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1 Running head: Trans-ecosystem biotransport of contaminants

2 **Biotransport of Metallic Trace Elements from Marine to**
3 **Terrestrial Ecosystems by Seabirds**

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Abstract

Physical systems, such as currents and winds, have traditionally been considered responsible for transporting contaminants. While evidence is mounting that animals play a role in this process through their movements, we still know little about how such contaminant biotransport occurs, and the extent of effects at deposition sites. Here, we address this question by studying how rhinoceros auklets (*Cerorhinca monocerata*), a seabird that occurs in immense colonies (~300,000 pairs at our study site, Teuri Island), affect contaminant levels at their colony, and at nearby sites. More specifically, we hypothesize that contaminants are transported and deposited by seabirds at their colony, and that these contaminants are passed on locally to the terrestrial ecosystem. To test this hypothesis, we analyzed the concentration of nine heavy metal and metalloids, as well as $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ stable isotopes, in bird tissues, plants and soil, both within and outside of the colony. Our results show that rhinoceros auklets transport marine-derived mercury, possibly from their wintering location, and deposit mercury *via* their feces at their breeding site, thereby contaminating plants and soils within the breeding colony. Our work confirms not only that animals can transport contaminants from marine to terrestrial ecosystems, potentially over unexpectedly long distances, but also that bird tissues contribute locally to plant contamination.

Keywords: Biological transport; rhinoceros auklet; mercury; bioaccumulation; machine learning

INTRODUCTION

Migratory species, such as marine apex predators, can transport significant quantities of nutrients across ecosystem boundaries into recipient food webs (Michelutti et al., 2009). These animals often carry high contaminant loads as a result of both bioaccumulation and biomagnification through marine foodwebs (Fisk et al., 2003; Mallory and Braune, 2012). Among top predators, recent studies have highlighted a major role played by seabirds in transferring contaminants from marine to terrestrial ecosystems (Sun et al., 2004; Evenset et al., 2007; Choy et al., 2010a,b). As seabirds nest in immense colonies that can be at the same location for centuries, they can create highly concentrated contaminated sites at their breeding site through the accumulation of feces, feathers, and carcasses (Blais et al., 2005; Ellis et al., 2006; Mulder et al., 2011; Bauer and Hoyer, 2014).

Among these contaminants, mercury is particularly problematic because this trace metal is a neurotoxin occurring in ecosystems throughout the world. In contrast to some organic contaminants, mercury pollution is difficult to remediate (Elliott and Elliott, 2016) because (i) mercury does not naturally degrade, and (ii) it is continually released by common industrial activities (Driscoll et al., 2013; Mason et al., 2012; Selin, 2009; Vo et al., 2011). As such, mercury levels in surface oceans have tripled since the Industrial Revolution (Driscoll et al., 2013; Lamborg et al., 2014; Poulain et al., 2015). Contaminants such as mercury are being added to recipient sites at concentrations far beyond those individually transported by physical processes (Blais et al., 2007). Yet, among these physical drivers, the relative roles of anthropogenic vs. biological vectors are difficult to tease apart (Lamborg et al., 2014), and, to date, received little attention. While the role of seabirds in transferring marine-derived nutrients (*e.g.*, Farina et al., 2003; Ellis et al., 2006) and contaminants (*e.g.*, Blais et al., 2007; Michelutti et al., 2010) across ecosystem boundaries is recognized, both the origin and the pathway from sea to land taken by these biotransported contaminants remains unclear.

Here, we assess the origin and pathway of allochthonous mercury transported and

deposited by seabirds at their breeding colony, and determine the impact of such biotransport on terrestrial ecosystems. To evaluate the role of seabirds as biovectors, we measured mercury concentrations in rhinoceros auklets (*Cerorhinca monocerata*) sampled at Teuri Island, in the Sea of Japan – one of the largest colonies of this species in the world, where *ca.* 300,000 pairs breed annually (Watanuki and Ito, 2012). As already advocated (Mallory et al. 2010), we simultaneously sampled multiple proxies of seabird health (tissues, feces), as well as soil and plants within and outside seabird colony to measure the impact of these sea-to-land biovectors. This allowed us to study metal biotransport in which auklets bioaccumulate mercury at sea through foraging, carry it in their tissues to their breeding site, and deposit it mainly in guano (auklets do not moult at breeding stage: Gaston and Dechesne, 1996), which degrades and is eventually transferred to plants. To characterize this biotransport process, we measured mercury concentrations in soil, plant roots and plant leaves, at both auklet-affected sites and control sites, where no seabirds have nested at least within the past 30 years (Y. Watanuki, unpublished data). Because mercury burden is often correlated with levels of other elements (Michelutti et al., 2010), we also measured levels of eight other metals and metalloids in the same samples of roots to evaluate the effects of auklets on the root metal profiles.

