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Beyond bulk $\delta^{15}N$: Combining a suite of stable isotopic measures improves the resolution of the food webs mediating contaminant signals across space, time and communities

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ABSTRACT

Top predators are used as indicators of contaminant trends across space and time. However, signals are integrated over complex food webs, and variation in diet may confound such signals. Trophic position, assessed by bulk $\delta^{15}N$, is widely used to infer the variation in diet relevant to contamination, yet a single variable cannot completely describe complex food webs. Thus, we examined relationships across three aquatic systems varying from a single species to a small food web using bulk values from four isotopes and 21 amino acid-specific values. Because variation in baseline ('source') $\delta^{15}N$ can confound estimates of trophic position , we calculated trophic position from the difference between $\delta^{15}N_{trophic}$ ($\delta^{15}N$ for amino acids that change with trophic position) and $\delta^{15}N_{source}$ ($\delta^{15}N$ for amino acids that do not change with trophic position). Across all three systems, variation in $\delta^{15}N_{source}$ explained over half of the variation in bulk $\delta^{15}N$, and stable isotope values that reflected the base of the food web ($\delta^{13}C$, $\delta^{18}O$, $\delta^{34}S$) predicted contaminants as well or better than $\delta^{15}N$—which was supported by a *meta*-analysis of other studies. In ospreys feeding in lakes, variation in $\delta^{15}N_{source}$ across space created a spurious relationship between ΣDDT and apparent trophic position, and masked a relationship between ΣPCB and trophic position. In a seabird guild, changes in diet over time obscured temporal variation in contaminants over five decades. In Arctic fish and invertebrates, more accurate trophic magnification factors were calculated using $\delta^{15}N_{trophic\text{-}source}$. Thus, (1) using $\delta^{15}N_{trophic\text{-}source}$, instead of bulk $\delta^{15}N$, avoided incorrect conclusions and improved accuracy of trophic magnification factors necessary to assess risk to top predators; and (2) diet assessed with multiple spatial isotopes, rather than $\delta^{15}N$ alone, was essential to understand patterns in contaminants across space, time and biological communities. Trophic position was most important for lipophilic 'legacy' contaminants (ΣDDT, ΣPCB) and habitat was most important for other contaminants (ΣPBDE, ΣPFAS, mercury). We argue that the use of amino acid-specific analysis of $\delta^{15}N$ alongside 'non-trophic' isotopes should be a core feature of any study that examines the influence of trophic position on chemical pollution, as required for a chemical to be added to international conventions such as the Stockholm Convention.

1. Introduction

Chemical pollution is one of the Anthropocene's great environmental challenges, and numerous international treaties have been ratified with the goal of containing chemical pollution within proposed planetary boundaries (Rockström et al., 2009). The Stockholm Convention on Persistent Organic Pollutants and the Minamata Convention on Mercury both include provisions to monitor pollution levels to determine the effectiveness of the conventions at meeting acceptable boundary limits. Indicator species are often used to monitor effectiveness of pollution regulations, both to track levels of known compounds and to screen for the emergence of novel chemical concerns (Phillips, 1977; Furness and Camphuysen, 1997; Elliott and Elliott, 2013). Top predators, including birds of prey and seabirds, are excellent indicator species for persistent bioaccumulative contaminants because (i) they integrate chemical signatures across space and time, including entire biological communities, (ii) have relatively high and easily measured contaminant concentrations and (iii) are consumed by humans or represent levels in human

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consumers of wild foods (Gilbertson et al., 1987; Burger and Gochfield, 2004; Elliott and Elliott, 2013). Moreover, population declines of top predators, including seabirds and birds of prey, have been among the most tangible impacts of chemical pollution, and have driven public pressure to enact treaties aimed at reducing such pollution (Blus et al., 1971; Grier 1982; Best et al., 2010; Bierregaard et al. 2014).

Although tissues of top predators can be used to monitor contaminant levels in the environment, variation in diet can confound such monitoring (Hebert et al. 2000; Braune et al., 2014a,b; McKinney et al. 2015; Dolgova et al. 2018). Many contaminants make their way into organisms via their diet, and biomagnify through food webs, reaching their highest levels in top predators (Atwell et al. 1998; Elliott 2005; Loseto et al. 2008). Stable isotope ratios are biogeochemical tracers that are incorporated from diet into predator tissues (Hobson 1999; Fisk et al. 2001; Kelly et al. 2007, 2008). Stable isotope ratios measured in the same tissue as contaminants offer the ability to track chemical pollution through the food chain to determine whether variation in contaminant levels in top predators is due to diet or other factors (Burgess et al. 2013; Elliott and Elliott 2013; Braune et al., 2014a,b). For example, nitrogen isotope ratios form the basis for calculating trophic magnification factors that allow risk assessments across taxa within food webs (Borgå et al., 2012; McLeod et al. 2015; Fremlin et al., 2020). Trophic magnification factors play an important role in determining which of the tens of thousands of chemicals on the market may be of most risk to top predators (Conder et al., 2012).

The ratio of ^{15}N to ^{14}N, represented in relative terms by $\delta^{15}N$, increases systematically from prey to predator, and can be used to estimate the 'trophic position' of a predator (Atwell et al., 1998). Authors often correct measured contaminant levels by $\delta^{15}N$, or by their trophic magnification factor, under the assumption that dietary variation in contaminants is primarily associated with biomagnification ('trophic position hypothesis'; Hobson et al., 2002; Eagles-Smith et al., 2009; Day et al., 2012a,b; Foster et al., 2012; Burgess et al., 2013; Braune et al., 2014a,b). Trophic magnification factors are defined as the increase in a contaminant per increase in trophic position across a food web inferred by bulk $\delta^{15}N$ (Broman et al., 1992; Rolff et al., 1993; Fisk et al., 2001; Hop et al., 2002; Borgå et al., 2012; McLeod et al., 2015; Fremlin et al., 2020). Trophic magnification factors are more transferable across systems than prey-predator biomagnification factors (the ratio of a contaminant in predator to prey) because they incorporate trophic variation occurring within species, allowing an estimate of change in contaminant concentration if a species changes trophic position without a need to re-estimate the prey basket. Out of wildlife studies using isotopes to explain variation in contaminants, published between 2013 and 2019 (i.e. recent enough that stable isotope analysis is widely used) in three representative journals, *Environmental Science & Technology*, *Journal of Applied Ecology* and *Ecological Applications*, 88% (42 out of 47) reported only bulk $\delta^{15}N$. Thus, there is a widespread assumption that food chain length (trophic position) captures most of the variation in food webs relevant to contaminants. However, trophic position may be less important within a single predator species (Verreault et al., 2010), and may not capture most of the variation in diet relevant to contaminants.

Many food webs are complex, and a single metric (food chain length) is unable to represent all variation in relationships. Contaminant levels often vary among habitats due to processes such as long-range transport, point source pollution, degradation of organic compounds by microbes or methylation of mercury by bacteria, and variation in processes at the base of food webs might be as important as biomagnification for understanding variation in contaminants ('habitat variation hypothesis'; Lavoie et al., 2015; Elliott and Elliott 2016). For example, variation in baseline $\delta^{15}N$ can obscure variation in bulk $\delta^{15}N$ associated with trophic position (Lorrain et al., 2009; Seminoff et al., 2012; Chikaraishi et al., 2014). Amino acid-specific stable isotope analysis provides the opportunity for measuring both baseline (used here as a synonym for 'source') and baseline-corrected trophic position because the $\delta^{15}N$ values in 'source' amino acids (i.e. phenylalanine, $\delta^{15}N_{phe}$), do not increase with trophic position and, therefore, measure baseline $\delta^{15}N$, while 'trophic' amino acids (i.e. glutamate, $\delta^{15}N_{glu}$) increase systematically with trophic position (McCelland and Montoya 2002; Lorrain et al., 2009; Walsh et al., 2014). The difference between these two values, $\delta^{15}N_{glu\text{-}phe}$, provides a baseline-corrected estimate of trophic position (Walsh et al., 2014; Hebert et al. 2016; Gagné et al., 2019). Because several additional amino acids are classified as 'source' or 'trophic', averaging $\delta^{15}N$ across baseline amino acids ($\delta^{15}N_{source}$) and trophic amino acids ($\delta^{15}N_{trophic}$) likely provides better indices than using single amino acids. Thus, $\delta^{15}N_{trophic\text{-}source}$ can be used as an indicator of trophic position (Fig. 1).

Additional dietary tracers associated with the spatial origin of diet ($\delta^{13}C$, $\delta^{18}O$, $\delta^{34}S$) could be used to refine diet reconstruction (Hobson 1999; Hebert et al., 2006; Elliott and Elliott 2016). Although all three isotopes are associated with habitat, $\delta^{13}C$ also varies systematically with trophic position and $\delta^{18}O$ reflects variable contributions from diet and body water. In contrast, $\delta^{34}S$ may be a particularly useful dietary tracer of spatial origin as $\delta^{34}S$ varies little from source to consumer (Florin et al., 2011; Vander Zanden et al., 2016). The combined use of multiple isotopes could provide a more nuanced description of food web structure in the context of environmental pollution. In particular, the use of amino acid $\delta^{15}N$ and $\delta^{13}C$ values, including both those that change for predator to prey ('trophic' and 'non-essential' amino acids, respectively, for $\delta^{15}N$ and $\delta^{13}C$) and those that do not ('source' and 'essential'), may provide a more refined estimate of diet.

