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Variation in bioaccumulation of persistent organic pollutants based on octanol–air partitioning: Influence of respiratory elimination in marine species

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ABSTRACT

Risk assessments of persistent organic pollutants (POPs) are often based on octanol–water (K_{OW}) partitioning dynamics and may not adequately reflect bioaccumulation in air-breathing organisms. It has been suggested that compounds with low K_{OW} and high octanol–air partitioning (K_{OA}) coefficients have the potential to bioaccumulate in air-breathing organisms, including marine mammals. Here we evaluate differences in concentrations of POPs for two trophically matched Arctic species, spotted seal (*Phoca largha*) and sheefish (*Stenodus leucichthys*). We compared concentrations of 108 POPs in matched tissues (liver and muscle) across three ranges of K_{OW}. We found a significant positive correlation between POP concentration and log K_{OA} in spotted seal tissues for low log K_{OW} compounds (log $K_{OW} < 5.5$, $p < 0.05$). This provides further evidence for empirical models and observed bioaccumulation patterns in air-breathing organisms, and highlights the potential for bioaccumulation of these compounds in Arctic marine mammals.

1. Introduction

Persistent organic pollutants (POPs), including organochlorines (OCs), have been detected in biota in otherwise relatively pristine regions such as the Arctic (AMAP, 2004; Wania and MacKay, 1996). Risks associated with these chemicals depend on their potential for long-range transport, environmental persistence, toxicity, and capacity to biomagnify and bioaccumulate (Letcher et al., 2010; Muir and de Wit, 2010). The Arctic is well recognized as a sink for a number of these persistent, bioaccumulative, and toxic (PBT) chemicals and is of particular interest for POP risk assessments due to their repeated deposition and remobilization (Burkow and Kallenborn, 2000). While international bans on certain industrial POPs have resulted in decreasing trends of some contaminants in Arctic biota since the 1990s, recent studies show that as the Arctic warms due to anthropogenic climate change, contaminants once trapped in ice and permafrost may be released and/or revolitalize (Ma et al., 2011; Moore et al., 2014).

The potential for bioaccumulation and biomagnification of lipophilic contaminants has been repeatedly demonstrated in Arctic environments (Kelly and Gobas, 2003; Muir et al., 1999; Van Oostdam et al., 2005). This is of particular concern in this region where the consumption of the lipid rich tissues of top predators, such as marine fish and mammals, is a vitally important energy source for subsistence populations (Johnson et al., 2009). The fish based food webs of marine mammals result in moderately to highly contaminated tissues of some pinnipeds and cetaceans, including those in Arctic regions (Letcher et al., 2010). Similarly, Arctic subsistence populations are exposed to high quantities of POPs through their diet, which has been associated with adverse health outcomes such as reduced gestation time and impaired fetal development (Dallaire et al., 2013; Laird et al., 2013).

Biomagnification potential is determined by an organism's ability to absorb, biotransform and eliminate a given compound, which is largely dependent upon its physical–chemical properties. The octanol–water (K_{OW}) partition coefficient is often used to assess the tendency for a particular compound to bioconcentrate in the lipid compartments of aquatic organisms (Gossett et al., 1983; Meylan et al., 1999). Understanding and predicting chemical behavior within food webs using quantitative structure activity relationships (QSARs) based on K_{OW} is an important aspect of the registration and management of POPs, and is crucial for identifying potential contaminants of concern. The bioaccumulation models most frequently used to define criteria for acceptable levels of POPs in food, water and sediments have been derived from data collected from aquatic organisms.

Generally, bioaccumulation is considered qualitatively in risk assessments based on a "cut-off" value. For example, in their persistent, bioaccumulative, and toxic (PBT) substance policy statement, the U.S. Environmental Protection Agency (EPA) defines bioaccumulative

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substances as those with a bioconcentration factor (BCF) >1000 (U.S. Environmental Protection Agency, 1999). The European REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals) guidelines define a bioaccumulative substance as one with BCF > 2000 (Environment Canada, 1999). If its production volume is <100 tons/year, demonstrating that a chemical has a log K_{OW} < 4.5 (measured or QSAR calculated) can be used in place of BCF data to establish that a compound is not bioaccumulative. The relationship between BCF/BAF and K_{OW} for neutral, lipophilic, organic chemicals has been demonstrated in numerous aquatic and marine food webs in both the laboratory and field (Arnot and Gobas, 2006; Fisk et al., 2001; Russell et al., 1999).

