM analyses identified significant differences between species for a number of elements.

244    Blood concentrations of Cd, Se, Rb, and Mo all differed significantly between species

245    (Table 6), and in feathers, significant differences between species were found in the levels

246    of Mn, Fe, Co, As, Mo, Se, and U.

247    Univariate GLMs demonstrated that there was a significant and positive association

248    between As and $\delta^{15}$N in blood, although the pattern of this relationship varied between

249    species, as demonstrated by the significant interaction term in the model (Table 6). Blood

250    As concentrations were negatively associated with $\delta^{13}$C across all species (i.e., at

251    community level, Table 6) but the exact nature of this relationship may also have varied

252    between species, the interaction term in the model approaching significance (Table 6).

253    There was a significant relationship between Se and $\delta^{13}$C in feathers (Table 6). There were

254    no other significant relationships between element concentrations and trophic position

255    and/or foraging location in either tissue type (Table 6).

256    The positive association between blood As concentrations and trophic position

257    ($\delta^{15}$N) was consistent with the concept that As is bioconcentrated along food chains. To

258    investigate this further, we examined how As concentrations in prey species varied with

259    $\delta^{15}$N and $\delta^{13}$C values (Table 7). Arsenic concentrations differed significantly between

260    taxonomic groups but also varied with $\delta^{15}$N (Table 7). However, associations between $\delta^{15}$N

261    and As concentrations were not consistent between prey groups (Table 7), indicating that

262    associations between the trophic position of species and As are complex (Fig. 3).

263

## 4. Discussion

264

265

266    The large number of elements analysed in this study required us to focus the

267    discussion only on those elements for which a significant relationship was identified among

268    the parameters in the univariate analyses. We have also focused particularly on those

269 elements for which a significant relationship occurred in blood as blood is the better tissue

270 for characterising trophic and spatial patterns in relation to element composition within

271 these birds. This is because feathers have the potential to acquire element burdens both

272 directly through diet and from remobilisation of elements from internal tissues (Lewis and

273 Furness, 1991; Monteiro, 1996). This may be why the relationships identified between

274 stable isotope ratios and elements in blood in the present study were not always mirrored

275 with feathers.

276

277 *4.1 Arsenic*

278

279 Of the elements analysed in seabird blood, only As showed significant interactions

280 with $\delta^{15}$N and $\delta^{13}$C at the community level (Table 6), suggesting that trophic position

281 and/or foraging range affect As assimilation by Procellariiformes during the breeding

282 season. The significant and positive association between blood As and $\delta^{15}$N may be due to

283 bioconcentration or reflect changes in isotopic baseline that coincide with variation in

284 environmental As concentrations. We found no evidence of bioconcentration of As through

285 the foodchain. Although As concentrations varied significantly between prey groups

286 (crustacea, cephalopods, and fish), there was no consistent relationship between As

287 concentrations and trophic position in prey (Table 7, Fig. 3).

288 Given the lack of clear evidence of bioconcentration, it is possible that foraging

289 location is the prime factor affecting As assimilation in Procellariiformes during the

290 breeding season, and any associations with $\delta^{15}$N may be coincidental. In this respect, As

291 strongly influenced the position of white-chinned petrel in the PCA ordination space (Fig.

13

292   2a), reflecting their high blood As concentrations (2500 ng g$^{-1}$ dry weight) relative both to

293   other species in this study and to other species elsewhere; reference values for blood As in

294   birds from uncontaminated areas is 20 ng g$^{-1}$ wet weight (Burger and Gochfeld, 1997),

295   approximately equivalent to a dry weight red blood cell concentration of 400 ng g$^{-1}$.

296   White-chinned petrels have amongst the greatest foraging distances of any seabird and

297   breeding birds from South Georgia regularly forage on the northern Patagonian shelf,

298   >2000km from the colony (Phillips et al., 2006). Prey food webs on the Patagonian shelf

299   have greater trophic complexity than those in waters around South Georgia (Forero et al.,

300   2004, 2005), and so $\delta^{15}$N levels in birds feeding in that region may be elevated, reflecting

301   longer food chain lengths. The Patagonian shelf region is also more likely to be subject to

302   greater anthropogenic inputs from riverine influxes, mineral activities, and industry than the

303   more remote regions of the Southern Ocean (González-Solís et al., 2002), which could

304   explain the elevated arsenic levels in birds that feed there. It is also possible, however, that

305   enhanced environmental exposure to As may stem from natural processes. For example,

306   disruption in the phosphorus/nitrogen ratio could lead to greater As accumulation by

307   phytoplankton in a particular region and subsequently enhance As levels in the food chain.

308       In our study, there was no relationship between feather As concentrations and $\delta^{15}$N.

309   Feather As concentrations were highest in Antarctic prion (APR), but only about half the

310   concentrations measured in black-footed albatross (*Phoebastria nigripes*) feathers from the

311   North Pacific (Fujihara et al., 2004). Low feather As concentrations in Antarctic

312   Procellariiformes may reflect exposure during the non-breeding season to relatively low-

313   level natural levels of As. In our study, As concentrations in prey tended to be highest in

314   benthic/demersal species (Table 5). This suggests that a major source of As in the prey of

315 Antarctic Procellariiformes around South Georgia may stem from metallic mineralisation of

316 the seabed rather than from atmospheric transport of particles. Indeed, marine sediments are

317 thought to be the largest geochemical reservoir of arsenic with residence times of ca. 100

318 million years (Maher and Butler 1988). Furthermore, manganese (Mn) concentrations were

319 also elevated in benthic species, again suggesting a natural source of As; elevated As levels

320 *without* matching high levels of Mn are thought to be indicative of anthropogenic As input

321 (Peterson and Carpenter, 1986).