Here, we use up to 25 isotopic values (bulk carbon, nitrogen, oxygen and sulfur isotopes, as well as carbon and nitrogen isotopes from amino acids) to quantitatively track chemical pollution throughout three systems: a bird of prey species (osprey, *Pandion haliaetus*) feeding in lakes across the vast landscape of western Canada, a guild of seabirds in the eastern Pacific, and a community of Arctic fish and invertebrates. The food webs were chosen from those available in the Environment and Climate Change Canada's National Wildlife Specimen Bank to reflect variation at the level of a single species (osprey), several species (seabirds) or a small food web (fish and invertebrates), and to examine variation across space, time and species. In each system, we measured mercury and organohalogen compounds, along with stable isotope ratios. We tested the trophic position hypothesis using amino acid-specific analyses, and predicted that $\delta^{15}N_{trophic\text{-}source}$ better predicts levels of biomagnifying contaminants than bulk $\delta^{15}N$ (Fig. 1). Based on the habitat variation hypothesis, we also predicted that we would observe a relationship between contaminants and $\delta^{13}C$, $\delta^{18}O$ or $\delta^{34}S$, with the direction depending on particular contaminants. We then used those isotopic relationships to examine how contaminants varied across space, time and biological communities. In ospreys, we tested whether, after correcting for diet, large lakes with small watersheds and lower proportion of glaciers and agriculture would have low contaminant concentrations. We expected such relationships due to dilution of input from agriculture or glaciers, the latter contaminated by long-range transport as reported in alpine fish, as found previously (Guigueno et al., 2011; Elliott et al., 2012; Grenier et al., 2020). In Pacific seabirds, we examined whether, after accounting for variation in diet, ΣDDT declined exponentially following restrictions on use in the 1960s, as would be expected based on a constant rate of degradation. We used the diet-corrected values to assess actual decline rates. In Arctic fish and invertebrates, we calculated isotope discrimination factors and trophic magnification factors using baseline-corrected isotope values. Finally, we conducted a *meta*-analysis of associations between isotopes and contaminant levels to examine whether these relationships were consistent across other systems.

2. Methods

We analyzed samples archived in Environment and Climate Change Canada's National Wildlife Specimen Bank, including osprey eggs and plasma from lakes across western Canada, seabird eggs from coastal

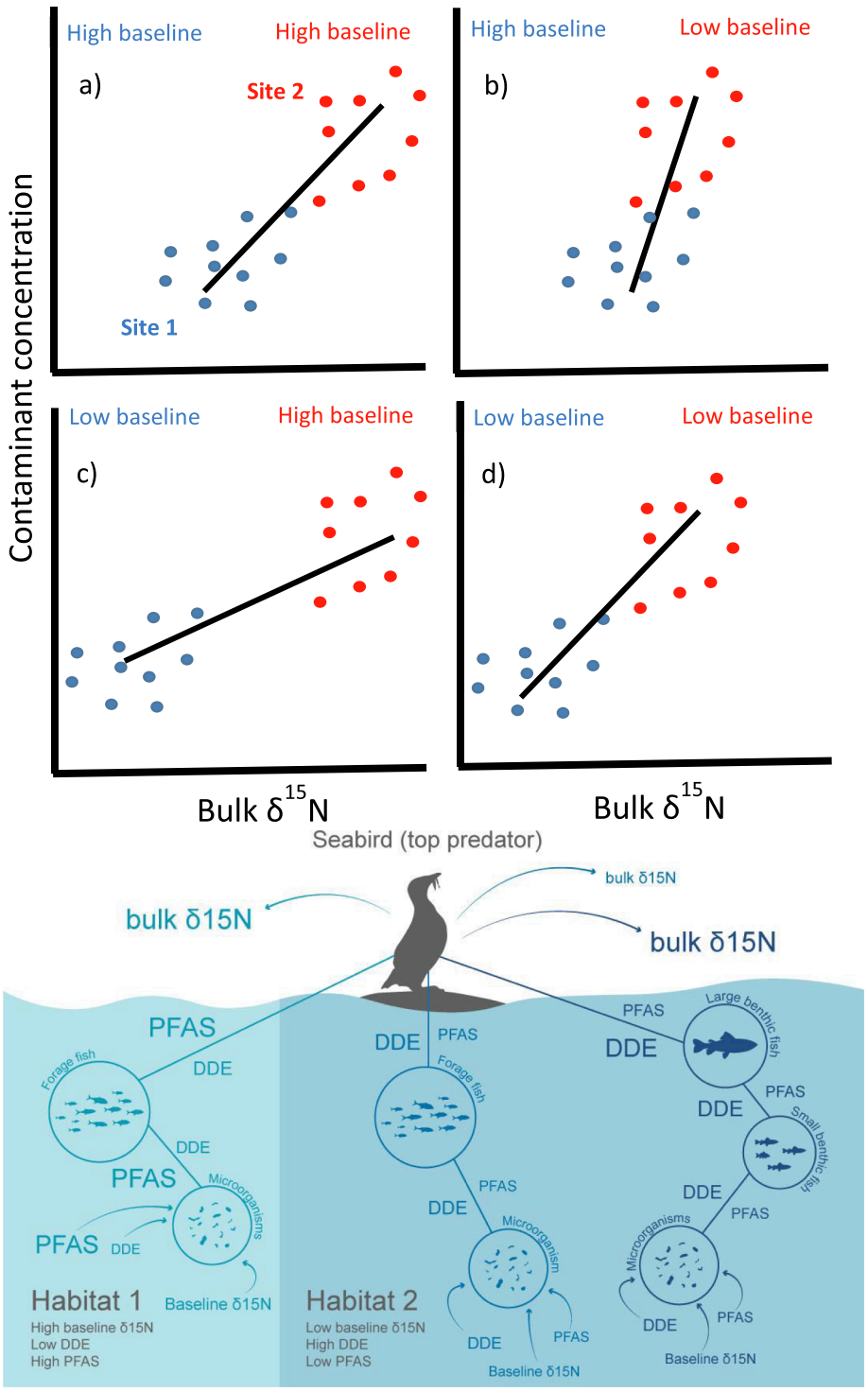

Fig. 1. A conceptual diagram showing how variation in habitat and baseline $\delta^{15}N$ can alter the bulk $\delta^{15}N$ -contaminant relationship. Upper panels (a-d) show variation in contaminant concentration with bulk $\delta^{15}N$ across two sites (or time periods or species) that vary in trophic position. As is typically found, trophic position correlates with contaminants within (weakly) and across (strongly) sites. The trophic magnification factor is correct where baseline $\delta^{15}N$ match (a, d), but not where they do not match (b, c). Lower panel (e) shows the same principles at work within a food web leading to a seabird, where the size of each label is proportional to its numerical value. In Habitat 1, individuals feeding in a high baseline environment have low levels of DDE despite having high $\delta^{15}N$. In Habitat 2, individuals feeding at a higher trophic position (large fish) have high DDE levels. PFAS (a non-legacy compound) concentrations vary with habitat but not trophic position.

British Columbia and fish muscle collected in northern Hudson Bay. From 1999 to 2003, we sampled osprey eggs (one per nest; N = 29), chick plasma (N = 42) and adult plasma (N = 10) from 15 watersheds of the Canadian Cordillera in Alberta, British Columbia and the Yukon (methods described in detail by Guigueno et al., 2011; Elliott et al., 2012). Seabird eggs, from ancient murrelets (*Synthliboramphus antiquus*; N = 10), rhinoceros auklets (*Cerorhinca monocerata*; N = 22), Leach's storm-petrels (*Oceanodroma leucorhoa*; N = 17), double-crested cormorants (*Phalacrocorax auritus*; N = 25), pelagic cormorants (*Phalacrocorax pelagicus*; N = 20) and great blue herons (*Ardea herodias*; N = 2), were collected at colonies along the British Columbia coast (methods described in detail by Miller et al., 2014, 2015; Elliott and Elliott 2016). Representative muscle samples of small fish and invertebrates were collected opportunistically from the breeding ledges of thick-billed murres (*Uria lomvia*) at Coats Island (62°98′N, 82°00′W) in northern Hudson Bay, Nunavut, Canada. Blood was also collected from ten murre chicks. Thirty-one individuals from 13 species were collected between 2007 and 2009: The samples were a subsample of those analyzed as part of Braune et al., 2014a,b), and contaminant and $\delta^{15}N$ data used in this study were presented in those papers. Species sampled included Arctic cod (*Boreogadus saida*; N = 5), capelin (*Mallotus villosus*; N = 2), sand lance (*Ammodytes* spp.; N = 4), Atlantic poacher (*Leptagonus decagonus*;

N = 1), Arctic shanny (*Stichaeus punctatus*; N = 1), daubed shanny (*Leptoclinus maculatus*; N = 1), banded gunnel (*Pholis fasciata*; N = 1), fish doctor (*Gymnelus viridis*; N = 1), fourline snake blenny (*Eumesogrammus praecisus*; N = 5), sculpin (*Triglops* sp.; N = 3), sea butterfly (*Clione limacine*; N = 1), gammarid (Gammaridae; N = 1) and jellyfish (Medusozoa; N = 1). All samples were frozen in the field and then homogenized before archival in the Specimen Bank.