However, using only K_{OW} to predict chemical partitioning under environmental conditions may not adequately reflect actual physical–chemical behavior, particularly in air-breathing species. Compared to aquatic systems, there has been relatively little research assessing the bioaccumulation potential of POPs in terrestrial ecosystems (Gobas et al., 2003; Kelly and Gobas, 2003). QSAR and bioaccumulation models generally do not consider the octanol–air partitioning coefficient (K_{OA}), which may characterize an important excretory route for air-respiring animals including terrestrial vertebrates and marine mammals. The few studies that do incorporate lipid–air partitioning behavior into models of bioaccumulation by terrestrial species or marine mammals suggest K_{OA} may be a better indicator of bioaccumulation potential in air-breathing species (Czub et al., 2008; Kelly et al., 2007; Kelly and Gobas, 2001, 2003).

It has been suggested that compounds with a relatively low K_{OW} (log K_{OW} < 5) can biomagnify in terrestrial food webs (i.e. lichen, wolf and reindeer) for compounds with a relatively high K_{OA} (log K_{OA} > 5) due to low rates of respiratory elimination (Gobas et al., 2003; Kelly and Gobas, 2003). Similarly, it was demonstrated that in a piscivorous marine food web, air-breathing organisms (i.e. marine mammals) display lower concentrations of low K_{OW}, high K_{OA} compounds (e.g. β-hexachlorocyclohexane or β-HCH) than water-respiring organisms at the same trophic position (Kelly et al., 2007). The chemical amplification of compounds with a log K_{OA} > 6 and 2 < log K_{OW} < 5 was estimated to be approximately 2000-fold from base concentrations of primary producers to humans (top predators). This represents a previously underestimated risk to human populations, especially residents of Northern communities that rely on subsistence diets.

Here we present further evidence for the bioaccumulation of low K_{OW}, high K_{OA} compounds in an air-respiring arctic marine mammal relative to a fish species from the same area and trophic level, both of which are important subsistence diet items for the local community. While we cannot account for variation between these two species in exposure to OCs, we have taken several steps including trophic matching, lipid normalization, and PCB 153 normalization in order to eliminate potential confounding variables. Our objective is to compare the concentrations of specific OCs in muscle and liver tissue from both species and assess their relative concentrations as compared to their physical–chemical properties.

2. Methods

2.1. Species selection

Vertebrate OC concentrations are influenced by multiple factors including location, season, year, feeding ecology, tissue and trophic level. We minimized the effects of these variables by carefully selecting study species, spotted seal (*Phoca largha*) and sheefish (*Stenodus leucichthys*), that are spatially, temporally, and trophically matched. Both species are highly piscivorous, occupy similar trophic positions, and were collected in Kotzebue Sound (Alaska) during the winters of 2004–2007. In addition, these species are important subsistence resources for rural communities in northern Alaska.

2.2. Sample collection and storage

Spotted seals were sampled in October of 2004–2007 and sheefish in March 2005 at Kotzebue, AK (66.90°N, 162.59°W) under Marine Mammal Health and Stranding Response Program (MMHSRP) permit #932-1489-07. Blubber, muscle and liver samples from spotted seals (n = 18) and muscle and liver from sheefish (n = 8) were collected for chemical analyses from legally subsistence harvested animals. All animals were assessed for gross general health prior to sampling to allow for data interpretation in the context of animal condition and were deemed to be in excellent physical condition. Collection of samples was performed as previously described (Hoekstra et al., 2002). Samples were immediately frozen at −20 °C, shipped to the University of Alaska Fairbanks (UAF), and stored at −80 °C until analysis.

2.3. Morphometrics and age estimation

Spotted seal and sheefish harvest date, sex, age, and morphometric information appear in Table S1. Seal length was measured as the straight line distance from the tip of the nose to both the base and the tip of the tail. Sheefish length was measured as the straight line distance from the tip of mandible to fork of tail. Seal age was estimated by counting annual growth layers in the cementum of teeth as described by Dehn et al. (2005). Sheefish were aged by counting otolith annual growth increments as described in Brown et al. (2007). Ages were read in triplicate by each of the three independent readers.

2.4. Stable isotopes and trophic level calculations

Nitrogen (N) stable isotope signatures were determined in sheefish and spotted seal muscle and liver. Isotopes were analyzed by the Alaska Stable Isotope Facility using an Elemental Analyzer (Costech Scientific) and a Delta Plus XL Isotope Ratio Mass Spectrometer with a Conflo III interface (Thermo-Finnegan) (EA-IRMS). Each sample was analyzed in duplicate and the mean value was used for all subsequent data analysis. Data are presented in delta notation, where $\delta^{15}N = (R_{sample}\text{-}R_{standard})/(R_{standard})^{*}1000‰$ and R is the ratio of the heavier (^{15}N) to the lighter (^{14}N) isotope and the international standard for N is atmospheric nitrogen (N_{atm}). QA/QC was evaluated via the standard deviations of the reference material, peptone.

