Electronic Supplementary Information (ESI)

A multi-pathway exposure assessment for polycyclic aromatic hydrocarbons among residents in the Athabasca oil sands region, Canada

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S1. Overview of New Organism Bioaccumulation Models

To mechanistically describe polycyclic aromatic hydrocarbon (PAH) bioaccumulation in indigenous Canadians residing in the Alberta oil sands region (AOSR), it was necessary to add additional species to the ACC-Human model. As per traditional food (TF) intake survey data from AOSR Indigenous Canadians (Chan et al. 2016), models representing whitefish (*Coregonus clupeaformis*), Northern pike (*Esox lucius*), moose (*Alces alces*), ruffed grouse (*Bonasa umbellus*), mallard duck (*Anas platyrhynchos*), Saskatoon berry (*Amelanchier alnifolia*) and mint (*Mentha spp.*), were integrated into the existing modelling framework, generating ACC-Human AOSR. These new species models required parameterization, namely body mass and lipid mass growth, feeding and egestion, respiration, birthing/egg-laying, and lactation (for moose only) for animals and root/leaf/fruit growth, transpiration, chemical deposition (gas and particle phases), and internal chemical transport and distribution for plants. For all species biotransformation rates were also included. Because the availability of the relevant information in the literature differed for each organism, parameterization schemes are presented separately for each species below. Table S1 through S4 contain equations for the calculation of critical rate parameters, fugacity capacities and *D*-values in the new organism models. Table S5 lists the fugacity calculations, and Table S6 describes the mass balance equations for each species. Following the tables, and a brief description of shared parameter approaches (absorption efficiency and biotransformation), are detailed explanations of equations for each new organism model (moose - S.4.1, ruffed grouse and mallard duck - S.4.2).

Model parameterizations for new fish species only required straightforward scaling of mechanistic functions previously developed for polar cod (Czub et al. 2008) according to differences in body mass, life span, lipid content, etc. Therefore, the different parameters for whitefish and Northern pike are simply compiled in Table S7.

Similarly, contaminant uptake, distribution, metabolism, and excretion in plants was modeled according to a previous framework developed by Undeman et al. (2009). Briefly, this model is fugacity-based and non-steady state, predicting plant bioaccumulation in 3 compartments: leaf, root, fruit. Leaves/fruit exchange chemicals via diffusion in air, uptake pollutants via dry and wet particle deposition, and also receive chemicals from soil pore water via their roots (Undeman et al. 2009). The key physical chemical properties that govern a compound's bioaccumulation in plants are its octanol-air (K_{OA}) and air-water (K_{AW}) partition coefficients. Similar to fish, this model only required simple scaling of critical parameters (ex. growth period, transpiration rate, leaf area index, etc.) for willow, cattail, lodgepole pine, Saskatoon berry, and mint; these variables are listed in Table S8.

S2. Chemical-Dependent Absorption Efficiency and Biotransformation in New Organism Models

S2.1 Absorption Efficiency. Absorption efficiency (E_0) is a dimensionless coefficient that describes the fraction of ingested chemical absorbed in the gastrointestinal tract. E_0 is dependent on K_{OW} as described in Table S2, and takes a numerical value from 0-1 (Czub and McLachlan 2004). Food was assumed to adopt the core body temperatures of moose, ruffed grouse, and mallard duck (37.0 °C) during absorption.

S2.2 Biotransformation. Considerable biotransformation of PAHs has been suggested in many of the species included in the model, including mallard (Honey et al. 2000), whitefish and northern pike (Ohiozebau et al. 2016), and humans (Viau et al. 1995). Because literature data on PAH biotransformation rate constants and enzyme induction pathways among these species are lacking, we assumed biotransformation to occur with first order kinetics.

For humans, we compared predictions of human biotransformation rate constants using four methods: 1) a model, Iterative Fragment Selection (IFS) (Arnot et al. 2014), 2) a second model, QSARINS (Papa et al. 2018), and 3a) allometric scaling based on reported biotransformation rate constants in rainbow trout (Arnot et al. 2008; Niimi and Palazzo 1986), and 3b) total elimination rate constants in rats (Withey et al. 1991; Ramesh et al. 2001; Moir et al. 1998; Moreau et al. 2015). The equation used for allometric scaling was as follows:

$$k_{\text{human}} = k_{\text{org}} \left(m_{\text{human}}/m_{\text{org}}\right)^{-0.25} \quad (16)$$

where k_{human} represents either the biotransformation or total elimination rate constant of humans (h^{-1}), k_{org} represents either the biotransformation rate constant of rainbow trout or total elimination rate constant of rat (h^{-1}), m_{human} represents total human body mass at physical maturity (male - 68 kg, female - 60 kg), and m_{org} represents mean body mass of rainbow trout (0.70 kg – Fishbase, same as average fish weight in Niimi and Palazzo 1986) or rat (0.25 kg - Withey et al. 1991; Ramesh et al. 2001; Moir et al. 1998; Moreau et al. 2015) at physical maturity. The rate constant predictions are presented in Table S1.

Table S1. Human biotransformation rate constant of three target PAHs obtained by different approaches.

	IFS (h^{-1}) 95% C.I.	QSARINS (h^{-1}) 95% C.I.	Scaling from Fish (h^{-1})	Scaling from Rat (h^{-1})
PHE	0.007–3.5	0.004–1.7	0.0007	No data
PYR	Out of domain	0.005–1.7	0.002	<0.02*
BaP	Out of domain	0.003–1.0	0.004	<0.01*

* These values are calculated by allometric scaling from the total elimination rate constant of rat instead of only biotransformation rate constants, so the biotransformation rate constants would be lower than these values.

IFS and QSARINS show large uncertainties in predicting the biotransformation rate constants of the target PAHs in humans, with 95% confidence intervals that range over three orders of magnitude. As shown in the table above, the allometrically scaled biotransformation rate constants for pyrene and benzo[a]pyrene based on rainbow trout are close to the lower 95% confidence limit predicted by QSARINS, but the rate constant for phenanthrene is one order of magnitude lower than the lower 95% confidence limits of the IFS and QSARINS predictions. The total elimination rate constants for pyrene and benzo[a]pyrene from allometric scaling based on rats are slightly higher than the lower 95% confidence limits for biotransformation rate constants predicted by QSARINS. Since the experimental rat data are total elimination rate constants, the biotransformation rate constants would likely be lower, and closer to the QSARINS lower 95% confidence limits. Therefore, the lower 95% confidence limits of the QSARINS predictions are used in this study.

Intrinsic biotransformation rate constants for each modeled PAH in whitefish, Northern pike, moose, ruffed grouse, and mallard duck were estimated using the allometric scaling method described above for humans, based on reported data for rainbow trout (no other measured data or prediction values available for these animals). In Eq. 16, k_{human} is replaced by the biotransformation rate constant of moose, whitefish, Northern pike, ruffed grouse, or mallard duck (h^{-1}), k_{org} is the reported biotransformation rate constant of rainbow trout (h^{-1}), m_{human} is replaced by the total body mass at physical maturity of moose (male - 360 kg, female 320 kg), whitefish (1.58 kg), Northern pike (2.65 kg), ruffed grouse (male - 0.50 kg, female - 0.48 kg), or mallard duck (male - 0.94 kg, female - 0.88 kg), and m_{org} represents mean body mass at rainbow trout physical maturity (0.70 kg – Fishbase, same as average fish weight in Niimi and Palazzo 1986). Note that all plants were assumed to biotransform PAHs at equivalent rates based on analysis of ryegrass (Gao et al. 2006). Whole-body elimination rate constants for each species and PAH congener combination are listed in Table S4.

S3. Model Parameterization Tables

Table S2. Derivation of transport parameters in the added organism modules.

Parameter		Units	Value	Ref.
Ingestion	G_U	[$m^3\ h^{-1}$]	$G_{Uwfish} = \vartheta_{wzoo} \cdot G_{Uwzoo}$ where ϑ_{wzoo} = diet proportion zooplankton, G_{Uwzoo} = uptake rate zooplankton. ϑ_{wzoo} and G_{Uwzoo} both varied with fish age and season.	1, 2, 3, 4, 5, 6, 7, 8,
			$G_{Upike} = \vartheta_{pzoo} \cdot G_{Upzoo} + \vartheta_{pwfish} \cdot G_{Upwfish} + \vartheta_{ppike} \cdot G_{Uppike} + \vartheta_{pbent} \cdot G_{Upbent}$ where ϑ_{pzoo} = diet proportion zooplankton, G_{Upzoo} = uptake rate zooplankton, ϑ_{pwfish} = diet proportion whitefish, and $G_{Upwfish}$ = uptake rate whitefish, ϑ_{ppike} = diet proportion juvenile pike, and G_{Uppike} = uptake rate juvenile pike, $\vartheta_{pbenthos}$ = diet proportion benthos, and $G_{Upbenthos}$ = uptake rate benthos. ϑ_{wzoo}, G_{Uwzoo}, ϑ_{pwfish}, $G_{Upwfish}$, ϑ_{ppike}, and G_{Uppike}, $\vartheta_{pbenthos}$, and $G_{Upbenthos}$ all varied with fish age and season.	
			$G_{Umoose} = \left(0.8 \cdot m_{\text{moose}}(t)^{0.75} / \frac{24\ h}{d}\right) / DN_{willow} + G_{Umoose,soil}$ where DN_{willow} = willow leaf density ($g \cdot m^{-3}$) and $G_{Umoose,soil} = SC_{moose} \cdot G_{Udw,moose} / DN_{soil}$ SC_{moose} = soil content in moose diet (g soil·g $diet^{-1}$) and DN_{soil} = soil density ($g \cdot m^{-3}$).	
			$G_{Ugrouse} = \left(ER_{grouse} / \frac{24\ h}{d}\right) / \left(ED_{lp} / DN_{lpdw}\right) + G_{Ugrouse,soil}$ where ER_{grouse} = grouse daily energy requirement ($kJ \cdot d^{-1}$), ED_{lp} = lodgepole pine energy density ($kJ \cdot g^{-1}$), DN_{gdw} = density of lodgepole pine ($g \cdot m^{-3}$) and $G_{Ugrouse,soil} = SC_{grouse} \cdot G_{Udw,grouse} / DN_{soil}$.	
			$G_{Umall} = \left(ER_{mall} / \frac{24\ h}{d}\right) / (ED_{mall} / DN_{mall}) + G_{Umall,soil}$	

			where ER_{mall} = mallard daily energy requirement (kJ·d^{-1}), ED_{mall} = mallard diet energy density (kJ·g^{-1}), DN_{gdw} = density of mallard diet (g·m^{-3}). Mallard diet density and energy density were calculated by summing the energy contents and densities of all mallard diet components (cattail, zooplankton) according to their dietary proportions (0.9, 0.1, respectively) and $G_{Umall,soil} = SC_{mall} \cdot G_{Udw,mall}/DN_{soil}$.	
Dry Wt. Ingestion	G_{Udw}	[g dw h^{-1}]	Calculated from moose, grouse, and mallard wet weight ingestion, assuming water contents of 0.80 in vegetation, zooplankton, and benthos.	1, 4, 9
Water Uptake / Urination	G_{UW} / G_{ur}	[m^3 h^{-1}]	Full Size: $G_{Umoose} = 0.0013$ $G_{Ugrouse} = 2.12 \cdot 10^{-6}$ $G_{Umall} = 2.73 \cdot 10^{-6}$ Juvenile: Rate increases to maximum following trend in body weight increase.	1, 4, 9, 10, 11
Respiration	G_R	[m^3 h^{-1}]	$G_{Rwfish} = m_{\mathrm{wfish}}(t) \cdot \frac{1.39 \cdot 10^{-8}}{m_{\mathrm{wfish}}(t)^{0.9} \cdot \left(\frac{1}{520} + \frac{0.1}{K_{OW}}\right)}$ $G_{Rpike} = m_{\mathrm{pike}}(t) \cdot \frac{1.39 \cdot 10^{-8}}{m_{\mathrm{pike}}(t)^{0.9} \cdot \left(\frac{1}{520} + \frac{0.1}{K_{OW}}\right)}$ $G_{Rmoose} = 0.0864 \cdot V_{\mathrm{moose}}(t)^{0.75}$	1, 3, 7, 10, 12
			Full Size: $G_{Rgrouse} = 0.014$ $G_{Rmall} = 0.096$ Juvenile: Rate increases to maximum following trend in body weight increase.	
Feces Dry Wt. Egestion	G_{FBdw}	[g dw h^{-1}]	$G_{FBdw} = 0.1 \cdot G_{Udw}$ (moose, grouse, mallard)	13
Birthing	G_{calf}	[m^3 h^{-1}]	Moose: $G_{calf} = \left(Vol_{calf\ lipid} + Vol_{calf\ water}\right)/dt$ Grouse, Mallard: $G_{egg} = n_{egg} \cdot \left(Vol_{egg\ lipid} + Vol_{egg\ water}\right)/dt$ where n_{egg} = number of eggs per clutch assumed for grouse (n_{egg} = 10) and mallard (n_{egg} = 9).	2, 4, 14, 15
Lactation	G_{lac}	[m^3 h^{-1}]	$G_{lac} = \frac{3000\ g}{d} \cdot \frac{m^3}{1.0 \cdot 10^6 g} \cdot \frac{d}{24\ h}$	1, 4, 16
Feces-Blood Partition Coefficient	K_{FB}	[m^3 blood lipid / g dry feces]	$8 \cdot 10^{-8}$ (moose, grouse, mallard)	1