2.1. Stable isotope analysis

Stable isotope analysis for seabird and osprey eggs was carried out using the same egg homogenate as used for chemical analyses. Samples were freeze-dried. Bulk $\delta^{13}C$ values were lipid normalized algebraically (Elliott et al., 2014), except for Arctic fish where analyses were conducted on bulk tissues with lipids extracted using 2:1 chloroform: methanol soak and rinse (Braune et al., 2014a,b). Stable isotope analyses for bulk $\delta^{34}S$, $\delta^{15}N$, and $\delta^{13}C$ were performed on the homogenate at the G. G. Hatch Stable Isotope Laboratory (Ottawa, ON). Homogenates were freeze-dried and powdered. Stable nitrogen and carbon isotope assays were performed on 1 mg subsamples of homogenized material loaded into tin cups. Samples were analyzed using an isotope cube elemental analyzer (Elementar, Germany) interfaced with a Delta Advantage continuous-flow isotope ratio mass spectrometer (Thermo, Germany) coupled with a ConFlo III (Thermo, Germany). A glutamic acid laboratory standard was included for every 10 unknown samples. Quality control was maintained by running sample duplicates. All measurements are reported in standard δ-notation in parts per thousand (‰) relative to the AIR international standard. Replicate measurements of internal laboratory standards [C-55 (glutamic acid)] indicated measurement error of ± 0.2‰. Stable sulfur isotope analyses were performed on 10 mg subsamples of the homogenized fish loaded into tin capsules (lipids were not extracted from homogenates as lipid extraction is known to alter $\delta^{34}S$ and lipids should not contain sulfur; Elliott et al., 2014). Samples were analyzed with an isotope cube elemental analyzer (Elementar, Germany) interfaced with a Finnigan DeltaPlus XP isotope ratio mass spectrometer (Thermo Germany) coupled with a ConFlo IV (Thermo Germany). All measurements are reported in parts per thousand (‰) relative to the VDCT international standard. Calibrated internal standards were used to normalize the data with a precision of ± 0.4‰. Solids were analyzed for ^{18}O isotopes using an elementar PyroCube (Elementar Analysensysteme GmbH, Hanau, Germany) interfaced to a PDZ Europa 20–20 isotope ratio mass spectrometer (Sercon Ltd., Cheshire, UK). Samples were thermally decomposed to CO in a glassy carbon reactor filled with glassy carbon, graphite felt, and lamp black at 1400 °C. An adsorption trap isolated CO from any interfering N_2. The final delta values, delivered to the customer, were expressed relative to international standard V-SMOW (Vienna Standard Mean Ocean Water).

Amino acid-specific analysis occurred at the UC Davis Stable Isotope Facility following the methods described by Yarnes et al. (2011). The following amino acids were analyzed: alanine (Ala), aspartate (Asp), glutamate (Glu), glycine (Gly), isoleucine (Ile), leucine (Leu), lysine (Lys), methionine (Met, not analyzed for seabirds), phenylalanine (Phe), proline (Pro), serine (Ser, not analyzed for carbon or for seabirds), threonine (Thr, not analyzed for seabirds) and valine (Val). Amino acids were liberated via acid hydrolysis and derived by methyl chloroformate. Methoxycarbonyl amino acid methyl esters were then injected in splitless (15 N) mode and separated on an Agilent DB-23 column (30 m × 0.25 mm ID, 0.25 μm film thickness). Once separated, the esters were converted to N2 in a combustion reactor at 1000 °C. Water was subsequently removed through a nafion dryer. During the final step of the analysis, N2 entered the IRMS. Pure reference N2 was used to calculate provisional δ- values of each sample peak. Next, isotopic values were adjusted to an internal standard (e.g. norleucine) of known isotopic composition. Final δ-values were obtained after adjusting the provisional values for changes in linearity and instrumental drift such that correct δ-values for laboratory standards were obtained. Laboratory standards were custom mixtures of commercially available amino acids that had been calibrated against IAEA-N1, IAEA-N2, IAEA-N3, USGS-40, and USGS-41.

2.2. Chemical analysis

Homogenated tissue was analysed at the National Wildlife Research Centre as per methods described previously (Braune et al., 2014a,b; Miller et al., 2014, 2015). For mercury quantification, samples were homogenized, freeze-dried, homogenized again, and weighed into nickel combustion boats. Total mercury was analyzed using an Advanced Mercury Analyzer (AMA-254) equipped with an ASS-254 autosampler for solid samples as described elsewhere (EPA Method 7473; see Braune et al., 2014a for details). Homogenized samples were analyzed for organochlorines (OCs) including chlorobenzenes, hexachlorocyclohexanes, chlordane-related compounds (ΣCHL = oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor and heptachlor epoxide), DDT and its metabolites (ΣDDT = *p,p*' -DDE, *p,p*' -DDD and *p,p*' -DDT), octachlorostyrene (OCS), mirex, photomirex, dieldrin and PCB congeners (ΣPCB). Chemical extraction, cleanup and analysis of PCBs and OC pesticides are described by Braune et al. (2014a,b) and Elliott et al. (2012). For each batch of samples, one duplicate extraction, one duplicate injection, two/three method blanks, one/two in-house reference materials (Reference Egg Pools DCCOQA-2011–01, or DCCOQA-2011–06 and DCCOQA-2011–07) and one certified reference material (Lake Michigan Fish Tissue NIST1947) were run for quality control. All reported residue levels were corrected for internal standard recoveries. The nominal detection limit was 0.1 ng/g wet weight. Homogenized samples were also analyzed for polybrominated diphenyl ethers (PBDEs), polybrominated biphenyls (PBBs) and total hexabromocyclododecane (HBCDD). Sample extraction and clean-up were the same as for the OCs except that tissue homogenates were spiked with internal standards (BDE-30, BDE-156, labeled $^{13}C_{12}$-BDE-209). Details are described by Braune et al. (2014a,b) and Miller et al. (2014, 2015). The extraction, cleanup and analysis of the per- and polyfluoroalkyl substances (PFASs) have been described by Braune et al. (2014a,b) and Miller et al. (2015), including use of appropriate standards. For every block of 10 samples, a blank sample and a NWRC in-house reference material (spiked pork liver) was analyzed.

2.3. Statistical analyses

Statistical analyses were conducted in R 3.2.1. Prior to statistical analysis, we log-transformed all variables to achieve normality. For calculation of summed contaminant values (ΣDDT, ΣPCB, ΣBDE, ΣPFAS and ΣPFCA) we assumed all congener values below the minimum detection level were zero. We used separate principal components analysis for $\delta^{15}N$ and $\delta^{13}C$ to investigate relationships among amino acids for each system. We used AIC to compare candidate models with contaminants (log-transformed) as the dependent variables and species, lipids (for organics), site, year and isotope values as the independent variables. For ospreys, we also included watershed properties (for organics) and modeled mercury deposition rate (for mercury; variables selected based on Guigueno et al., 2011, Elliott et al., 2012). We assumed that large lakes with small watersheds and lower proportion of glaciers and agriculture would have low levels of contaminants due to dilution of contaminants input from agriculture or deposition in glaciers (Guigueno et al., 2011, Elliott et al., 2012). For principal components analyses and general linear models, we used half the detection limit for any value that was zero. Discrimination factors were calculated by subtracting amino acid-stable isotope values in murre chick red blood cells from the values from prey muscle, based on known average prey diet in 2007 (the year of sampling; Gaston and Elliott 2014). Biomagnification factors (*BMF*; Braune et al., 2014a) were calculated as the ratio of contaminant concentrations in the predator (murres, C_{Murre}) to the contaminant concentration in prey (n; concentration: C_n) weighted

by the proportion, by weight, of prey item n in the diet (P_n):

$$BMF = \frac{C_{Murre}}{\sum C_n * P_n}$$

Trophic magnification factors (*TMF*) were calculated by fitting a linear function to $\log_e$ contaminant values and $\delta^{15}N$ values and determining the slope, b (Borgå et al., 2012):

$$TMF = e^b$$

We calculated the slope, b, for both the relationship between contaminants and bulk $\delta^{15}N$ (the traditional definition of TMF, bulk $\delta^{15}N$ *TMF*) and between contaminants and $\delta^{15}N_{trophic\text{-}source}$ ($\delta^{15}N_{trophic\text{-}source}$ *TMF*). In the traditional definition, a species with a different baseline than the rest of the assemblage, could lead to a biased slope, which is accounted for in the $\delta^{15}N_{trophic\text{-}source}$ *TMF*.