Trophic level was estimated for each species using $\delta^{15}N$ based on the following equation:

$$TL = 2 + \left[\left(\delta^{15}N_{consumer}\text{-}\delta^{15}N_{primary\text{-}consumer}\right)/3.8‰\right]$$

where, 2 is the assumed trophic level of the primary consumers of the food web, $\delta^{15}N_{primary\text{-}consumer}$ is 9.8‰ as determined for *Calanus* spp. and 3.8‰ is the trophic enrichment factor for $\delta^{15}N$ in an Arctic marine food web in the Bering Sea of Alaska (Hobson et al., 2002).

2.5. POP analysis

Organochlorines (OCs) were determined in spotted seal and sheefish tissues according to previously described methods (Dietz et al., 2004). Briefly, samples were extracted with dichloromethane (DCM) and quantified using high resolution, single-column capillary gas chromatography with electron capture detection. Standard reference materials (SRM 1588a: organics in cod liver oil) from the National Institutes of Standards and Technology (Gaithersburg, MD, USA) were used to confirm the accuracy and reproducibility of the analytical methods. A calibration check standard was run every six samples, followed by a spike or SRM for quality assurance and control (QA/QC). Method blanks were included to blank correct contaminant concentrations and calculate method detection limits (MDL). The MDL for OCs in tissues varied depending upon analyte and sample size, ranging from

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0.001 to 14 ng/g wet weight (ww). A full list of OCs examined in this study is available in the Supplemental Materials.

2.6. Assignment of K_{OW} and K_{OA} values to OCs

Physical–chemical values for the OCs analyzed in this study were drawn from several sources. When available, log K_{OW} and K_{OA} values were extracted from published final adjusted values (FAVs) of K_{OW} and K_{OA} at 25 °C (Beyer et al., 2002; Li et al., 2003; Shen and Wania, 2005). Log K_{OW} and K_{OA} values for PCB congeners which were not specifically analyzed in previous literature were calculated using the equation provided in Table 21 of Li et al. (2003), which is a relationship designed to predict physical–chemical properties for PCBs based on molecular mass and number of chlorine substitutions in the ortho position. Non-PCB OCs (i.e. β-HCH) were assigned log K_{OW} and K_{OA} values based on measurements reported by Mackay et al. (2006) by determining the median of all reported values at 25 °C. To assign a single K_{OW}/K_{OA} value in the case of homologous pairs (i.e., co-eluting pairs with the same degree of chlorination), the reported K_{OW}/K_{OA} values of both compounds were combined and an overall median taken. When co-eluting pairs were non-homologous, any congener present that was more highly chlorinated with 2,3,4,5-, 2,4,5- or 2,3,4-substitution was assumed to be the predominate congener and the median K_{OW}/K_{OA} value for that congener was used to represent the pair. For the remaining co-eluting pairs, the congener least likely to undergo cytochrome P450 biotransformation, as classified by Boon et al. (1994), was assumed to be the predominate congener and the median K_{OW}/K_{OA} value for this compound was used to represent the pair.

2.7. PCB 153 normalization

Concentrations of OCs were normalized to PCB 153 prior to comparisons between species as [OC]/[CB153] (Muir et al., 1988). PCB 153 is a common congener in a number of industrial mixtures, is highly bioaccumulative, and is not readily metabolized in model organisms (Committee on Remediation of PCB-Contaminated Sediments, 2001). This method for normalization relies on several assumptions – one of which is that PCB 153 is not metabolized in these species. Different organisms have been shown to have different capacities to metabolize PCB congeners, and without extensive research into PCB metabolism in either sheefish or spotted seal we cannot rule out the possibility that they have different capacities to metabolize PCB 153 (Tanabe et al., 1988). However normalization to PCB 153 allows us to compare differences in concentrations of OCs between two species across ranges of physical–chemical properties.

2.7.1. Statistical analysis

Statistically significant ($p < 0.05$) differences in trophic levels calculated from $\delta^{15}N$ values of spotted seal and sheefish muscle and liver were detected via single-factor analysis of variance (ANOVA). The significance of regressions (i.e., whether slopes were >0) was determined via a 1-tailed F-test ($p < 0.05$). These statistical analyses were carried out using R Programming Language (R Core Team, Version 3.1.2).

3. Results/discussion

3.1. Trophic level

Spotted seals had a slightly higher trophic level (4.0/4.2, as calculated from $\delta^{15}N$ values in muscle and liver, respectively) than sheefish (3.7/3.6). Although the $\delta^{15}N$ values were significantly different between species, a difference of 0.3–0.6‰ in $\delta^{15}N$ values likely does not represent a substantial difference in trophic feeding ecology. This does not imply that these two species are consuming the same prey items, however trophic similarity as well as spatial and temporal similarity allows for a tissue matched comparison of OC dynamics in two upper trophic level organisms with vastly different respiratory physiology.

