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LETTER

Coupled trophic and contaminant analysis in seabirds through space and time

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Abstract

Wildlife contaminant loads are often used to indicate ecosystem health, but their interpretation is complicated by the dynamics affecting the trophic transfer of toxins. Yet, coupled analyses of trophic position and contaminants may provide insights that help resolve the underlying signal of contaminants in ecosystems. Here, we analyze heavy metal concentrations and trophic positions for pelagic seabirds across time and space. We derive metal-specific trophic transfer coefficients from the literature and use them to interpret the changes in raw heavy metal concentrations in two settings: (i) for eight seabird species across a 125-year timeline in Hawaii, and (ii) for contemporary specimens of two tern species across three ocean basins. While previous studies report how trophic position varies in these two settings, here we investigate how trophic downgrading may affect the observed raw changes in contaminants. Using this approach, we find the highly-toxic metal elements (Hg, As, Pb) decline after 1980. However, several other metals (Cu, Mn, Mo, Cd, Fe) increase from 1990–2015. Though simultaneous biomagnification and trophic downgrading may obscure contaminant analyses across space and time, the trophic declines we observed ($\leqslant$0.5 trophic level) are likely not sufficient to influence such comparisons. In addition, as extrapolating contaminant concentrations across broad ranges of trophic levels may be prone to large uncertainties, careful selection of the focal species for analysis is required. While high trophic level species, such as long-lived, fish-eating seabirds, are ideal for monitoring environmental contaminants across large spatial or time scales, lower trophic level species, like primary producers and consumers, may be more suitable for quantifying the concentrations of bio-available contaminants entering the marine ecosystem and the base of the marine food webs. Monitoring low and high trophic levels simultaneously may provide an integrated perspective that is needed to quantify the contaminants entering and bio-magnifying through marine ecosystems.

Introduction

Heavy metals have increasingly accumulated in marine ecosystems due to their widespread industrial use and subsequent leakage into the environment (Rainbow 1995, Järup 2003). While some mineral micronutrients are vital to key biochemical processes, both in plants and animals, they may become toxic when their concentrations are elevated. Zn for example is necessary for hydroxylation, Mn is important for enzyme regulation, Fe and Cu

are critical for oxygen transport, and several are involved in protein stabilization (Nieboer and Richardson 1980, Hänsch and Mendel 2009). Other metals have fewer known biological functions and are disruptive even at low concentrations (Booth and Zeller 2005, Sunderland and Mason 2007, Dietz *et al* 2009, Sonne 2010). Elements like Pb, Cd, and Hg, broadly impact digestive, immune, reproductive, neurological, and developmental processes across a wide range of organisms (Babich and Stotzky 1985, Barceló and Poschenrieder 1990, Tchounwou *et al* 2012), and as a result their concentrations are often strictly monitored and regulated.

Despite their various levels of toxicity, density or atomic weight, many such elements are collectively classified as 'heavy metals' (Duruibe *et al* 2007), a categorization which does not account for these highly variable processes and impacts on wild ecosystems. Improved frameworks for describing how different metals propagate through natural systems will help alleviate this problem of generalization and more accurately inform environmental hazards. To this end, monitoring the transport and availability of heavy metals in marine food webs is critical for resolving the threats to protected and commercial marine life and any subsequent risks to humans.

Seabirds are often highly-migratory, fish-eating predators that broadly integrate food webs across large geographic distances into their tissues (Furness and Camphuysen 1997, Finkelstein *et al* 2007, Abbasi *et al* 2015, Bond *et al* 2015, Gagne *et al* 2018b). As a result, many seabird species are useful indicators of the dynamics of marine food webs. Seabirds are also readily accessible for study, either in large nesting colony aggregations, through ongoing stranding programs, or in natural history repositories. Museums and other collections, for example, may contain large historical specimen archives, and these specimens can help generate a long-term record of ecosystem dynamics (Vo *et al* 2011, Wiley *et al* 2012, Wiley *et al* 2013, Ostrom *et al* 2017, Gagne *et al* 2018a, 2018b). The changes in heavy metal concentrations in seabird tissues over time could document broad changes in environmental contamination, either from the proliferation of industrial applications or in response to regulatory efforts to reduce their pollution (Hosono *et al* 2010, Hosono *et al* 2011).