METHODS AND MATERIALS

Ethical note

All work was conducted under the permits of the Ministry of the Environment, the Agency of Cultural Affairs, and Hokkaido University's Ethical Review Board. To avoid bird disturbance, handling of auklets was kept to a minimum.

Study site and field collections

Teuri Island (44°24'N, 141°17'E) lies 38 km off Haborocho harbor along the northwestern coast of Hokkaido, in the northern Sea of Japan. In 2016, adult rhinoceros auklets were

caught by hand during incubation (May) and chick-provisioning (June-July) periods. Blood (1 mL) was drawn from the brachial vein using 25G syringes. Upon collection, the blood samples were immediately centrifuged to separate plasma from red blood cells, and stored in a freezer at -20°C. To collect fecal samples, birds were placed in a box before blood collection. We released the birds typically within 10 min, regardless of whether fecal samples were successfully collected. No individual was sampled more than once. Whole plants were collected during incubation and chick-provisioning periods from both the auklet affected (*i.e.*, center of the colony) and unaffected sites (*i.e.*, outside of colony; Fig 1). We set up study plots at two auklet-affected sites (approximately 100 m apart) and at three control sites to examine the effect of auklets on the entire island. We focused vegetation collection on the Scandinavian small-reed (*Calamagrostis purpurea* (Trin.) Trin. subsp. *langsдорffii* (Link) Tzvelev; *Iwanogariyasu* [イワノガリヤス] in Japanese), the dominant plant species at both the auklet-affected and control sites. Surface soil samples were simultaneously taken near the plant sample sites using a trowel. Additionally, soil core samples were taken near the plant and soil sample sites using a 4.4 cm internal diameter custom-made corer that was pushed directly into the soil to identify the longitudinal effect of auklet deposition on soil profiles. The core tubes were pushed until they met strong resistance (~9-12 cm), indicating that the entire sedimentary record was retrieved. The cores were sectioned at 1 cm intervals on site using a vertical extruder. We finally note that while the area occupied by the colony has expanded or contracted since the early 1970s, the sites that we selected remained well within or well outside of the colony (respectively) over the past ~50 years (Y. Watanuki, pers. obs.).

Laboratory analyses

All samples were shipped frozen to the Department of Environmental Veterinary Sciences, Hokkaido University. We then followed standard laboratory procedures for each analysis (see below). More specifically, for plant samples, the whole plants were rinsed in distilled water and the roots and leaves were extracted. Feces and the extracted plant samples were air-dried,

weighed, dried at 50°C in an oven for approximately 48 h, and manually homogenized prior to analysis of each sample. For animal tissues, red blood cells were analyzed as wet weight for mercury analysis, but freeze-dried for stable isotope analysis prior to the analyses. Soil and plant samples were divided in half, with one half used for measuring metal and metalloid concentrations, and the rest used for measuring stable isotope ratios of carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$), which are proxies for feeding habitat and trophic level, respectively, and to examine the impacts of those in soil and plant samples (Newsome et al., 2007). Blood and fecal samples were analyzed separately for measuring metal and metalloid concentrations, and stable isotope ratios of carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$), due to small quantity of samples. Sample sizes for each analysis are shown in Table S1.

Total mercury analyses

Concentrations of total mercury (denoted Hg hereafter, unless otherwise stated) in red blood cells, feces, soil, root and leaf samples were determined using a Direct Thermal Decomposition Mercury Analyzer (MA-3000, Nippon Instruments). After preparation of the calibration standards, the concentration of Hg was measured by thermal decomposition. Analytical accuracy for Hg was determined by analyzing one or two blank samples with each sample set, as well as Standard Reference Materials (SRMs): BCR-320, DOLT-4 and tomato leaves obtained from the Canadian National Research Council. Recoveries of all SRMs were within the certified range of values (BCR-320: $98 \pm 4\%$ ($n = 2$); DOLT-4: $101 \pm 8\%$ ($n = 2$); tomato leaves: $98 \pm 2\%$ ($n = 12$), Average $\pm$ SD). The detection limit of the analyzer was at 0.001 ng. Hg concentrations for blood are reported in $\mu\text{g g}^{-1}$ wet weight (ww), and for the remaining matrices in $\mu\text{g g}^{-1}$ dry weight (dw).