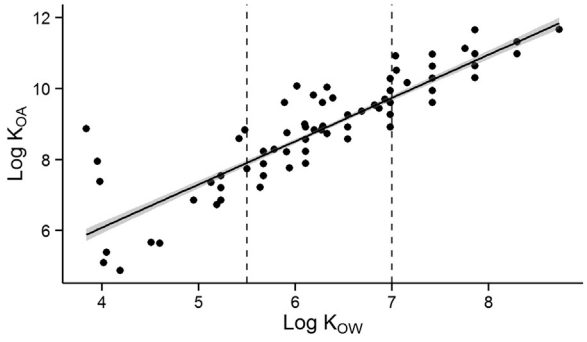

Fig. 1. Relationship between K_{OW} and K_{OA} for organochlorines (OCs) quantified in spotted seal and sheefish tissues. The dashed lines at log $K_{OW} = 5.5$ and log $K_{OW} = 7$ show the boundaries of Low, Moderate, and High K_{OW} compounds considered in this study.

3.2. Concentrations of ΣOCs

Log K_{OW} of the compounds analyzed ranged from 3.84 for γ-hexachlorocyclohexane (γ-HCH) to 8.73 for PCB 209 and log K_{OA} from 4.87 for 1,3,5-trichlorobenzene (1,3,5-TCB) to 11.66 for PCB 209. As expected based on the relationship between K_{OW} and K_{OA} (Henry's Law), there was a significant positive correlation between log K_{OW} and log K_{OA} ($p < 0.001$, Fig. 1), but the relationship weakens as K_{OW} and K_{OA} decrease, with $R^2 < 0.15$ for compounds with Kow < 5.5.

OCs were measured in muscle and liver of both species, and in seal blubber. Concentrations (Tables S2 and S3) were within previously reported ranges for similar arctic fish and marine mammals (AMAP, 2004; Gobas et al., 1989). Seal and fish had similar ΣOC concentrations (sum of 108 analytes) in matched tissues. Mean ΣOC levels were 12.3 (±8.8) ng/g (wet weight, ww) in fish muscle, 15.7 (±9.7) ng/g in seal muscle, 15.7 (±8.8) ng/g in fish liver, and 31.2 (±16.6) ng/g in seal liver. In seal blubber (no analogous tissue in fish) the mean ΣOC concentration was 837 (±245) ng/g.

3.3. Relationship of OC concentrations to K_{OW} and K_{OA}

The effects of K_{OW} and K_{OA} on PCB 153 normalized concentrations were assessed independently over three K_{OW} ranges, low (log $K_{OW} \leq 5.5$), moderate ($5.5 < \log K_{OW} \leq 7$) and high ($\log K_{OW} > 7$). For compounds with $K_{OW} < 5.5$, K_{OW} and K_{OA} are not correlated, allowing for an independent assessment of the effects of each partition coefficient on contaminant concentrations in spotted seals and sheefish tissues.

3.4. High K_{OW} compounds

When a chemical's log K_{OW} exceeds ~7, absorption efficiency from food is reduced due to increased molecular size and decreased diffusion across the gastrointestinal tract (Fisk et al., 1998; Gobas et al., 1989). Thus, these compounds should exhibit reduced dietary uptake (absorption) and increased concentration in feces. K_{OW} and K_{OA} are highly correlated for the high K_{OW} chemicals ($\log K_{OW} > 7$, $n = 23$). For these compounds, concentrations (ww, PCB 153 normalized) displayed a decreasing trend in both sheefish and spotted seal tissue with increasing K_{OW}, although the relationship was only significant for sheefish liver ($p = 0.04$). No relationships were significant between concentration (ww, PCB 153 normalized) and log K_{OA} for this subset of compounds. PCB 209 was determined to be a statistical outlier and removed from these analyses due to the potential for disproportionate leveraging of models.

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3.5. Moderate K_{OW} compounds

It was anticipated that concentrations of moderate K_{OW} compounds ($5.5 < \log K_{OW} \leq 7$; $n = 68$) would increase with K_{OW} in fish and K_{OA} in seals. Since these variables are highly correlated in this range, we expected similar relationships between OC concentration and both K_{OW} and K_{OA} in spotted seals and sheefish. However, we observed no statistically significant relationships between K_{OW} nor K_{OA} and concentrations (ww, PCB 153 normalized) across this range of compounds.

To test if biotransformation could potentially explain the observed lack of relationship between K_{OW} and concentration in this region, the analysis was repeated with only polychlorinated biphenyls (PCBS) classified as non-metabolizable by cytochrome P450 enzymes (Boon et al, 1994). Correlations were actually weaker for this subset of compounds than for the entire suite of compounds, suggesting that complex interactions and/or factors other than K_{OW}, K_{OA}, and biotransformation are major drivers determining chemical concentration profiles in these species.