As heavy metals pass through species in the food chain they often amplify, increasing in concentration at higher trophic levels ("TL", Suedel *et al* 1994). While TLs may be expected to vary between species, TLs are also dynamic within species and can vary across space and time. Recent studies, for example, show that the TLs at which seabirds now feed are significantly below their historical preferences (Gagne *et al* 2018a, 2018b), reflecting much broader trends of ecosystem chain length compression (Estes *et al* 2011). The TL of seabirds in the Hawaiian archipelago has declined broadly over 125 years. These TL trends from fishery-independent data align with fisheries-dependent studies (Pauly *et al* 1998, Pauly and Watson 2005, Essington *et al* 2006), and appear driven by commercial fisheries extraction and climate change (Gagne *et al* 2018a, 2018b). Beyond chronological shifts, contemporary comparisons of two tropical tern populations across three ocean basins highlight geographical within-species variability, and confirm that seabirds within areas of greatest fishing intensity feed at lower TLs (Gagne *et al* 2018a).

Given the evidence of trophic shifts over time amongst seabirds, it is important to understand to what extent TL dynamics may confound the interpretation of changing heavy metal concentrations. Here, we explore the coupled dynamics of TL and metal concentrations in seabirds. Using feather samples from museum specimens spanning 125 years, we examined whether recorded declines in seabird TLs impact heavy metal trends both in feather tissue and when extrapolated to other TLs in the marine food web. We began by reviewing previous literature to model trophic dynamics of heavy metals and estimate where TL and metal concentration relationships in food webs converge, differ, or demonstrate inconsistency. We then used these estimates of trophic transfer to calculate heavy metal concentrations in seabirds corrected to a constant, reference TL.

Methods

Sample collection and heavy metal assay

We sampled feathers from seabirds in three regions: the Florida Keys (Dry Tortugas National Park), the Hawaiian Islands (multiple sites), and from American Samoa (Rose Atoll Marine National Monument). The Hawaii series consisted of Laysan albatross (*Phoebastria immutabilis*), Bulwer's petrel (*Bulweria bulwerii*), wedge-tailed shearwater (*Ardenna pacifica*), white-tailed tropicbird (*Phaethon lepturus*), brown booby (*Sula leucogaster*), brown noddy (*Anous stolidus*), white tern (*Gygis alba*), and sooty tern (*Onychoprion fuscatus*). The American Samoa and Florida Keys series only included brown noddy and sooty tern. We obtained samples from museum specimens, stranding occurrences, and nesting colonies of wild birds (USFWS permits MB052060-0, MB180283-1). We minimized impacts on historical specimens by accessing body contour feathers along the flanks. For contemporary specimens, we collected senesced, fully-emerged flight feathers. Sample abundance was constrained by specimen availability ($10 \leqslant n \leqslant 24$ feathers species^{-1}, ave $= 17$). Table S1 is available online at stacks.iop.org/ERC/1/111006/mmedia provides the full details on each specimen and its origin.

Published studies (Gagne *et al* 2018a, 2018b) and their open-access repositories (osf.io/h8bmg/, osf.io/4s9ty/) provide extensive details on the habitats and TLs for all species as well as the raw data used from those studies.

We debrided feathers with compressed air, stored them in heavyweight polyethylene bags (ULINE^{TM}, 4 mil) and stabilized them with desiccant (Fisher^{TM} grade 48, 4–10 mesh). To obtain the 100 mg required for contaminant tests from historical specimens (e.g., Sobhakumari *et al* 2019) we combined individual samples from within six 20-year eras (1890–1909, 1910–1929, 1930–1949, 1950–1969, 1970–1989, 1990–2009). Individual samples from contemporary birds (2012–2015) each had sufficient mass and avoided ensembling. We homogenized samples into powder and sent them to the University of California Davis Animal Health and Food Safety laboratory (CAHFS) for standard heavy metal screens. CAHFS used inductively coupled plasma-optical emission spectrometry ("ICP-OES", Sobhakumari *et al* 2019) to quantify the sample concentrations of nine metal elements [copper (Cu), zinc (Zn), iron (Fe), lead (Pb), manganese (Mn), arsenic (As), cadmium (Cd), molybdenum (Mb), and mercury (Hg)]. ICP-OES requires that each raw analyte is digest with HNO_3 at 190 °C and then stabilized with HCL and ultrapure (18 Mohm) water. To ensure standards and accuracy throughout the analyses of all samples, CAHFS ran blanks, controls, and sensor drift checks regularly. This method is relatively cost-effective and requires a modest amount of specimen material (see above). As archiving techniques before 1980 used As and Hg, we analyzed these elements from 1980 to the present (Palmer 2001, Sirois 2001, Marte *et al* 2006) to ensure our analyses described ecosystem contamination. Previous studies have measured bioaccumulated methylmercury to avoid preservation artifacts (e.g., Vo *et al* 2011), however this would have more than quadrupled our laboratory costs and doubled our specimen extraction requirements.