Extraction and analysis of heavy metals and metalloids

We measured concentrations of eight elements (Cd, Cr, Co, Ni, Cu, Zn, Pb, and As) as per Nakata et al. (2016), using an inductively coupled plasma-mass spectrometer (ICP-MS; 7700 series, Agilent technologies, Tokyo, Japan). Briefly, all laboratory materials and instruments used in the heavy metal analysis were washed with 2% nitric acid (HNO₃), and rinsed at least twice with distilled water. Samples of approximately 1.0 g of plant roots were dried for 48 h in an oven at 50°C. The dried samples were placed in pre-washed digestion vessels, followed by acid digestion using nitric acid (atomic absorption spectrometry grade, 60%, Kanto Chemical Corp., Tokyo) and hydrogen peroxide (Cica reagent, 30%, Kanto Chemical Corp.) in a microwave digestion system (Speed Wave MWS-2, Berghof, Germany). After cooling, each mixture was transferred into a plastic tube, and various elements were determined using an ICP-MS. Analytical quality control was performed using the DORM-3, and DOLT-4 certified reference materials (National Research Council of Canada, Ottawa, ON). Replicate analysis of these reference materials showed good recoveries (95–105%, $n = 5$, respectively). The instrument detection limits for Cd, Cr, Co, Ni, Cu, Zn, Pb and As were 0.2, 0.5, 0.5, 0.5, 1.0, 0.1, 1.0 and 2.0 $\mu\text{g kg}^{-1}$, respectively.

Stable isotope analyses

All processed and homogenized samples were shipped to the Port and Airport Research Institute in Yokosuka, Japan. To remove inorganic carbon, all samples except for blood were acidified with 1N HCl and dried at 60°C. Lipids were not removed from blood and plant samples due to expected low lipid levels. Isotopically fractionated metabolites, such as urea and ammonium as well as inorganic carbon were removed from fecal samples using a 2:1 chloroform:methanol soak and rinse (Kuwaie et al., 2008). Isotope fractionation in catabolism occurs when nitrogen in amino acid is deaminated to produce metabolites depleted in ¹⁵N (Fry, 2006). Thus, uric acid, which may be a major nitrogen metabolite in fecal samples, is not fractionated because uric acid is not produced through deamination. Both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ were

measured with an isotope ratio mass spectrometer (Delta Plus Advantage, Thermo Electron, Bremen, Germany) coupled with an elemental analyzer (Flash EA 1112; Thermo Electron). Results were reported in delta notation in parts per thousand (‰) relative to VPDB ($\delta^{13}\text{C}$) and Air ($\delta^{15}\text{N}$). L-Histidine ($\delta^{13}\text{C}$ –PDB (‰) = -10.18; $\delta^{15}\text{N}$ –Air (‰) = -7.81; Shoko Co., Ltd., Minato-ku, Tokyo) was included as an internal standard every 5th sample to check analytical accuracy. Based on within-run replicate measurements of multiple standards (L-Histidine; L-Alanine ($\delta^{13}\text{C}$ –VPDB (‰) = -19.6; $\delta^{15}\text{N}$ –Air (‰) = 10.1; Shoko Science Co., Ltd., Yokohama, Kanagawa); L-Alanine ($\delta^{13}\text{C}$ –VPDB (‰) = -19.6; $\delta^{15}\text{N}$ –Air (‰) = 26.1; Shoko Science Co., Ltd.), measurement precision for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values was estimated to be always at or below 0.3‰.

Statistical analyses

Elemental compositions were log₁₀-transformed prior to analysis. We examined the effect of site (auklet-affected vs. control), stage (incubation vs. chick-provisioning), $\delta^{15}\text{N}$, $\delta^{13}\text{C}$ in roots and an interaction term of stage $\times$ $\delta^{15}\text{N}$ on Hg in roots when using linear models. Model selection was based on Akaike's information criteria (AIC). The model that received the lowest AIC was designated as the best model. Models within 2.0 units of the best model were considered as indistinguishable from the best model (Burnham and Anderson, 2010). To account for uncertainty in the model selection process, a model averaging approach was used, in which the parameter estimates of factors included in the adequate models were weighed with the corresponding Akaike weights, and averaged. To visualize if the auklet-affected site could be teased apart from control sites, a principal component analysis (PCA) was performed with nine heavy metal (Cd, Pb, Hg, Cu, Zn, Cr, Co, Ni) and metalloid (As) concentrations in roots. To assess the predictive power of metal and metalloid concentrations on determining the presence of auklets, we employed a supervised machine-learning algorithm based on adaptive boosting (Freund et al. 1996). The classifier's accuracy was determined by 10-fold cross-validation (CV₁₀), where the algorithm is trained on nine tenths of the data and the last

decile is used to compute a confusion matrix; this process was repeated 100 times. Finally, significance between the two types of sites for nine element concentrations was assessed based on the Dunn test, and general linear models were fitted to the data. All statistical analyses were performed in R 3.4.2 (R Core Team, 2017). For comparison purposes in the Discussion, we assumed a moisture of 65% in red blood cells (Tartu et al., 2014; Bond and Robertson, 2015) and 79% in whole blood (Eagles-Smith et al., 2008), and also that red blood cells consist of 45% of the whole blood, while most mercury is accumulated in red blood cells (Bond and Robertson 2015).