Absorption Efficiency	E_0	$\frac{1}{E_0} = 1.9 + 5.6 \cdot 10^{-9} \cdot K_{OW}$ (whitefish, pike) $\frac{1}{E_0} = 1.295 + 3.226 \cdot 10^{-8} \cdot K_{OW}$ (moose) $\frac{1}{E_0} = 1.04 + 2.4 \cdot 10^{-9} \cdot K_{OW}$ (grouse, mallard)	1, 17, 18

References: 1) Czub and McLachlan 2004 2) Fishbase - *Coregonus clupeaformis* 3) Fishbase - *Esox lucius* 4) Binnington et al. 2016 5) Shipley 2010 6) Norstrom et al. 2007 7) Pendergrast and Boag 1970 8) Intrinsik Corp. 2010 9) Belovsky and Jordan 1981 10) Pekins et al. 1994a 11) Kroner and Cozzie 1999 12) Keijer and Butler 1982 13) Moser and McLachlan 2002 14) Leupin 2003 15) Caldwell and Cornwell 1975 16) Reese and Robbins 1994 17) Lo et al. 2016 18) Drouillard and Norstrom 2000

Table S3. Equations used for the calculation of fugacity capacities Z in mol·m^{-3}·Pa^{-1}.

Phase	Equation
Air	$Z_A(T) = (R \cdot T)^{-1}$ where R is the gas constant (8.314 J·mol^{-1}·K^{-1}) and T is the temperature in K.
Water	$Z_W(T) = H^{-1}(T)$ where H is the Henry's Law constant (Pa·m^{-3}·mol^{-1}).
Soil, Sediment	$Z_S(T) = vf_W \cdot Z_W(T) + vf_A \cdot Z_A(T) + (1 - vf_W - vf_A) \cdot vf_{OC} \cdot Z_{OC}(T)$ $Z_{sed}(T) = vf_{SS} \cdot vf_{OC} \cdot Z_{OC}(T) + (1 - vf_{SS}) * Z_W(T)$ where vf_W, vf_A, vf_{OC}, and vf_{SS} represent volume fractions of water, air, organic carbon, and suspended solids, and Z_{OC} is the fugacity capacity of organic carbon.
Octanol	$Z_O(T) = Z_W(T) \cdot K_{OW}(T)$ where K_{OW} is the octanol-water partition coefficient.
Cattail, Willow, Lodgepole pine, Saskatoon berry, Mint	$Z_{plant}(T) = [vf_W \cdot (1/DN_W) + vf_{lip} \cdot K_{OW}(T) \cdot (1/DN_{lip}) + vf_{pro} \cdot K_{proW} \cdot (1/DN_{pro}) + vf_A \cdot K_{AW}(T) \cdot (1/DN_A) + vf_{carb} \cdot K_{carbW} \cdot (1/DN_{carb})] \cdot DN_{plant} \cdot Z_W(T)$ where vf_W, vf_{lip}, vf_{pro}, vf_A, and vf_{carb} represent plant volume fractions of water, lipid, protein, air, and carbohydrates, respectively; DN_W, DN_{lip}, DN_{pro}, DN_A, and DN_{carb}, represent the densities of water, lipid, protein, air, and carbohydrate, respectively, and K_{proW}, K_{AW}, and K_{carbW} represent partition coefficients between protein-water, air-water, and carbohydrate-water, respectively.
Zooplankton	$Z_{zoo}(T) = Z_W(T) \cdot BAF_{zoo} \cdot (DN_{zoolip}/DN_W)$ where DN_{zoolip} and DN_W represent the densities of zooplankton lipid and water.
Benthos	$Z_{bent}(T) = \left(vf_{lip} + vf_{nlom} \cdot SC_{\frac{nlom}{lip}}\right) \cdot Z_O(T) + vf_W \cdot Z_W(T)$ where vf_{lip}, vf_{nlom}, and vf_W represent the benthos volume fractions of lipid, non-lipid organic matter, and water, respectively, and $SC_{nlom/lip}$ represents a proportionality constant expressing sorption capacity of non-lipid organic matter relative to octanol (Undeman et al. 2014).
Whitefish, Pike	$Z_{fish}(T) = vf_{lip} \cdot DN_{fish} \cdot Z_O(T)/DN_{lip}$ where vf_{lip} represents the fish volume fraction of lipid, DN_{fish} represents total fish density, and DN_{lip} represents fish lipid density.
Moose, Grouse, Mallard	$Z_{org}(T) = vf_{lip} \cdot Z_O(T) + vf_W \cdot Z_W(T)$ where vf_{lip} and vf_W represent the organism volume fractions of lipid and water.
Moose Milk	$Z_{moose\ milk}(T) = vf_{milk\ lip} \cdot DN_{milk}/DN_{milk\ lip} \cdot Z_{moose}(T) + vf_{milk\ W} \cdot DN_{milk}/DN_W \cdot Z_W(T)$ where $vf_{milk\ lip}$ and $vf_{milk\ W}$ represent the organism volume fractions of lipid and water, respectively, and DN_{milk} and $DN_{milk\ lip}$ represent the densities of moose milk and moose milk lipids, respectively.

Table S4. Equations used for the calculation of D values in mol·h^{-1}·Pa^{-1}.

Process	Equation
Food Intake	$D_{Uwfish} = G_{Uwfish} \cdot \vartheta_{wzoo} \cdot Z_{zoo}(T) \cdot E_{0wfish}$ $D_{Upike} = G_{Upike} \cdot [\vartheta_{pzoo} \cdot Z_{zoo}(T) + \vartheta_{pwfish} \cdot Z_{wfish}(T) + \vartheta_{ppike} \cdot Z_{pike}(T) + \vartheta_{pbent} \cdot Z_{bent}(T)] \cdot E_{0pike}$ $D_{Umoose} = [G_{Umoose} \cdot Z_{willow}(T) + G_{Usoil,moose} \cdot Z_{soil}(T)] \cdot E_0$ $D_{Ugrouse} = [G_{Ugrouse} \cdot Z_{pine}(T) + G_{Usoil,grouse} \cdot Z_{soil}(T)] \cdot E_0$ $D_{Umall} = \{G_{Umall} \cdot [\vartheta_{cat} \cdot Z_{cat}(T) + \vartheta_{mzoo} \cdot Z_{zoo}(T) +] + G_{Usoil,mall} \cdot Z_{soil}(T)\} \cdot E_0$
Water Intake	$D_{UW\ org} = G_{UW\ org} \cdot Z_W(T)$
Respiration	$D_{R\ org} = G_{R\ org} \cdot Z_A(T_{org})$
Egestion	$D_{E\ org} = G_{FBdw\ org} \cdot [w_{ex} Z_W(T_{org}) + K_{FB} Z_O(T_{org})]$ where w_{ex} is a correction term introduced to consider water as an additional sorbing matrix; w_{ex} was set to $1.35 \cdot 10^{-5}$ [34].
Biotransformation	$D_{M\ org} = Vol_{total\ org} \cdot Z_{org}(T_{org}) \cdot k_{org}$

	k_{wfish}	k_{pike}	k_{moose}	k_{grouse}	k_{mall}
PHE	$1.66 \cdot 10^{-3}$	$1.46 \cdot 10^{-3}$	$4.33 \cdot 10^{-4}$	$2.22 \cdot 10^{-3}$	$1.90 \cdot 10^{-3}$
PYR	$5.07 \cdot 10^{-3}$	$4.46 \cdot 10^{-3}$	$1.33 \cdot 10^{-3}$	$6.80 \cdot 10^{-3}$	$5.82 \cdot 10^{-3}$
BaP	$8.49 \cdot 10^{-3}$	$7.46 \cdot 10^{-3}$	$2.22 \cdot 10^{-3}$	$1.14 \cdot 10^{-2}$	$9.74 \cdot 10^{-3}$

Lactation	$D_{lac} = G_{lac} \cdot Z_{moose\ milk}(T_{moose})$
Birthing/ Egg-laying	$D_{calf} = G_{calf} \cdot Z_{moose}(T_{moose}) - at\ age\ 0\ h$ $D_{egg\ bird} = G_{egg\ bird} \cdot K_{ME} \cdot Z_{bird}(T_{bird}) - at\ age\ 0\ h$ where K_{ME} represents the chemical partition coefficient between mother grouse/mallard and their eggs.
Urination	$D_{ur\ org} = G_{ur\ org} \cdot Z_W(T_{org})$

Table S5. Equations used for the calculation of fugacities in Pa.