We winsorized raw concentrations above the metal and species-specific 95% quantile, and detections below the reference limit were set to half the reference limit according to published practice (Wong *et al* 2002). Available specimens across species and years did not always overlap in years. To facilitate comparisons between years we linearly interpolated tissue concentrations across time between each available raw sample estimate by metal and species. Standard deviation was estimated from the linearly interpolated estimates through the entire time span. The standard deviation was then used to calculate the metal specific concentration normal 95% confidence intervals. Intervals estimating a lower limit concentration of less than zero are truncated to zero.

Trophic transfer coefficient estimation

Previous studies have often employed log-linear models to explain the trophic transfer rates of heavy metals (Campbell *et al* 2005, Cui *et al* 2011). Such approaches assume a controlled food web, a steady state of metal availability, and a metal concentration that follows a log-linear relationship with TL. As none of these assumptions may hold in an uncontrolled food web or across metals (Suedel *et al* 1994), we used a spline regression model with defined constraints that can estimate log-linear and nonlinear relationships. This modeling approach fit a smoothing spline with three degrees of freedom. The model was fixed to the concentration observed at the lowest TL using a weighted vector (R, Core Team 2014). The model then was fit to best predict the observed empirical concentrations across all TLs.

To collect empirical data from which we could estimate the trophic transfer models, we obtained metal concentration data from varied food web studies across aquatic systems. We only used studies that were conducted in an unbounded field-based setting, spanned at least TLs 1–3, and measured $\geqslant$5 of the 9 metals in our contaminant screens (Suedel *et al* 1994, Campbell *et al* 2005). For metals not included in a study (Fe), we used the next metal available (Mn) with the closest atomic mass. Fitted models estimate the trophic transfer coefficient (TTC) for each metal at any TL. All final models were calibrated so the y-intercept (the TTC when $TL = 0$, the food web base) is equal to 1. As a result, biodilution occurs when $0 < TTC < 1$ and biomagnification is when $TTC > 1$ (figure 1). Script and code is available in the online repository.

Tissue concentration correction

We used modelled TTC relationships to assess how declining TLs influence heavy metal concentrations. We plotted the raw metal concentrations over time and across spatial populations as a series of uncorrected tissue contaminant values. Since TL is dynamic, we used the TTC models to adjust the raw concentrations to reflect the observed chronology of trophic changes (figure 2). To do this, we first cross-referenced each feather sample to an associated TL for that specific species in time. Trophic estimates were drawn from a previous study that used feathers sampled from the same locations and dates we analyzed here for contaminants (Gagne *et al* 2018a, 2018b). Then we corrected each sample series to the base of the ecosystem ($TL = 0$) and the maximum observed seabird TL ($TL_{max} = 4.3$). We corrected the metal concentrations to $TL = 0$ (equation (2), $TTC_{TL=constant}$) by dividing the tissue concentration by the TTC estimated for the time- and space-associated TLs (equation (2), $TTC_{TL=tissue}$). Temporal TLs were derived in earlier published work using equation (1) (Gagne *et al* 2018a). For correction to a constant TL, the multiplier was the metal-specific TTC at the desired TL divided by the metal-specific TTC given the TL of the tissue, following equation (2).

