# **ELECTRONIC SUPPLEMENTARY INFORMATION (ESI)**

# Exploring the potential influence of climate change and particulate organic carbon scenarios on the long-range transport and behavior of neutral organic contaminants in the Arctic environment

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### Section S1. Additional details on the GloboPOP model and previous model evaluations

For reference, the name of each zone of the GloboPOP model and approximate range in latitude is shown below.

Name of Zone	<u>Approximate Range in Latitude</u>
North Polar	64–90 °N
North Boreal	54–64 °N
North Temperate	38–54 °N
North Subtropic	25–54 °N
North Tropic	0–25 °N
South Tropic	0 - 25 °S
South Subtropic	25 – 39 °S
South Temperate	39 – 48 °S
South Subpolar	$48 - 67 \ ^{\circ}S$
South Polar	67 – 90 °S

Each zone includes 4 atmospheric layers (atmospheric boundary layer, lower/middle troposphere, middle/upper troposphere, stratosphere), surface ocean water, fresh water, fresh water sediments, cultivated soil, uncultivated soil, forest canopy and forest soil. In addition, the North Polar, South Subpolar and South Polar zones have seasonally-changing sea-ice cover. Shelf sediments are also included in the North Polar zone. Where appropriate (i.e., for zones where temperatures get low enough), seasonal snowpack is tracked over winter and into the spring melt period. Chemicals can partition between the gas phase and aerosols in the atmosphere, between suspended solids and water in the aqueous phases, between air, water and solids in the soil compartments and between water and solids in sediments. See Armitage et al.<sup>1</sup> for more details on the current version of the GloboPOP model.

The vertical resolution of the atmosphere (i.e. 4 layers) follows from the simplified 2-D model developed by Strand and Hov.<sup>2</sup> Strand and Hov compared model output from a 'coarse' low resolution 2-D atmospheric model (6 latitudinal bands, 4 vertical layers) and a more highly resolved 2-D model (75 latitudinal bands, 33 vertical layers).<sup>2</sup> Simulations with idealized and real conservative atmospheric tracers (<sup>222</sup>Rn, <sup>85</sup>Kr) revealed some differences between the two models with respect to vertical mixing and horizontal dispersion but in general, agreement between the two modeling tools was good. Based on these results, Strand and Hov concluded

that the low resolution model was able to reproduce, "in an acceptable manner most of the large scale transport properties of the comprehensive model".<sup>2</sup> While there are still limitations related to the zonal averaging and coarse spatial resolution of the GloboPOP model (e.g. specific point sources and episodic transport events cannot be represented), the model is deemed acceptable for long-term simulations of contaminants (particularly those with widely-dispersed emissions).

Using realistic emission estimates, the GloboPOP model has been used previously to simulate the global-scale fate and transport of hexachlorocyclohexane (HCHs), polychlorinated biphenyls (PCBs) and perfluorooctanoate (PFO<sup>-</sup>).<sup>3-7</sup> The global-scale fate and transport of dichlorodiphenyl-trichloroethane (DDT), perfluorooctane sulfonate (PFOS) and endosulfan has been simulated using the CliMoChem model,<sup>8-10</sup> a tool similar to GloboPOP with respect to spatial resolution and process description. While agreement between model output and measurements is never 'perfect' in these exercises, it is frequently within a factor of 2–10 of the available empirical data. More importantly, spatial (i.e. latitudinal gradients) and temporal trends (e.g. relative change over time in different latitudinal bands) are captured reasonably well. For example, global fractionation of PCBs modeled by the GloboPOP tool agree well with key observations.<sup>5</sup> Given that the performance of more highly-resolved models is not greatly improved when considering long-term simulations (e.g. model output for atmospheric levels typically within a factor of 2-5 of observations, see Scheringer<sup>11</sup> and references therein), the general consensus is that the models such as GloboPOP and CliMoChem are suitable for certain applications (e.g., long-term simulations using realistic emission estimates, screening assessments of long-range transport potential to remote regions) but not others (e.g. short-term, site-specific evaluations, episodic transport events).