322 Although there was a significant relationship between $\delta^{13}C$ and blood As

323 concentrations in the Procellariiformes in our study, no such relationship occurred among

324 prey species. This may be because prey were collected from a small geographic area,

325 relative to the foraging areas of the birds. However, there was still considerable variation in

326 $\delta^{13}C$ ratios between prey species (Fig. 1) and this probably reflects the influence of depth

327 on prey $\delta^{13}C$ ratios. The lack of association between $\delta^{13}C$ and As in prey indicates that As

328 uptake by seabirds is unlikely to be influenced by the proportion of mesopelagic prey in the

329 diet, unlike patterns of Hg uptake which are heavily influenced by prey type (see Monteiro

330 et al., 1996; Thompson et al., 1998). The association observed between $\delta^{13}C$ and As in

331 Procellariiformes is most likely due to geographical variation in background As

332 concentrations within the marine environment.

333

334 *4.2 Cadmium*

335

336 Blood cadmium concentrations in seabirds varied significantly between species but

337 not with isotopic signatures, indicating that neither trophic position nor foraging location

338    dictated Cd burdens during the breeding season. Our results match those of Gonzalez-Solis

339    et al. (2002) in that Cd concentrations were greater in northern than southern giant petrels

340    (Table 3). Gonzalez-Solis et al. (2002) postulated that the former are exposed to higher

341    levels through foraging over the Patagonian shelf, as the region is thought to be more

342    exposed to heavy metal contamination than the waters immediately adjacent to South

343    Georgia. However, if latitudinal variation in foraging location was the major factor

344    affecting Cd assimilation in Antarctic Procellariiformes, we would anticipate a correlation

345    between Cd concentrations in birds and $\delta^{13}$C. It may be that foraging location does dictate

346    Cd uptake, but that any such influence reflects the existence of Cd 'hot spots' rather than a

347    latitudinal gradient *per se*. In fact, Nygård et al. (2001) linked deep ocean upwelling, which

348    is often naturally Cd-rich (Holm-Hansen, 1985), to high Cd concentrations in Antarctic krill

349    *Euphausia superba* and other prey of Procellariiform seabirds (Petri and Zauke, 1993).

350        Previous studies have attributed high Cd concentrations in Procellariiformes to

351    consumption of squid (Muirhead and Furness, 1988; Honda and Marcovecchio, 1990;

352    Elliott, 2005; Stewart et al., 1999). Our results support this, in that higher Cd levels were

353    found in the blood of grey-headed albatrosses and white-chinned petrels (Table 3). Both

354    white-chinned petrels and grey-headed albatrosses from South Georgia consume large

355    amounts of squid, the latter feeding in particular on *Martialia hyadesi*, and, also on lamprey

356    *Geotria australis* (Berrow and Croxall, 1999, Xavier et al., 2003). In our study, both these

357    prey types had relatively elevated Cd concentrations (Table 5). Hence, high Cd burdens in

358    Procellariiformes do not necessarily indicate anthropogenic contamination. Levels of

359    cadmium in feathers, for example, were largely similar to those in similar species, e.g.

360  black-footed and Laysan albatrosses (*Phoebastria immutabilis*) in the more contaminated

361  North Pacific (Burger and Gochfeld, 2000).

362      Squid are thought to accumulate high Cd concentrations naturally (Kurihara et al.,

363  1993; Gerpe et al., 2000). Elevated burdens in Procellariiformes can also result from

364  amphipod consumption (Cheng et al., 1984; Elliott and Scheuhammer, 1997), and in our

365  study, it was notable that we recorded the highest Cd levels of any prey item in *Themisto*

366  *gaudichaudii* (Table 5). Rainbow (1989) concluded that *Themisto* species are a significant

367  source of Cd in the diets of some seabirds, and our findings are consistent with this as the

368  highest blood Cd levels in birds in our study were in blue petrels, a known predator of *T.*

369  *gaudichaudii* (Prince, 1980). Moreover, hyperiid amphipods may also contribute to the diet

370  of some squid species (Croxall and Prince, 1980) which can then be eaten by seabirds.

371  Thus, dietary preferences, rather than foraging area, may comprise the dominant influence

372  on Cd burdens in Procellariiformes.

373

374  *4.3 Selenium*

375

376      Blood (hence breeding season) concentrations of Se, an essential element, varied

377  significantly between seabird species, but did not show a significant relationship with

378  trophic position ($\delta^{15}$N) and foraging location ($\delta^{13}$C) (Table 6). Therefore, it seems probable

379  that Se does not biomagnify among Procellariiformes. Additionally, our data suggest that

380  foraging location does not dictate Se burdens in Southern Ocean Procellariiformes,

381  although this may in fact reflect the constrained foraging ranges of many species during the

382  breeding season rather than a true absence of association between the two parameters.

