Supporting Information

Modeling historical budget for β-Hexachlorocyclohexane (HCH) in the Arctic Ocean: A contrast to α-HCH

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Table

Table S1 Parameter A_{AW}, B_{AW}, A_{OA}, B_{OA}, and half-lives for α-HCH and β-HCH

Chemical	A_{AW}[a]	B_{AW}[a]	A_{OA}[b]	B_{OA}[b]	$t_{1/2air}$[c]	$t_{1/2water}$[c]	$t_{1/2soil}$[c]
α-HCH	10.2	-3990	-3.38	3230	1418	3500	14000
β-HCH	8.58	-3950	-5.99	4390	2127	5250	21000

Note:

a. A_{AW} and B_{AW} are used for temperature-dependent (T) air-water partition coefficient calculation (K_{AW}) as

$$\log K_{AW} = A_{AW}+B_{AW}/T$$

A_{AW} and B_{AW} are calculated as:

$$B_{AW} = \Delta U_{AW}/\ln(10)R$$

$$A_{AW} = \log H_0/RT_0 - B_{AW}/T_0$$

where, ΔU_{AW} is the internal energy of phase transfer between air and water in J mol^{-1}. Abraham et al. estimated the value using the LEFRs method [1]. R is gas constant, R = 8.314 kg m^2 s^{-2} K^{-1} mol^{-1}. H_0 is Henry's law constant at a specific temperature [2], T_0 is the specific temperature, T_0 = 273.15K.

b. A_{OA} and B_{OA} are used for temperature-dependent (T) octanol-air partition coefficient calculation (K_{OA}) as

$$\log K_{OA} = A_{OA}+B_{OA}/T$$

A_{OA} and B_{OA} are calculated as [3]:

$$B_{OA} = \Delta U_{AW}/\ln(10)R$$

$$A_{OA} = \log K_0 - B_{AW}/T_0$$

where, ΔU_{OA} is the internal energy of phase transfer between octanol and air in J mol^{-1}. K_{OA0} is octanol-air partition coefficient at a specific temperature.

c. $t_{1/2air}$, $t_{1/2water}$, $t_{1/2soil}$ are half-lives for α-HCH and β-HCH in air, water, and soil (h). considering half-life data of β-HCH is lacking. The common treatment is to relate the half-life of β-HCH to α-HCH with a factor [4]. According to the measure of Bidleman et al. [5], the half-life of β-HCH is three times larger than α-HCH's in lakes. However, the natural observed half-lives is not only related to the persistence to degradation in a phase but also related to the distribution among media because half-lives are different among media and chemical in different media can exchange with each other. For most chemicals (including HCHs), the half-life in water is longer than that in air. Because of their difference in K_{AW}, most of α-HCH distribute in air but β-HCH in water [2, 6]. For

HCHs, half-life in water is more than two times larger than that in air [4]. Thus, we roughly assumed the factor between half-lives of α-HCH and β-HCH is 1.5 ($t_{1/2\_Air}$=2127 h, $t_{1/2\_Ocean}$=5250 h, $t_{1/2\_Soil}$=21000 h). The corresponding half-life of α-HCH is collected from Wöhrnschimmel et al [4]. Degradation in snowpack and ice would not be considered in this model.

Table S2 Parameters of Mass transfer coefficients (MTCs)

Description	Symbol	Value	Unit
MTC for air's transfer to stratophere	MTC_{Air_Strat}	0.064 [7]	m/h
Air side air-water MTC	MTC_{AW}	30 [7]	m/h
Water side air-water MTC	MTC_{WA}	0.03 [7]	m/h
Soil side air-soil MTC	MTC_{AS}	1 [7]	m/h
Soil side air phase air-soil MTC	MTC_{SA1}	0.04 [7]	m/h
Soil side water phase air-soil MTC	MTC_{SA2}	1E-5 [7]	m/h
Soil side solid phase air-soil MTC	MTC_{SA3}	4.54E-5 [7]	m/h
Air side air-snow MTC	$MTC_{Air\text{-}Snow}$	5 [8]	m/h
Air side air-ice MTC	$MTC_{Air\text{-}Ice}$	5 [8]	m/h
Snow side air-snow MTC	$MTC_{Snow\text{-}Air}$	50[a]	m/h
Ice side air-ice MTC	$MTC_{Ice\text{-}Air}$	0.5[a]	m/h
Rain rate in Source region	MTC_{Rain_Source}	9.7E-5 [9]	m/h
Rain rate in Arctic region	MTC_{Rain_Arctic}	1.25E-5 [10]	m/h
Snow rate in Arctic region	MTC_{Snow_Arctic}	1.25E-5 [10]	m/h