### Section S2. Additional details on global climate change (GCC) scenarios

*Temperature and Precipitation Rate.* Changes in temperature (surface air, surface ocean) and precipitation rate (over land, over ocean) on a monthly basis for each model zone assumed for the GCC scenarios are summarized in Tables S1–S5. No changes were made to the frequency of precipitation events, only the amount of precipitation falling over the year. All terrestrial surface compartments follow the surface air temperature except during periods of snow cover (where surface temperature is maintained at a constant 0 °C). Freshwater also follows surface air

temperature until onset of winter/ice cover. Note that the minimum temperature of sea-water is set at  $\sim -2$  °C.

The GloboPOP model includes a surface air layer (1000–835 hPa) and three additional upper layers (835–500 hPa, 500–110 hPa, 110–10 hPa). Mean annual temperature changes summarized in the IPCC 4<sup>th</sup> assessment<sup>12-14</sup> indicate relatively modest uniform increases globally between approximately 800–400 hPa. Beyond 200 hPa, more intense warming is projected for latitudes between approximately 60 °N and 60 °S while cooling is projected for the more polar latitudes. This GCC scenario was represented by assuming that temperatures in the 2<sup>nd</sup> and 3<sup>rd</sup> layer increase uniformly by 2 °C (all model zones, all months). The temperature scenario for the top layer is presented in Table S3.

Note that the scenarios presented in Tables S1–S5 are based on consideration of IPCC and other projections<sup>12-14</sup> for the latter part of the 21<sup>st</sup> century (2080–2099) but do not correspond to a specific IPCC emission scenario (e.g. the A1 or B2 family) or formal calculations (e.g. spatial aggregation) of the corresponding climate projections. Given the coarse spatial resolution of the GloboPOP model (see above), the main goal of the GCC scenario development for the current application was to derive a parameter set of a 'future climate' that is broadly consistent with the IPCC projections in terms of latitudinal trends. Overall, the temperature increases applied to the monthly means in the default parameterization (Tables S1–S3) are most consistent with the averages of projections from a set of different model simulations (i.e., the multimodel dataset, MMD) for the A1B scenario. Specifically, the temperature changes refer to a comparison of MMD-A1B scenario projections, the GCC scenario employed here can be considered 'intermediate' with respect to the magnitude of temperature increases.

Region	Jan	Feb	March	April	May	June	July	Aug	Sept	Oct	Nov	Dec
N.Polar	8.0	8.0	4.0	4.0	4.0	1.0	1.0	1.0	10.0	10.0	10.0	8.0
N.Boreal	6.0	6.0	3.5	3.5	3.5	2.5	2.5	2.5	4.5	4.5	4.5	6.0
N.Temperate	3.5	3.5	3.0	3.0	3.0	3.8	3.8	3.8	3.5	3.5	3.5	3.5
N. Subtropic	3.0	3.0	3.0	3.0	3.0	3.5	3.5	3.5	3.0	3.0	3.0	3.0
N. Tropic	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0
S. Tropic	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0
S. Subtropic	3.0	3.0	3.0	3.0	3.0	2.5	2.5	2.5	2.5	2.5	2.5	3.0
S. Temperate	3.0	3.0	2.5	2.5	2.5	2.5	2.5	2.5	3.0	3.0	3.0	3.0
S. Subpolar	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0
S. Polar	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0

Table S1. Changes in average monthly temperature (°C) in surface air for the future climate scenario compared to the default.

Table S2. Changes in average monthly temperature (°C) surface ocean water for the future climate scenario compared to the default.

Region	Jan	Feb	March	April	May	June	July	Aug	Sept	Oct	Nov	Dec
N.Polar	0.0	0.0	0.0	0.0	1.0	1.0	1.0	1.0	1.0	0.0	0.0	0.0
N.Boreal	0.0	0.0	0.0	0.0	1.0	1.0	1.0	1.0	1.0	0.0	0.0	0.0
N.Temperate	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2
N. Subtropic	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2
N. Tropic	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
S. Tropic	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
S. Subtropic	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2
S. Temperate	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2
S. Subpolar	1.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.0	1.0
S. Polar	1.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.0	1.0

Region	Jan	Feb	March	April	May	June	July	Aug	Sept	Oct	Nov	Dec
N.Polar	-1.5	-1.5	-1.5	-1.5	-1.5	-1.5	-1.5	-1.5	-1.5	-1.5	-1.5	-1.5
N.Boreal	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
N.Temperate	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
N. Subtropic	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
N. Tropic	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0
S. Tropic	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0
S. Subtropic	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
S. Temperate	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
S. Subpolar	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
S. Polar	-2.0	-2.0	-2.0	-2.0	-2.0	-2.0	-2.0	-2.0	-2.0	-2.0	-2.0	-2.0

Table S3. Changes in average monthly temperature (°C) in top atmospheric layer (110–10 hPa) for the future climate scenario compared to the default.