383    Selenium concentrations were the highest of any element in Procellariiformes blood except

384    Fe. Moreover, blood Se levels in both giant petrel species were consistent with those

385    previously recorded by González-Solís et al. (2002). Blue petrels and Antarctic prions had

386    the highest blood Se concentrations of the birds we analysed and both species feed

387    predominantly on crustacea, including Antarctic krill (Prince, 1980; Croxall et al., 1997).

388    Of the prey species we analysed, crustacea, especially Antarctic krill and *T. gaudichaudii*,

389    had high Se concentrations relative to other prey species and groups. The physico-chemical

390    form in which Se is sequestered by different prey may also vary and affect subsequent

391    bioavailability. Hence, it would appear that inter-specific variation in blood Se

392    concentrations in seabirds most likely reflects species differences in prey selection.

393        Selenium concentrations in feathers varied considerably between species and were

394    also significantly and negatively related to foraging location (i.e. $\delta^{13}$C) (Table 6). This was

395    unexpected as concentrations of Se might be expected to increase with decreasing latitude,

396    as birds move into potentially more contaminated waters north of the sub-Antarctic polar

397    front. The negative relationship in this instance is difficult to explain, but could potentially

398    stem from natural sources of Se being more elevated in specific regions closer to the

399    Antarctic continent. In other words, Se burdens accumulated by Procellariiformes in the

400    non-breeding period may be dictated by naturally occurring Se in the marine environment,

401    potentially stemming from Antarctic intermediate and bottom waters which are sporadically

402    rich in organic Se (Cutter and Cutter, 2001). Evidence to support this hypothesis, comes

403    from the comparable levels of Se in the species covered by this study, to those of other

404    studies. Burger and Gochfeld (2000) examined Se in feathers from black-footed and Laysan

405 albatrosses in the North Pacific, and identified levels comparable to those found in this

406 study.

407

408 *4.4  Rubidium and Molybdenum*

409

410     Two other elements, Rb and Mo, also varied between seabird species, but not with

411 either $\delta^{15}N$ or $\delta^{13}C$, indicating that trophic position and foraging location were not key

412 determinants in the distribution of these two  elements within Procellariiformes. Rb is

413 known to bioaccumulate through the foodchain (Nyholm and Tyler, 2000), and in fact,

414 Campbell et al. (2005) reported a significant correlation between Rb and $\delta^{15}N$ in marine

415 organisms, potentially suggesting biomagnification. While our results were not significant

416 in the presence of other parameters, there was a significant, albeit weak, positive

417 relationship between Rb and $\delta^{15}N$ in blood (linear regression, $F_{1,140} = 9.9$, $p = 0.002$, $r^2 =$

418 0.07). However, whether this is evidence of biomagnification of Rb within this particular

419 system is doubtful given there was no significant relationship between Rb concentrations

420 and $\delta^{15}N$ in prey (linear regression, $F_{1,82} = 1.5$, $p = 0.229$, $r^2 = 0.02$). Given that the

421 accumulated evidence concerning Rb bioconcentration through foodchains is unclear,

422 further investigations into Rb distributions and bioaccumulation in the marine environment

423 appear warranted.

424

**5.  Conclusions**

425

426

427   This study presents new data on the distributions and dynamics of heavy metals,

428 essential metals and metalloids in Southern Ocean seabirds. Our results indicate that there

429 are considerable differences in residues accumulated by different species. For arsenic,

430 residues in Procellariiformes appear to be partially explained by changes in foraging range

431 and trophic positioning, although these patterns were not always consistent across species.

432 Arsenic was accumulated at higher concentrations in Procellariiformes feeding at higher

433 trophic levels, although this may be confounded to some extent by individuals foraging

434 across different food webs with varying $\delta$ $^{15}$N baselines. There was little or no evidence that

435 trophic level was important in the accumulation of other elements. Our data suggest that

436 differences between species in feeding preferences for prey and the location of the foraging

437 sites are the likely major factors determining element uptake by seabirds in the South

438 Atlantic.

439

## 440 **6. Acknowledgements**

441

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633    **Fig. 1.** Range plots for $\delta^{15}N$ (a) and $\delta^{13}C$ (b) values for all Procellariiformes (blood) and

634    prey species. Species are ordered by increasing $\delta^{15}N$ values within each of the four

635    groupings. Data is shown for Procellariiform blood samples and prey muscle samples.

636

637    **Fig. 2.** Biplot of Procellariiform species mean scores extracted by principal component

638    analyses (PCA) and element loadings on the two principal axes (PC1 and PC2), with

639    standardised element and isotope values in (a) blood and (b) feathers from ten species.

640    APR=Antarctic prion, BBA=Black-browed albatross, BLP=Blue petrel, CDP=Common

641    diving petrel, GDP=South Georgian diving petrel, GHA=Grey-headed albatross,

642    NGP=Northern giant petrel, SGP=Southern giant petrel, WCP=White-chinned petrel,

643    WNA=Wandering albatross.

644

645    **Fig. 3.** Patterns in $\log_{10}$ arsenic concentrations in muscle tissue samples from three

646    Procellariiform prey groups in relation to (a) $\delta^{15}N$ values and (b) $\delta^{13}C$ values from the same

647    tissue samples.