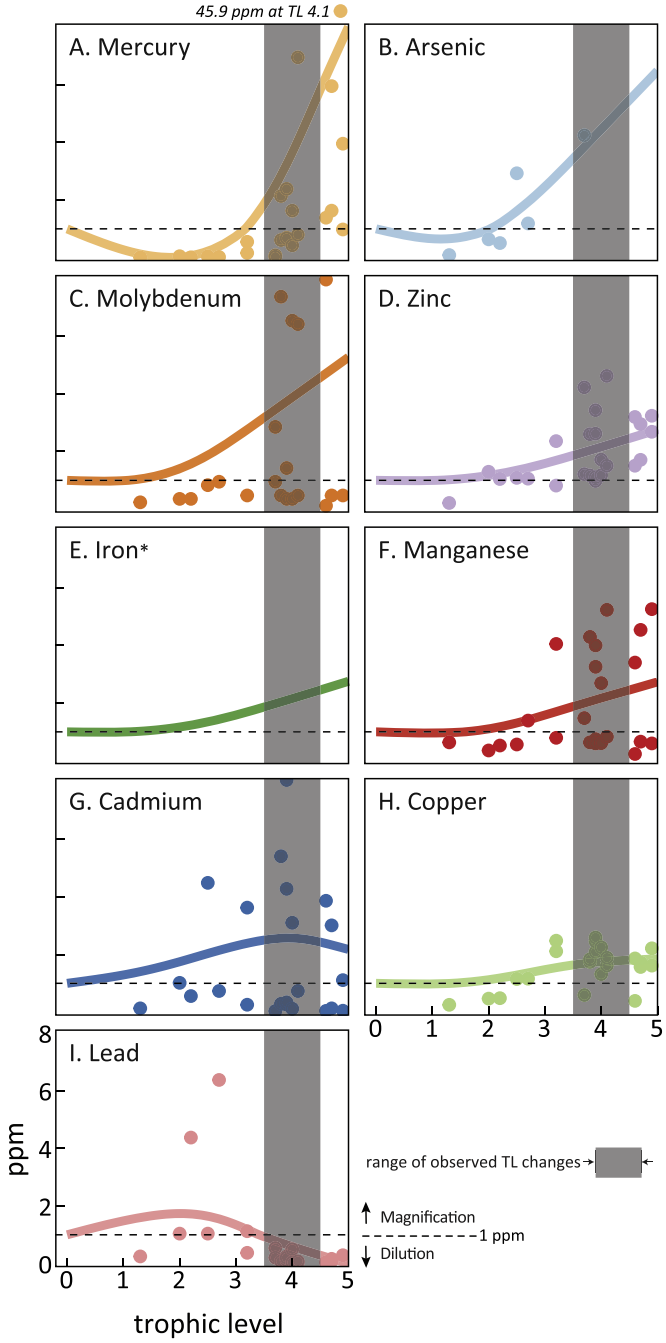

Figure 1. Metal concentrations against trophic level in a marine ecosystem. Data from Campbell *et al* 2005 show specimen metal concentrations (ppm) for nine elements against the specimen TLs sampled from a North Atlantic pelagic community. We standardized the ppm data to reference value of 1 at the base of the food web where primary producer species uptake available metals (TL = 0). Here, ppm values >1 indicate bio-magnification, where values <1 indicates trophic dilution. Grey shaded intervals show the range of TL change observed in Hawaii from 1890–2015 in eight species of seabirds. Solid lines describe trophic transfer spline models that describe the general trophic pattern of each element. Panels are sorted by decreasing metal magnification. Iron (E) was not measured in the study, and therefore we use the trophic transfer form of the next closest element on the periodic table (Mn) as a proxy.

$$TL = \frac{(\delta^{15}N_{Trp} - \delta^{15}N_{Src} - \beta)}{TEF} + 1 \quad (1)$$

$$TTC_{correc} = \frac{TTC_{TL=\text{constant}}}{TTC_{TL=tissue}} \quad (2)$$

All analyses and data visualizations were performed in the R environment (R, Core Team 2014).