RESULTS

Mercury and stable isotope ratios in red blood cells and feces

We detected quantifiable concentrations of Hg in blood and feces in all individuals. Mean [Hg] in auklet red blood cells during the study period [mean $\pm$ 1SD (range)] was 0.86 ± 0.27 (0.47 - 1.6) $\mu\text{g g}^{-1}$ ww (Fig. 2a), and was similar between incubation and chick-provisioning periods (Table 1; $P = 0.06$). Mean [Hg] in auklet feces during the entire study period was 0.08 ± 0.10 $\mu\text{g g}^{-1}$ dw and decreased from incubation to chick rearing (Table 1; $t = 2.25$, $P = 0.04$, $\text{df} = 8$). Stable isotope ratios, $\delta^{15}\text{N}$ ($\Delta\text{AIC} = 1.71$) or $\delta^{13}\text{C}$ ($\Delta\text{AIC} = 1.99$), varied independently of Hg in red blood cells, but were stage-dependent (Fig. S1), through time (from incubation to chick-rearing), $\delta^{15}\text{N}$ increased, while $\delta^{13}\text{C}$ decreased (Table 1, Fig. S1).

Mercury and stable isotope results in plants and soil

Mean [Hg] in roots at the auklet-affected site was higher than those at the control site ($P < 0.0001$; Fig 2d), but mean concentrations in leaves were marginally detectable and similar between the two sites ($P = 0.08$; Table 2). Thus, only roots were considered for further statistical analyses. Mean [Hg], $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ in soil were similar between auklet-affected and control sites (Table 2, Fig. 2c). We did not find any auklet effects on [Hg], $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ in soil core samples at any given depth (Table S2). Although we did not compare identical depths, because values did not vary with depth at either site, we concluded that levels were

likely similar among sites. Among root samples, the fitting of linear models explaining root [Hg] showed that site (auklet affected vs. control), $\delta^{15}\text{N}$ (positive), and breeding stage (incubation vs. chick-provisioning) were variables included in the best models (with $\Delta\text{AIC} < 2$; Table 3).

Concentration of metals and metalloids in roots

Based on a PCA, the concentrations of the nine elements (Cd, Pb, Hg, Cu, Zn, Cr, Co, Ni, and As) allow discrimination of the two types of sites, with some overlap (Fig. 3). To differentiate affected from non-affected sites based on these nine concentrations, the trained adaptive boosting classifier showed that only three of them (As, Cd, and Co) are critical (importance values > 1 ; CV_{10} classification success rate = 80%; Fig S2), and hence provide a distinct signature for auklet presence. Indeed, all three have significantly different concentrations among sites (at the 0.5% level) with, however, [Cd] being higher at the control site (Fig. 4). Hg is not directly a critical feature for classification for three reasons: (i) [Hg] covaries with [As] and [Co], with all three elements explaining most of the concentration variance among sites (50%; Fig. 3), (ii) [As] and [Co] are both orthogonal to (independent of) [Cd]; and (iii) all three concentrations, [As], [Co], and [Cd], are an order of magnitude higher than [Hg], whose signal is hence overwhelmed (Fig. 4). Most remarkably, although Hg is not critical in distinguishing auklet presence at a given site, Hg is the element that, as shown above, is the most affected by auklets ($P < 10^{-3}$) at their breeding site.

DISCUSSION

Seabirds are critical biovectors in ecosystems due to their frequent commutes between sea and land during the breeding season (Blais et al., 2007; Mallory et al., 2015). Based on a quasi-experimental approach, we evaluated the possible role of seabirds as biovectors of allochthonous mercury from oceanic into terrestrial ecosystems, by focusing on a highly mobile marine top predator, the rhinoceros auklet. We show that seabirds deposited allochthonous mercury from sea to land *via* their feces, at their breeding colony, and that

254 allochthonous mercury was then transferred to terrestrial plants, thereby crossing ecosystem
255 boundaries.
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Hg fluxes in seabirds through their breeding season