648 **Table 1.** Limits of Detection (LoD) for prey, blood, and feather samples (ng $g^{-1}$).

| Element | Prey LoD | Blood LoD | Feather LoD |
|---|---|---|---|
| Al | 199 | 1357 | 2365 |
| As | 3 | 18 | 32 |
| Ba | 20 | 136 | 237 |
| Be | 1 | 7 | 12 |
| Cd | 1 | 5 | 8 |
| Co | 2 | 14 | 24 |
| Cs | 1 | 5 | 8 |
| Cu | 7 | 45 | 79 |
| Fe | 332 | 2261 | 3942 |
| Li | 1 | 7 | 12 |
| Mn | 2 | 14 | 24 |
| Mo | 10 | 68 | 118 |
| Ni | 3 | 23 | 39 |
| Pb | 20 | 136 | 237 |
| Rb | 1 | 5 | 8 |
| Sb | 3 | 23 | 39 |
| Sc | 17 | 113 | 197 |
| Se | 10 | 68 | 118 |
| U | 1 | 5 | 8 |
| V | 7 | 45 | 79 |
| W | 3 | 23 | 39 |
| Zn | 332 | 2261 | 3942 |

649

650 **Table 2.** TORT-2 lobster hepatopancreas reference material for elements; expected and

651 obtained CRM values ($\mu g\ g^{-1}$ dry wt.). All elements, where possible, were CRM checked

652 against an expected value. However, not all elements tested had a reported certified

653 concentration in the CRM. For these elements only the obtain CRM value is reported.

| Element | Expected CRM Value | ± 95% CI | Obtained CRM Value | ± 95% CI |
|---|---|---|---|---|
| Al | | | 17.0 | 0.9 |
| As | 21.6 | 1.8 | 26.4 | 0.8 |
| Ba | | | 2.0 | 0.1 |
| Be | | | 0.01 | 0.0 |
| Cd | 26.7 | 0.6 | 28.8 | 0.8 |
| Co | 0.5 | 0.1 | 0.5 | 0.1 |
| Cs | | | 0.0.2 | 0.0 |
| Cu | 106.0 | 10.0 | 93.3 | 3.0 |
| Fe | 105.0 | 13.0 | 126.0 | 10.8 |
| Li | | | 0.3 | 0.0 |
| Mn | 13.6 | 1.2 | 12.8 | 0.4 |
| Mo | 0.9 | 0.1 | 1.1 | 0.1 |
| Ni | 2.5 | 0.2 | 2.4 | 0.1 |
| Pb | | | 0.2 | 0.0 |
| Rb | | | 2.7 | 0.1 |
| Sb | | | 0.1 | 0.0 |
| Sc | | | 0.1 | 0.0 |
| Se | 5.6 | 0.7 | 7.5 | 0.3 |
| U | | | 0.1 | 0.0 |
| V | 1.6 | 0.2 | 1.9 | 0.1 |
| W | | | 0.01 | 0.0 |
| Zn | 180.0 | 6.0 | 191.7 | 12.2 |

Table 3. Mean concentrations of elements (ng g$^{-1}$ dry wt.) ± SD in blood from 10 species of Procellariiformes.