<table>
<tr><th>Process</th><th>Equation</th></tr>
<tr><td>Air,
Water,
Soil,
Sediment</td><td>$f_A(T) = C_A/(Z_A(T) \cdot MW_{PAH})$ $\quad f_S(T) = C_S/(Z_S(T) \cdot MW_{PAH})$
$f_W(T) = C_W/(Z_W(T) \cdot MW_{PAH})$ $\quad f_{sed}(T) = C_{sed}/(Z_{sed}(T) \cdot MW_{PAH})$
where C_A, C_W, C_S, C_{sed} are air, freshwater, and soil PAH concentrations in g·m^{-3}, respectively, and MW_{PAH} is the PAH molecular weight in g·mol^{-1}.</td></tr>
<tr><td>Plants</td><td>$f_{root}(T) = f_S(T) \cdot D_{xyroot} + f_{leaf}(T) \cdot D_{phroot} - f_{root}(T) \cdot (D_{xyleaf} + D_{xyfruit} + D_{transroot})$
$f_{leaf}(T) = f_{root}(T) \cdot D_{xyleaf} + f_A(T) \cdot (D_{diffleaf} + D_{part} + D_{rain}) - f_{leaf}(T) \cdot (D_{diffleaf} + D_{phfruit} + D_{phroot} + D_{transleaf})$
$f_{fruit}(T) = f_{leaf}(T) \cdot D_{phfruit} + f_A(T) \cdot D_{difffruit} + f_{root} \cdot D_{xyfruit} - f_{fruit}(T) \cdot (D_{difffruit} + D_{transfruit})$
where all D-values are defined according to Undeman et al. (2009) below:
<table>
<tr><th>D-value</th><th>Definition</th></tr>
<tr><td>D_{xyroot}</td><td>Chemical transport from soil pore water into root</td></tr>
<tr><td>D_{phroot}</td><td>Phloem transport to root</td></tr>
<tr><td>$D_{transroot}$</td><td>Biotransformation in root</td></tr>
<tr><td>D_{xyleaf}</td><td>Xylem transport to leaf</td></tr>
<tr><td>$D_{diffleaf}$</td><td>Diffusive exchange between leaf and air</td></tr>
<tr><td>D_{part}</td><td>Dry and wet particle-bound deposition</td></tr>
<tr><td>D_{rain}</td><td>Rain dissolution</td></tr>
<tr><td>$D_{transleaf}$</td><td>Biotransformation in leaf</td></tr>
<tr><td>$D_{phfruit}$</td><td>Phloem transport to fruit</td></tr>
<tr><td>$D_{difffruit}$</td><td>Diffusive exchange between fruit and air</td></tr>
<tr><td>$D_{xyfruit}$</td><td>Xylem transport to fruit</td></tr>
<tr><td>$D_{transfruit}$</td><td>Biotransformation in fruit</td></tr>
</table></td></tr>
<tr><td>Zooplankton</td><td>$f_{zoo}(T) = f_W(T)$</td></tr>
<tr><td>Benthos</td><td>$f_{bent}(T) = [\rho_{sed} \cdot f_{sed}(T) + (1 - \rho_{sed}) \cdot f_W(T)] \cdot D_{Rbent} + f_{zoo}(T) \cdot D_{Ubent} - f_{bent}(T) \cdot (D_{Rbent} + D_{Ebent} + D_{Mbent})$
where ρ_{sed} is benthos time in sediment, and all D-values are defined according to Undeman et al. (2014, personal correspondence) below:
<table>
<tr><th>D-value</th><th>Definition</th></tr>
<tr><td>$D_{R\ bent}$</td><td>Respiration</td></tr>
<tr><td>$D_{U\ bent}$</td><td>Dietary uptake</td></tr>
<tr><td>$D_{E\ bent}$</td><td>Egestion</td></tr>
<tr><td>$D_{M\ bent}$</td><td>Biotransformation</td></tr>
</table></td></tr>
</table>

Whitefish, Pike	$f_{fish}(t) = \left[M_{fish}(t-\Delta t) + \Delta M_{fish}(t)\right] / V_{fish}(t) \cdot Z_{fish}(t)$ where f_{fish} is calculated as a function of time t, and all fish-related parameters are calculated at their respective body temperatures. Note that $M_W(t\text{-}\Delta t)$ represents the PAH mass within the fish calculated at the conclusion of the previous time step, and $\Delta M_W(t)$ represents the change in PAH mass within the fish calculated during the current time step (see below): $M_{fish}(t-1) = f_{fish}(t-\Delta t) \cdot V_{fish}(t-\Delta t) \cdot Z_{fish}(t-\Delta t)$ $\Delta M_{fish}(t) = \begin{bmatrix} E_{0\,fish} \cdot D_{U\,fish} \cdot f_{U\,fish}(t-\Delta t) + D_{R\,f} \cdot f_W(t-\Delta t) - \\ \left(E_{0\,fish} \cdot D_{E\,fish} + D_V + D_{M\,fish}\right) \cdot f_{fish}(t-\Delta t) \end{bmatrix} \cdot \Delta t$
Moose, Grouse, Mallard	$f_{org}(t) = \left[M_{org\,h}(t-\Delta t) + \Delta M_{org\,h}(t)\right] / V_{org\,h}(t) \cdot Z_{org\,h}(t)$ where similar to fish above, f_{org} is calculated as a function of time t at respective organism core temperatures. Again, as in fish $M_{org}(t\text{-}\Delta t)$ and $\Delta M_{org}(t)$ represent the previous calculated mass of PAH within the organism, and change to the mass of PAH, respectively (see below): $M_{org\,h}(t-1) = f_{org\,h}(t-\Delta t) \cdot V_{org\,h}(t-\Delta t) \cdot Z_{org\,h}(t-\Delta t)$ $\Delta M_{org\,h}(t) = [D_U \cdot f_U(t-\Delta t) + D_{R\,org} \cdot f_A(t-\Delta t) + D_{UW} \cdot f_W(t-\Delta t) - D_{E\,org} + D_{M\,org} + D_{Rep\,org} + D_{R\,org} + D_{ur\,org}) \cdot f_{Wh}(t-\Delta t)] \cdot \Delta t$ where f_U refers to the fugacity of an organism's diet and $D_{Rep\,org}$ refers to each species respective D value for chemical loss via reproduction: $D_{R\,moose} = D_{lac} + D_{calf}$, $D_{R\,grouse} = D_{grouse\,egg}$, and $D_{R\,mall} = D_{mall\,egg}$

Table S6. Contaminant mass balance equations for the animals added to the ACC-human model.

Equation
$\frac{dM_{wfish}}{dt} = \left[D_{U\ wfish} \cdot f_{wfish\ diet}(T) + D_{R\ wfish} \cdot f_W(T)\right] - \left[D_{E\ wfish} + D_{M\ wfish} + D_{R\ wfish}\right] \cdot f_{wfish}(T)$
$\frac{dM_{pike}}{dt} = \left[D_{U\ pike} \cdot f_{pike\ diet}(T) + D_{R\ pike} \cdot f_W(T)\right] - \left[D_{E\ pike} + D_{M\ pike} + D_{R\ pike}\right] \cdot f_{pike}(T)$
$\frac{dM_{moose}}{dt} = \left[D_{U\ moose} \cdot f_{willow}(T) + D_{R\ moose} \cdot f_A(T) + D_{UW\ moose} \cdot f_W(T)\right] - \left[D_{E\ moose} + D_{R\ moose} + D_{ur\ moose} + D_{M\ moose} + D_{calf} + D_{lac}\right] \cdot f_{moose}(T)$
$\frac{dM_{grouse}}{dt} = \left[D_{U\ grouse} \cdot f_{pine}(T) + D_{R\ grouse} \cdot f_A(T) + D_{UW\ grouse} \cdot f_W(T)\right] - \left[D_{E\ grouse} + D_{R\ grouse} + D_{ur\ grouse} + D_{M\ grouse} + D_{egg\ grouse}\right] \cdot f_{grouse}(T)$
$\frac{dM_{mall}}{dt} = \left[D_{U\ mall} \cdot f_{mall\ diet}(T) + D_{R\ mall} \cdot f_A(T) + D_{UW\ mall} \cdot f_W(T)\right] - \left[D_{E\ mall} + D_{R\ mall} + D_{ur\ mall} + D_{M\ mall} + D_{egg\ mall}\right] \cdot f_{mall}(T)$

Table S7. Physiological parameters for the modeled fish species.

Species	Life span	Growth Rate[1]	Max Weight	Max Volume	Whole-body lipid content	Diet	Ref.
	a	g	g	m^3	g lip g^{-1}		
Whitefish	20	$W_{wfish}(t) = 0.0067 \cdot \left[58.7 \cdot (1 - e^{(-0.21 \cdot (t/8760 + 0.35))})\right]^{3.181}$	2700	$2.7 \cdot 10^{-2}$	0.09	zooplankton, juvenile whitefish	1, 2, 3
Northern pike	20	$W_{cape}(t) = 0.0045 \cdot \left[95.47 \cdot (1 - e^{(-0.16 \cdot (t/8760 + 0.73))})\right]^{3.08}$	5030	$5.03 \cdot 10^{-2}$	0.02	zooplankton, whitefish, juvenile pike	4, 5

[1]Where *t* represents lifetime measured in hours.

References: 1) Fishbase – *Coregonus clupeaformis*, 2) Scott and Crossman 1998, 3) Law et al. 2009 4) Fishbase – *Esox lucius*, 5) Medford and Mackay 1977

Table S8. Parameters describing the modeled plant species.

Species	Growth Period	Air/Soil Temperature	Max Leaf Area Index	Transpiration Rate	Leaf Growth Function	Leaf Thickness	Maximum Fruit Mass	Fruit Yield	Ref.
	h	K	$m^2_{leaf}\ m^{-2}_{land}$	$m^3\ h^{-1}\ m^{-2}_{leaf}$	-	m	kg $fruit^{-1}$	$n_{fruit}\ m^{-2}_{land}$	
Rye grass	varied	289/289	4.5 - 7	$1.0 \cdot 10^{-5}$	linear	$4.0 \cdot 10^{-4}$	-	-	1
Willow	2208	288/287	4.4	$1.3 \cdot 10^{-4}$	linear	$2.0 \cdot 10^{-4}$	-	-	2, 3, 4, 5, 6
Cattail	1512	295/294	0.08	$1.6 \cdot 10^{-4}$	exponential	$1.5 \cdot 10^{-3}$	-	-	1, 5
Lodgepole pine	1512	288/287	5.0	$4.5 \cdot 10^{-5}$	linear	$5.4 \cdot 10^{-4}$	-	-	7, 8, 9
Saskatoon berry	1512	288/287	0.3	$1.9 \cdot 10^{-4}$	linear	$1.2 \cdot 10^{-4}$	$4.0 \cdot 10^{-3}$	215	10, 11, 12, 13
Mint	2208	288/287	2.0	$1.9 \cdot 10^{-4}$	exponential	$2.0 \cdot 10^{-4}$	-	-	1, 14 15, 16

References: 1) Undeman et al. 2009 2) Tharakan et al. 2005 3) Tharakan et al. 2008 4) Cooper et al. 2004 5) Irmak et al. 2013 7) Krol et al. 19998) O'Reilly & Owens 1998 9) Reid et al. 2006 10) Chen et al. 2009 11) Kim et al. 2011 12) Safley et al. 2009 13) Fallovo et al. 2008 14) Búfalo et al. 2016 15) Maffei et al. 1994 16) Shazia Erum et al. 2012. 17) Kabenge et al. 2012.

Note: Unavailable parameters for Saskatoon berry were replaced with those of the morphologically similar blueberry (*Vaccinium spp.*).

S4. Parameterization of Bioaccumulation Models for Moose, Grouse and Mallard

S4.1 Moose Model Parameterization

S4.1.1 Moose Mass & Volume. Male and female moose exhibit sexual dimorphism in body size. Thus, by adapting Gompertz growth equations from Binnington et al. (2016) and Kelly (2000) for caribou, and relying on moose body weight data from AnAge (the Animal Ageing and Longevity Database - http://genomics.senescence.info/species/entry.php?species=Alces_alces) adult moose masses (kg) were calculated according to:

Male Moose: $$M_{\text{mooseM}}(t) = 385 \cdot exp^{(-exp^{(-0.0039 \cdot t)})} \quad (1a)$$

Female Moose: $$M_{\text{mooseF}}(t) = 345 \cdot exp^{(-exp^{(-0.0039 \cdot t)})} \quad (1b)$$

where t represents age in years. Seasonal lipid content for males ranged from 0.02-0.11 g lipid·g^{-1} throughout the year, while for females this range was 0.07 to 0.16 g lipid·g^{-1} (Korea National Oil 2009). As a result, total body mass for male adult moose reached a maximum of 400 kg in autumn and a minimum of 375 kg in spring, while total body mass for female adult moose varied from 350 kg in autumn to 330 kg in spring. Moose water content was assumed to be similar to humans (71% of non-lipid mass) (Czub and McLachlan 2004; Czub et al. 2008). Masses were converted to volumes using lipid and water densities of 900 g·L^{-1} and 1000 g·L^{-1}, respectively.