We detected mercury in both red blood cells and feces of all individuals assayed, confirming that auklets carry detectable amounts of mercury not only in their tissues such as liver and kidney (Ishii et al., 2014), but also in their fecal matter. Average blood [Hg] measured at Teuri (2.46 ± 0.76 1SD $\mu\text{g g}^{-1}$ dw, Table 1) was higher to that reported at $1.75 \mu\text{g g}^{-1}$ dw on average (note: this is measured in whole blood; Hipfner et al., 2011) in a Canadian auklet population in the eastern Pacific, or in other piscivorous species such as the Antarctic petrels (at $0.84 \mu\text{g g}^{-1}$ dw on average: *Thalassoica antarctica*; Carravieri et al., 2017). While other work reported higher concentrations in seabirds (at $8.22 \mu\text{g g}^{-1}$ dw on average in brown skuas *Stercorarius antarcticus*; Goutte et al., 2014; $2.7 \mu\text{g g}^{-1}$ dw in snow petrels *Pagodroma nivea*; Tartu et al., 2014, see also review in Ackerman et al., 2016), the lowest [Hg] that we detected in red blood cell ($1.34 \mu\text{g g}^{-1}$ dw, converted from ww) still exceeds the avian affect threshold ($1.20 \mu\text{g g}^{-1}$ dw; in double-crested cormorant *Phalacrocorax auratus* Gibson et al., 2014; $1.20 \mu\text{g g}^{-1}$ dw; in black-legged kittiwake *Rissa tridactyla* Tartu et al., 2015). It is however unclear whether or to what extent such elevated Hg contamination levels in Teuri auklets are possibly detrimental to their health.

Such detrimental effects potentially occur throughout the entire breeding season of auklets, as their mean blood mercury concentrations were essentially identical between the two breeding stages. The timeframe for integration of mercury into the blood system (half-life of 30-65 days reported in great skuas; Bearhop et al., 2000) and of $\delta^{15}\text{N}$ / $\delta^{13}\text{C}$ (half-life of roughly 20 days in this system; Carleton and Martinez del Rio, 2005) are roughly the same as the length of their breeding season, hereby ruling out the possibility that high tissue [Hg] and isotopic ratio changes were due to proximal (*i.e.*, at colony) causes, and suggesting that most of the Hg measured in auklets comes from their wintering grounds. While it is likely that foraging habitats and/or prey of auklets changed as breeding progressed (*i.e.*, average $\delta^{13}\text{C}$ values and $\delta^{15}\text{N}$ changed over the course of breeding; Fig S1), blood [Hg] was constant throughout the breeding season, further suggesting that either no Hg intake occurred during

breeding, or that [Hg] in auklets was at a dynamic equilibrium. However, the reason why blood [Hg] was stage-independent while blood $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ are stage-dependent is unexpected. Indeed, previous work on rhinoceros auklets at Teuri showed that, as these birds shift from smaller crustaceans such as copepods (for self-feeding during incubation) to larger fish (for chick-provisioning), their $\delta^{15}\text{N}$ values increase (Ito et al., 2010). As Hg levels in copepods are lower than in fish, this dietary shift should lead to a concomitant increase in blood Hg (Davoren and Burger, 1999), which we did not observe here. While unexpected, our results are however not unique, as Hg levels do not always systematically increase with trophic levels (Hipfner et al., 2011), and other studies also failed to show clear patterns between $\delta^{15}\text{N}$ and Hg within single seabird populations (Elliott et al., 1992, 2016; Hipfner et al., 2011; Tartu et al., 2014). These conflicting results suggest that blood [Hg] during breeding results from a dynamic equilibrium, between Hg intake and offloading, and that a lack of increase in blood [Hg] concomitant to an increase in $\delta^{15}\text{N}$ might be due to Hg contents being either higher than expected in copepods, or lower than expected in larger fish, possibly due to a shift in availability of preys that are usually harvested by auklets during chick provisioning.

However, a constant blood [Hg] may not just be due to extrinsic factors, but also result from intrinsic features such as the long-term persistence of Hg in auklets' tissues (Monteiro and Furness, 1995). Age may also be a confounding variable, since Hg may bioaccumulate in body tissues with age in marine vertebrates (Thompson, 1990 – but see Furness et al., 1990). Experimental studies showed that excretion of Hg *via* feces was about 22% of the intake in black-headed gull chicks (*Chroicocephalus ridibundus*; Lewis and Furness, 1991). While we could not control mercury intake, our recorded mean Hg levels in feces was 3% of the recorded blood Hg when data were pooled. Auklet species do not molt during the breeding phase (Pyle, 2009; Sorensen et al., 2010), and thus, biological transport by auklets occurs *via* feces and potentially carcasses and/or abandoned eggs rather than *via* feathers. However, fecal [Hg] decreased between the two breeding stages, and as we showed above that Hg in consumed prey most likely increases through breeding stages, an alternative explanation must

be sought. As sample sizes for feces analyses are quite small (see Table 2), the interpretation of these results requires caution.

Hg transfer from contaminated seabirds to plants and soil

The roots of the plants at the colony were found to have higher Hg levels than those at the control sites (Table 2), hereby suggesting that the origin of this contamination was likely to be seabirds. Although Hg levels in roots are much lower than in auklet tissues and feces, the detected Hg is likely to be organic Hg as a result of biotransport, and should not be detected under Japanese Environmental Guidelines (Ministry of Environment Japan).