| Species | n | Al | As | Ba | Be | Cd | Co | Cs | Cu | Fe | Li | Mn | Mo | Ni | Pb | Rb | Sb | Sc | Se | U | V | W | Zn |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Antarctic prion | 16 | <LoD | 431 ± 186 | 345 ± 289 | <LoD | 48 ± 58 | <LoD | 5 ± 4 | 414 ± 366 | 2604580 ± 168540 | 1990 ± 7849 | 132 ± 66 | 226 ± 172 | 2140 ± 723 | 1052 ± 3203 | 4161 ± 523 | <LoD | <LoD | 198425 ± 59904 | 8 ± 5 | 85 ± 130 | <LoD | <LoD |
| Black-browed albatross | 15 | <LoD | 373 ± 267 | 192 ± 234 | <LoD | 86 ± 177 | <LoD | 15 ± 10 | 815 ± 579 | 4330039 ± 6571356 | 6 ± 10 | 150 ± 128 | 84 ± 113 | 1728 ± 1286 | 140 ± 172 | 4185 ± 1898 | <LoD | <LoD | 102335 ± 85049 | 11 ± 15 | 140 ± 126 | <LoD | <LoD |
| Blue petrel | 2 | <LoD | 388 ± 47 | 191 ± 174 | <LoD | 346 ± 27 | <LoD | <LoD | 71 ± 68 | 2440536 ± 75575 | 3 ± 0 | 91 ± 3 | 225 ± 46 | 1652 ± 276 | 68 ± 0 | 4368 ± 178 | <LoD | <LoD | 261757 ± 131638 | 12 ± 3 | <LoD | <LoD | <LoD |
| Common diving petrel | 15 | <LoD | 389 ± 129 | 187 ± 170 | <LoD | 19 ± 29 | <LoD | <LoD | 639 ± 1010 | 2265035 ± 355674 | 3 ± 0 | 128 ± 78 | 217 ± 131 | 1442 ± 589 | 322 ± 718 | 3612 ± 625 | <LoD | <LoD | 84842 ± 29419 | 9 ± 6 | 162 ± 285 | <LoD | <LoD |
| S. Georgian diving petrel | 15 | <LoD | 476 ± 197 | 310 ± 324 | <LoD | 27 ± 32 | <LoD | 14 ± 6 | 196 ± 186 | 2570513 ± 582271 | 29 ± 20 | 190 ± 76 | 88 ± 68 | 2007 ± 553 | 281 ± 446 | 3824 ± 967 | <LoD | <LoD | 118897 ± 60846 | 10 ± 5 | 60 ± 97 | <LoD | <LoD |
| Grey-headed albatross | 15 | <LoD | 896 ± 380 | 303 ± 198 | <LoD | 228 ± 81 | <LoD | 15 ± 10 | 670 ± 905 | 2483096 ± 239518 | 106 ± 46 | 252 ± 430 | 68 ± 80 | 1936 ± 530 | 235 ± 648 | 4526 ± 777 | <LoD | <LoD | 194381 ± 43946 | 6 ± 5 | <LoD | <LoD | <LoD |
| Northern giant petrel | 16 | <LoD | 417 ± 787 | <LoD | <LoD | 88 ± 213 | <LoD | 7 ± 4 | 638 ± 246 | 2333908 ± 659977 | 60 ± 23 | 97 ± 38 | 94 ± 26 | 1131 ± 966 | <LoD | 4991 ± 819 | <LoD | <LoD | 151170 ± 39472 | <LoD | 75 ± 188 | <LoD | <LoD |
| Southern giant petrel | 16 | <LoD | 184 ± 156 | <LoD | <LoD | 37 ± 105 | <LoD | 5 ± 3 | 690 ± 109 | 2447292 ± 698505 | 111 ± 70 | 202 ± 494 | 90 ± 13 | 1480 ± 990 | <LoD | 5052 ± 712 | <LoD | <LoD | 151432 ± 77353 | <LoD | <LoD | <LoD | <LoD |
| White-chinned petrel | 16 | <LoD | 2492 ± 1375 | <LoD | <LoD | 179 ± 60 | <LoD | 10 ± 5 | 241 ± 250 | 2482217 ± 219636 | 46 ± 31 | 192 ± 69 | 127 ± 71 | 1866 ± 561 | <LoD | 4532 ± 904 | <LoD | <LoD | 97951 ± 40289 | <LoD | <LoD | <LoD | <LoD |
| Wandering albatross | 15 | <LoD | 686 ± 377 | <LoD | <LoD | 50 ± 60 | <LoD | 17 ± 6 | 889 ± 455 | 2398513 ± 681723 | 98 ± 47 | 86 ± 17 | 92 ± 50 | 1312 ± 970 | <LoD | 3913 ± 653 | <LoD | <LoD | 80187 ± 21270 | <LoD | <LoD | <LoD | <LoD |

654

655

656

657 **Table 4.** Mean concentrations of elements (ng g$^{-1}$ dry wt.) ± SD in feathers from 10 species of Procellariiformes.