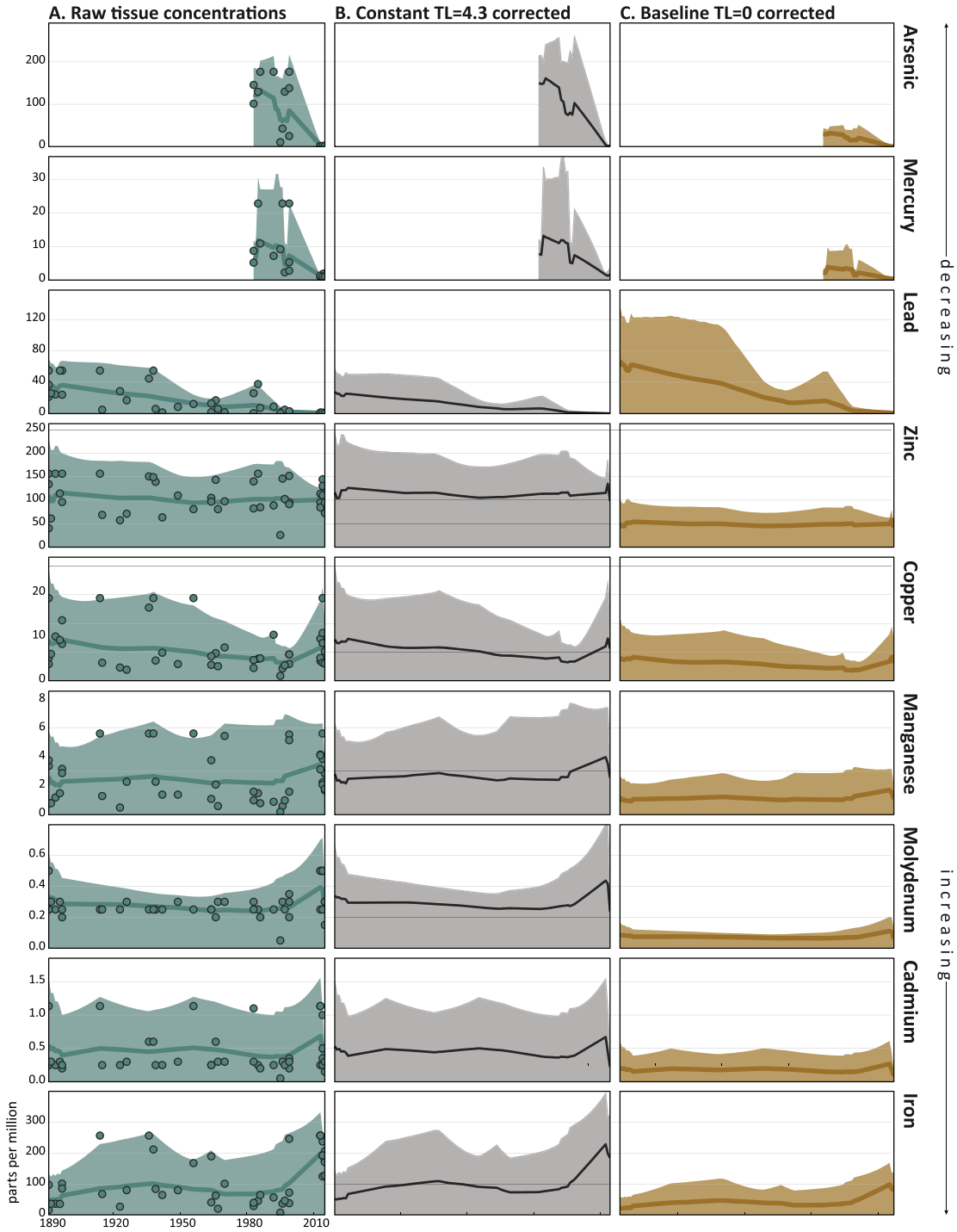

Figure 2. Some highly-toxic metals decline over time, independent of coupled trophic dynamics. A-C show the longitudinal trend and magnitude of heavy metals under three different trophic corrections. Open circles represent the raw metal concentrations, solid lines represent mean concentration across species, and shaded bands are the 95% CI (zero-truncated). All series are an eight-species ensemble from the Hawaiian Islands. The small difference between raw (A, teal) and constant (B, gray) TL correction shows the small effect that declining TL has on tissue concentrations over time. The baseline correction (C, tan) shows the large impact that adjustment to TL = 0 has on magnitude. Panels are sorted top-to-bottom according to concentration trend over time.

Results

We obtained heavy metal concentrations from 98 samples from Hawaii (67), Florida (15), and American Samoa (16). From these data, we compared metal concentrations within Hawaii from 1891–2015, and across all locations from 1990-present.

TTCs vary considerably between elements and across ecosystem types. Figure 1 shows the transfer coefficients we used to correct our tissue concentrations. We used the TTCs from the pelagic study to correct our tissues because it most closely reflected the ecosystem from which we obtained our feathers. figure S2

Table 1. Definitions of terms pertaining to trophic dynamics of contaminants.

Term	Definition
Bioconcentration	The uptake of a contaminant by an aquatic organism where water is the sole contaminant source
Bioaccumulation	The uptake of a contaminant from both water and sources
Biomagnification	The process of both bioconcentration and bioaccumulation that result in increased tissue concentrations of a contaminant as it passes through two or more trophic levels
Biodilution	The decrease in concentration of a pollutant with an increase in trophic level
Trophic Transfer Coefficient	Defined herein as the concentration of a contaminant in consumer tissue divided by the concentration contaminant in preceding trophic level. Simply, a measure of the potential for a contaminant to bio-magnify or dilute

All definitions taken from Macek *et al* 1979 and Campbell *et al* 2005

summarizes the results of three additional studies that span large portions of food webs ($0 \leqslant TL \leqslant 5$) in three aquatic ecosystem types. The metal concentrations documented from pelagic (figure 1), coastal (figure S2(b)), and freshwater (figure S2(c)) studies reflect the combined influence of metabolic and environmental uptake and loss (table 1). Across all studies, total Hg concentrations have a positive log-linear form, increasing with TL across the entire trophic web. This aligns with the documented bioaccumulation of Hg in large, predatory fish (Bodaly *et al* 1993, Branco *et al* 2007). In all studies, As magnifies across a range of TLs, though it dilutes when $TL > 3$ in the coastal study (figure S2(b)). Only in the pelagic study (figure 1(i)) does Pb magnify across a portion of the food web, and then only when $TL < 3$. The variability in all other metals suggests no consistent relationships exist, underscoring the need to characterize the local dynamics of food webs.