S4.1.2 Moose Feeding Preferences and Feeding/Drinking Rates. Moose were assumed to consume only willow leaves (wet weight) (Renecker and Hudson 1986) at a daily body mass-weighted rate equivalent to that of adult caribou (Binnington et al. 2016; Miller 1976):

$$G_{U\ moose} = 0.8 \cdot m_{\text{moose}}(t)^{0.75} \quad (2)$$

Based on adaptation of the plant bioaccumulation model framework by Undeman et al. (2009), willow leaf fugacity was calculated according to the f_{leaf} equation listed for plants in Table S5. The maximal water uptake rate of adult moose was set equal to 32 L·d^{-1} (Belovsky and Jordan 1981), where water uptake by juvenile moose was assumed to increase proportionally to the rate of increase in juvenile moose body mass growth.

S4.1.3 Moose Respiration. Moose respiration rates were calculated according to the same relationship with body volume as used for caribou in Kelly and Gobas (2003):

$$G_{R\ moose} = 86.4 \cdot V_{\text{moose}}(t)^{0.75} \quad (3)$$

where $V_{\text{moose}}(t)$ represents the volume of the moose at time t. Note that 70% of inhaled air was assumed to equilibrate with the moose at a temperature of 37.0 °C.

S4.1.4 Moose Reproduction. Female moose reached sexual maturity at 2 years of age and gave birth to 1 calf annually during late May (Reese and Robbins 1994). Female moose were assumed to gain body and lipid mass equal to that of the calf [14 kg ww, 1.12 kg lipid] during the second half of their pregnancy, and subsequently shed this added weight immediately following birth. Moose nursing period (130 d), lactation rate (3 $L \cdot d^{-1}$), and milk lipid content (0.12 g lipid·g $milk^{-1}$) were adapted from Reese and Robbins (1994) and Kelly and Gobas (2003). Calves at birth and milk were assumed to be at chemical equilibrium with adult female moose, and moose milk temperature was assumed to be 37 °C (Czub and McLachlan 2004; Czub et al. 2008).

S4.2 Grouse and Mallard Model Parameterization

S4.2.1 Grouse and Mallard Mass & Volume. Rusch and Keith (1971) list body mass data for ruffed grouse in Alberta during fall, while Remington and Braun (1988) detailed lipid content in adult sage grouse. Mallard duck body mass and lipid content data was adapted from seasonal measurements by Moorman et al. (1992). Grouse and mallards also exhibit sexual dimorphism, where female birds typically represent the heavier sexes. Also, seasonal variability in adult weights throughout the year was modeled according to data for comparator bird species by Raveling (1979). We assumed male and female grouse and mallard non-lipid body weights at physical maturity (kg) to be represented by adult weights, and that values in physically immature birds increased allometrically until reaching these physically mature values according to the four equations below, adapted from Gompertz functions defining body mass growth for marine mammals by age (Binnington and Wania 2014; Czub and McLachlan 2007):

Male Grouse: $M_{\mathrm{grouseM}}(t) = [(1 - vf_{lip}) + vf_{lip}] \cdot [(0.48 \cdot [1 - \exp(-0.46 \cdot t)]^{0.34}) \cdot \theta_S(JD)]$ (4a)

Fem. Grouse: $M_{\mathrm{grouseF}}(t) = [(1 - vf_{lip}) + vf_{lip}] \cdot [(0.57 \cdot [1 - \exp(-0.46 \cdot t)]^{0.34}) \cdot \theta_S(JD)]$ (4b)

Male Mallard: $M_{\mathrm{mallM}}(t) = [(1 - vf_{lip}) + vf_{lip}] \cdot [(0.96 \cdot [1 - \exp(-0.46 \cdot t)]^{0.34}) \cdot \theta_S(JD)]$ (4c)

Fem. Mallard: $M_{\mathrm{mallF}}(t) = [(1 - vf_{lip}) + vf_{lip}] \cdot [(1.00 \cdot [1 - \exp(-0.46 \cdot t)]^{0.34}) \cdot \theta_S(JD)]$ (4d)

where t represents age in years, θ_S represents a multiplier defined by adapting seasonal variability in cackling goose body mass from Raveling (1979), and JD represents Julian Day. Seasonal variation in modeled grouse and mallard male and female non-lipid weights and lipid contents were derived by assuming linear transitions between minimum body mass values at the end of spring (-9.5% from assumed total adult mass) and maximum values at the onset of winter (+9.5%). The seasonally variable non-lipid and lipid weights derived from these values were then summed to produce total body weights throughout grouse and mallard lifetimes.

S4.2.2 Grouse and Mallard Energy Demands and Feeding/Drinking Rate. Grouse and mallard energy demands were defined according to the framework outlined by Norstrom et al. (2007) in their Avian BioAccumulation Model (ABAM), a deterministic POP bioaccumulation model in wild birds. The ABAM scheme defined total Daily Energy Expenditure (DEE, with units $kJ \cdot d^{-1}$ - i.e. daily energy requirement) for their test species, the non-passerine herring gull (*Larus argentatus*), according to:

$$\mathrm{DEE} = \left(Q_{\mathrm{em}} + Q_{\mathrm{L}} + Q_{\mathrm{egg}} + Q_{\mathrm{for}}\right) / F_{\mathrm{d}} \quad (5)$$

where Q_{em} represents daily existence metabolic rate, Q_L represents a cost/benefit term for the production/utilization of adipose tissue (i.e. positive/negative value when the bird generates/consumes lipids), Q_{egg} represents the energy required for female birds to produce eggs, Q_{for} represents energy expenditures associated with foraging, and F_D represents dietary energy assimilation efficiency (Norstrom et al. 2007). We adapted this equation for grouse and mallard to account for an absence of information on foraging costs in these species per the suggestion of (Norstrom et al. 2007) to:

$$\mathrm{DEE} = \left(\mathrm{FMR} + Q_{\mathrm{L}} + Q_{\mathrm{egg}}\right) / F_{\mathrm{d}} \quad (6)$$

where FMR represents the measured field metabolic rate of grouse and mallard, which includes any energy cost associated with foraging, and therefore Q_{for} is omitted from this version of the equation. Grouse and mallard FMR calculations were assumed to be similar to those of Canada geese (McWilliams and Raveling 2004), as specified in equation 7 below. Grouse diet was assumed to be exclusively composed of Lodgepole pine (Pendergrast and Boag 1970), while mallard diet was composed of a combination of aquatic vegetation (10%), zooplankton (60%), and benthic organisms (30%) as per Intrinsik Corp. (2010). Functions for Q_L (equation 8) and Q_{egg} (equation 9) were adapted from Norstrom et al. (2007), and as per ABAM F_d was set equal to 0.85 for grouse and mallard:

$$\mathrm{FMR} = 8.47 \cdot m_{\mathrm{bird}}(t)^{0.704} \quad (7)$$

$$Q_L = 39.3\ \mathrm{kJ} \cdot \mathrm{g\ lipid}^{-1} \cdot \Delta\, m_{\mathrm{bird\ lip}}(t) \quad (8)$$

$$Q_L = E_{\mathrm{egg}} \cdot N_{\mathrm{egg}} / F_{\mathrm{con}} \quad (9)$$

where $\Delta\, m_{\mathrm{bird\ lip}}\ (t)$ represents daily change in the mass of grouse or mallard lipid, E_{egg} represents the energy content of each egg, N_{egg} represents the number of eggs laid per clutch, and F_{con} represents grouse or mallard efficiency in converting maternal energy to egg mass. Based on an E_{egg} value of 602 kJ cited by Norstrom et al. (2007) in ABAM for herring gulls, respective values of 221 kJ and 401 kJ were assumed for grouse and mallard based on relative differences in sexually mature female body mass between herring gulls (~ 1.5 kg), grouse (0.55 kg), and mallard (1.0 kg) (Moorman et al. 1992; Norstrom et al. 2007; Rusch and Keith 1971). The ABAM value for F_{con} (0.75) was assumed for grouse and mallard, and the energy cost of feeding chicks was assumed to be negligible (Norstrom et al. 2007).

The maximal water uptake rates of adult grouse and mallard were set equal to 0.05 $L \cdot d^{-1}$ and 0.07 $L \cdot d^{-1}$, respectively based on scaling by body mass from ringed seal (Czub and McLachlan 2007). Correspondingly, water uptake rates in physically immature grouse and mallard were assumed to increase at a rate directly proportional to the rate of increase in immature grouse and mallard body mass growth.

S4.2.3 Grouse and Mallard Respiration. Adult grouse and mallard respiration rates of 0.34 $m^3 \cdot d^{-1}$ and 2.34 $m^3 \cdot d^{-1}$, respectively, were calculated based on data from Pekins et al. (1994b) for grouse and Keijer and Butler (1982) for mallard. Juvenile grouse and mallard inhalation rates increased toward these maximum values at a rate proportional to increasing body mass with age. 70% of inhaled air was assumed to equilibrate with the bird's core at a temperature of 37.0 °C.

S4.2.4 Grouse and Mallard Reproduction. Female grouse and mallard were assumed to reach sexual maturity at age 2, with annual clutch sizes of 9 and 10 eggs, respectively (Caldwell and Cornwell 1975; Leupin 2003). From egg wet weight and lipid weight data for herring gulls used by Norstrom et al. (2007) in ABAM, and based on ratios of sexually mature female body mass, grouse eggs were assumed to weigh 53.0 g and possess a lipid weight of 6.7 g while mallard eggs were assumed to weigh 64.2 g with lipid weights of 8.1 g. As the assumption of equifugacity between mother birds and their egg(s) has yet to be demonstrated in the literature, we relied on empirical egg/mother whole body lipid-normalized POP concentration ratios (congener-specific) published by Braune and Norstrom (1989) to simulate gestational contaminant exposure. Adapting ratios for PCBs, newborn chicks were assumed to possess contaminant concentrations half of those of their mothers at birth.

S5. Evaluation of New Bioaccumulation Models

To assess the capability of our expanded ACC-Human AOSR model to accurately reflect PAH exposures in the newly added species, we compared modeled concentrations of phenanthrene (PHE), pyrene (PYR), and benzo(*a*)pyrene (BaP) to biomonitoring data. Few compound-specific monitoring data were found for moose and birds in the Alberta oil sands region. For these organisms, a search was also performed for data from other locations, for related organisms, and related organisms in other areas. Although the importance of direct oil sands PAH sources are likely not represented through monitoring of similar populations living elsewhere, such data do at least provide some benchmark for the plausibility of the model predictions.

As biomonitoring typically yields mean exposure values from populations of animals of variable ages (or lengths) sampled at comparable points in time, we produced analogous model concentration predictions by deriving cross-sectional body burden-age trends, or CBATs (Binnington and Wania 2014; Lorber 2002; Quinn and Wania 2012), from our model results. Briefly, our primary model output is the lifetime PAH exposure estimate for individual organisms representing a birth cohort, termed longitudinal body burden-age trends (LBATs), which can be collectively "sampled" at one point in time to produce measures of average population exposure for a group of distinct wildlife birth cohorts (CBATs). The temporal resolution of CBATs varied with the lifespan of the organism. Thus, whitefish, Northern pike, and moose CBATs (lifespans of 20 years) included a maximum of 21 animal birth cohorts each born 1 - 2 years apart, while grouse and mallard CBATs (lifespan of 10 years) included a maximum of 11 birth cohorts each born 1 year apart. Below we compare modeled CBATs to biomonitoring data for each group of organisms separately.

S5.1 Whitefish and Northern Pike Model Evaluations

We evaluated our whitefish and Northern pike sub-models by comparing predictions with two sets of biomonitoring data. Ohiozebau et al. (2017) measured PHE, PYR, and BaP concentrations in the muscle of whitefish (n=93) and Northern Pike (n=104) from sites in northern Alberta and the Northwest Territories from 2011 to 2012 (in order of decreasing proximity from the oil sands area: Fort McMurray, Fort McKay, Fort Chipewyan, Fort Smith (Alberta), and Fort Resolution (Northwest Territories)). Golzadeh et al. (2021) measured concentrations of PHE, PYR, and BaP in muscle of whitefish (n=6) sampled by the Bigstone Cree Nation community not far from Fort McMurray, from June through September 2015. Since neither study reported fish length or age, we compared measured concentrations to the arithmetic mean PHE, PYR, and BaP concentrations of either all whitefish and Northern pike age

classes (age 0-20 y) or only those aged 0-10 y. As both studies were conducted some years after the end of our model simulation period, concentrations from the last year of the simulation period were used for comparison.