Region	Jan	Feb	March	April	May	June	July	Aug	Sept	Oct	Nov	Dec
N.Polar	25%	25%	15%	15%	15%	10%	10%	10%	20%	20%	20%	25%
N.Boreal	25%	25%	15%	15%	15%	10%	10%	10%	15%	15%	15%	25%
N.Temperate	10%	10%	5 <mark>%</mark>	5 <mark>%</mark>	5 <mark>%</mark>	<mark>-1</mark> 0%	<mark>-1</mark> 0%	<mark>-1</mark> 0%	10%	10%	10%	10%
N. Subtropic	<mark>-1</mark> 0%	<mark>-1</mark> 0%	<mark>-1</mark> 0%	0%	0%	<mark>-2</mark> 0%	<mark>-2</mark> 0%	<mark>-2</mark> 0%	10%	10%	10%	<mark>-1</mark> 0%
N. Tropic	<mark>-1</mark> 0%	<mark>-1</mark> 0%	<mark>-1</mark> 0%	0%	0%	<mark>-2</mark> 0%	<mark>-2</mark> 0%	<mark>-2</mark> 0%	0%	0%	0%	<mark>-1</mark> 0%
S. Tropic	<mark>-1</mark> 0%	<mark>-1</mark> 0%	<mark>-1</mark> 0%	0%	0%	<mark>-2</mark> 0%	<mark>-2</mark> 0%	<mark>-2</mark> 0%	0%	0%	0%	<mark>-1</mark> 0%
S. Subtropic	5 <mark>%</mark>	5 <mark>%</mark>	<mark>-5</mark> %	<b>-</b> 5%	<b>-5</b> %	<mark>-1</mark> 0%	<mark>-1</mark> 0%	<mark>-1</mark> 0%	<b>5</b> %	<mark>-5</mark> %	<mark>-5</mark> %	5 <mark>%</mark>
S. Temperate	5 <mark>%</mark>	5 <mark>%</mark>	<mark>-5</mark> %	<mark>-5</mark> %	<mark>-5</mark> %	<mark>-1</mark> 0%	<mark>-1</mark> 0%	<mark>-1</mark> 0%	<mark>-5</mark> %	<mark>-5</mark> %	<mark>-5</mark> %	5 <mark>%</mark>
S. Subpolar	10%	10%	12%	1 <mark>2%</mark>	12%	20%	20%	20%	1 <b>2%</b>	1 <b>2%</b>	1 <b>2%</b>	10%
S. Polar	10%	10%	12%	12%	1 <b>2%</b>	20%	20%	20%	1 <b>2%</b>	1 <b>2%</b>	1 <b>2%</b>	10%

Table S4. Percentage increase or decrease in precipitation rate over land in the future climate scenario compared to the default.

Table S5. Percentage increase or decrease in precipitation rate over oceans in the future climate scenario compared to the default.

Region	Jan	Feb	March	April	May	June	July	Aug	Sept	Oct	Nov	Dec
N.Polar	35%	35%	<mark>15%</mark>	<mark>15%</mark>	<mark>15%</mark>	<u>10</u> %	10%	<mark>10</mark> %	20%	20%	20%	35%
N.Boreal	<b>15%</b>	15%	15%	<mark>15%</mark>	15%	15%	<b>15%</b>	15%	<mark>15%</mark>	15%	1 <u>5%</u>	<b>15%</b>
N.Temperate	<b>5</b> %	<mark>5</mark> %	10%	10%	10%	10%	<b>10%</b>	<b>1</b> 0%	<b>5</b> %	<b>5</b> %	<b>5</b> %	5%
N. Subtropic	<b>10%</b>	10%	10%	10%	10%	10%	<b>10%</b>	10%	10%	<b>10%</b>	<b>1</b> 0%	10%
N. Tropic	15%	15%	15%	15%	15%	15%	15%	15%	<mark>15%</mark>	15%	15 <u>%</u>	<b>15%</b>
S. Tropic	15%	15%	15%	15%	15%	15%	15%	15%	15%	15%	15%	-15%
S. Subtropic	15%	15%	15%	15%	15%	15%	15%	15%	15%	15%	15%	-15%
S. Temperate	15%	15%	15%	15%	15%	15%	15%	15%	15%	15%	15%	-15%
S. Subpolar	<mark>10</mark> %	<u>10</u> %	<mark>12</mark> %	<mark>12</mark> %	<mark>12</mark> %	20%	20%	20%	<mark>12</mark> %	<mark>12</mark> %	<mark>12</mark> %	<u>10</u> %
S. Polar	10%	10%	<mark>12</mark> %	<mark>12</mark> %	12%	20%	20%	20%	12%	12%	12%	<mark>10</mark> %