3.6. Low K_{OW} compounds

No significant correlation was found in any tissue between OC concentration (ww, PCB 153 normalized) and log K_{OW} for the subset of low K_{OW} compounds ($n = 16$). Fish efficiently excrete these compounds to the aqueous environment across the gills, therefore they do not bioaccumulate (Gobas et al., 1989; Thomann, 1989). Although these compounds may accumulate in seals, since respiratory elimination in seals is to air rather than water, concentration should not be correlated with K_{OW}, but rather K_{OA}. It is important to note that both fish and seals utilize biliary and urinary routes of elimination, and since we did not measure concentrations in urine or feces here we cannot rule out the possibility that differences in relative concentrations of OCs related to K_{OW}/K_{OA} could be due to differences in kidney and liver physiology.

We observed a significant ($p < 0.05$) positive correlation between log K_{OA} and both OC concentration and ratios of OC/PCB153 in all three seal tissues for the low K_{OW} OCs (Fig. 2, blubber data not shown). This strongly supports theoretical models and empirical data described by Gobas et al. (2003) and Kelly and Gobas (2003) that air-breathing organisms bioaccumulate compounds with a log $K_{OW} < 5$ if K_{OA} is sufficiently high and biotransformation is relatively low. Bioaccumulation of low K_{OW} compounds, such as α-HCH, endosulfan, and chlorobenzenes, has been reported in arctic marine mammals and seabirds, however these compounds are less frequently reported or present at lower levels in aquatic organisms (Hoferkamp et al., 2010; Vorkamp et al., 2004; Weber et al., 2010).

There was also a significant positive relationship between log K_{OA} and concentration (wet weight and lipid adjusted) in the liver, but not muscle, of sheefish, however the slope of the relationship was significantly lower than matched relationships in spotted seal. Low K_{OW}, high K_{OA} compounds (e.g. dieldrin, heptachlor epoxide, and β-HCH) were at concentrations 2–8 time higher (ww) in seal than fish for matched tissues and 5–14 times higher on a lipid weight basis. Age associated differences are unlikely since these fish are older than the seals in this study (Table S1).

We recognize that the relationship between K_{OA} and normalized OC concentration in spotted seal appears to be largely driven by the three compounds with log $K_{OA} > 8$; β-HCH (log $K_{OW} = 3.84$, log $K_{OA} = 8.87$), dieldrin (log $K_{OW} = 5.48$, log $K_{OA} = 7.37$), heptachlor epoxide (log $K_{OW} = 5.42$, log $K_{OA} = 8.59$). Unfortunately we did not analyze for any other compounds that fit the low K_{OW} and high K_{OA} criteria. However, based on empirical evidence and theoretical models of bioaccumulation, we are confident that our results are consistent with the

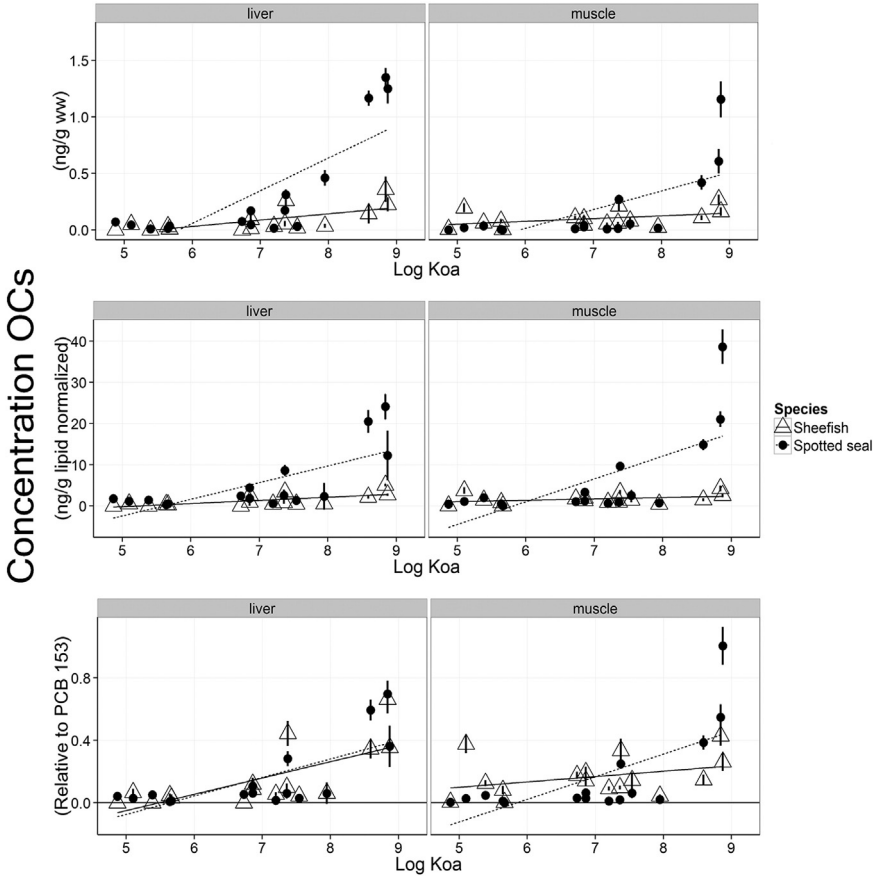

Fig. 2. Organochlorine concentration (wet weight, lipid weight and relative to PCB 153) versus log K_{OA} in sheefish (●, dashed line) and spotted seal (Δ, solid line) tissues. Data represent geometric means ± standard error. The slope of the regression line is significantly >0 ($p < 0.05$) in all cases for seal muscle and liver and sheefish liver.