Note:

a. $MTC_{Snow\text{-}Air}$ and $MTC_{Ice\text{-}Air}$ are given by Darcy's law:

$$MTC_{S\&I\_Air} = -\frac{P}{\mu}\frac{p_0}{ln2\ z_0} \times 3600$$

where P is snow and ice permeability, $P_{Snowpack}$ =5×10^{-9} m^2 and P_{Ice} =1×10^{-9} m^2 [8]. μ is air viscosity. In the Arctic, μ=1.58×10^{-5} Pa·s [8]. z_0 is roughness length. $z_0 = A_{sr}/30$. While A_{sr} is surface roughness amplitude, set to 0.1 m [8]. p_0 the amplitude of the pressure, Pa.

$$p_0 = \frac{3\rho_{Air} v^2 A_{sr}}{\lambda}$$

where ρ_{Air} is the density of air, ρ_{Air} = 1.3 kg/m^3. λ is the wavelength of the surface roughness, λ = 5 m [8].

Table S3 K values used in the AMBBM 2.0 model

Symbol	Formula
$K_{AW\_Media\_Region\_Season}$ [a]	$10^{A_{AW}+B_{AW}/T_{Media\_Region\_Season}}$
$K_{OA\_Media\_Region\_Season}$ [a]	$10^{A_{OA}+B_{OA}/T_{Media\_Region\_Season}}$
K_{GP} [b]	$10^{-2.91}\rho_{Particle}f_{OM\_Particle}K_{OA\_Air}$
K_{SA} [c]	$f_{OM\_Soil}K_{OA\_Air}$
$K_{Snowpack\_Air}$ [d]	$SA_{Snowpack}\rho_{Snowpack}K_{Ice\_Air\_dim}$
$K_{Snowflake\_Air}$ [d]	$SA_{Snowflake}\rho_{Snowflake}K_{Ice\_Air\_dim}$
K_{Ice_Air} [d]	$SA_{Ice}\rho_{Ice}K_{Ice\_Air\_dim}$

Note:

a. Temperature-dependent air-water partition coefficient (K_{AW}) and octanol-air partition coefficient (K_{OA}) are the most basic and important. $K_{AW\_Media\_Region\_Season}$ and $K_{OA\_Media\_Region\_Season}$ are K_{AW} and K_{OA} values in the specific media(air or ocean) at specific region (Source or Arctic) in specific season (warm season or cold season if the region is the Arctic). In the following equations, placeholders like "Media", "Region", and "Season" will be omitted if no specific value is set. A_{AW}, B_{AW}, A_{OA}, and B_{OA} are unique parameters for each chemical's K values calculation, given in Table S1. T refers to the temperature in the specific environment.

b. Harner-Bidleman equation [11] was used to describe gas/particle partition coefficient (dimensionless). $\rho_{Particle}$ is the density of particle, equals to 1500 kg/m^3. $f_{OM\_Particle}$ is fraction of the organic matter in the particle in volume, was set to 0.2.

c. K_{SA} is the partition coefficient between soil and gas [12]. f_{OM_Soil} is the fraction of organic matter in soil, $f_{OM\_Soil} = 0.04$.

d. This is a highlight to include snow and ice into Arcitc's mutilmedia model. The calculation process of snowflake-air, snowpack-air and ice-air partition coefficient is a bit more complicated. According to the work by Steinlin et al. [13]. These coefficients are calculated as the following:

$$K_{Snow\&Ice\_Air} = SA_{Snow\&Ice}\rho_{Snow\&Ice}K_{Ice\_Air\_dim}$$

$K_{Snow\&Ice\_Air}$ eliminate dimension of $K_{Ice\_Air\_dim}$ through multiply by specific surface area and density of ice or snow. The $SA_{S\&I}$ is the specific surface area of snowflake, snowpack, and sea ice. Based on their different structure, $SA_{Snowflake} = 96$ m^2/kg, $SA_{Snowpack} = 46$ m^2/kg, and $SA_{Sea\_ice} = 10$ m^2/kg [13, 14]. Be relavant to the SA, their densities are also different. $\rho_{Snowflake} = 150$ kg/m^3, $\rho_{Snowpack} = 400$ kg/m^3, and $\rho_{Sea\_Ice} = 900$ kg/m^3. The