For Pacific seabirds, where we had long-term trend data for ΣDDT and Hg for one species (double-crested cormorant, *Phalacrocorax auritus*), we analyzed the relationship between isotopes, contaminants and year. We assumed a degradation effect for ΣDDT which was phased out prior to our time series, and consequently modeled ΣDDT with an exponential function relative to year. We compared relationships with and without accounting for diet by adding the predicted residual of ΣDDT or Hg onto the time series value based on the relationship between those contaminants and isotopic values. We report $\delta^{15}N_{glu\text{-}phe}$ as the difference in $\delta^{15}N$ between glutamate and phenylalanine and $\delta^{15}N_{trophic\text{-}source}$ as the difference in $\delta^{15}N$ between the average values for trophic (Asp, Glu, Ile, Leu, Pro) and source (Met, Lys, Phe) amino acids.

2.4. Meta-analysis

We searched for all studies that reported relationships between at least two isotopes (typically $\delta^{15}N$ and $\delta^{13}C$, but sometimes also $\delta^{34}S$) and four commonly-studied contaminants (DDT, PCB, mercury and PBDE), using Web of Science and the search filters ("DDT" OR "PCB" OR "mercury" OR "PBDE") AND "isotope". We only included studies on wild animals. We then examined all papers that cited, or were cited by, any of the papers containing such relationships. We calculated the effect size for each study (slope / SE of slope) where slope is the slope of the relationship between the contaminant and each isotope. Where values were reported for multiple individual congeners but not summed values, we averaged effect size across congeners. Where multiple studies occurred on the same system, we selected only the most recent study. We reported the absolute average values because relationships can be positive or negative across habitats, i.e. with $\delta^{13}C$ or $\delta^{34}S$. We then used a pairwise *t*-test to test the hypothesis that the strength of relationships is stronger for some isotopes than other isotopes.

3. Results

3.1. Osprey eggs

The variation in $\delta^{15}N$ for 'trophic' amino acids was similar to the variation in 'source' amino acids such that any trophic signal in bulk $\delta^{15}N$ was swamped by variation in $\delta^{15}N$ baseline (i.e., variation in $\delta^{15}N$ in source amino acids; Table 1). Indeed, bulk $\delta^{15}N$ was independent of $\delta^{15}N_{trophic\text{-}source}$ (regression: $t_{27} = 0.45$, $P = 0.66$; $R^2 = 0.01$). Both phenylalanine and 'source' $\delta^{15}N$ (average of Met, Lys, Phe) correlated with the amount of anthropogenic activity in the surrounding area (Fig. 2). Due to lack of marine inputs, we did not measure $\delta^{34}S$ or $\delta^{18}O$ in osprey. Eggs from ospreys nesting in alpine environments had relatively low $\delta^{15}N$ while those nesting in agricultural environments had relatively high $\delta^{15}N$, with the remaining watersheds (Columbia, Ootsa, Yukon) being intermediate but showing distinct clusters (Fig. 3). Thus, the assumption that the baseline should be the same to compare $\delta^{15}N$ across habitats is violated as both baseline $\delta^{15}N$ and baseline POP exposure vary across habitats. Egg $\delta^{13}C$ showed no clusters associated with the different watersheds. All amino acids except Thr loaded heavily on the first principal component for nitrogen, with the second principal component weakly discriminating source from trophic amino acids (Fig. 3). All amino acids also heavily loaded on the first principal component for carbon with no strong separation among essential and non-essential amino acids (Fig. 3).

Table 1
The range (SD in parentheses) of average $\delta^{15}N$ values for trophic (Asp, Glu, Ile, Leu and Pro) and source (Met, Lys and Phe) amino acids for three different systems, as well as for Glu, Phe and bulk tissue. Note that the pattern in the variation might be different even if the magnitude is the same.

System	Trophic (‰)	Glu (‰)	Bulk (‰)	Source (‰)	Phe (‰)
Osprey eggs	18.4–26.0 (1.94)	17.1–24.4 (1.92)	8.6–14.3 (1.66)	2.5–9.8 (2.00)	2.1–9.3 (1.92)
Seabird eggs	18.6–29.3 (1.92)	18.8–30.6 (2.53)	10.4–17.3 (1.26)	3.7–9.0 (1.00)	3.9–9.3 (1.17)
Arctic fish	17.8–28.9 (2.44)	20.1–30.4 (2.46)	9.2–17.9 (2.10)	6.8–11.8 (1.21)	5.7–10.3 (1.19)

We considered the effect of diet on contaminants after accounting for watershed features that might influence contaminant levels. After accounting for confounding variables (lake size; $t_{39} = -3.36$, $P = 0.002$), $\delta^{15}N_{trophic\text{-}source}$ ($t_{39} = 4.50$, $P < 0.0001$) was significantly related to ΣPCB in chicks, while bulk $\delta^{13}C$, bulk $\delta^{15}N$, other watershed features (including proportion glacial coverage) and % lipids were not (all $P > 0.05$). ΣPCB was higher in small lakes likely due to lower dilution. In adults, $\delta^{15}N_{trophic\text{-}source}$ ($t_9 = 2.05$, $P = 0.04$), but not bulk $\delta^{15}N$ ($P > 0.3$), also correlated with ΣPCB (Fig. 4). Bulk $\delta^{15}N$ ($t_{39} = 2.73$, $P = 0.009$) and elevation ($t_{39} = -3.48$, $P = 0.001$) were significantly related to ΣDDT in chicks, while bulk $\delta^{13}C$, $\delta^{15}N_{trophic\text{-}source}$, proportion of glacial coverage, watershed area, % lipids and lake area were not (all $P > 0.05$). Highest levels of ΣDDT were in agricultural areas (Oliver, lowland Alberta) that had high $\delta^{15}N_{source}$. Thus, interpretation of relationships between ΣPCB or ΣDDT and was complicated by variation in both baseline contaminant values and $\delta^{15}N_{source}$. Isotope values were not related to either ΣPCB or ΣDDT in eggs; contaminants in eggs represent a complex signal of wintering, migratory and pre-breeding habitats (Elliott et al., 2007). No relationships were found with mercury levels, which are related to deposition rates in particular lakes rather than diet in our study system (Guigueno et al., 2011).

3.2. Pacific seabirds

Roughly half of the variation in 'trophic' $\delta^{15}N$ was associated with variation in baseline (Table 1). All amino acids loaded heavily on the first principal component for nitrogen, with the second principal component discriminating source from trophic amino acids (Fig. 3). Similarly, all amino acids loaded heavily on the first principal component for carbon with the second component discriminating essential from non-essential amino acids (Fig. 3). Seabird species showed clear clusters in both carbon and nitrogen plots (Fig. 3). Bulk $\delta^{15}N$ increased with $\delta^{15}N_{trophic\text{-}source}$ ($t_{95} = 2.17$, $P = 0.02$; $R^2 = 0.13$).

Log ΣDDT ($t_{11} = 3.02$, $P = 0.01$) and log ΣPCB ($t_{11} = 2.87$, $P = 0.02$) correlated with $\delta^{15}N_{trophic\text{-}source}$, but not bulk $\delta^{15}N$, $\delta^{34}S$, $\delta^{18}O$ or $\delta^{13}C$ (all $P > 0.25$ in full model; Fig. 4). For double-crested cormorants, the species with the longest time trend, log ΣDDT ($t_{12} = -2.61$, $P = 0.02$; half-life: 18.7 y), log ΣPCB ($t_{12} = -5.21$, $P = 0.0008$; half-life = 7.1 y), log Hg ($t_{12} = -3.64$, $P = 0.0006$) and $\delta^{34}S$ ($t_{12} = -7.36$, $P = 0.0003$) declined between 1970 and 2011. However, log ΣDDT, the contaminant with the most complete time series, increased ($t_{10} = 1.93$, $P = 0.04$) between 1985 and 2002. Baseline $\delta^{15}N$ also increased between 1973 and 2006, when 1979 (a year of anomalously high baseline and bulk $\delta^{15}N$ shortly after the 1977 North Pacific regime shift) was excluded ($t_5 = 3.57$, $P = 0.04$). When adjusted for $\delta^{15}N_{trophic\text{-}source}$, log ΣDDT showed a