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prediction that air-breathing organisms will accumulate greater concentrations of these compounds than trophically matched water-respiring organisms in the same region.

These findings support the idea that dynamics associated with octanol–air partitioning should be considered in air breathing organisms, including marine mammals. Predators of arctic marine mammals, including polar bears and humans, may be especially sensitive to bioaccumulation of high K_{OA} compounds. Chemical management policies that focus only on eliminating the use of compounds with a K_{OW} greater than an assigned threshold value are too simplistic and do not apply to air-breathing species. Refined models that account for lipid–air chemical partitioning behavior, such as that proposed by Kelly and Gobas (Kelly and Gobas, 2003), are clearly needed to address risks in food webs that include birds and mammals.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.marpolbul.2015.09.020.

References

AMAP, 2004. AMAP Assessment 2002: Persistent Organic Pollutants in the Arctic (Oslo, Norway).

Arnot, J.A., Gobas, F.A., 2006. A review of bioconcentration factor (BCF) and bioaccumulation factor (BAF) assessments for organic chemicals in aquatic organisms. Environ. Rev. 14, 257–297. http://dx.doi.org/10.1139/a06-005.

Beyer, A., Wania, F., Gouin, T., Mackay, D., Matthies, M., 2002. Selecting internally consistent physicochemical properties of organic compounds. Environ. Toxicol. Chem. 21, 941–953. http://dx.doi.org/10.1002/etc.5620210508.

Boon, J.P., Oostingh, I., van der Meer, J., Hillebrand, M.T.J., 1994. A model for the bioaccumulation of chlorobiphenyl congeners in marine mammals. Eur. J. Pharmacol. Environ. Toxicol. Pharmacol. 270, 237–251. http://dx.doi.org/10.1016/0926-6917(94)90068-X.

Brown, R.J., Bickford, N., Severin, K., 2007. Otolith trace element chemistry as an indicator of anadromy in Yukon River drainage coregonine fishes. Trans. Am. Fish. Soc. 136, 678–690. http://dx.doi.org/10.1577/T06-040.1.

Burkow, I.C., Kallenborn, R., 2000. Sources and transport of persistent pollutants to the Arctic. Toxicol. Lett. 112–113, 87–92. http://dx.doi.org/10.1016/S0378-4274(99)00254-4.

Committee on Remediation of PCB-Contaminated Sediments, 2001. A Risk Management Strategy for PCB-Contaminated Sediments. National Academic Press.

Czub, G., Wania, F., McLachlan, M.S., 2008. Combining long-range transport and bioaccumulation considerations to identify potential arctic contaminants. Environ. Sci. Technol. 42, 3704–3709. http://dx.doi.org/10.1021/es7028679.

Dallaire, R., Dewailly, É., Ayotte, P., Forget-Dubois, N., Jacobson, S.W., Jacobson, J.L., Muckle, G., 2013. Exposure to organochlorines and mercury through fish and marine mammal consumption: associations with growth and duration of gestation among Inuit newborns. Environ. Int. 54, 85–91. http://dx.doi.org/10.1016/j.envint.2013.01.013.

Dehn, L.-A., Sheffield, G.G., Follmann, E.H., Duffy, L.K., Thomas, D.L., Bratton, G.R., Taylor, R.J., O'Hara, T.M., 2005. Trace elements in tissues of phocid seals harvested in the Alaskan and Canadian Arctic: influence of age and feeding ecology. Can. J. Zool. 83, 726–746. http://dx.doi.org/10.1139/z05-053.

Dietz, R., Riget, F.F., Sonne, C., Letcher, R., Born, E.W., Muir, D.C.G., 2004. Seasonal and temporal trends in polychlorinated biphenyls and organochlorine pesticides in East Greenland polar bears (*Ursus maritimus*), 1990–2001. Sci. Total Environ. 331, 107–124. http://dx.doi.org/10.1016/j.scitotenv.2004.03.025.

Environment Canada, 1999. Canadian Environmental Protection Act. Environment Canada.

Environmental Protection Agency, U.S., 1999. Category for Persistent, Bioaccumulative, and Toxic New Chemical Substances (No. Publication 64 FR 60194). US EPA.