| Species | n | Al | As | Ba | Be | Cd | Co | Cs | Cu | Fe | Li | Mn | Mo | Ni | Pb | Rb | Sb | Sc | Se | U | V | W | Zn |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Antarctic prion | 16 | 28396 ± 25222 | 411 ± 227 | 1654 ± 1930 | <LoD | 59 ± 110 | 605 ± 1295 | <LoD | 185292 ± 644966 | 1010868 ± 3457595 | <LoD | 5254 ± 16921 | 3411 ± 12163 | 36011 ± 68146 | <LoD | 153 ± 79 | 120 ± 116 | <LoD | 7319 ± 2192 | 19 ± 9 | 610 ± 2060 | 285 ± 571 | 113658 ± 364227 |
| Black-browed albatross | 16 | 17924 ± 13028 | 121 ± 39 | 1481 ± 1802 | <LoD | 578 ± 246 | 270 ± 481 | <LoD | 8613 ± 11995 | 610216 ± 1657667 | <LoD | 2704 ± 6669 | 1686 ± 4057 | 3284 ± 6669 | <LoD | 194 ± 27 | 84 ± 43 | <LoD | 3381 ± 923 | 17 ± 3 | 321 ± 782 | 128 ± 160 | 39592 ± 25311 |
| Blue petrel | 5 | 18704 ± 22291 | 164 ± 88 | 1065 ± 291 | <LoD | 74 ± 90 | 498 ± 329 | <LoD | 8745 ± 1953 | 968705 ± 1285689 | <LoD | 4972 ± 5981 | 3462 ± 4877 | 7862 ± 9365 | <LoD | 123 ± 110 | 121 ± 82 | <LoD | 6597 ± 2029 | 21 ± 7 | 545 ± 799 | 47 ± 40 | 6953 ± 10470 |
| Common diving petrel | 16 | 37468 ± 70275 | 177 ± 126 | 1226 ± 1108 | <LoD | 376 ± 349 | 140 ± 115 | <LoD | 31254 ± 58194 | 130911 ± 119824 | <LoD | 954 ± 837 | 581 ± 437 | 6534 ± 15075 | <LoD | 140 ± 157 | 192 ± 283 | <LoD | 5050 ± 3878 | 19 ± 14 | 207 ± 525 | 21 ± 4 | 301098 ± 481053 |
| S. Georgian diving petrel | 16 | 65277 ± 34649 | 261 ± 163 | 1402 ± 981 | <LoD | 300 ± 353 | 419 ± 241 | <LoD | 20176 ± 15166 | 791151 ± 572700 | <LoD | 5077 ± 3490 | 2979 ± 2133 | 25442 ± 55886 | <LoD | 162 ± 115 | 80 ± 84 | <LoD | 9797 ± 4917 | 42 ± 15 | 865 ± 504 | 95 ± 94 | 22283 ± 37587 |
| Grey-headed albatross | 16 | 21372 ± 27043 | 125 ± 29 | 769 ± 605 | <LoD | 196 ± 163 | 161 ± 101 | <LoD | 5661 ± 1820 | 229219 ± 190078 | <LoD | 1291 ± 940 | 893 ± 730 | 12683 ± 14224 | <LoD | 220 ± 46 | 61 ± 59 | <LoD | 5395 ± 1467 | 17 ± 5 | 163 ± 113 | 56 ± 46 | 50115 ± 20426 |
| Northern giant petrel | 16 | 11944 ± 8969 | 145 ± 72 | 641 ± 454 | <LoD | 83 ± 51 | 90 ± 31 | <LoD | 6211 ± 1559 | 103719 ± 54442 | <LoD | 608 ± 244 | 552 ± 229 | 3811 ± 10126 | <LoD | 243 ± 55 | 59 ± 45 | <LoD | 8377 ± 2346 | 16 ± 4 | 124 ± 71 | 25 ± 14 | 67557 ± 28067 |
| Southern giant petrel | 17 | 13939 ± 14005 | 112 ± 36 | 815 ± 631 | <LoD | 289 ± 93 | 101 ± 60 | <LoD | 6877 ± 3788 | 95208 ± 42181 | <LoD | 587 ± 184 | 509 ± 142 | 930 ± 1118 | <LoD | 246 ± 35 | <LoD | <LoD | 9757 ± 3539 | 17 ± 6 | 111 ± 65 | 25 ± 10 | 90208 ± 23541 |
| White-chinned petrel | 16 | 13355 ± 15522 | 127 ± 69 | 502 ± 407 | <LoD | 138 ± 132 | 101 ± 56 | <LoD | 13110 ± 17785 | 262076 ± 195334 | <LoD | 1399 ± 992 | 1007 ± 646 | 19449 ± 25581 | <LoD | 116 ± 21 | <LoD | <LoD | 4819 ± 1202 | 8 ± 2 | 177 ± 120 | 50 ± 38 | 77646 ± 17983 |
| Wandering albatross | 16 | 7126 ± 7482 | 93 ± 30 | 547 ± 344 | <LoD | 317 ± 478 | 114 ± 123 | <LoD | 6032 ± 2175 | 166858 ± 276706 | <LoD | 991 ± 1514 | 772 ± 1275 | 1720 ± 2043 | <LoD | 151 ± 66 | <LoD | <LoD | 4574 ± 878 | <LoD | 149 ± 176 | 40 ± 30 | 58160 ± 52039 |

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Table 5. Mean concentrations of elements (ng g$^{-1}$ dry wt.) ± SD in muscle tissue of 17 prey species.