While some of the known toxic metals (Hg, As, Pb) decline across the 125 year chronology in Hawaiian seabirds, others (Zn) appear relatively constant (figure 2). Concentrations for the remaining metals (Fe, Cu, Mn, Cd, Mo) are relatively stable from 1890–1990, but increase thereafter. Significance was tested on reciprocal log transformed concentrations to validate these observed trends. A significant increase through time was detected in Fe, whereas declines were significant in As, Hg, and Pb (table S1). In figure 1, we plot concentrations in their raw form, and also when corrected according to the observed TL declines and the associated TTC for pelagic systems (figure 2(a), Campbell *et al* 2005). For all metals, the impact of trophic corrections affects the absolute concentration. However, while seabird TLs did change over time (figure 1) and TL does influence metal concentrations in oceanic systems (figures 2(a), (b)), the observed TL changes (~0.5 TL) were not large enough to broadly alter the chronology of raw metal concentrations.

Independent of temporal trend direction, Zn is the highest (102.00 ppm, sd = 5.80) long term average raw concentration in Hawaiian tissues, followed by Fe (89.70 ppm, sd = 29.70), As (70.20 ppm, sd = 42.30), Pb (16.70 ppm, sd = 11.10), Cu (6.70 ppm, sd = 1.40), Hg (6.50 ppm, sd = 3.40), Mg (2.50 ppm, sd = 0.30), Cd (0.50 ppm, sd = 0.05), and Mb (0.30 ppm, sd = 0.03). Based on previously published thresholds established from adverse effects on seabirds (Burger and Gochfeld 2004), on average Pb is above the adverse effects threshold of 4 ppm, Hg is just below the threshold of 5 ppm, and Cd is below the adverse effect threshold of 2 ppm. Pb does not drop below the threshold until 1997, Hg does in the 1980s, and Cd appears to have been below the threshold during the entire study period.

Having addressed metal concentrations across time, figure 3 plots their concentrations across three spatially-distant locations for two tropical terns. As with the time series data, we again corrected raw values based on the observed TL differences between sites (figure 3(j)). Unexpectedly, several metals had the highest concentrations at the remote and protected location, Rose Atoll: Pb = 3.4 ppm, Cd = 3.2 ppm, and Hg = 8.3 ppm. As a comparison, earlier work on Johnston Atoll in 1990 showed overall lower means in the same metals: Pb = 2 ppm, Cd = .22 ppm, and Hg = 1.9 ppm [26, 28].

Discussion

Corroborating previous studies, we find that the highest TL states in time and space can have some of the highest contaminant loads (figures 2–3). This relationship is not simple and may not take the same form throughout TLs, with varying uncertainty among metals and TLs (figure 1). Therefore, estimating how TL magnifies or dilutes contaminants requires careful derivation, on a case-specific basis with respect to species and ecosystem types. Regardless, we find that some of the most toxic metals decline over time, perhaps an encouraging response to their regulation. However, several other metals increase after 1990 (figure 2), indicating that potential ecosystem accumulation and organismal threats remain. In addition, we find seabirds sampled at the most remote site (Rose Atoll) had the highest detected concentrations for several metals (figure 3) confirming the