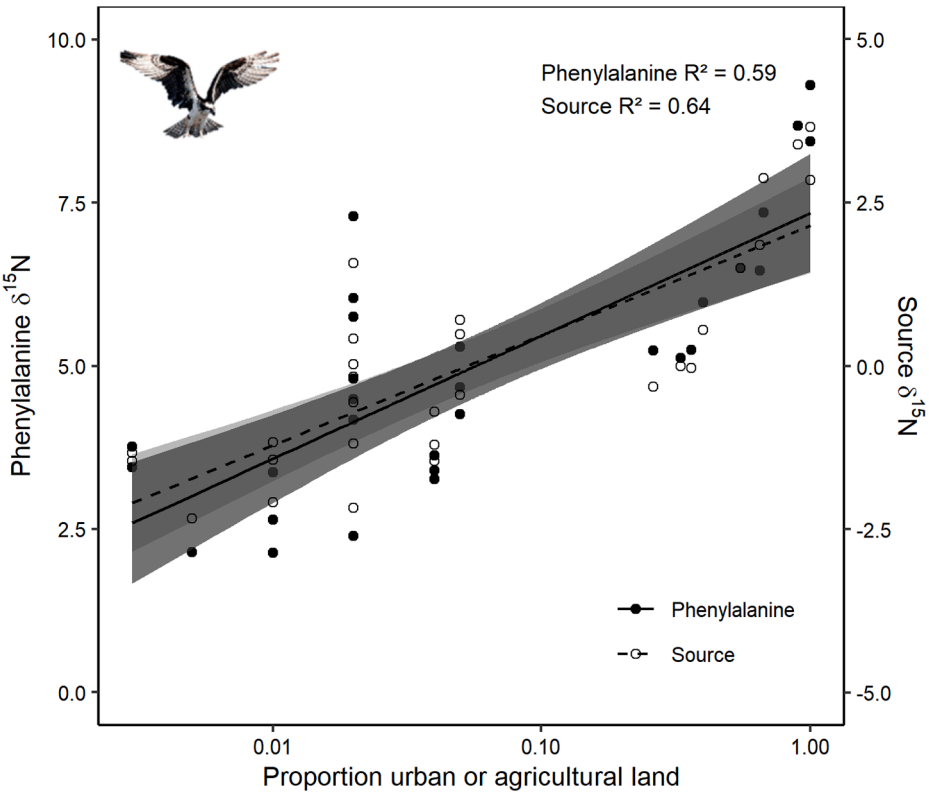

Fig. 2. Phenylalanine $\delta^{15}N$ (black line) and the average of $\delta^{15}N$ for the three source amino acids (Phe, Met, Lys; dashed line) increase with urban or agricultural land cover within 25 km upstream of osprey nests. Shaded area represents the 95% confidence interval for phenylalanine (dark grey) and source (light grey). Source amino acids provided a slightly better correlation than using phenylalanine alone.

stronger relationship with year than non-adjusted log ΣDDT ($t_{12} = -5.42$, $P = 0.0002$; half-life: 11.8 y; ΔAIC 7.71 compared with non-corrected values; Fig. 5). When adjusted for $\delta^{34}S$, Hg no longer showed a declining trend. Log ΣPBDE decreased with $\delta^{34}S$ ($t_{10} = -4.23$, $P = 0.002$) while Hg ($t_{10} = 7.45$, $P < 0.0001$) and log HBCD ($t_{10} = 2.48$, $P = 0.03$) increased with $\delta^{34}S$; bulk $\delta^{15}N$ and $\delta^{15}N_{trophic\text{-}source}$, bulk $\delta^{18}O$ and bulk $\delta^{13}C$ were independent of those contaminants (all $P > 0.25$ in full model). Log ΣPFCA and log ΣPFAS were independent of all isotope values while PFuDA increased with $\delta^{15}N_{trophic\text{-}source}$ ($t_{10} = 2.18$, $P = 0.04$), but not other isotope values (all $P > 0.2$ in full model). Thus, diet changed over time, leading to alternative sources for contaminants, such as benthic food webs potentially higher in DDT and Hg (Fig. 5).

3.3. Arctic fish and invertebrates

Roughly half of the variation in 'trophic' $\delta^{15}N$ was associated with variation in baseline (Table 1). All amino acids loaded heavily on the first principal component for nitrogen, with the second principal component discriminating source from trophic amino acids (Fig. 3). Taxonomic groups showed clear clusters, with invertebrates low in $\delta^{15}N$ and benthic fish high in $\delta^{15}N$. Similarly, all amino acids loaded heavily on the first principal component for carbon with no strong separation among essential and non-essential amino acids (Fig. 3). The fourth component (9.2% of variance) discriminated essential from non-essential amino acids. Bulk $\delta^{15}N$ increased with $\delta^{15}N_{trophic\text{-}source}$ ($t_{29} = 3.38$, $P = 0.002$; $R^2 = 0.28$). Log ΣPCB ($t_{12} = 4.47$, $P = 0.001$) and log ΣDDT ($t_{12} = 4.32$, $P = 0.001$) correlated with $\delta^{15}N_{trophic\text{-}source}$, but not bulk $\delta^{15}N$, $\delta^{34}S$, $\delta^{13}C$ or percent lipids (all $P > 0.05$ in full model; Fig. 4). Log Hg correlated with $\delta^{34}S$ ($t_{25} = -2.03$, $P = 0.03$; Fig. 4) and $\delta^{15}N_{trophic\text{-}source}$ ($t_{25} = -1.82$, $P = 0.04$), but not bulk $\delta^{15}N$ ($t_{25} = 1.33$, $P = 0.10$) or $\delta^{13}C$ ($t_{25} = 0.85$, $P = 0.40$). Log ΣBDE, Log ΣPFAS and log ΣPFCA were not significantly correlated with any of the five parameters (all $P > 0.05$ in full model).

The $\delta^{15}N$ discrimination factor between prey and consumer (seabird chick) showed the classic pattern of little discrimination in the source amino acids (not significantly different from zero) and strong discrimination in the trophic amino acids (Table 2). The remaining amino acids were intermediate while the metabolic amino acid Thr was strongly negatively discriminated. These relationships coincided with principal components analyses for $\delta^{15}N$, which showed similar separation between the source and trophic amino acids, after ignoring the first principal component (the first component showed strong loading for all amino acids due to similar baselines; Fig. 3). Except for ospreys, the principal components analyses for $\delta^{13}C$ showed separation between essential and non-essential amino acids after ignoring the first component. Except for chlordane and ΣPBDE, trophic magnification factors were lower than biomagnification factors (Table 3). Biomagnification factors from $\delta^{15}N_{trophic\text{-}source}$ correlated with those estimated from murre-prey relationships ($t_5 = 3.52$, $P = 0.02$, $R = 0.87$) while those from bulk $\delta^{15}N$ did not ($t_5 = 1.81$, $P = 0.13$, $R = -0.51$).

3.4. Meta-analysis

ΣDDT did not correlate any better with bulk $\delta^{15}N$ than bulk $\delta^{13}C$ (pairwise t-test: $t_9 = 0.30$, $P = 0.77$) or $\delta^{34}S$ ($t_3 = 0.60$, $P = 0.59$; Table 4). ΣPCB did not correlate any better with bulk $\delta^{15}N$ than bulk $\delta^{13}C$ ($t_8 = 0.86$, $P = 0.41$) or $\delta^{34}S$ ($t_2 = 0.13$, $P = 0.91$). ΣPBDE did not correlate any better with bulk $\delta^{15}N$ than bulk $\delta^{13}C$ ($t_{10} = 1.88$, $P = 0.09$) or $\delta^{34}S$ ($t_3 = 1.21$, $P = 0.31$). Hg did not correlate any better with bulk $\delta^{15}N$ than bulk $\delta^{13}C$ ($t_{15} = 0.18$, $P = 0.86$) but correlated better with $\delta^{34}S$ than bulk $\delta^{15}N$ ($t_7 = 2.78$, $P = 0.03$).

4. Discussion

Across space, time and species, variation in the pathway of a contaminant within the food web was critical in determining