*Arctic sea-ice cover, freshwater ice and seasonal snow pack.* Ranges of estimated sea-ice cover for the ACIA baseline period (1981–2000) and projections for the future (B2 scenario) are summarized in Table S6.<sup>15</sup>

Time Period	Average or Projected Values					
B2 Scenario (IPCC)	Sea-ice extent (10 <sup>6</sup> ki	$m^2$ )				
	March	September				
1981–2000	16.14–16.32	7.02–7.41				
2011–2030	15.00-15.62	3.33-7.00				
2041–2060	13.94–14.97	0.55–6.72				
2071–2090	13.26–14.52	0.05–6.59				

Table S6. Range of projected sea-ice extent compared to baseline (1981–2000) presented in ACIA<sup>15</sup> for the B2 scenario.

As apparent from Table S6, there is substantial variability in model output for these projections. Given that estimated sea-ice extents (2000–2011) for September are already well below  $5 \times 10^{6}$  km<sup>2</sup> (National Snow and Ice Data Center, www.nsidc.org), it is reasonable to adopt the lower bound of the projections to serve as guidance for scenario development. Accordingly, sea-ice extent was assumed to reach negligible levels by August/September for these simulations (minimum) but similar maximum extents over the winter period. The GloboPOP default and future sea-ice cover scenarios are compared in Figure S1. Note that multi-year ice is not explicitly considered in either scenario.



Figure S1. Comparison of % sea-ice cover (SIC) in the North Polar zone over the course of the year for the default and future climate scenario. Month 1 = January, Month 12 = December

Dibike et al.<sup>16</sup> recently published a modelling study aiming to characterize changes in lake-ice cover and lake-water thermal structure patterns in response to GCC. The A2 scenario was used to generate projections of changes in these parameters for the period 2040–2079 as a function of latitude. The period of open water was projected to increase by 15–50 days with the largest changes found in parts of the northern Boreal and high Arctic latitudes. For the GCC scenario, the period of open ice in freshwater system (linked to temperature change) increases by 35, 40 and 55 days in the North Temperate, Boreal and Polar zones respectively, broadly consistent with these projections.

Seasonal snow pack in the GloboPOP model is linked to precipitation rate in the winter (i.e. when air temperatures fall below zero). While not directly linked to air temperature, it was assumed that the snow melt period in the GCC scenario is 10 days shorter (i.e. faster melt). The evolution of the seasonal snow pack linked to the GloboPOP default and GCC scenarios over the annual cycle is compared in Figure S2. No changes to other snow properties (e.g. specific surface area, density) were included.



Figure S2. Comparison of seasonal snow-pack development (depth in m) in the a) North Polar and b) North Boreal zones for the default and future climate scenario

*Enhanced primary production and other organic carbon inputs in Arctic freshwater environments.* As noted in the main text, changes in diatom community structure have already been observed in high Arctic lakes.<sup>17-20</sup> A common observation is the relative enhancement of planktonic (pelagic) taxa at the expense of benthic taxa.

The GCC+OC scenario assumes a 50% increase in the volume fraction of particulate organic carbon (POC) in the freshwater compartment and a 50% increase in the organic carbon fraction of the active sediment layer in the underlying sediments as a surrogate for these responses. All scenarios (i.e., marine and freshwater) assume no change to OC mineralization rate in the water column and sediments and also no change in the sorption capacity of OC with respect to interactions with organic contaminants. As in the marine compartment, the mass transport coefficient (MTC) characterizing particle settling out of the water column was increased by the same factor as the volume fraction of POC. Note that volume fractions of POC in surface waters and sediments and particle settling MTCs in all other model regions were not altered.