Simulated PAH concentrations in whitefish and Northern pike were regularly within the range of measured values (on a fresh-weight basis) provided for 5 locations by Ohiozebau et al. (2017), and were closer to concentrations measured in fish from sites closer to the AOSR (Fort McKay, Fort McMurray) than in those from more distant sites (Table S9). This was true for each of the three compounds and both fish species. If estimated PAH concentrations exceeded the highest measured value, they were always within a factor of approximately 2 of that value. Simulated PHE concentrations were an order of magnitude greater than measured concentrations reported by Golzadeh et al. (2021); PYR and BaP were not detected by Golzadeh et al. (2021). The relatively small sample size of the latter study (n=6) must be noted. In addition, discrepancies between modeled concentrations and reported concentrations in both studies may be due to the analysis of only muscle tissue in both studies, which contains lower lipid levels than whole-body tissue for which concentrations are calculated by the model. Overall, the modeled values agree well with the measured concentrations.

S5.2 Moose Model Evaluation

We used two reports on PAH levels in moose residing in northern Alberta (Golzadeh et al. 2021 and Lundin et al. 2015) to evaluate the capability of our new sub-model to estimate exposure in moose (Table S10). Lundin et al. (2015) report median PAH levels in scat samples collected within a 2500 km^2 area south of Fort McMurray during 2008-2009 on a compound-specific basis but did not provide any measurements for BaP. Therefore, we were limited to comparisons for PHE and PYR. Golzadeh et al. (2021) report mean PHE, PYR, and BaP levels in moose muscle (n=3) collected near the Fort McMurray area from June to September 2015. Because the age of the animals could not be determined we compared sex-matched CBATs to these data using two approaches: either including all modeled moose (age 0 to 20 y), or only those up to the age of 10. Concentrations in feces were reported by Lundin et al. (2015) in units of $ng_{PAH}\ g_{dw}^{-1}$, while Golzadeh et al. (2021) report concentrations in $ng_{PAH}\ g_{ww}^{-1}$. In order to compare these values to our modeled lipid-normalized whole-body concentrations, we converted the measured data into lipid-normalized values by assuming a lipid content of 2% in dry moose feces (reported for dairy cow feces by Møller et al. (2014)), and 2.2% for moose muscle (mean lipid content in moose muscle reported by Golzadeh et al. (2021)).

Model predictions were below the range of measured concentrations (Table S10). The predicted median PHE and PYR concentrations in moose were within one order of magnitude of the medians measured in the scat samples. Of the three samples analyzed by Golzadeh et al. (2021), only PYR was detected, in a single sample. Model mean concentrations of PYR were half of the reported mean. In general, considering the ranges of measurements including non-detects, the model predictions either fall below or overlap with the minimal end of the limited set of measurements. An extended search for compound-specific data did not yield any useful results.

There are two major possible explanations for the under-predictions. First, the number of validation samples is extremely limited and may not be a reliable indicator of moose PAH body burdens in the region. Second, concentrations in the major component of the modeled moose diet, willow, may have been under-predicted. Model willow concentrations were compared to concentrations measured in pooled samples of vegetation taken from 22 sites in the Fort McMurray and surrounding areas (Boutin & Carpenter 2017) (Table S11). The exact species of vegetation in the pooled samples were not specified, though the authors attempted to collect dominant species at each site, and report observing 13 species of willow during their vegetation surveys. Only maximum concentrations were reported in the study. The maximum modeled concentrations in willow were lower (within an order of magnitude) than the maximum concentrations measured in the pooled plant samples. This may reflect the model not accounting for certain substantial PAH exposure pathways for plants; namely, deposition of contaminated fugitive dust from petroleum coke stockpiles (Jautzy et al. 2015; Landis et al. 2019).

S5.3 Grouse and Mallard Model Evaluation

Similar to moose, the pool of available compound-specific validation data for birds in the study area was limited. Grouse and mallard evaluation data were available from Environment and Climate Change Canada (ECCC, 2018) and Golzadeh et al. (2021). ECCC (2018) report PAH concentrations in mallard liver samples taken north and south of the oil sands development area in 2013. Golzadeh et al. (2021) reported PAH concentrations in mallard (n=7) and grouse muscle (n=10) samples taken in the Fort McMurray area from June to September of 2015. Data were lipid-normalized using the mean lipid contents reported by Golzadeh et al. (2021) for grouse muscle (0.52%), mallard duck muscle (2.72%), and mallard duck liver (5.53%).

Model predictions fall under or overlap with the lower end of the measured range of concentrations for samples in which there were detections (Table S12). PHE and PYR concentrations in the mallard liver samples ranged from below detection limits to 72.5 and 16.5 ng/g lipid, respectively, while BaP was

below detection limits in all liver samples. Mean PYR concentrations in mallard and grouse muscle were 2.57 and 15.4 ng/g lipid in mallard and grouse muscle, respectively, while PHE and BaP were not detected. Similar to moose, the major component of the model bird diets are plants (mallard: cattail; grouse: pine), Therefore, the organism under-predictions may be a result of under-predictions in the plants. The under-prediction is less for mallard, which has a more varied diet than the grouse, consisting also of aquatic organisms.

An extended search for compound-specific data yielded one study of PAHs in the carcasses of 17 male lesser scaup ducks sampled in Indiana, USA, from January to March 1994, in a heavily polluted area considered at the time to be one of the most concentrated steel and petrochemical complexes in the USA (Custer et al. 2000). PYR concentrations were not presented in the study as it was detected in less than 6 samples. PHE was detected in 10 samples, with concentrations ranging from below detection limits to 0.02 μg/g ww. BaP was detected in 9 samples, with concentrations ranging from below detection limits to 0.04 μg/g ww. Assuming a 4.1% whole body lipid content (an average of the lipid content in muscle and liver reported by Golzadeh et al. (2021)), the maximum detected concentrations of PHE and BaP were 488 and 976 ng/g lipid, respectively. These concentrations may be considered a "worst-case" reference scenario for a heavily polluted industrial environment, and are far above the model predictions in Table S12.

S5.4 Human Model Evaluation

There were no available reports of PHE, PYR, or BaP concentrations in human tissues in the model region nor in Canada to which we could compare our model results. Thus, as a rough means of validating the human food chain, we compared modeled concentrations in female humans to a diverse set of lipid-normalized concentrations reported in human breast milk in Turkey (Çok et al. 2012), the Czech Republic (Pulkrabova et al. 2016), and the United States (Kim et al. 2008) (Table S13). Our modeled values fell within or just below the range of measured values. This may be due to several factors including differences in contamination of the local environment and food supply, differences in diet, and differences in typical food preparation methods. In general, the small disparity between our modeled values and the range of measured values in various locations across the globe imparts a certain degree of confidence to our model's structure and performance.

S6. Model Evaluation Tables

Table S9. Comparisons of modeled whitefish and Northern pike PAH exposures to literature data from Ohiozebau et al. (2017) and Golzadeh et al. (2021). Modeled arithmetic mean, wet weight whole-body phenanthrene (PHE), pyrene (PYR), and benzo(*a*)pyrene (BaP) concentrations (ng·g ww^{-1}) from sampling date-matched CBATs were compared to reported measured values for fish muscle. Given that in all manuscripts fish ages or lengths were not provided we have included 2 sets of modeled CBAT data for whitefish and Northern pike, ranging in age from either 0-10 or 0-20 years. Note that our model does not account for fish breeding behaviours, and thus all whitefish and Northern pike are functionally male.

	Year	Season	Region	Species	n^a	PHE (mean ± SD)	PYR (mean ± SD)	BaP (mean ± SD)
Ohiozebau *et al.* 2017	2011	Summer	Fort Resolution	Whitefish (muscle)	26	n.d.[b]	0.6 ± 0.7	0.1 ± 0.1
			Fort Smith		18	n.d.	0.2 ± 0.3	0.1 ± 0.2
			Fort Chipewyan		18	0.5 ± 0.7	5.6 ± 5.9	4.7 ± 8.6
			Fort McKay		20	3.0 ± 3.4	4.5 ± 7.8	3.9 ± 4.7
			Fort McMurray		11	n.a.[c]	n.a.	n.a.
Model 0-10 y	2008	Summer	Alberta Oil Sands Region	Whitefish	5	3.5 ± 0.9	3.2 ± 1.7	3.8 ± 0.9
Model 0-20 y					10	2.9 ± 0.9	2.4 ± 1.4	3.4 ± 0.8
Ohiozebau *et al.* 2017	2011	Fall	Fort Resolution	Whitefish (muscle)	26	0.5 ± 0.6	0.1 ± 0.1	0.6 ± 0.5
			Fort Smith		18	3.9 ± 4.6	0.5 ± 1.1	0.4 ± 0.4
			Fort Chipewyan		18	0.9 ± 1.1	3.0 ± 3.3	3.3 ± 6.6
			Fort McKay		20	2.0 ± 1.7	2.6 ± 3.9	3.4 ± 7.1
			Fort McMurray		11	1.7 ± 2.3	2.8 ± 3.6	1.1 ± 1.3
Model 0-10 y	2008	Fall	Alberta Oil Sands Region	Whitefish	5	6.1 ± 2.4	3.6 ± 1.7	4.2 ± 0.9
Model 0-20 y					10	5.3 ± 2.1	2.8 ± 1.4	3.7 ± 0.8
Ohiozebau *et al.* 2017	2012	Spring	Fort Resolution	Whitefish (muscle)	26	1.8 ± 1.9	n.d.	0.4 ± 0.5
			Fort Smith		18	0.1 ± 0.1	1.8 ± 3.0	0.7 ± 0.9
			Fort Chipewyan		18	0.8 ± 0.9	1.0 ± 1.5	0.1 ± 0.2
			Fort McKay		20	1.3 ± 0.0	2.1 ± 2.9	0.7 ± 0.5
			Fort McMurray		11	3.3 ± 2.7	1.2 ± 0.6	0.8 ± 0.6
Model 0-10 y	2008	Spring	Alberta Oil Sands Region	Whitefish	5	4.5 ± 1.8	5.1 ± 3.8	4.5 ± 1.2
Model 0-20 y					10	3.5± 1.6	3.6 ± 3.0	3.9 ± 1.0

	Year	Season	Region	Species	*n*	PHE	PYR	BaP
						ng g ww^{-1} / mean ± SD		
Ohiozebau *et al.* 2017	2011	Summer	Fort Resolution	Northern Pike (muscle)	24	n.d.	0.2 ± 0.2	0.6 ± 1.5
			Fort Smith		19	0.5 ± 0.7	0.1 ± 0.1	0.3 ± 0.5
			Fort Chipewyan		20	n.d.	1.0 ± 1.4	0.1 ± 0.1
			Fort McKay		20	2.0 ± 3.0	1.6 ± 2.5	0.3 ± 0.4
			Fort McMurray		21	2.3 ± 4.0	4.4 ± 4.4	1.2 ± 0.9
Model 0-10 y	2008	Summer	Alberta Oil Sands Region	Northern Pike	5	1.4 ± 0.1	2.1 ± 1.0	2.0 ± 1.1
Model 0-20 y					10	1.4 ± 0.1	1.6 ± 0.8	1.5 ± 0.9
Ohiozebau *et al.* 2017	2011	Fall	Fort Resolution	Northern Pike (muscle)	24	n.d.	n.d.	0.3 ± 0.3
			Fort Smith		19	n.d.	0.3 ± 0.8	0.3 ± 0.3
			Fort Chipewyan		20	n.d.	0.2 ± 0.3	0.6 ± 0.6
			Fort McKay		20	3.3 ± 4.7	3.6 ± 2.0	3.3 ± 3.2
			Fort McMurray		21	1.1 ± 0.2	0.3 ± 0.1	0.9 ± 1.1
Model 0-10 y	2008	Fall	Alberta Oil Sands Region	Northern Pike	5	3.2 ± 0.6	2.6 ± 1.2	2.2 ± 1.1
Model 0-20 y					10	2.9 ± 0.6	2.0 ± 1.1	1.6 ± 0.9
Ohiozebau *et al.* 2017	2012	Spring	Fort Resolution	Northern Pike (muscle)	24	0.6 ± 0.5	0.4 ± 1.0	0.4 ± 0.4
			Fort Smith		19	0.9 ± 0.6	0.2 ± 0.1	0.1 ± 0.1
			Fort Chipewyan		20	1.0 ± 1.6	0.1 ± 0.1	0.8 ± 1.7
			Fort McKay		20	3.7 ± 1.6	0.7 ± 0.6	1.0 ± 0.4
			Fort McMurray		21	3.3 ± 1.3	1.2 ± 0.6	1.6 ± 0.4
Model 0-10 y	2008	Spring	Alberta Oil Sands Region	Northern Pike	5	1.9 ± 0.2	3.2 ± 1.8	2.2 ± 1.1
Model 0-20 y					10	1.7 ± 0.2	2.3 ± 1.5	1.6 ± 1.0