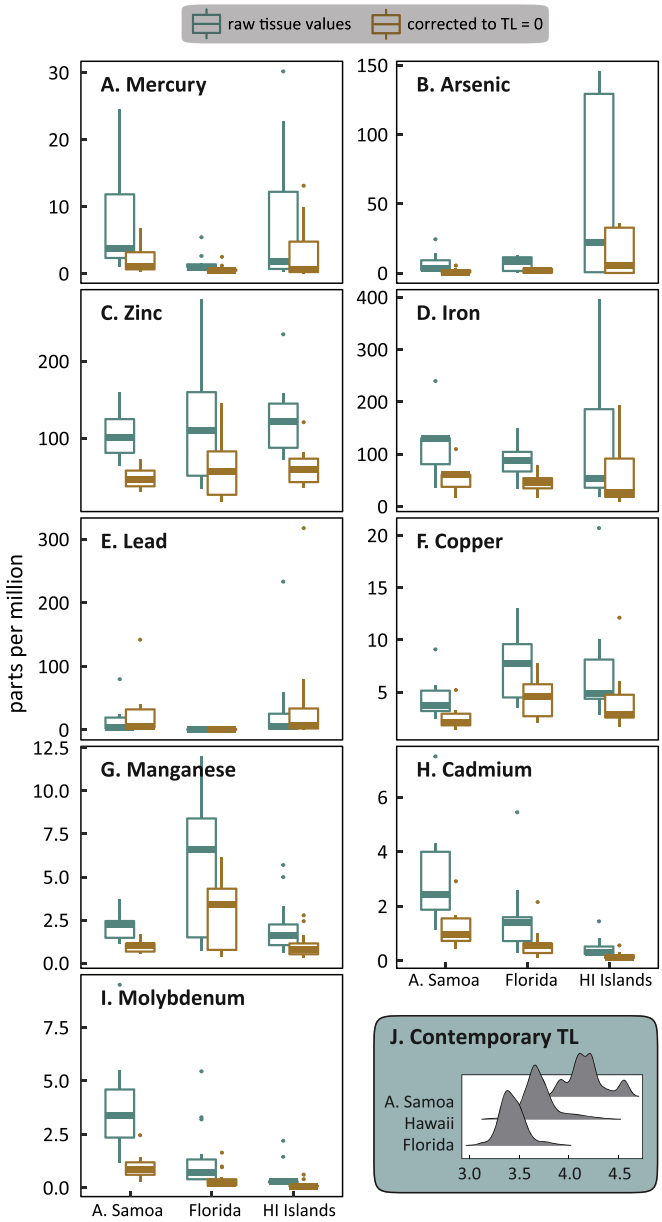

Figure 3. Ensemble comparison of concentrations in three ocean regions. A-I, Ensemble metal concentrations for two tropical terns sampled in the South Pacific (Rose Atoll, American Samoa), North Atlantic (Dry Tortugas, Florida), and North Pacific (Main Hawaiian Islands). Box plots represent the raw tissue concentration and the trophic correction to TL = 0, given observed geographic variation in TL (J). Median values of Hg, Fe, Mo, and Cd were highest at remote Rose Atoll (American Samoa), perhaps a response to point source pollution from a major ship grounding. Zn, Mg, and Cu values were highest in Florida, while As peaked in Hawaii.

distant reach of some pollutants. We discuss the implications of our coupled models on the magnitude, temporal trends, and the geographic comparisons of heavy metals below.

Correcting for trophic transfer

The degree to which changing TL affects metal concentrations is linked to the range of TL shift that an organism experiences over time. For trophic impacts to affect contaminant signals measurably in seabirds across time, observed TL declines may need to be on or greater than 1–2 TLs. Such a shift, say from 4 to 2, is equivalent to switching from a high-level predator to herbivore habits. Due to physiological constraints, this is unlikely to be a broad pattern observed in wild organisms. In Hawaii, we observed an average 125-year decline of 0.4 TL (Gagne *et al* 2018a), with trophic models suggesting dietary replacement of fish with squids. It appears that while those dietary changes reflect significant ecosystem shifts, such TL declines may not measurably alter raw contaminant trends (figure 3). However, when correcting metal concentrations to the base of the food web, larger multipliers will inflate uncertainty more than if correcting to a nearby constant TL. For example, if we extrapolate outside the range of TLs observed, when correcting to TL = 0 from TL = 4, our TTC estimates range by two and a half orders of magnitude. This result highlights the importance of carefully selecting the tissues sampled from

relevant TLs of interest, rather than relying on large back calculations to correct to the desired TL. Beyond the impacts of TL and bioaccumulation, foraging depth and other trophic dynamics are increasingly shown to affect Hg loadings, in particular, within species (Ramos *et al* 2013, Elliott and Elliott 2016, Madigan *et al* 2018) and deserve more attention.

Reconciling trends in time and space with other studies

Pb, As, and Hg showed steeper declining trends over time than any other metal in the assay. Though, in comparison to earlier studies in 1990 and 1998, our own uncorrected Hawaiian Pb and Hg was higher (1990: Pb = 5.8 ppm, Hg = 9.9 ppm, 1998: Pb = 2.2, Hg = 6.2 ppm) than Manana and Midway Islands in 1990 and 1998 (1990: Pb = 2 ppm, Hg = 1.6 ppm, 1998: Pb = 1.6 ppm, Hg = 2.1 ppm) (Burger and Gochfeld 2000). Though higher in magnitude, our observed decline in total Hg through time agrees with similar work on albatross in the North Pacific (Vo *et al* 2011) that showed a temporal declining trend in total Hg. It may be possible that these metals have longer records of use in these locations due to military presence and global industrial use. Furthermore, legislation to reduce their use and leakage into marine systems may have influenced these declines, much of it enacted in the late 20th century (e.g., Clean Air Act, Clean Water Act, and Mercury Export Ban). However, without a well-documented historical record of sources of metals, it is unclear whether these levels are a product of an acute event or point source (e.g., a shipwreck) or a legacy of persistent accumulation from nonpoint source pollution over time.