In this study, we set up three control plots at Teuri where auklets or other seabirds are rarely observed flying over or perch on the sites (A. Shoji, pers. obs.). Although we did not experimentally manipulate auklet activity, or even their contamination levels, there is growing evidence supporting a causal relationship between contaminated seabirds and contaminated plants, as allochthonous inputs from seabirds significantly affect the chemical properties of both soil and flora in the proximity of their colonies (Breuning-Masden et al., 2008; Zwolicki et al., 2013, 2015; Ziolek and Melke, 2014). While seabirds can efficiently transfer heavy metals to both sediments and aquatic plants (Godzik, 1991; Blais et al., 2005; Evenset, 2007), our results are based on a novel approach that allowed us to show evidence for a direct transfer from a marine to a terrestrial ecosystem, by simultaneously collecting samples along the biotransport chain, from the biovectors to the terrestrial plants. Indeed, as our study sites are separated by at most 2 km, and are all located in similar soil, geologic, and atmospheric contexts, it is unlikely that variation in soil, geology, or atmospheric deposition rates explain the differences in [Hg] or in the other metals that we measured. Therefore, the most likely source of Hg in roots is from contaminated seabirds, which as discussed above, most likely bring Hg from outside of the colony area, from their wintering grounds.

Mercury in roots does not seem to be transferred to the leaves of contaminated plants. Indeed, while Hg in leaves was similar between auklet-affected and control sites, it was remarkably lower than in roots (Table 2). Although few studies have examined Hg uptake by plant tissues in wild populations, Tomiyasu et al. (2003) reported that Hg in leaves is lower than in roots in goldenrod (*Solidago altissima*), and showed that Hg does not seem to move from roots to leaves. While their results suggest that Hg in leaves may originate from the air, and not from the roots, it may also explain why our Hg levels in leaves are lower than in roots – and are also site-independent – if the Hg intake pathways differ in leaves and roots. We suggest that Hg in roots results from plant uptake from the soil, where Hg was deposited by contaminated seabirds *via* their fecal matter.

Under this scenario, it could be expected that Hg in the soil of auklet-affected sites would be higher than at our control sites. Contrary to this expectation however, we found that Hg in soil was not different between auklet-affected and control sites, while still being significantly higher than in contaminated roots (Fig. 2). Note that here, we measured Hg as *total mercury* concentration. A possible explanation of this counter-intuitive result is that inorganic mercury contained in the soil may have overwhelmed the signal from biovectors (Lubick and Malakoff, 2013). For instance, Asian dust storms containing Hg from China occur year round, but are more intense in the auklet's spring breeding season (Japan Meteorological Agency, 2017). Indeed, our soil Hg concentrations exceeded soil quality guidelines (Ministry of Environment Japan, 2008). We propose that organic Hg (methylmercury: the form of Hg that can pass through biological membranes; Mason et al., 1996), which is carried and deposited by auklets, is more efficiently absorbed by the roots than inorganic Hg. As no information on the efficiency of this process seems to be available, further quantification of organic Hg and tracing of Hg in soil with stable Hg isotope ratios ($\delta^{200}\text{Hg}$; Lepak et al., 2015) would help resolve some of these issues.

While we found that biotransport significantly affected total mercury, other metals that we measured were not affected in the same way or extent. Indeed, while higher

concentrations of Hg, As, Pb, Co were found in roots at the auklet-affected site, the reverse was found for Cd. The reason why Cd concentration in roots at the control site was higher than at the auklet affected site is unclear, but may not be biologically meaningful due to the large variance at each site.

We demonstrated that the concentration profile of metal and metalloid contaminants alone creates a signature for plants impacted by seabirds, and that rhinoceros auklets act as a major biovector of such contaminants, transferring mercury and other metalloids from oceanic to terrestrial ecosystems. In theory, the source of this mercury can either be local, resulting from their foraging activity during the breeding season, or come from the distant wintering grounds. However, we provide some evidence downplaying the importance of local foraging sources of Hg transferred to local plants, suggesting instead that transferred Hg potentially resulted from their migratory behavior. Our work illustrates the importance of accounting for this source of contamination in terrestrial habitats when assessing the environmental risk of bioaccumulated contaminants.

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Table 1. Total mercury concentrations (Hg in $\mu\text{g g}^{-1}$), stable isotope ratios ($\delta^{15}\text{N}$, $\delta^{13}\text{C}$ in ‰) and sample sizes (N) in red blood cells (RBC in ww) and fecal samples (feces in dw) during incubation and chick-provisioning periods. Assuming moisture content of 65% in RBC, converted values in RBC in dw were provided for comparable purpose.