| Species | n | Al | As | Ba | Be | Cd | Co | Cs | Cu | Fe | Li | Mn | Mo | Ni | Pb | Rb | Sb | Sc | Se | U | V | W | Zn |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Crustaceans** | | | | | | | | | | | | | | | | | | | | | | | |
| *Euphausia superba* | 10 | 240691 ± 194587 | 2344 ± 251 | 46063 ± 27443 | <LoD | 176 ± 40 | 155 ± 83 | 33 ± 16 | 22123 ± 5644 | 372020 ± 230259 | 463 ± 284 | 7797 ± 4172 | 142 ± 49 | 1192 ± 605 | 167 ± 123 | 1960 ± 515 | 14 ± 14 | 299 ± 75 | 5486 ± 1141 | 61 ± 20 | 1178 ± 761 | 37 ± 101 | 49485 ± 18771 |
| *Themisto gaudichaudii* | 7 | 162821 ± 56608 | 4119 ± 465 | 10519 ± 1519 | <LoD | 19229 ± 2453 | 4269 ± 10614 | 10 ± 2 | 18279 ± 4106 | 395128 ± 152043 | 389 ± 154 | 3927 ± 992 | 692 ± 919 | 1397 ± 1362 | 255 ± 391 | 724 ± 116 | 6 ± 10 | 516 ± 169 | 5142 ± 711 | 145 ± 34 | 971 ± 314 | 2 ± 0 | 51472 ± 25824 |
| **Fish** | | | | | | | | | | | | | | | | | | | | | | | |
| *Champsocephalus gunnari* | 16 | 8046 ± 7507 | 3795 ± 2288 | 1257 ± 1266 | <LoD | 29 ± 16 | 38 ± 28 | 65 ± 40 | 2053 ± 1734 | 82261 ± 50544 | 284 ± 221 | 1108 ± 796 | 41 ± 45 | 281 ± 360 | 33 ± 88 | 1830 ± 874 | 7 ± 5 | 65 ± 19 | 3043 ± 518 | 10 ± 9 | 189 ± 141 | 4 ± 5 | 43330 ± 11102 |
| *Dissostichus eleginoides* | 2 | 1036 ± 118 | 9226 ± 2770 | 168 ± 31 | <LoD | 1 ± 0 | 14 ± 1 | 117 ± 21 | 924 ± 495 | 47900 ± 6075 | 113 ± 17 | 923 ± 180 | 8 ± 2 | 137 ± 110 | 14 ± 6 | 1716 ± 224 | 5 ± 2 | 63 ± 3 | 2224 ± 258 | 2 ± 0 | 11 ± 2 | 2 ± 0 | 11550 ± 2978 |
| *Geotria australis* | 11 | 5787 ± 3719 | 1698 ± 822 | 1060 ± 856 | <LoD | 1948 ± 1151 | 30 ± 31 | 4 ± 3 | 4241 ± 1370 | 43360 ± 21458 | 39 ± 31 | 558 ± 242 | 49 ± 24 | 86 ± 78 | 19 ± 15 | 486 ± 184 | 5 ± 2 | 40 ± 21 | 1815 ± 337 | 4 ± 4 | 207 ± 146 | 3 ± 3 | 26612 ± 12891 |
| *Parachaenichthys georgianus* | 1 | 4348[a] | 4161[a] | 2241[a] | <LoD | 426[a] | 77[a] | 103[a] | 3089[a] | 92029[a] | 241[a] | 2641[a] | 34[a] | 212[a] | 10[a] | 2081[a] | 7[a] | 48[a] | 2801[a] | 6[a] | 168[a] | 4[a] | 41845[a] |
| *Patagonotothen guntheri* | 8 | 10750 ± 11364 | 5968 ± 6388 | 2965 ± 2770 | <LoD | 168 ± 148 | 91 ± 115 | 79 ± 88 | 2480 ± 3288 | 285291 ± 369982 | 930 ± 1170 | 7854 ± 9008 | 42 ± 43 | 476 ± 705 | 38 ± 47 | 2791 ± 3064 | 14 ± 19 | 131 ± 158 | 4100 ± 4509 | 33 ± 41 | 442 ± 564 | 11 ± 10 | 63892 ± 68792 |
| *Pseudochaenichthys georgianus* | 5 | 4151 ± 1720 | 5152 ± 882 | 426 ± 226 | <LoD | 14 ± 14 | 31 ± 18 | 155 ± 37 | 1011 ± 161 | 104317 ± 75579 | 190 ± 67 | 449 ± 181 | 9 ± 3 | 118 ± 67 | 20 ± 12 | 2065 ± 144 | 8 ± 2 | 69 ± 23 | 2622 ± 203 | 4 ± 3 | 128 ± 180 | 2 ± 1 | 33603 ± 12931 |
| **Cephalopods** | | | | | | | | | | | | | | | | | | | | | | | |
| *Galiteuthis glacialis* | 3 | 19409 ± 8627 | 1434 ± 509 | 14951 ± 10737 | <LoD | 2105 ± 1745 | 44 ± 3 | 8 ± 3 | 61979 ± 31048 | 66581 ± 11678 | 121 ± 102 | 1161 ± 213 | 793 ± 1109 | 704 ± 446 | 682 ± 547 | 578 ± 230 | 55 ± 11 | 117 ± 100 | 3703 ± 346 | 231 ± 109 | 379 ± 374 | 17 ± 2 | 85928 ± 34899 |
| *Gonatus antarcticus* | 2 | 6378 ± 165 | 2065 ± 596 | 1378 ± 835 | <LoD | 901 ± 676 | 15 ± 1 | 6 ± 0 | 16853 ± 13774 | 19097 ± 10696 | 18 ± 23 | 1058 ± 471 | 1423 ± 1894 | 549 ± 329 | 10 ± 0 | 353 ± 39 | 39 ± 33 | 26 ± 25 | 3162 ± 271 | 75 ± 48 | 401 ± 409 | 2 ± 0 | 94001 ± 111988 |
| *Kondakovia longimana* | 2 | 7472 ± 3945 | 1706 ± 113 | 2133 ± 1636 | <LoD | 9216 ± 6413 | 30 ± 4 | 23 ± 14 | 13216 ± 1778 | 46122 ± 11729 | 122 ± 28 | 1021 ± 117 | 181 ± 176 | 473 ± 262 | 24 ± 20 | 1212 ± 52 | 13 ± 10 | <LoD | 4734 ± 381 | 118 ± 129 | 246 ± 184 | 2 ± 0 | 91585 ± 35517 |
| *Martialia hyadesi* | 2 | 4056 ± 1167 | 4828 ± 1290 | 82 ± 14 | <LoD | 14524 ± 1805 | 46 ± 5 | 18 ± 2 | 28745 ± 10952 | 19018 ± 400 | 139 ± 25 | 1083 ± 2 | 68 ± 3 | 353 ± 14 | 51 ± 31 | 4997 ± 464 | 1 ± 0 | 55 ± 8 | 3498 ± 347 | 8 ± 2 | 109 ± 34 | 2 ± 0 | 69918 ± 10882 |
| *Moroteuthis knipovitchi* | 4 | 4830 ± 3376 | 2944 ± 1689 | 976 ± 844 | <LoD | 10009 ± 10296 | 32 ± 17 | 12 ± 5 | 17517 ± 16023 | 46102 ± 40016 | 129 ± 106 | 1652 ± 231 | 345 ± 551 | 328 ± 142 | 90 ± 148 | 1888 ± 1203 | 22 ± 37 | 46 ± 26 | 4709 ± 1817 | 47 ± 63 | 222 ± 183 | 17 ± 31 | 95067 ± 36352 |
| *Psychroteuthis glacialis* | 2 | 9130 ± 5520 | 1397 ± 94 | 4598 ± 4137 | <LoD | 2248 ± 460 | 37 ± 13 | 2 ± 2 | 16654 ± 5875 | 27134 ± 2526 | 93 ± 130 | 774 ± 383 | 1176 ± 1447 | 297 ± 107 | 61 ± 72 | 513 ± 339 | 10 ± | 51 ± 62 | 4024 ± 617 | 153 ± 152 | 577 ± 166 | 2 ± 0 | 24226 ± 34026 |

[a] No SD available due to low sample size (included in table for completeness but excluded in statistical analyses).