Fisk, A.T., Norstrom, R.J., Cymbalisty, C.D., Muir, D.C.G., 1998. Dietary accumulation and depuration of hydrophobic organochlorines: bioaccumulation parameters and their relationship with the octanol/water partition coefficient. Environ. Toxicol. Chem. 17, 951–961. http://dx.doi.org/10.1002/etc.5620170526.

Fisk, A.T., Hobson, K.A., Norstrom, R.J., 2001. Influence of chemical and biological factors on trophic transfer of persistent organic pollutants in the Northwater Polynya marine food web. Environ. Sci. Technol. 35, 732–738. http://dx.doi.org/10.1021/es001459w.

Gobas, F.A.P.C., Clark, K.E., Shiu, W.Y., Mackay, D., 1989. Bioconcentration of polybrominated benzenes and biphenyls and related superhydrophobic chemicals in fish: role of bioavailability and elimination into the feces. Environ. Toxicol. Chem. 8, 231–245. http://dx.doi.org/10.1002/etc.5620080306.

Gobas, F.A.P.C., Kelly, B.C., Arnot, J.A., 2003. Quantitative structure activity relationships for predicting the bioaccumulation of POPs in terrestrial food-webs. QSAR Comb. Sci. 22, 329–336. http://dx.doi.org/10.1002/qsar.200390022.

Gossett, R.W., Brown, D.A., Young, D.R., 1983. Predicting the bioaccumulation of organic compounds in marine organisms using octanol/water partition coefficients. Mar. Pollut. Bull. 14, 387–392. http://dx.doi.org/10.1016/0025-326X(83)90604-5.

Hobson, K.A., Fisk, A., Karnovsky, N., Holst, M., Gagnon, J.-M., Fortier, M., 2002. A stable isotope ($\delta^{13}C$, $\delta^{15}N$) model for the North Water food web: implications for evaluating trophodynamics and the flow of energy and contaminants. The International North Water Polynya Study. Deep Sea Res. Part II Top. Stud. Oceanogr. 49, pp. 5131–5150. http://dx.doi.org/10.1016/S0967-0645(02)00182-0.

Hoekstra, P.F., O'Hara, T.M., Pallant, S.J., Solomon, K.R., Muir, D.C.G., 2002. Bioaccumulation of organochlorine contaminants in Bowhead Whales (*Balaena mysticetus*) from Barrow, Alaska. Arch. Environ. Contam. Toxicol. 42, 497–507. http://dx.doi.org/10.1007/s00244-001-0046-x.

Hoferkamp, L., Hermanson, M.H., Muir, D.C.G., 2010. Current use pesticides in Arctic media; 2000–2007. Sci. Total Environ. 408, 2985–2994. http://dx.doi.org/10.1016/j.scitotenv.2009.11.038.

Johnson, J.S., Nobmann, E.D., Asay, E., Lanier, A.P., 2009. Dietary intake of Alaska Native people in two regions and implications for health: the Alaska Native Dietary and Subsistence Food Assessment Project. Int. J. Circumpolar Health 68. http://dx.doi.org/10.3402/ijch.v68i2.18320.

Kelly, B.C., Gobas, F.A.P.C., 2001. Bioaccumulation of persistent organic pollutants in Lichen–Caribou–Wolf food chains of Canada's Central and Western Arctic. Environ. Sci. Technol. 35, 325–334. http://dx.doi.org/10.1021/es0011966.

Kelly, B.C., Gobas, F.A.P.C., 2003. An Arctic Terrestrial Food-Chain Bioaccumulation Model for Persistent Organic Pollutants. 37, 2966–2974.

Kelly, B.C., Ikonomou, M.G., Blair, J.D., Morin, A.E., Gobas, F.A.P.C., 2007. Food web-specific biomagnification of persistent organic pollutants. Science 317, 236–239. http://dx.doi.org/10.1126/science.1138275.

Laird, B.D., Goncharov, A.B., Chan, H.M., 2013. Body burden of metals and persistent organic pollutants among Inuit in the Canadian Arctic. Environ. Int. 59, 33–40. http://dx.doi.org/10.1016/j.envint.2013.05.010.

Letcher, R.J., Bustnes, J.O., Dietz, R., Jenssen, B.M., Jørgensen, E.H., Sonne, C., Verreault, J., Vijayan, M.M., Gabrielsen, G.W., 2010. Exposure and effects assessment of persistent organohalogen contaminants in arctic wildlife and fish. Levels, trends and effects of legacy and new persistent organic pollutants in the Arctic: an AMAP assessment. Sci. Total Environ. 408, pp. 2995–3043. http://dx.doi.org/10.1016/j.scitotenv.2009.10.038.

Li, N., Wania, F., Lei, Y.D., Daly, G.L., 2003. A comprehensive and critical compilation, evaluation, and selection of physical–chemical property data for selected polychlorinated biphenyls. J. Phys. Chem. Ref. Data 32, 1545–1590. http://dx.doi.org/10.1063/1.1562632.