	Sample	Incubation	converted in <i>dw</i>	<i>N</i>	Chick- provisioning	converted in <i>dw</i>	<i>N</i>
Hg	RBC	0.99 ± 0.32	$\approx 2.83 \pm 0.32$	9	0.77 ± 0.18	$\approx 2.20 \pm 0.18$	12
	Feces	0.22 ± 0.17		2	0.05 ± 0.01		7
$\delta^{15}\text{N}$	RBC	10.90 ± 0.30		24	11.9 ± 0.40		28
	Feces	11.60 ± 1.60		4	12.7 ± 0.60		12
$\delta^{13}\text{C}$	RBC	-19.00 ± 0.30		24	-20.00 ± 0.40		28
	Feces	-21.60 ± 1.10		4	-23.80 ± 0.60		12

Table 2. Total mercury concentration (Hg), stable isotope ratio ($\delta^{15}\text{N}$, $\delta^{13}\text{C}$), sample size (N) in roots and leaves of Scandinavian small-reed, and soil, at auklet-affected (affected) and control (control) sites.

	Sample	Incubation				Chick-provisioning			
		Affected	N	Control	N	Affected	N	Control	N
Hg	Roots	0.04 ± 0.01	6	0.02 ± 0.01	9	0.07 ± 0.07	7	0.02 ± 0.01	9
	Leaves	0.0077 ± 0.0023	6	0.0051 ± 0.0022	9	0.0097 ± 0.0048	6	0.0099 ± 0.0049	17
	Soil	0.06 ± 0.01	2	0.09 ± 0.07	3	0.06 ± 0.05	9	0.09 ± 0.05	7
$\delta^{15}\text{N}$	Roots	6.0 ± 1.5	8	6.15 ± 2.8	7	7.6 ± 2.3	10	7.1 ± 2.4	7
	Leaves	4.7 ± 1.9	8	4.7 ± 1.9	9	8.4 ± 3.5	6	6.7 ± 2.8	18
	Soil	10.0 ± 2.4	2	9.2 ± 2.3	3	10.5 ± 1.9	10	9.4 ± 2.0	9
$\delta^{13}\text{C}$	Roots	-27.4 ± 1.1	8	-28.0 ± 0.5	7	-27.2 ± 1.1	10	-27.3 ± 1.1	7
	Leaves	-27.5 ± 1.3	8	-28.3 ± 0.3	9	-28.3 ± 1.2	6	-28.1 ± 1.1	18
	Soil	-25.6 ± 0.2	2	-27.3 ± 0.7	3	-25.5 ± 0.3	10	-26.4 ± 1.1	9

Table 3 Model selection results by ΔAIC based on linear models explaining $\log_{10}$ -transformed Hg in roots of Scandinavian small-reed at Teuri in 2016 ($\log_{10}(\text{Hg}) \sim \Sigma$ variables). The best model is in boldface. The number of parameters (df) and Akaike weight (w_i) are also shown. Potential factors included are Site (auklet affected *vs.* control), Stage (incubation *vs.* chick-provisioning), $\delta^{15}\text{N}$, $\delta^{13}\text{C}$ in the roots and an interaction term of Stage $\times$ $\delta^{15}\text{N}$. Models within 2 AIC units of the best model are shaded in light gray.

	Model (Σ variables)	df	ΔAIC	w_i
Hg Roots	Site + $\delta^{15}\text{N}$	22	0.00	0.39
	Site + Stage + $\delta^{15}\text{N}$	21	1.07	0.23
	Site + Stage + $\delta^{13}\text{C}$	21	2.89	0.09
	Site + Stage + $\delta^{15}\text{N}$ + $\delta^{13}\text{C}$ + Stage $\times$ $\delta^{15}\text{N}$	19	3.02	0.09
	Site + Stage	27	3.03	0.09
	Site + Stage + $\delta^{15}\text{N}$ + $\delta^{13}\text{C}$	20	3.06	0.09
	Site	28	5.38	0.03
	$\delta^{15}\text{N}$	23	13.82	0.00
	Stage $\times$ $\delta^{15}\text{N}$	21	14.14	0.00
	$\delta^{13}\text{C}$	23	15.95	0.00
	Stage	28	21.06	0.00

Figure captions

Figure 1. Map showing the location of Teuri Island in the North Pacific. An unfilled square symbol indicates the location of Teuri Island. Black filled circle: auklet affected sites; gray filled circle: control sites

Figure 2. Schematic simplified mercury transfer pathway via seabirds. Mercury concentration (a) in red blood cells (ww)($n = 21$), (b) in auklet feces (dw) during incubation ($n = 2$) and chick-provisioning periods ($n = 7$), (c) in soil ($n = 10$) and (d) in roots at control ($n = 15$) and Auklet affected sites ($n = 11$) collected from Teuri Island during auklet breeding season in 2016. Boxplots show the first quartile, median, third quartile, and range of $\log_{10}$ -transformed concentrations.