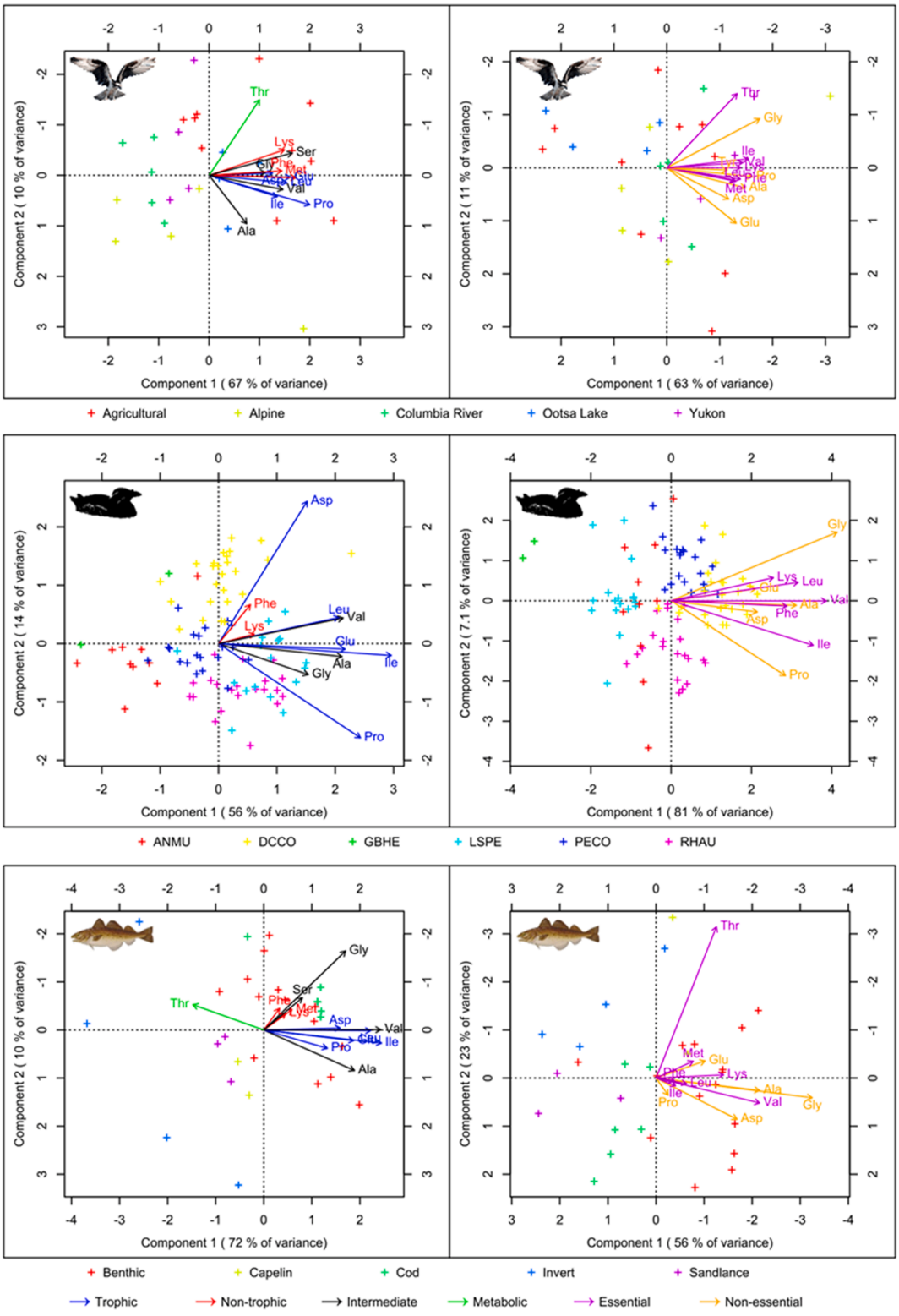

Fig. 3. Principal components analysis of amino acids for $\delta^{15}N$ (left) and $\delta^{13}C$ (right) in (a,b) Arctic fish, (c,d) osprey eggs, and (e,f) Pacific seabird eggs.

contaminant levels of top predators, levels that were many orders of magnitude higher than in surrounding waters. In ospreys, variation in baseline $\delta^{15}N$ across space created a spurious relationship between DDE and trophic position while masking a relationship between PCBs and trophic position. This contradiction occurred because both baseline $\delta^{15}N$ and ΣDDT correlated with the degree of agricultural input (percent of watershed used in agriculture). In Pacific seabirds, changes in diet over time created the appearance of an increase in DDE over 17 years (1985–2002) and a decrease in mercury (1973–2006) that disappeared once diet was accounted for. The diet switch (decreasing $\delta^{34}S$ and increasing baseline $\delta^{15}N$) towards benthic prey could be driven by a local decline in forage fish, especially herring (Elliott and Elliott 2016). Benthic fish are part of a detrivore food web, sometimes consuming dead animals, leading to high baseline $\delta^{15}N$. In Arctic fish, a relationship

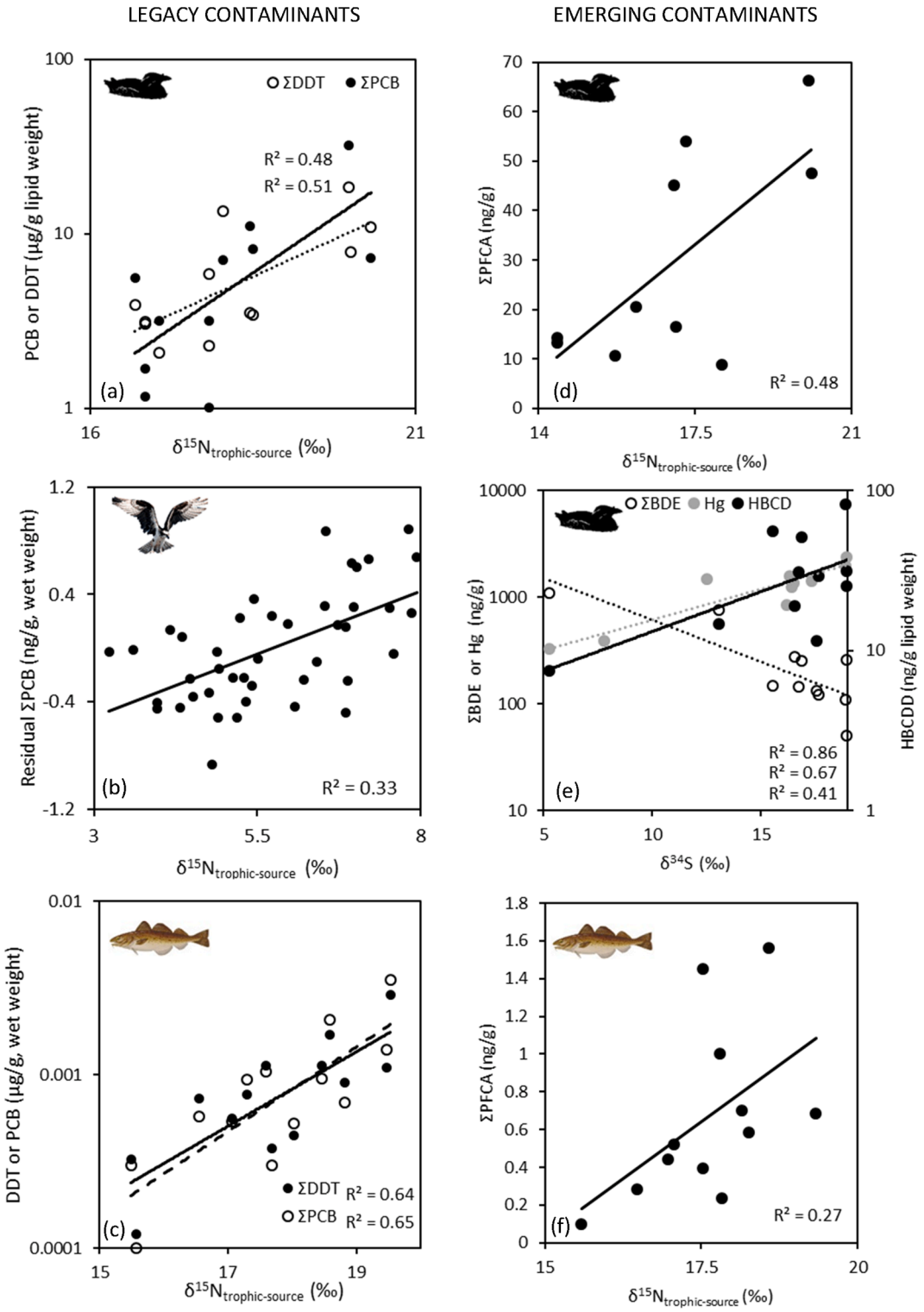

Fig. 4. An index of baseline-corrected trophic position ($\delta^{15}N_{trophic\text{-}source}$) correlates with contaminant concentration for legacy contaminants in (a) seabirds, (b) osprey and (c) Arctic fish. Bulk $\delta^{15}N$ showed non-significant (seabirds, osprey) or weak (Arctic fish) relationships. With the exception of (d) PFuDA, emerging contaminants showed relationships with (e) spatial isotopes ($\delta^{34}S$) but not (f) $\delta^{15}N_{trophic\text{-}source}$.

between trophic position and mercury across species was only apparent after accounting for habitat, and trophic magnification factors for multiple contaminants correlated better with biomagnification factors once corrected for baselines. Thus, accounting for variation at the base of the food web ($\delta^{34}S$, $\delta^{15}N_{source}$) is crucial to account for the effect of diet on contaminant levels in top predators.