664 **Table 6.** Univariate GLM results for element concentrations in blood and feathers of 10

665 Procellariiform species. Model components were selected by AIC from all possible model sub-sets

666 with two factorial levels. Parameter estimates (β) shown as Na if non-significant or from multi-level

667 parameters. Significance values represent values for the parameter within the overall model.

| Tissue | Element | Parameter | df | F | $p$ | β | s.e. |
|---|---|---|---|---|---|---|---|
| Blood | LogAs[a] | $\delta^{15}N$ | 1 | 13.8 | <0.001*** | 4.2 | 1.1 |
| | | $\delta^{13}C$ | 1 | 13.9 | <0.001*** | -2.8 | 0.7 |
| | | Species | 8 | 1.9 | 0.061 | Na | Na |
| | | Species* $\delta^{13}C$ | 8 | 1.9 | 0.067 | Na | Na |
| | | Species* $\delta^{15}N$ | 8 | 2.9 | 0.006** | Na | Na |
| | | $\delta^{13}C$* $\delta^{15}N$ | 1 | 13.4 | <0.001*** | 0.2 | 0.1 |
| | LogCd[b] | Species | 9 | 13.1 | <0.001*** | Na | Na |
| | Se[c] | $\delta^{13}C$ | 1 | 2.8 | 0.103 | -19944.2 | 23857.4 |
| | | Species | 9 | 2.4 | 0.014* | Na | Na |
| | | Species* $\delta^{13}C$ | 1 | 2.5 | 0.011* | Na | Na |
| | LogRb[d] | Species | 9 | 4.9 | <0.001*** | Na | Na |
| | LogMo[e] | $\delta^{13}C$ | 1 | 3.9 | 0.052 | -0.1 | 0.03 |
| | | Species | 9 | 7.5 | <0.001*** | Na | Na |
| Feathers | LogMn[f] | Species | 9 | 4.6 | <0.001*** | Na | Na |
| | LogFe[g] | Species | 9 | 5.0 | <0.001*** | Na | Na |
| | LogCo[h] | Species | 9 | 7.45 | <0.001*** | Na | Na |
| | LogAs[i] | Species | 9 | 5.8 | <0.001*** | Na | Na |
| | | $\delta^{15}N$ | 1 | 2.3 | 0.129 | 0.03 | 0.02 |
| | LogMo[j] | Species | 9 | 2.2 | 0.029* | Na | Na |
| | | $\delta^{15}N$ | 1 | 2.6 | 0.112 | 0.2 | 0.1 |
| | | Species* $\delta^{15}N$ | 9 | 2.0 | 0.043* | Na | Na |
| | Se[k] | $\delta^{13}C$ | 1 | 5.1 | 0.025* | -551.4 | 243.2 |
| | | Species | 9 | 6.9 | <0.001*** | Na | Na |
| | LogU[l] | Species | 9 | 14.2 | <0.001*** | Na | Na |

668 [a] $R^2 = 0.69$  [g] $R^2 = 0.24$

669 [b] $R^2 = 0.48$  [h] $R^2 = 0.32$

670 [c] $R^2 = 0.51$  [i] $R^2 = 0.30$

671 [d] $R^2 = 0.25$  [j] $R^2 = 0.34$

672 [e] $R^2 = 0.37$  [k] $R^2 = 0.44$

673 [f] $R^2 = 0.23$  [l] $R^2 = 0.48$

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676 **Table 7.** Univariate GLM results for $\log_{10}$ transformed arsenic concentrations of tissue samples from

677 prey groups (see Table 5 for groups). Model components specified in table as selected by AIC from all

678 possible model sub-sets with two factorial levels. Parameter estimates (β) shown as Na for multi-level

679 parameters.

| Parameter | df | F | p | β | s.e. |
|---|---|---|---|---|---|
| Group | 2 | 8.9 | <0.001*** | Na | Na |
| $\delta^{15}N$ | 1 | 5.6 | 0.021* | -0.2 | 0.1 |
| $\delta^{13}C$ | 1 | 3.8 | 0.054 | 0.2 | 0.1 |
| Group* $\delta^{15}N$ | 2 | 9.4 | <0.001*** | Na | Na |
| Cephalopod* $\delta^{15}N$ | 1 | Na | <0.001*** | -0.3 | 0.1 |
| Crustacea* $\delta^{15}N$ | 1 | Na | 0.010* | -0.3 | 0.1 |
| Fish* $\delta^{15}N$ | 1 | 0[b] | 0[b] | 0[b] | 0[b] |
| $\delta^{13}C$* $\delta^{15}N$ | 1 | 5.6 | 0.021* | -0.02 | 0.01 |

680 [a] $R^2 = 0.43$
681 [b] This parameter is set to zero because it is redundant.
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