For the purposes of this exercise, no distinction is drawn between the various sources of (particulate) organic carbon and the bulk levels reflect inputs via primary productivity (i.e. algal, planktonic), run-off/erosion from terrestrial sources and suspended fecal or other decomposing

organic matter (and the associated microbial communities). Information on how these inputs are changing now and may change in the future is summarized below.

# Section S3. Temperature-dependence of partitioning properties and reaction kinetics assumed for current simulations

*Temperature-dependence of partitioning properties.* The temperature-dependence of the three partitioning properties described above is a key consideration in the context of simulating the potential influence of GCC on chemical fate and transport. This temperature-dependence is accounted for in the GloboPOP model using the Van't Hoff equation and internal energies of phase transition ( $dU_{ij}$ , kJ mol<sup>-1</sup>). The general form of the equation is shown below.

$$K_{ij-2} = K_{ij-1} \exp\left[\frac{dU_{ij}}{R} \left(\frac{1}{T_1} - \frac{1}{T_2}\right)\right]$$
(S1)

where  $T_1$  and  $K_{ij-1}$  are the reference temperature (K) and the partition coefficient at  $T_1$  respectively,  $T_2$  and  $K_{ij-2}$  are environmental temperature and adjusted partition coefficient value respectively and *R* is the gas law constant.

As with the partition coefficients, the internal energies of phase transition must also be internally consistent<sup>21</sup> and hence only two of three are defined for the set of hypothetical chemicals. In general, the octanol-water partition coefficient ( $K_{OW}$ ) is expected to be the least sensitive to temperature. A generic value of -20 kJ mol<sup>-1</sup> for the internal energy of phase transition ( $dU_{OW}$ ) is often assumed in the absence of empirical data.<sup>21</sup> This default value for  $dU_{OW}$  is based on assuming a  $dU_O$  value of 0 kJ mol<sup>-1</sup> (i.e. negligible temperature-dependence in octanol) and a  $dU_W$  value of 20 kJ mol<sup>-1</sup>. Using this assumption and data on the temperature-dependence of octanol-air partitioning ( $dU_{OA}$ ) as a function of log  $K_{OA}$  for various POPs,<sup>21-24</sup> the following approach was adopted for these simulations:

$$dU_{OW} = -20 \text{ kJ mol}^{-1}$$
  $dU_{OA} = \text{see Table S7}$   $dU_{AW} = dU_{OW} - dU_{OA}$  (S2)

log <i>K</i> <sub>OA</sub> (at 25 °C)	<i>dU</i> <sub>0A</sub> ( <b>kJ mol</b> <sup>-1</sup> )	log K <sub>OA</sub> (at 25 °C)	dU <sub>OA</sub> (kJ mol <sup>-1</sup> )
4	-47	10	-95
5	-55	11	-103
6	-63	12	-111
7	-71	13	-119
8	-79	14	-127
9	-87	15	-135

Table S7. Temperature-dependence of the octanol-air partition coefficient ( $K_{OA}$ ) assumed for these simulations.

These temperature-dependency assumptions are likely to be most reliable for the portion of chemical space overlapping with the properties of the chemicals used to derive the relationship between  $dU_{OA}$  and log  $K_{OA}$  (e.g. similar to PCBs, some organochlorine pesticides). The validity of these assumptions for chemical space outside of this domain is uncertain. Assuming the same temperature-dependence for all hypothetical chemicals, an approach adopted in previous investigations using GloboPOP is deemed less preferable, given the available data.<sup>21-24</sup>

*Temperature-dependence of reaction kinetics.* Degradation rate constants are adjusted using activation energy (kJ mol<sup>-1</sup>) and a similar equation as shown above for the partitioning properties. The activation energies assumed for the current simulations are presented in Table S8. Note that an activation energy of 30 kJ mol<sup>-1</sup> corresponds to a factor of approximately 1.5 change in reaction kinetics for a 10 °C change in temperature.

Compartment	Activation Energy (kJ mol <sup>-1</sup> )					
Atmosphere (gas phase)	10					
Water (fresh, marine)	30					
Snow pack	30					
Forest canopy	30					
Sediments (fresh, shelf)	30					
Soil (all)	30					

Table S8. Activation energies assumed to apply to the degradation reactions of all hypothetical chemicals.

Degradation reactions in the atmosphere are assumed to occur only in the gas phase (i.e. the fraction of chemical sorbed to aerosols is shielded). All other reaction half-lives apply to the bulk phase.