[a]n: total number of samples across all seasons as season-specific n were not stated
[b]n.a. - no specimens available in season at location
[c]n.d. - below detection limits

	Year	Season	Region	Species	*n*	PHE	PYR	BaP
						ng g ww^{-1} / mean ± SD [a]		
Golzadeh *et al.* 2021	2015	Summer	Alberta Oil Sands Region	Whitefish (muscle)	6	0.24 ± 0.17	n.d.	n.d.
Model 0-10 y	2008	Summer	Alberta Oil Sands Region	Whitefish	5	3.5 ± 0.9	3.2 ± 1.7	3.8 ± 0.9
Model 0-20 y					10	2.9 ± 0.9	2.4 ± 1.4	3.4 ± 0.8

[a]method detection limits ranged from 0.06 to 0.1 ng/g ww for 10 g of sample

Table S10. Comparisons of modeled moose PAH exposures to measured concentrations in moose feces from Lundin et al. (2015) and moose muscle from Golzadeh et al. (2021). Modeled median or mean whole-body PHE, PYR, and BaP concentrations (ng·g lipid^{-1}) from sampling date- and sex-matched CBATs were compared to reported measured values. Measured concentrations were lipid-normalized, assuming 2% lipid content of dry moose feces (Møller et al. 2014), and 2.20% lipid content of moose muscle reported by Golzadeh et al. (2021).

	Year	Season	Region	Species	n_{male}	n_{fem}	PHE	PYR
							ng g lipid^{-1} / median ± SD	
Lundin *et al.* 2016	2008-09	Winter	Algar	Moose (feces)	7	13	16 ± 10	1.5 ± 7.0
			Egg Pony		14	14	11 ± 7.0	10 ± 7.5
			Wiau		6	14	22 ± 8.5	8.5 ± 4.5
Model 0-10	2007-08	Winter	Alberta Oil Sands Region	Moose	10	10	2.5 ± 0.2	0.5 ± 0.04
Model 0-20					20	20	2.5 ± 0.2	0.5 ± 0.03

	Year	Season	Region	Species	*n*	PHE	PYR	BaP
						ng g lipid^{-1} [a] / mean ± SD		
Golzadeh *et al.* 2021	2015	Summer	Alberta Oil Sands Region	Moose (muscle)	3	n.d.	4.09[b]	n.d.
Model 0-10	2008	Summer	Alberta Oil Sands Region	Moose	10	2.0 ± 0.3	0.4 ± 0.09	
Model 0-20					20	2.0 ± 0.2	0.4 ± 0.07	

[a] Method detection limits ranged from 0.06 to 0.1 ng/g ww (or 2.7 to 4.5 ng/g lipid assuming 2.20% lipid content of muscle) for 10 g of sample

[b] Concentrations are reported in reference as mean ± standard error. Considering report of single value, this is not a mean but rather the PYR concentration of the sole detection.

Table S11. Comparisons of modeled plant PAH concentrations to measured concentrations in pooled upland and wetland plant samples from 22 sites in the Alberta Oil Sands Region reported by Boutin & Carpenter (2017). Exact plant species in pooled samples were not specified, and only maximum concentrations were reported. The authors attempted to collect dominant species at each site. ~10 types of berries, cattail, five pine species, and more than 10 species of willow were included in the list of species observed during vegetation surveys.

	Year	Season	Region	Species	*n*	PHE	PYR	BaP
						ng g fresh weight^{-1} / maximum[a] (above ground, below ground)		
Boutin & Carpenter 2017	2013-14	Not given	Ft. McMurray	Several (pooled samples)	22 sites	17.2, 18.2	12.4, 7.1	34.6, 4.2
			East of FM			6.2, 12.8	3.3, 3.3	3.0, 2.2
			West of FM			8.6, 10.7	5.7, 3.8	n.d., n.d.
Model	2008	Spring, Summer, Fall	Alberta Oil Sands Region	Cattail	n/a	0.49, 0.49	0.23, 0.32	0.01, 0.73
				Willow		2.01, 0.98	1.04, 1.08	0.07, 4.3
				Mint		3.63, 0.56	2.24, 0.87	0.15, 4.95
				Blueberry[b]		4.46, 0.63, 0.11	3.05, 0.90, 0.02	0.22, 5.75, 0.003
				Pine		0.41, 0.99	0.20, 0.37	0.01, 1.10

[a] Unclear if measured concentrations in Boutin & Carpenter (2017) are fresh weight or dry weight.
[b] Modeled blueberry concentrations are reported in root, leaf, and fruit.

Table S12. Comparisons of modeled grouse and mallard PAH exposures to literature data from ECCC (2018) and Golzadeh et al. (2021). Modeled arithmetic mean whole-body PHE, PYR, and BaP concentrations (ng·g lipid^{-1}) from sampling date- and sex-matched CBATs were compared to measured values.

	Year	Season	Region	Species	*n*	ng g lipid^{-1} / range [a]		
						PHE	PYR	BaP
ECCC 2018	2013	Not stated	Northern Alberta, NWT	Mallard (liver)	11	n.d. – 72.5	n.d. – 16.5	n.d.
Model 0-10 y	2007-08	All	Alberta Oil Sands Region	Mallard	10 male 10 fem	0.8 – 1.3	0.6 – 1.2	1.5 – 2.9

[a] Assume values reported "0" are non-detects. Detection limits were not provided.

	Year	Season	Region	Species	*n*	ng g lipid^{-1} / mean ± SE [a]		
						PHE	PYR	BaP
Golzadeh *et al.* 2021	2015	Summer	Alberta Oil Sands Region	Mallard (muscle)	7	n.d.	2.57 ± 2.21	n.d.
Model 0-10 y	2007-08	All	Alberta Oil Sands Region	Mallard	10 male 10 fem	1.0 ± 0.2	0.8 ± 0.2	2.0 ± 0.5
Golzadeh *et al.* 2021	2015	Summer	Alberta Oil Sands Region	Grouse (muscle)	10	n.d.	15.4 ± 13.5	n.d.
Model 0-10 y	2007-08	All	Alberta Oil Sands Region	Grouse	10 male 10 fem	1.1 ± 0.03	0.07 ± 0.003	0.02 ± 0.001

[a] Method detection limits ranged from 0.06 to 0.1 ng/g ww for 10 g of sample (or 2.2 to 3.7 ng/g lipid for mallard muscle assuming 2.72% lipid content, or 11.5 to 19.2 ng/g lipid for grouse muscle assuming 0.52% lipid content)

Table S13. Comparisons of modeled, age-matched female human PAH exposures to previous measurements of PAHs in human breastmilk in various locations (ng·g lipid^{-1}). Note that studies in Hong Kong (Tsang et al. 2011) and China (Wang et al. 2018, Yu et al. 2011) were excluded from the comparison as concentrations were mostly at least a few times higher than those measured by Çok et al. 2012.

	Average Age	Sampling Date	Region	PHE mean (range)	*df*	PYR mean (range)	*df*	BaP mean (range)	*df*
Çok *et al.* 2012	28	Sep-Oct 2009	Mersin, Turkey (Smokers)	24 (5.4 – 93)	7/7	8.2 (2.8 – 27)	7/7	0.27 (0.094 – 0.76)	7/7
			Mersin, Turkey (Non-smokers)	15 (2.9 – 74)	40/40	5.4 (1.0 – 20)	40/40	0.24 (0.036 – 0.93)	40/40
Model	28	Sep-Oct 2008	AOSR (Smokers)	1.36 (1.34 – 1.37)		0.36 ()		0.16	
			AOSR (Non-smokers)	0.70 (0.69 – 0.71)		0.17 (0.16 – 0.17)		0.079 (0.076 – 0.081)	
Pulkrabova *et al.* 2016[a]	30	Aug-Sep 2013	Karvina, Czech Republic (Industrial, Non-smokers)	14 (bdl – 47)	62/66	0.70 (bdl – 6.1)	49/66	0.27 (bdl – 0.52)	18/66
	30	Jan-Apr 2014	Karvina, Czech Republic (Industrial, Non-smokers)	20 (bdl – 85)	66/67	1.2 (bdl – 9.8)	57/67	0.24 (bdl – 0.80)	26/67
	32	Aug-Sep 2013	České Budějovice, Czech Republic (Control, Non-smokers)	9.4 (bdl – 38)	76/93	0.67 (bdl – 9.2)	63/93	0.17 (bdl – 0.72)	25/93
	33	Jan-Apr 2014	České Budějovice, Czech Republic (Control, Non-smokers)	12 (bdl – 71)	71/95	0.8 (bdl – 15)	62/95	0.14 (bdl – 0.71)	29/95
Model	30	Jan-Apr 2008	AOSR (Non-smokers)	0.73 (0.71– 0.75)		0.16		0.053 (0.051 – 0.055)	
		Aug-Sep 2008	AOSR (Non-smokers)	0.72		0.17		0.08 (0.078 – 0.083)	
Kim *et al.* 2008	25[b]	2005	Baltimore & Various, United States (Non-smokers)	13 (6.3 – 23)	12/12	1.3 (0.7 – 3.0)	12/12	not measured	--
Model	25	2005	AOSR (Non-smokers)	0.7 (0.64 – 0.73)		0.16 (0.14 – 0.19)		0.069 (0.048 – 0.109)	

[a] bdl: below detection limit

[b] Age range given as 15-25 for 3 out of 12 mothers, ages of remaining mothers not given.

S7. Market Food and Cigarette Smoking Parameterization

Concentrations in domestic and imported raw potatoes, other vegetables, fruit, and meat were taken from Canadian Food Inspection Agency reports (2014; 2015; 2016). Concentrations for coffee (Houessou et al. 2007; Kayali-Sayadi et al. 1999; Orecchio et al. 2009; Tfouni et al. 2013) and tea (Girelli et al. 2017; Kayali-Sayadi et al. 1998; Lin & Zhu 2006; Viñas et al. 2007) were for brewed liquids prepared in different locations with various tea and coffee origins. Data for pasta/noodles (Kazerouni et al. 2001; Lodovici et al. 1995) and soup (Kazerouni et al. 2001) were only available for BaP, in cooked forms of these foods. The average ratios of PHE:BaP and PYR:BaP in typical soup constituents, i.e. chicken, potatoes, and vegetables, were used to estimate PHE and PYR concentrations in soup. Similarly, average ratios of PHE:BaP and PYR:BaP in vegetables, which are cultivated in a manner similar to grains, were used to estimate PHE and PYR concentrations in pasta/noodles.