Geographic comparisons propose that remote atolls are not immune to persistent pollutants. Our contemporary (2012–2015) data suggested that Rose Atoll has the highest mean concentrations for several detrimental heavy metals: Pb, Cd, and Hg. For comparison, previously published work on the similar Johnston Atoll around the same time had lower means but the same rank order for those metals (Burger *et al* 1992, Burger and Gochfeld 2000). These high concentrations may, in small part, be influenced by a major ship grounding event at Rose Atoll, where metal contaminants have remained persistent (Schroeder *et al* 2008). In addition to this point source, these high concentrations could also be a function of the atmospheric convergence bands that this atoll often resides in, which can magnify concentrations stemming from high output activities, such as industrialization in Asia (Cheng 2003).

Interestingly, this gradient of higher contaminant concentrations in the South Pacific contradicts published work about anthropogenic impacts on food web characteristics, measured via seabirds' feather tissues (Gagne *et al* 2018a). Rose Atoll appears to be the least impacted via commercial fishing pressures, but the most impacted via heavy metal contaminants. This dichotomy highlights the independence and lack of correlation among various measures of anthropogenic impacts rather than the general degradation of marine ecosystems. Nonetheless, our geographic comparison of contemporary specimens is a small dataset and future studies will improve the certainty of these results.

Heavy metal screens

Numerous contaminants threaten ocean wildlife and can indicate the extent of environmental pollution in marine systems. Here, we chose heavy metal screens as their analysis is logistically feasible (e.g., cost, tissue preparation, tissue amount), their pathologies and hazardous limits are often described (Burger and Gochfeld 2004), and they are easily linked to anthropogenic activities (Furness 2017). As a result, we could examine a large sample size (~100 specimens) that included historical samples providing a 125-year longitudinal analysis. However, debate exists about the classification of these nine elements as heavy metals and their utility in evaluating environmental toxins (Nieboer and Richardson 1980, Pourret and Bollinger 2017). Nominally, heavy metals are defined as metals with a specific density >5 g cm^{-3}, though this has never been authoritatively defined (Pourret and Bollinger 2017). This results in Fe being grouped with Hg, though the two elements have remarkably different biological concentrations and consequences (Nieboer and Richardson 1980). Therefore, it appears that future metal toxin assays should consider basing screen selection on impacts and biologically significant binding preferences to O, N, or S (Nieboer and Richardson 1980), as opposed to their atomic mass. To understand the ultimate impacts of metal contaminants to the organisms themselves, additional tissue quantifications of selenium may be useful, as this element may play a significant role in reducing heavy metal toxicity (Ikemoto *et al* 2004).

Conclusion

Building on previous work documenting changing seabird TL in time and space, this study investigated how that dynamic interfaces with heavy metal signatures in seabird feathers. Two main contributions emerge from this work. Documenting possible trends in seabird tissues temporally and spatially sheds light on the intricacies of trophic transfer, and sets up a framework for the interpretation of temporal environmental signatures of metals.

Based on this analysis, it appears that the decline in TL in most seabirds is not large enough to drastically impact the estimated metal concentration trends in their tissues. Additionally, we show that estimating metal concentrations for other TLs far from those of seabirds is prone to large uncertainty, and we caution against using trophic transfer coefficients to extrapolate to other food web components >1 TL from the sampled TL. In addition, as extrapolating contaminant concentrations across broad ranges of TLs may be prone to large uncertainties, careful selection of the focal species for analysis is required. While high TL species, such as fish-eating seabirds, may be more suitable for monitoring environmental contaminants across large spatial or time scales, lower TL species, like primary producers and consumers, may be more suitable for quantifying the concentrations of bio-available contaminants at the base of the marine food webs. Together, these two perspectives can provide insights into the pathways and mechanisms metals follow when entering and traveling through the marine ecosystem. Namely, the documented declines in some adverse metals indicate that heavy metal concentrations may reflect changes in legislation and reductions in anthropogenic contaminant accumulation. Thus, we suggest future studies continue to illuminate the coupled spatial complexity and temporal gradient of metal availability and food web dynamics.

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University of California Davis Animal Health and Food Safety Laboratory performed the heavy metal screens, A Copenhaver improved earlier versions of the manuscript and logistical support. S L Pimm improved earlier analyses incorporated in the manuscript.

Data availability statement

Datasets analyzed for this study are included in the data repository at osf.io/4rcsk/.

Authors' contributions

K V and S P designed the study. All authors collected and prepared data; E J, T G, and K V analyzed data, prepared figures and wrote the manuscript; All authors reviewed the manuscript.

Ethics statement

This research was performed under USFWS permits #MB052060-0, MB180283-1 to KH. All research followed the Institutional Animal Care and Use Committee criteria under NOAA and HPU. The authors declare no conflicts of interest.

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Declaration of interests

We have no competing interests

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