### Section S4. Additional details on global emission scenario

The spatial distribution of primary emissions assumed for all hypothetical chemicals is presented in Table S9. Note that in this scenario, which is based on the estimated spatial distribution of PCB emissions, approximately 95% of the total annual emissions occur in the Northern hemisphere, the majority in the North Temperate zone (38-54 °N).

Zone	% of total annual	emissions
North Polar	0.00	
North Boreal	5.86	
North Temperate	59.85	~ 95%
North Subtropic	23.33	
North Tropic	6.46	
South Tropic	2.29	
South Subtropic	2.13	
South Temperate	0.07	~ 5%
South Subpolar	0.01	
South Polar	0.00	

Table S9. Spatial distribution of emissions assumed for uptake period.

Emissions were assumed to reach peak annual levels in June in the North Hemisphere and January in the South Hemisphere, with the maximum emissions being approximately three times higher than the minimum.

## Section S5. Model output under different assumptions for POC levels in Arctic Ocean

Comparisons between default and GCC+OC scenario model output (atmosphere, freelydissolved concentration in surface ocean water) for the primary emission phase (Year 0–20) are shown for simulations assuming default POC concentrations of 20  $\mu$ g L<sup>-1</sup> and 200  $\mu$ g L<sup>-1</sup> in Figure S3.



Figure S3. Comparison of model output from the GCC+OC scenarios to the default scenario in Year 20 for average concentration in surface air ( $C_A$ ) and iii) average freely-dissolved concentration in surface ocean water ( $C_O$ ) assuming the concentration of POC in the water column of 20 µg L<sup>-1</sup> (~ 1.5 µM) and 200 µg L<sup>-1</sup> (~ 15 µM). The yellow dashed circle and red arrows highlight the role of hydrophobicity (i.e. log  $K_{OW}$ ) in determining the response at a given level of POC in the water column.

As can be seen in Figure S3, responses in the atmosphere are essentially identical in both POC concentration scenarios. The main difference between the two POC scenarios with respect to the response of the freely-dissolved concentration in surface ocean water ( $C_0$ ) is seen for chemicals with log  $K_{OW}$  between 5 and 8 and log  $K_{AW} < -1$ . As POC levels are reduced in the default scenario, an equivalent response to doubling of POC levels requires a shift towards more hydrophobic chemicals (i.e. higher log  $K_{OW}$ ). These results imply that sensitivity/responses to POC-related changes in the Arctic marine for chemicals with such partitioning properties may vary regionally (e.g. high POC shelf locations vs. low POC central basin locations).

#### Section S6. Summary of general fate and transport behavior

*Long-range transport and accumulation potential.* The Arctic Contamination Potential ( $eACP_{10}$ ) of sets of hypothetical chemicals as a function of partitioning properties and susceptibility to degradation is displayed in Figure S4.<sup>1</sup> As in the main model application, emissions were directed 100% to the atmosphere. The three degradation scenarios are (i) perfect persistence (i.e. negligible degradation rates), (ii) 10:20:100 year baseline degradation half-lives in water, soil and sediments respectively and a baseline  $k_{OH}$  of  $1\cdot10^{-13}$  cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup> and (iii) 1:2:10 year baseline degradation half-lives in water, soil and sediments respectively and a baseline *k*<sub>OH</sub> of  $1\cdot10^{-12}$  cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup>. The scenario assuming perfect persistence can be viewed as the long-range transport potential of these hypothetical chemicals that stems solely from partitioning properties. A detailed discussion of the relationships between partitioning properties, susceptibility to degradation and eACP<sub>10</sub> can be found in two publications by Wania.<sup>25,26</sup>



Figure S4. Arctic Contamination Potential  $(eACP_{10})$  assuming (A) perfect persistence, (B) 10:20:100 year baseline degradation half-lives in water, soil and sediments respectively and a baseline  $k_{OH}$  of  $1 \cdot 10^{-13}$  cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup> and (C) 1:2:10 year baseline degradation half-lives in water, soil and sediments respectively and a baseline  $k_{OH}$  of  $1 \cdot 10^{-12}$  cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup>.  $eACP_{10}$  values are normalized to the maximum value for each degradation scenario. Log  $K_{OW}$  values of 5 and 8 are indicated by the white diagonal lines.