Figure 3. A principal components analysis of nine metals (As, Cd, Pb, Hg, Cu, Zn, Cr, Co, Ni) in roots between auklet-affected and control sites. Ellipses are shown in 95% concentration of points.

Figure 4. Metal concentrations in roots (dw) at auklet-affected and control sites. The response variables are each analysed element that was $\log_{10}$ transformed. Control indicates samples collected at the control site ($n = 12$) and Auklet affected indicates samples collected at the auklet affected sites ($n = 8$). Individual values are shown as dots. Boxplots show the first quartile, median, third quartile, and range of $\log_{10}$ -transformed concentrations.

FIGURE 1

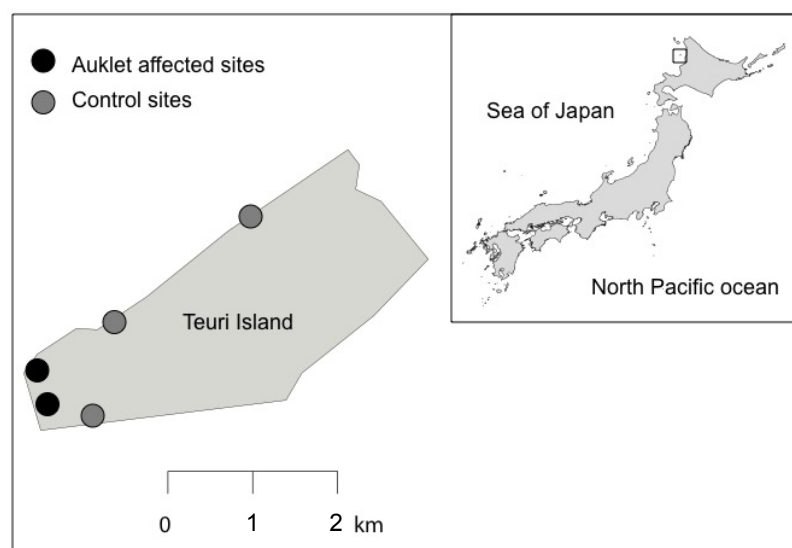


FIGURE 2

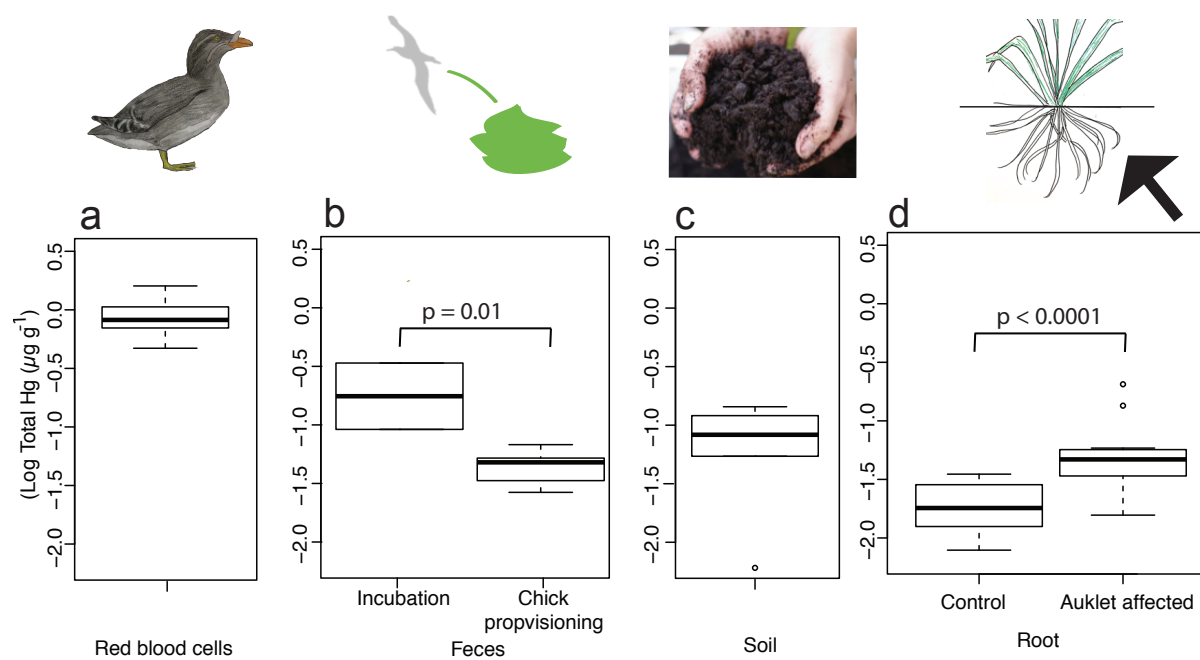


FIGURE 3