Ma, J., Hung, H., Tian, C., Kallenborn, R., 2011. Revolatilization of persistent organic pollutants in the Arctic induced by climate change. Nat. Clim. Chang. 1, 255–260. http://dx.doi.org/10.1038/nclimate1167.

Mackay, D., Shiu, W.-Y., Ma, K.-C., Lee, S.C., 2006. Handbook of Physical–Chemical Properties and Environmental Fate for Organic Chemicals. Second edition. CRC Press.

Meylan, W.M., Howard, P.H., Boethling, R.S., Aronson, D., Printup, H., Gouchie, S., 1999. Improved method for estimating bioconcentration/bioaccumulation factor from octanol/water partition coefficient. Environ. Toxicol. Chem. 18, 664–672. http://dx.doi.org/10.1002/etc.5620180412.

Moore, C.W., Obrist, D., Steffen, A., Staebler, R.M., Douglas, T.A., Richter, A., Nghiem, S.V., 2014. Convective forcing of mercury and ozone in the Arctic boundary layer induced by leads in sea ice. Nature 506, 81–84. http://dx.doi.org/10.1038/nature12924.

Muir, D.C.G., de Wit, C.A., 2010. Trends of legacy and new persistent organic pollutants in the circumpolar arctic: overview, conclusions, and recommendations. Levels, trends and effects of legacy and new persistent organic pollutants in the Arctic: an AMAP Assessment. Sci. Total Environ. 408, pp. 3044–3051. http://dx.doi.org/10.1016/j.scitotenv.2009.11.032.

Muir, D.C.G., Norstrom, R.J., Simon, M., 1988. Organochlorine contaminants in arctic marine food chains: accumulation of specific polychlorinated biphenyls and chlordane-related compounds. Environ. Sci. Technol. 22, 1071–1079. http://dx.doi.org/10.1021/es00174a012.

Muir, D., Braune, B., DeMarch, B., Norstrom, R., Wagemann, R., Lockhart, L., Hargrave, B., Bright, D., Addison, R., Payne, J., Reimer, K., 1999. Spatial and temporal trends and effects of contaminants in the Canadian Arctic marine ecosystem: a review. Sci. Total Environ. 230, 83–144. http://dx.doi.org/10.1016/S0048-9697(99)00037-6.

Russell, R.W., Gobas, F.A.P.C., Haffner, G.D., 1999. Role of chemical and ecological factors in trophic transfer of organic chemicals in aquatic food webs. Environ. Toxicol. Chem. 18, 1250–1257. http://dx.doi.org/10.1002/etc.5620180626.

Shen, L., Wania, F., 2005. Compilation, evaluation, and selection of physical–chemical property data for organochlorine pesticides. J. Chem. Eng. Data 742–768.

Tanabe, S., Watanabe, S., Kan, H., Tatsukawa, R., 1988. Capacity and mode of PCB metabolism in small cetaceans. Mar. Mammal Sci. 4, 103–124. http://dx.doi.org/10.1111/j.1748-7692.1988.tb00191.x.

Thomann, R.V., 1989. Bioaccumulation model of organic chemical distribution in aquatic food chains. Environ. Sci. Technol. 23, 699–707. http://dx.doi.org/10.1021/es00064a008.

Van Oostdam, J., Donaldson, S.G., Feeley, M., Arnold, D., Ayotte, P., Bondy, G., Chan, L., Dewaily, É., Furgal, C.M., Kuhnlein, H., Loring, E., Muckle, G., Myles, E., Receveur, O., Tracy, B., Gill, U., Kalhok, S., 2005. Human health implications of environmental contaminants in Arctic Canada: a review. Contaminants in Canadian Arctic Biota and Implications for Human Health. Sci. Total Environ. 351–352, pp. 165–246. http://dx.doi.org/10.1016/j.scitotenv.2005.03.034.

Vorkamp, K., Christensen, J.H., Riget, F., 2004. Polybrominated diphenyl ethers and organochlorine compounds in biota from the marine environment of East Greenland. Sci. Total Environ. 331, 143–155. http://dx.doi.org/10.1016/j.scitotenv.2004.03.026.

Wania, F., MacKay, D., 1996. Tracking the distribution of persistent organic pollutants. Environ. Sci. Technol. 30, 390–396. http://dx.doi.org/10.1021/es962399q.

Weber, J., Halsall, C.J., Muir, D., Teixeira, C., Small, J., Solomon, K., Hermanson, M., Hung, H., Bidleman, T., 2010. Endosulfan, a global pesticide: a review of its fate in the environment and occurrence in the Arctic. Sci. Total Environ. 408, 2966–2984. http://dx.doi.org/10.1016/j.scitotenv.2009.10.077.

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