Across the three ecosystems, baseline-corrected trophic position ($\delta^{15}N_{trophic\text{-}source}$) was consistently more important for explaining patterns of legacy contaminants (i.e. PCB, DDE) than 'emerging' contaminants (i.e. PFCAs, PBDEs). For example, $\delta^{15}N_{trophic\text{-}source}$ correlated with ΣDDT, ΣPCB and mercury (after accounting for $\delta^{34}S$) in Arctic fish and Pacific seabirds, but not with ΣPBDE, ΣPFCA and ΣPFAS (except PFuDA in seabirds). This pattern is unsurprising, as the deleterious effects of legacy contaminants were evident over 50 years ago due to their high

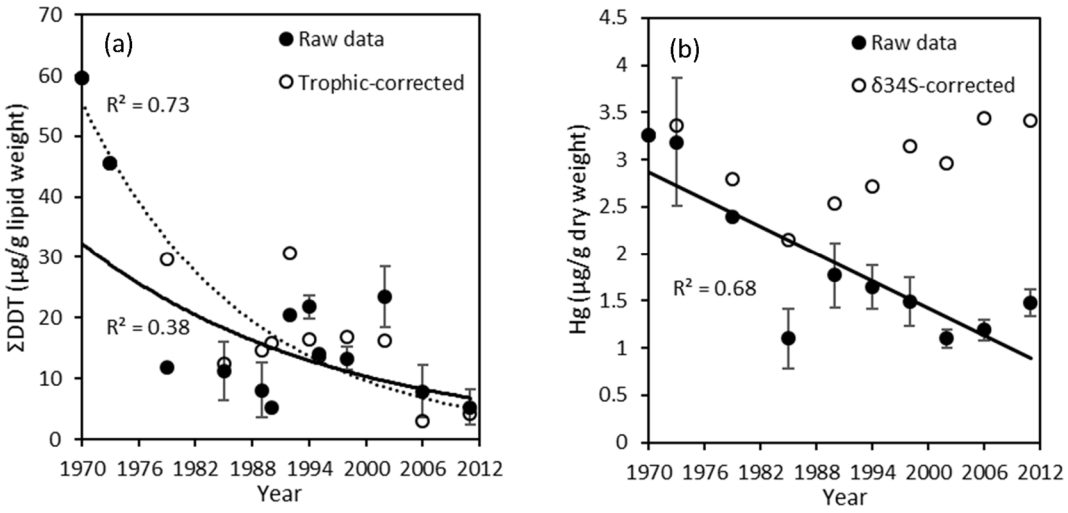

Fig. 5. (a) ΣDDT in double-crested cormorants declined during 1970–2011 (solid line), but the decline was explained better by date after accounting for $\delta^{15}N_{trophic\text{-}source}$ ('trophic-corrected'; dotted line); (b) Hg in double-crested cormorants declined during 1970–2011, but the decline disappeared after accounting for $\delta^{34}S$.

Table 2
Discrimination factors (±SE) between consumer (murre chick red blood cells) and prey, denoted by delta notation (Δ). Trophic amino acids are shown in bold and source amino acids are shown in italics (under $\Delta^{15}N$). The remaining amino acids are "intermediate" between trophic and non-trophic. Essential amino acids are shown in bold (under $\Delta^{13}C$).

Amino acid	$\Delta^{13}C$	$\Delta^{15}N$
Bulk	0.7 ± 0.5	3.1 ± 0.9
Ala	1.6 ± 0.9	2.9 ± 1.0
Asp	**0.0 ± 0.8**	**5.4 ± 0.8**
Glu	**−0.2 ± 1.1**	**4.2 ± 1.2**
Gly	1.9 ± 0.6	1.3 ± 1.0
Ile	**0.1 ± 0.5**	**5.8 ± 1.5**
Leu	**−0.1 ± 0.2**	**4.3 ± 0.6**
Lys		*−0.8 ± 0.9*
Met		*0.1 ± 0.4*
Phe	**0.1 ± 0.5**	*−0.3 ± 0.6*
Pro	**0.0 ± 0.4**	**6.1 ± 0.6**
Ser	1.8 ± 1.0	1.8 ± 1.1
Thr	**0.3 ± 0.4**	−4.4 ± 0.9
Val	**0.2 ± 0.4**	3.4 ± 1.1
Trophic (averaged)		**5.2 ± 0.9**
Source (averaged)		*−0.3 ± 0.6*

Table 3
Comparison of biomagnification and trophic magnification factors. Biomagnification factors (lipid weight basis, except Hg) were calculated as the ratio in consumer (murre livers) relative to levels in prey muscle, weighted by proportion of prey species in diet of murre chicks. Trophic magnification factors (TMFs) are based on the slope of the Ln (contaminant) - $\delta^{15}N$ relationship across prey items. All values represent log-transformed BMF and TMF, with the equation for BMF and TMF described in the text.

Contaminant	ΣCHL	ΣDDT	ΣPCB	ΣPBDE	ΣPFCA	Hg
BMF	1.1	3.9	4.2	0.1	2.6	3.4
Bulk $\delta^{15}N$ *TMF*	3.3	2.1	1.9	3.0	1.5	1.3
$\delta^{15}N_{trophic\text{-}source}$ *TMF*	3.4	8.9	8.0	1.4	7.3	4.3

levels in top predators, which is directly related to their strong rates of biomagnification (Atwell et al., et al., 1998; Fisk et al., 2001). Meanwhile, spatial variation (especially $\delta^{34}S$) was more important for those contaminants more recently incorporated into existing international conventions (some of which are classified as 'emerging'), and for which biomagnification rates are often lower (Kelly et al., 2007, 2008; Elliott et al., 2009; Newsome et al., 2010). For example, HBCD, Hg and ΣPBDE in seabirds were primarily associated with variation in habitat, as assessed by $\delta^{34}S$. Birds foraging farther offshore, closer to Asia, had higher levels of HBCD, while birds foraging nearshore to North America had higher levels of ΣPBDE (Miller et al., 2014, 2015). Other studies have likewise failed to find strong evidence for biomagnification of emerging contaminants, including highly brominated diphenyl ethers, ΣPFCA, ΣPFAS, at the upper levels of Arctic food chains (Kelly et al., 2007). Presumably, they are either broken down or adsorb tightly to organic particles and are eliminated, or have limited lipid solubility compared with legacy compounds (Kelly et al., 2007, Buck et al., 2011). As is the case in many other studies (Table 4), "spatial" stable isotope ratios ($\delta^{34}S$ or $\delta^{13}C$) better predicted concentrations than "trophic" stable isotope ratios ($\delta^{15}N$) for both legacy and emerging contaminants, at least at the species or guild level where trophic position was relatively constant. Many of the emerging compounds were associated with marine input ($\delta^{34}S$), as the oceans may be the ultimate sink for those compounds. We encourage greater use of spatial isotopes in contaminant models, which can often add to the variation explained by $\delta^{15}N$ alone (McMeans et al., 2010). In some studies, only $\delta^{15}N$ is examined even though $\delta^{13}C$ is also routinely measured (foxes [Fuglei et al., 2007]; lake food webs [Kiriluk et al., 1995; Lescord et al., 2015]; dolphins [Wilson et al., 2012]), or $\delta^{13}C$ is only used to distinguish among habitats (bears [Christensen et al., 2005]; fish [Páez et al., 2012; Carr et al., 2017]; wolves [McGrew et al., 2014]; cormorants [Ofukany et al., 2012]; sharks [McMeans et al., 2015]); we argue that including other isotopes quantitatively could improve studies.

Baseline $\delta^{15}N$ also played a role in obscuring trends in trophic position. Across all three systems, baseline variation in $\delta^{15}N$ (5–7‰) is only slightly smaller than variation in bulk $\delta^{15}N$ (8-11‰; Table 1). Thus, variation in baseline $\delta^{15}N$ obscures much of the variation in trophic position associated with bulk $\delta^{15}N$. The correlation between bulk $\delta^{15}N$ and $\delta^{15}N_{t\text{-}s}$ is weak, with R^2 values varying from 0.01 to 0.28. Studies routinely "correct" for trophic position by examining the residual variation in contamination after accounting for bulk $\delta^{15}N$ as an index of trophic position (i.e. Day et al., 2012a,b; Braune et al., 2014a,b; Burgess et al., 2013). Without correcting for baselines, such attempts at correction may lead to erroneous conclusions, such as those demonstrated in our study. Such issues may also be compounded if $\delta^{15}N$ varies with condition (e.g. Hobson 2008). For top predators feeding across multiple, but often unknown, habitats spread out across hundreds of kilometers, determining a baseline value by measuring $\delta^{15}N$ in primary consumers is challenging. Amino acid-specific stable isotope analysis overcomes that challenge by measuring baselines in the same tissues where contaminant levels are measured.

Table 4
Studies of single species or species guilds (i.e. entire food webs excluded) that reported both $\delta^{15}N$ and $\delta^{13}C$ in conjunction with contamination. The table is representative rather than comprehensive. We reported the absolute average values because relationships can be positive or negative across habitats, i.e. with $\delta^{13}C$ or $\delta^{34}S$.