Maximum  $eACP_{10}$  values for the Perfect Persistence scenario are observed for chemicals with relatively low hydrophobicity (i.e.  $\log K_{OW} < 5.5$ ) and relatively high affinity for the water phase compared to air (i.e.  $\log K_{AW} < -1$ ). These chemicals have a low affinity for organic matter and

hence are less susceptible to "permanent" losses from the global environment such as sediment burial and particle settling to the deep ocean. Chemicals that are highly bound to aerosols in the atmosphere (log  $K_{OA} > 11$ ) exhibit relatively low  $eACP_{10}$  because of efficient deposition (wet, dry) and sequestration in soils and sediments during transport. These hydrophobic chemicals are also efficiently transported to the deep ocean if deposited to surface waters through particle settling.

The pattern in  $eACP_{10}$  is similar when assuming 10:20:100 year baseline degradation half-lives in water, soil and sediments respectively and a baseline  $k_{OH}$  of 1.10<sup>-13</sup> cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup> except that the maximum value shifts to chemicals which are more water–soluble (i.e., lower log  $K_{AW}$ ). Such chemicals are transported primarily via oceanic exchange and hence are shielded from degradation in the atmosphere (which occurs in the gas phase with a baseline half-life of approximately 80 days).

The shift in maximum  $eACP_{10}$  towards more water-soluble (i.e. low log  $K_{AW}$  chemicals) is even more evident when assuming 1:2:10 year baseline degradation half-lives in water, soil and sediments respectively and a baseline  $k_{OH}$  of  $1 \cdot 10^{-12}$  cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup>. However, chemicals with a high affinity for aerosols (i.e. log  $K_{OA} > 11$ ) exhibit a relatively high  $eACP_{10}$  compared to the maximum for this scenario. This behavior results from the fact that the mass of chemical bound to aerosols is assumed to be shielded from reactions with OH radicals in the atmosphere (which degrade the chemical with a baseline half-life of approximately 8 days).

*Mass distribution in Arctic Environment*. The pattern in mass distribution (bulk phase) in the Arctic environment after 20 years of constant emissions is displayed in Figure S5.



Figure S5. Mass distribution of the hypothetical chemicals in the Arctic environment following 20-years of constant emissions. Colored areas (e.g. light blue, dark blue, dark green) indicate partitioning property combinations where  $\geq 2/3^{rd}$ s of the total mass is present in a single medium.

As shown in Figure S5, the mass distribution broadly follows expectations based on partitioning properties. For example, volatile chemicals with low affinity for organic matter (i.e., high log  $K_{AW}$ , low  $K_{OA}$ ) are found predominantly in the atmosphere whereas hydrophilic chemicals with low affinity for organic matter (i.e., low log  $K_{AW}$ , log  $K_{OA}$ , low  $K_{OW}$ ) are found predominantly in the surface ocean water. Chemicals with higher log  $K_{OA}$  and/or log  $K_{OW}$  values (e.g. hydrophobic, non-volatiles) are associated predominantly with organic matter either in soils or shelf sediments.

Apparent dissipation half-lives (depuration phase). The apparent dissipation half-lifes (see main text) during the depuration phase (Year 20–40) as displayed as a function of log  $K_{AW}$  and log  $K_{OA}$  in Figure S6. Note that the apparent dissipation half-lives in Figure S6 were generated assuming the shorter degradation half-lives (i.e., faster degradation rates, see main text). However, all values were normalized to the lowest value (e.g. a value of 10 means that the apparent dissipation life is 10-fold higher than the lowest value) in order to generalize the pattern in apparent dissipation half-lives.



Figure S6. Apparent dissipation half-lives as a function of log  $K_{AW}$  and log  $K_{OA}$ , normalized to the lowest value.

As illustrated in Figure S6, chemicals predominantly associated with organic matter in terrestrial soils and shelf sediments exhibit the longest apparent dissipation half-lives over Year 20–40 of the simulation. The slow response to the complete cessation of emissions is related to the degradation half-lives assumed in these compartments relative to others (i.e. the 1:2:10 ratio for water, soil and sediments respectively) and also because organic matter associated with soils and shelf sediments is less mobile than other media, particularly the atmosphere. Other transport processes influencing the mass of chemical in soils and shelf sediments are also relatively inefficient. In other words, the overall residence time associated with degradation, intermedia exchange (e.g. revolatilization, sediment resuspension) and advection out of the model zone is highest for chemicals predominantly distributed to the soil and shelf sediment compartments.

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