Table S14. PHE, PYR, and BaP contamination of market foods and inhaled cigarette smoke.

	PHE Concentration	PYR Concentration	BaP Concentration
Market Food Consumption *[ng/g]*			
Coffee	0.26	0.069	0.0094
Tea	1.3	0.28	0.0084
Soup (canned + ramen)	1.8	0.44	0.020
Potatoes	0.26	0.072	0.0032
Vegetables	0.41	0.16	0.0089
Pasta & Noodles	4.5	1.8	0.099
Pork	1.2	0.23	0.0010
Chicken	1.9	0.33	0.014
Fruit	0.32	0.074	0.0092
Cigarette Smoking *[ng/cig]*[a]	230	66	15

[a] taken from Vu et al. (2015)

Table S15. Equations describing PAH uptake via soil ingestion, market food ingestion, and firsthand cigarette smoke.

New Human Uptake Flux Equations (mol · h^{-1})	
Soil Ingestion, N_S	$N_S = IR_S \cdot Z_S \cdot f_S \cdot E_{0,GIT} \cdot AgeCorr$ where: IR_S is the volumetric soil ingestion rate Z_S is the fugacity capacity of the soil f_S is the fugacity of the soil $E_{0,GIT}$ is the chemical absorption efficiency in the gastrointestinal tract, is gender-specific, and depends on age, K_{OW}, $K_{feces\text{-}blood}$, and feces egestion rates as described in Czub et al. 2004 $AgeCorr$ is a unitless age scaling factor for ingestion rates, specific to gender, and greatest for a 25 year old MW_{PAH} is PAH molecular weight
Market Food Consumption, N_{market}	$N_{market} = N_{market,intake} \cdot E_{0,GIT} \cdot AgeCorr$ where: $N_{market,intake} = C_{PAH_{market}} \cdot IR_{market} \cdot MW_{PAH}$ $E_{0,GIT}$ is as above for soil ingestion $AgeCorr$ is as above for soil ingestion $C_{PAH_{market}}$ is the sum of the PAH content of all market foods IR_{market} is the sum of mass intake rates of all market foods MW_{PAH} is PAH molecular weight
Cigarette Smoking, N_{smo}	$N_{smo} = N_{smo,intake} \cdot E_{0,lung} \cdot AgeCorr$ where: $N_{smo,intake} = C_{PAH_{cig}} \cdot IR_{cig} \cdot MW_{PAH}$ $E_{0,lung}$ is a constant, unitless absorption efficiency in the lungs $AgeCorr$ is as above, but for cigarette smoking rate $C_{PAH_{cig}}$ is the mass of PAH taken in per cigarette IR_{cig} is the cigarette smoking rate MW_{PAH} is PAH molecular weight

S8. References

Arnot JA, Mackay D, Parkerton TF, Bonnell M. 2008. A database of fish biotransformation rates for organic chemicals. Environ Toxicol Chem 27: 2263–2270.

Arnot, J. A., Brown, T. N., Wania, F. 2014. Estimating screening-level organic chemical half-lives in humans. Environ Sci Technol 48(1), 723-730.

Belovsky GE, Jordan PA. 1981. Sodium dynamics and adaptations of a moose population. J Mammal 62: 613–621.

Binnington MJ, Curren MS, Quinn CL, Armitage JM, Arnot JA, Chan HM, et al. 2016. Mechanistic polychlorinated biphenyl exposure modeling of mothers in the Canadian Arctic: the challenge of reliably establishing dietary composition. Environ Int 92-93: 256-268.

Binnington MJ, Wania F. 2014. Clarifying relationships between persistent organic pollutant concentrations and age in wildlife biomonitoring: individuals, cross-sections, and the roles of lifespan and sex. Environ Toxicol Chem 33: 1415-1426.

Boutin C, Carpenter DJ. 2017. Assessment of wetland/upland vegetation communities and evaluation of soil-plant contamination by polycyclic aromatic hydrocarbons and trace metals in regions near oil sands mining in Alberta. Sci Total Environ 576: 829-839.

Braune BM, Norstrom RJ. 1989. Dynamics of organochlorine compounds in herring gulls: III. Tissue distribution and bioaccumulation in Lake Ontario gulls. Environ Toxicol Chem 8: 957-968.

Brown MG, Black TA, Nesic Z, Foord VN, Spittlehouse DL, Fredeen AL, et al. 2013. Evapotranspiration and canopy characteristics of two lodgepole pine stands following mountain pine beetle attack. Hydrol Process 28: 3326-3340.

Búfalo J, Rodrigues RM, Rolim de Almeida LF, dos Santos Tozin LR, Mayo Marques MO, Fernandes Boaro CS. 2016. PEG-induced osmotic stress in *Mentha x piperita* L.: Structural features and metabolic responses. Plant Physiol Biochem 105: 174-184.

Cable JM, Ogle K, Bolton WR, Bentley LP, Romanovsky V, Iwata H, et al. 2014. Permafrost thaw affects boreal deciduous plant transpiration through increased soil water, deeper thaw, and warmer soils. Ecohydrol 7: 982-997.

Caldwell PJ, Cornwell GW. 1975. Incubation behavior and temperatures of the mallard duck. The Auk 92: 706–731.

Canadian Food Inspection Agency. No date. National chemical residue monitoring program: 2014-2015 Report. Accessed August 3, 2018 at http://www.inspection.gc.ca/food/chemical-residues-microbiology/chemical-residues/eng/1324258929171/1324264923941.

Canadian Food Inspection Agency. No date. National chemical residue monitoring program: 2013-2014 Report. Accessed August 3, 2018 at http://www.inspection.gc.ca/food/chemical-residues-microbiology/chemical-residues/eng/1324258929171/1324264923941.

Canadian Food Inspection Agency. No date. National chemical residue monitoring program: 2012-2013 Report. Accessed August 3, 2018 at http://www.inspection.gc.ca/food/chemical-residues-microbiology/chemical-residues/eng/1324258929171/1324264923941.

Chan LHM, Receveur O, Batal M, David W, Schwartz H, Ing A, et al. 2016. First Nations Food, Nutrition, & Environment Study (FNFNES): Results from Alberta 2013. University of Ottawa. Ottawa, ON. pp. 1–178.

Chen W, Li J, Zhang Y, Zhou F, Koehler K, Leblanc S. 2009. Relating biomass and leaf area index to non-destructive measurements in order to monitor changes in arctic vegetation. Arctic 62: 281-294.

Çok I, Mazmanci B, Mazmanci MA, Turgut C, Henkelmann B, Schramm K-W. 2012. Analysis of human milk to assess exposure to PAHs, PCBs, and organochlorine pesticides in the vicinity Mediterranean city Mersin, Turkey. Environ Int 40: 63-69.

Cooper RL, Ware JV, Cass DD. 2004. Leaf thickness of Salix spp. (*Salicaceae*) from the Athabasca sand dunes of northern Saskatchewan, Canada. Can J Bot. 82: 1682–1686.

Custer TW, Custer CM, Hines RK, Sparks DW. 2000. Trace elements, organochlorines, polycyclic aromatic hydrocarbons, dioxins and furans in lesser scaup wintering on the Indiana Harbor Canal. Environ Pollut 110(3): 469-482.

Czub G, McLachlan MS. 2004. A food chain model to predict the levels of lipophilic organic contaminants in humans. Environ Toxicol Chem 23: 2356–2366.

Czub G, McLachlan MS. 2007. Influence of the temperature gradient in blubber on the bioaccumulation of persistent lipophilic organic chemicals in seals. Environ Toxicol Chem 26: 1600–1605.

Czub G, Wania F, McLachlan MS. 2008. Combining long-range transport and bioaccumulation considerations to identify potential Arctic contaminants. Environ Sci Technol 42: 3704–3709.

Drouillard K, Norstrom RJ. 2000. Dietary absorption efficiencies and toxicokinetics of polychlorinated biphenyls in ring doves following exposure to alochlor® mixtures. Environ Toxicol Chem 19(11): 2707-2714.

Environment and Climate Change Canada, 2018. Hunter-Trapper Harvested Wildlife Contaminants and Toxicology, Oil Sands Region. Downloaded on 2021-Jan-08 from http://donnees.ec.gc.ca/data/species/assess/wild-bird-and-hunter-trapper-harvested-wildlife-toxicology-and-contaminants-oil-sands-region/Hunter-Trapper-harvested-wildlife-contaminants-and-toxicology-oil-sands-region/?lang=en

Fallovo C, Cristofori V, Mendoza de-Gyves E, Rivera CM, Rea R, Fanasca S, et al. 2008. Leaf area estimation model for small fruits from linear measurements. HortScience 43: 2263-2267.

Fishbase. *Esox lucius* - Northern pike. http://www.fishbase.ca/summary/258. Accessed online 04/20/17.

Fishbase. *Coregonus clupeaformis* - Whitefish. http://www.fishbase.ca/Summary/SpeciesSummary.php?ID=234&AT=whitefish. Accessed online 03/11/17.

Gao Y, Ling W, Wong MH. 2006. Plant-accelerated dissipation of phenanthrene and pyrene from water in the presence of a nonionic-surfactant. Chemosphere 63: 1560–1567.

Girelli AM. Apriceno A, Tarola AM, Tortora F. 2017. Determination of polycyclic aromatic hydrocarbons in tea infusions samples by high performance liquid chromatography with fluorimetric detection. *J. Food Qual.* **2017**: 7 pp.

Golzadeh N, Barst BD, Baker JM, Auger JC, McKinney MA. 2021. Alkylated polycyclic aromatic hydrocarbons are the largest contributor to polycyclic aromatic compound concentrations in traditional foods of the Bigstone Cree Nation in Alberta, Canada. Environ Pollut 275: 116625.

Health Canada. 2018. Canadian Nutrient File - Search by food. Available at https://food-nutrition.canada.ca/cnf-fce/index-eng.jsp. Accessed September 28, 2018

Honey S, O'Keefe P, Drahushuk AT, Olson JR, Kumar S, Sikka HC. 2000. Metabolism of benzo(a)pyrene by duck liver microsomes. Comp Biochem Physiol C Toxicol Pharmacol 126: 285-

292.

Houessou JK, Maloug S, Leveque A-S, Delteil C, Heyd B, Camel V. 2007. Effect of roasting conditions on the polycyclic aromatic hydrocarbon content in ground *Arabica* coffee and coffee brew. *J. Food Agric. Food Chem.* **55**(23): 9719-9726.

Intrinsik Corp. 2010. Human health risk assessment of Flin Flon, Manitoba, and Creighton, Saskatchewan. Mississauga, ON. pp. 1–29.

Irmak S, Kabenge I, Rudnick D, Knezevic S, Woodward D, Moravek M. 2013. Evapotranspiration crop coefficients for mixed riparian plant community and transpiration crop coefficients for Common reed, Cottonwood and Peach-leaf willow in the Platte River Basin, Nebraska-USA. J Hydrol 481: 177–190.

Jautzy JJ, Ahad JME, Gobeil C, Smirnoff A, Barst BD, Savard MM. 2015. Isotopic evidence for oil sands petroleum coke in the Peace–Athabasca Delta. Environ Sci Technol 49(20): 12062-12070.

Kabenge I, Irmak S. Evaporative losses from a common reed ‐ dominated peachleaf willow and cottonwood riparian plant community. Water Resources Research, 2012, 48(9).

Kayali-Sayadi MN, Rubio-Barroso S, Cuesta-Jimenez MP, Polo-Díez LM. 1998. Rapid determination of polycyclic aromatic hydrocarbons in tea infusion samples by high-performance liquid chromatography and fluorimetric detection based on solid-phase extraction. *Analyst* **123**: 2145-2148.

Kayali-Sayadi MN, Rubio-Barroso S, Cuesta-Jiminez MP, Polo-Díez LM. 1999. A new method for the determination of selected PAHs in coffee brew samples by HPLC with fluorimetric detection and solid-phase extraction. *J. Liq. Chromatogr. Relat. Technol.* **22**(4): 615-627.