Species	$\delta^{15}N$	$\delta^{13}C$	$\delta^{34}S$	Contaminant	Reference
American dipper	<1.29	1.29		ΣPCB	Morrissey et al. (2011)
Bald eagle	−0.17	4.48		ΣPCB	Elliott et al. (2009)
Shearwater species	2.91	0.71		ΣPCB	Roscales et al. (2010)
Whale species	2.16	−1.25		ΣPCB	Pinzone et al. (2015)
Osprey	1.81	2.89		ΣPCB	Our study
Seabird species	0.70	−0.40	−1.12	ΣPCB	Our study
Arctic fish species	1.85	−0.88	−1.97	ΣPCB	Our study
Bird of prey species	1.85	−2.49		PCB-153	Bustnes et al. (2013)
Gull species	0.54	0.17	2.67	PCB-77/81	Roscales et al. (2016)
White-tailed eagle	1.63	1.10	<d13C	ΣPCB	Eulaers et al. (2014)
Average absolute value	**1.43 ± 0.31**	**1.57 ± 0.47**	**1.58 ± 0.45**		
American dipper	<2.05	2.87		ΣDDT	Morrissey et al. (2011)
Shearwater species	2.87	1.66		ΣDDT	Roscales et al. (2010)
Whale species	1.28	−1.44		ΣDDT	Pinzone et al. (2015)
Osprey	1.32	−1.79		ΣDDT	Our study
Seabird species	0.39	−0.38	−0.94	ΣDDT	Our study
Arctic fish species	1.72	−0.32	−1.86	ΣDDT	Our study
Bird of prey species	2.70	−3.74		DDE	Bustnes et al. (2013)
White-tailed eagle	2.85	<d15N	<d15N	DDE	Eulaers et al. (2015)
Bald eagle	−1.69	3.42		DDE	Elliott et al. (2009)
Average absolute value	**1.87 ± 0.26**	**1.95 ± 0.34**	**1.57 ± 0.39**		
Seabird species	1.03	0.09	7.45	Hg	Our study
Arctic fish	2.96	1.40	−6.20	Hg	Our study
Murre species	3.54	5.09		Hg	Day et al. (2012a,b)
Ringed seal	3.56	<d15N		Hg	Masbou et al. (2015)
American dipper	2.76	<d15N		Hg	Morrissey et al. (2011)
Fish species	1.88	3.94	1.63	Hg	Ethier et al. (2008)
Waterbird species	6.64	4.12	7.56	Hg	Lavoie et al. (2015)
Shark species	2.99	<d15N		Hg	Domi et al. (2005)
Freshwater fish species	~1.93	1.93		Hg	Eagles-Smith et al. (2008)
Waterbird species	1.84	4.18		Hg	Ramos et al. (2013)
Herptile and spider species	1.57	2.19		Hg	Bryan et al. (2012)
Yellow-legged gull	<d34S	<d34S	7.88	Hg	Walters et al. (2008)
Freshwater species	<d34S	<d34S	5.64	Hg	Schmitt et al. (2011)
Fish and invertebrate species	0.47	2.80	4.62	Hg	Reinhart et al. (2018)
Shark species	3.80	3.41		Hg	McKinney et al. (2016)
Fish and invertebrate species	2.10	1.81	<d13C	Hg	Clayden et al. (2017)
Polar bear	−0.02	1.82	4.00	Hg	Lippold et al. (2020)
Average absolute value	**2.58 ± 0.38**	**2.60 ± 0.32**	**5.10 ± 0.84**		
Seabird species	0.53	1.01	−4.23	ΣBDE	Our study
Arctic fish	1.97	−2.21	2.39	ΣBDE	Our study
Gull species	0.84	0.80		ΣBDE	Chen et al. (2012)
Whale species	1.23	−1.39		ΣBDE	62
American dipper	2.30	3.07		ΣBDE	Morrissey et al. (2011)
Bald eagle	1.58	4.68		ΣBDE	Elliott et al. (2009)
Waterbird species	0.93	0.87		ΣBDE	Zhang et al. (2010)
White-tailed eagle	2.45	1.62	1.18	ΣBDE	65
Gull species	−0.16	−1.61	−2.28	ΣBDE	64
Herring gull	−2.48	1.90		ΣBDE	Sørmo et al. (2011)
Peregrine falcon	−2.60	8.18		ΣBDE	Newsome et al. (2010)
Average absolute value	**1.55 ± 0.26**	**2.49 ± 0.66**	**2.52 ± 0.63**		

The incorporation of additional stable isotope ratios and isotopic baselines would allow better estimates of trophic magnification factors separate from spatial components (Hobson 1999; Hebert et al., 2006, 2016). Indeed, as baseline $\delta^{15}N$ can change by several "trophic" levels over relevant spatial scales, correlations between bulk $\delta^{15}N$ and contaminants may be partly explained by baseline variation and researchers are therefore already accounting for variation at the base of the food web when they attempting to correct for trophic position. For example, trophic magnification factors corrected for baselines were more similar to murre biomagnification factors than trophic magnification factors using bulk $\delta^{15}N$ alone (Table 3). In contrast, incorporating $\delta^{15}N_{Glu\text{-}Phe}$ did not improve trophic magnification factors for fish in Tokyo Bay compared with bulk $\delta^{15}N$ (Kobayashi et al., 2019).

Although most researchers consider trophic position—as inferred by $\delta^{15}N$—as the major predictor of contaminants (i.e. 88% of studies, as reported in the Introduction), our *meta*-analysis clearly showed that other stable isotopes can be equally important (Table 4). For no contaminant was $\delta^{15}N$ a significantly better predictor than $\delta^{13}C$ or $\delta^{34}S$, and for mercury $\delta^{34}S$ was significantly better and for PBDEs $\delta^{13}C$ was marginally better ($P = 0.08$). Thus, there is little empirical evidence, at the level of a species or small assemblage, that $\delta^{15}N$ is the best predictor of contaminants. We argue that food webs are complex, and incorporating other isotopes may provide better resolution of diet, thereby improving prediction of contaminants.

Our amino acid-specific $\delta^{15}N$ discrimination values were remarkably similar to those reported for other bird muscle, feather and red blood cells (Lorrain et al., 2009; McMahon et al., 2015; Hebert et al., 2016). The standard deviation across the three studies, averaged across amino acids, is 0.15 for $\delta^{13}C$ and 0.40 for $\delta^{15}N$, compared with 1.51 and 0.71, respectively, across eight studies of bulk $\delta^{15}N$ in feathers (Becker et al., 2007). That we have such similar values across variable tissues and species, in contrast to studies involving a single tissue (feather) of relatively uniform protein content (keratin), implies that the amino acid composition due to variable diet may be a major reason why bulk discrimination factors vary among species, tissues, sex, age and other factors. The reduction of variation in discrimination factors using amino

acids provides a promising avenue for future investigations of diet. The incorporation of isotopic baselines, and repeatable discrimination factors, improves our ability to quantitatively assess the role of each dietary component in contamination, which is rarely achieved in contaminants research (Jardine et al., 2006).

We recommend two methodological improvements over earlier work. First, given the current high cost of amino acid-specific analyses, costs can be reduced by running such analyses on only a pooled subsample (Dolgova et al., 2018). Obtaining the baseline for each lake (ospreys), colony (seabirds) or species (fish) from sub-samples (N = 3) provided robust statistical power for our analyses. While baselines for all individuals would be ideal (costing ~$4000 for 20 individuals), obtaining bulk values from 20 individuals and using a single lake-, colony- or species-wide baseline calculated from three pooled samples (costing ~$760) would have provided sufficient power to explain variation in contaminants. Second, relationships were marginally, but consistently, stronger using several baseline and trophic amino acids, rather than just phenylalanine and glutamate. We recommend averaging across several amino acids to improve relationships. Across all three communities, the three baseline amino acids (Phe, Met and Lys) tended to load together on the $\delta^{15}N$ principal components analysis while four trophic amino acids (Glu, Ile, Leu, Pro) tended to load together with Thr in the opposing direction (Fig. 3). In contrast, Asp, a trophic amino acid, loaded with either group. An approach using the first principal component of Glu, Ile, Leu, Pro and Thr and first principal component of Phe, Met and Lys could provide an even more robust metric.

In conclusion, across several systems, using bulk $\delta^{15}N$ alone would have led to inaccurate conclusions about the factors influencing the spatial patterns determining ΣPCB and ΣDDT in osprey, the temporal trend of Hg and ΣDDT in seabirds, and the rate of biomagnification of contaminants through an Arctic food web. By coupling amino acid-specific isotope analyses with spatial isotopes, we avoided assuming the uniformity of baselines that have confounded estimates of chemical bioaccumulation and biomagnification in past studies (Borgå et al., 2012; McLeod et al., 2015). We argue that the use of amino acid-specific analysis of $\delta^{15}N$ alongside 'non-trophic' isotopes should be a core feature of any study that examines the influence of trophic position on chemical pollution, as required for a chemical to be added to international conventions such as the Stockholm Convention.

CRediT authorship contribution statement

Kyle H. Elliott: Funding acquisition, Writing - original draft. **Birgit M. Braune:** Funding acquisition, Writing - review & editing. **John E. Elliott:** Funding acquisition, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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