Kazerouni N, Sinha R, Hsu C-H, Greenberg A, Rothman N. 2001. Analysis of 200 food items for benzo[a]pyrene and estimation of its intake in an epidemiologic study. *Food Chem. Toxicol.* **39**: 423-436.

Keijer E, Butler PJ. 1982. Volumes of the respiratory and circulatory systems in tufted and mallard ducks. J Exp Biol 101: 213–220.

Kelly BC. 2000. Trophic Transfer of Persistent Organic Pollutants in an Arctic Tundra Ecosystem. F.A.P.C. Gobas. Simon Fraser University. Vancouver, BC. pp. 1-229.

Kelly BC, Gobas FAPC. 2003. An arctic terrestrial food-chain bioaccumulation model for persistent organic pollutants. Environ Sci Technol 37: 2966–2974.

Kim SR, Halden RU, Buckley TJ. 2008. Polycyclic aromatic hydrocarbons in human milk of nonsmoking U.S. women. Environ Sci Technol 42(7): 2663-2667.

Kim SJ, Yu DJ, Kim T-C, Lee HJ. 2011. Growth and photosynthetic characteristics of blueberry (*Vaccinium corymbosum cv. Bluecrop*) under various shade levels. Scientia Horticulturae 129: 486–492.

Korea National Oil Corporation. 2009. Application for Approval of the Blackgold Expansion Project. Calgary, AB. pp. 1–3715.

Krol PM, Ormrod DP, Binder WD, L'hirondelle SJ. 1999. Effects of Ultraviolet-B radiation on needle anatomy and morphology of western larch, interior spruce and lodgepole pine. J Sustain Forest 10:141-148.

Kroner SM, Cozzie DA. 1999. Data Collection for the Hazardous Waste Identification Rule. 1–49. U.S Environmental Protection Agency, Office of Solid Waste. Washington, DC. pp. 1-49.

Landis MS, Studabaker WB, Pancras JP, Graney JR, Puckett K, White EM, et al. 2019. Source apportionment of an epiphytic lichen biomonitor to elucidate the sources and spatial distribution of polycyclic aromatic hydrocarbons in the Athabasca Oil Sands Region, Alberta, Canada. Sci Total Environ. 654: 1241-1257.

Law K, Halldorson T, Danell R, Stern G, Gewurtz S, Alaee M, et al. 2006. Bioaccumulation and trophic transfer of some brominated flame retardants in a Lake Winnipeg (Canada) food web. Environ Toxicol Chem 25: 2177-2186.

Leupin EE. 2003. Status of the Sharp-Tailed Grouse (*Tympanuchus phasianellus*) in British Columbia. BC Ministry of Sustainable Resource Management, Conservation Data Centre and BC Ministry of Water, Land, and Air Protection, Biodiversity Branch. Victoria, BC. Wildlife Bull. B-104: 25 pp.

Lin D, Zhu L. 2006. Factors affecting transfer of polycyclic aromatic hydrocarbons from made tea to tea infusion. *J. Agric. Food Chem.* **54**(12): 4350-4354.

Lo JC, Letinski DJ, Parkerton TF, Campbell DA, Gobas FPC. 2016. In vivo biotransformation rates of organic chemicals in fish: Relationship with bioconcentration and biomagnification factors. Environ Sci Technol 50: 13299-13308.

Lodovici M, Dolara P, Casalini C, Ciappellano S, Testolin G. 1995. Polycyclic aromatic hydrocarbon contamination in the Italian diet. *Food Addit. Contam.* **12**(5): 703-713.

Lorber M. 2002. A pharmacokinetic model for estimating exposure of Americans to dioxin-like compounds in the past, present, and future. Sci Total Environ 288: 81–95.

Lundin JI, Riffell JA, Wasser SK. 2015. Polycyclic aromatic hydrocarbons in caribou, moose, and wolf scat samples from three areas of the Alberta oil sands. Environ Pollut 206: 527–534.

Maffei M, Mucciarelli M, Scannerini S. 1994. Area leaf area index (LAI) and flowering related to oil productivity in peppermint? Flavour Fragr J 9: 119-124.

McWilliams SR, Raveling DG. 2004. Energetics and time allocation of cackling Canada geese during spring. Biology and Management of Canada Geese. Proc. 2nd International Canada Goose Symposium. Milwaukee, WI. pp. 99–110.

Medford BA, Mackay WC. 1977. Protein and lipid content of gonads, liver, and muscle of Northern pike (*Esox lucius*) in relation to gonad growth. J Fish Res Board Can 35: 213–219.

Miller DR. 1976. Biology of the Kaminuriak population of barren-ground caribou: Part 3. pp. 1–25.

Moir D, Viau A, Chu I, Withey J, McMullen E. 1998. Pharmacokinetics of benzo [a] pyrene in the rat. J. Toxicol. Env. Health Part A 53(7), 507-530.Moorman TE, Baldassarre GA, Richard DM. 1992. Carcass mass, composition and gut morphology dynamics of mottled ducks in fall and winter in Louisiana. Condor 94: 407–417.

Møller HB, Moset V, Brask M, Weisbjerg MR, Lund P. 2014. Feces composition and manure derived methane yield from dairy cows: Influence of diet with focus on fat supplement and roughage type. Atmos Environ 94: 36-43.

Moreau, M., Bouchard, M. 2015. Comparison of the kinetics of various biomarkers of benzo [a] pyrene exposure following different routes of entry in rats. J Appl Toxicol 35(7): 781-790.

Moser G, McLachlan MS. 2002. Modeling digestive tract absorption and desorption of lipophilic organic contaminants in humans. Environ Sci Technol 36: 3318–3325.

Niimi AJ, Palazzo V. 1986. Biological half-lives of eight polycyclic aromatic hydrocarbons (PAHs) in

rainbow trout (*Salmo gairdneri*). Water Res 20: 503–507.

Norstrom RJ, Clark TP, Enright M, Leung B, Drouillard KG, Macdonald CR. 2007. ABAM, a model for bioaccumulation of POPs in birds: validation for adult herring gulls and their eggs in lake Ontario. Environ Sci Technol 41: 4339–4347.

Ohiozebau E, Tendler B, Codling G, Kelly E, Giesy JP, Jones PD. 2017. Potential health risks posed by polycyclic aromatic hydrocarbons in muscle tissues of fishes from the Athabasca and Slave Rivers, Canada. Environ Geochem Health 39: 139–160.

Ohiozebau E, Tendler B, Hill A, Codling G, Kelly E, Giesy JP, et al. 2016. Products of biotransformation of polycyclic aromatic hydrocarbons in fishes of the Athabasca/Slave river system, Canada. Environ Geochem Health 38: 577–591.

Orecchio S, Paradiso Ciotti V, Culotta L. 2009. Polycyclic aromatic hydrocarbons (PAHs) in coffee brew samples: Analytical method by GC-MS, profile, levels and sources. *Food Chem. Toxicol.* **47**(4): 819-826.

O'Reilly C, Owens JN. 1988. Shoot, needle, and cambial growth phenology and branch tracheid dimensions in provenances of lodgepole pine. Can J Forest Res 19: 599-605.

Papa, E., Sangion, A., Arnot, J. A., Gramatica, P. 2018. Development of human biotransformation QSARs and application for PBT assessment refinement. Food Chem Toxicol 112: 535-543.

Pekins PJ, Gessaman JA, Lindzey FG. 1994a. Field metabolic rate of blue grouse during winter. Can J Zool 72: 227–231.

Pendergrast BA, Boag DA. 1970. Seasonal changes in diet of spruce grouse in central Alberta. J Wildlife Manage 34: 605–611.

Pulkrabova J, Stupak M, Svarcova A, Rossner P, Rossnerova A, Ambroz A, et al. 2016. Relationship between atmospheric pollution in the residential area and concentrations of polycyclic aromatic hydrocarbons (PAHs) in human breast milk. Sci Total Environ 562: 640-647.

Quinn CL, Wania F. 2012. Understanding differences in the body burden-age relationships of bioaccumulating contaminants based on population cross sections versus individuals. Environ Health Perspect 120: 554–559.

Ramesh, A., Inyang, F., Hood, D. B., Archibong, A. E., Knuckles, M. E., Nyanda, A. M. 2001. Metabolism, bioavailability, and toxicokinetics of Benzo (α) pyrene in F-344 rats following oral administration. Exp Toxicol Pathol 53(4): 275-290.

Raveling DG. 1979. The annual cycle of body composition of Canada Geese with special reference to control of reproduction. The Auk 96: 234–252.

Reese EO, Robbins CT. 1994. Characteristics of moose lactation and neonatal growth. Can J Zool 72: 953–957.

Reid DEB, Silins U, Lieffers VJ. 2006. Sapwood hydraulic recovery following thinning in lodgepole pine. Ann For Sci 63:329–338.

Remington TE, Braun CE. 1988. Carcass composition and energy reserves of sage grouse during winter. Condor 90: 15–19.

Renecker LA, Hudson RJ. 1986. Seasonal foraging rates of free-ranging moose. J Wildlife Manage 50: 143–147.

Rusch DH, Keith LB. 1971. Seasonal and annual trends in numbers of Alberta ruffed grouse. J Wildlife

Manage 35: 803.

Safley CD, Cline WO, Mainland CM. 2009. Evaluating the profitability of blueberry production. 1–18.

Scott WB, Crossman EJ. 1998. *Freshwater fishes of Canada*. Galt House Publications, Oakville, Ontario, Canada.

Shipley LA. 2010. Fifty years of food and foraging in moose: lessons in ecology from a model herbivore. ALCES 46: 1–13.

Tfouni SAV, Serrate CS, Leme FM, Camargo MCR, Teles CRA, Cipolli KMVAB, et al. 2013. Polycyclic aromatic hydrocarbons in coffee brew: Influence of roasting and brewing procedures in two *Coffea* cultivars. *LWT – Food Sci. Technol.* **50**(2): 526-530.

Tharakan PJ, Volk TA, Nowak CA, Abrahamson LP. 2005. Morphological traits of 30 willow clones and their relationship to biomass production. Can J For Res 35: 421–431.

Tharakan PJ, Volk TA, Nowak CA, Ofezu GJ. 2008. Assessment of canopy structure, light interception, and light-use efficiency of first year regrowth of shrub willow (*Salix sp.*). Bioenerg Res 1: 229–238.

Undeman E, Czub G, McLachlan MS. 2009. Addressing temporal variability when modeling bioaccumulation in plants. Environ Sci Technol 43: 3751–3756.

Undeman E, Gustafsson E, Gustafsson BG. 2014. A novel modeling tool with multi-stressor functionality for organic contaminant transport and fate in the Baltic Sea. Sci Total Environ 497-498: 382–391.

Viau C, Carrier G, Vyskočil, Dodd C. 1995. Urinary excretion kinetics of 1-hydroxypyrene in volunteers exposed to pyrene by the oral and dermal route. Sci Total Environ 163: 179-186.

Viñas P, Campillo N, Aguinaga N, Pérez-Cánovas E, Hernández-Córdoba M. 2007. Use of headspace solid-phase microextraction coupled liquid chromatography for the analysis of polycyclic aromatic hydrocarbons in tea infusions. *J. Chromatogr. A.* **1164**(1-2): 10-17.

Wang L, Liu A, Zhao Y, Mu X, Huang T, Gao H, et al. 2018. The levels of polycyclic aromatic hydrocarbons (PAHs) in human milk and exposure risk to breastfed infants in petrochemical industrialized Lanzhou Valley, Northwest China. Environ Sci Pollut Res 25: 16754-16766.

Withey, J. R., Law, F. C. P., Endrenyi, L. 1991. Pharmacokinetics and bioavailability of pyrene in the rat. J Toxicol Env Health Part A 32(4): 429-447.