$K_{\text{Ice\_Air\_dim}}$ is calculated as [15]:

$$K_{\text{Ice\_Air\_dim}} = K_{\text{Ice\_Air\_dim\_c}}\, exp\left[\frac{-\Delta H + RT_{\text{av}}}{R}\left(\frac{1}{T_{\text{Air\_Arctic\_Cold}}} - \frac{1}{T_{\text{cc}}}\right)\right]$$

where, $K_{\text{S\&I\_Air\_dim\_c}}$ is $K_{\text{S\&I\_Air\_dim}}$ at T_{cc} (-6.8°C) [15]. T_{av} is the average temperature in the considered temperature range, which equals to $T_{\text{Air\_Arctic\_Cold}}$. R is gas constant, R = 8.314 kg m^2 s^{-2} K^{-1} mol^{-1}. ΔH the enthalpy of sorption, which is also a function of $K_{\text{S\&I\_Air\_dim\_c}}$ [15]. $K_{\text{S\&I\_Air\_dim\_c}}$ can be calculated through pp-LEFR as [16]:

$$K_{\text{S\&I\_Air\_dim\_c}} = 10^{0.639L+3.53A+3.38B-6.85}$$

And,

$$\Delta H = -4.43 ln K_{\text{Ice\_Air\_dim \_c}} - 92.4$$

where L is logarithm of hexadecane–air partition coefficient, A is Electron acceptor property, and B is electron donor property.

Table S4 *Z* values used in the AMBBM 2.0 model

Symbol	Formula
Z_{Air}	$1/RT_{\mathrm{Air}}$
Z_{Paticle}	$K_{\mathrm{GP}}Z_{\mathrm{Air}}$
Z_{Water}	$Z_{\mathrm{Air}}/K_{\mathrm{AW\_Water}}$
$Z_{\mathrm{Snowflake}}$	$Z_{\mathrm{Air\_Arctic\_Cold}}K_{\mathrm{Snowflake\_Air}}$
Z_{Droplets}	$Z_{\mathrm{Air}}/K_{\mathrm{AW\_Air}}$
Z_{Soil}	$Z_{\mathrm{Air}}K_{\mathrm{SA}}$
$Z_{\mathrm{Snowflake}}$	$Z_{\mathrm{Air\_Arctic\_Cold}}K_{\mathrm{Snowflake\_Air}}$
Z_{Snowpack}	$Z_{\mathrm{Air\_Arctic\_Cold}}K_{\mathrm{Snowpack\_Air}}$
$Z_{\mathrm{Sea\_Ice}}$	$Z_{\mathrm{Air\_Arctic\_Cold}}K_{\mathrm{Ice\_Air}}$

Table S5 *D* values used in the AMBBM 2.0 model

Process	Symbol	Formula	Units
Advection			
Atmopheric inflow[a]	$D_{\mathrm{Tran\_Air\_SA}}$	$\frac{v}{\sqrt{A_{\mathrm{Arctic}}\pi/4}}V_{\mathrm{Arctic\_Air}}Z_{\mathrm{Air\_Arctic}}\cdot 24$	mol/(d·Pa)
Atmopheric outflow[a]	$D_{\mathrm{Tran\_Air\_AS}}$	$\frac{A_{\mathrm{Arctic}}}{A_{\mathrm{Source}}}\frac{v}{\sqrt{A_{\mathrm{Arctic}}\pi/4}}V_{\mathrm{Source\_Air}}Z_{\mathrm{Air}}\cdot 24$	mol/(d·Pa)
Current inflow[b]	$D_{\mathrm{Tran\_Current\_SA}}$	$Flow_{\mathrm{Current\_In}}Z_{\mathrm{Water\_Source}}\cdot 24$	mol/(d·Pa)
Current outflow[b]	$D_{\mathrm{Tran\_Current\_AS}}$	$Flow_{\mathrm{Current\_Out}}Z_{\mathrm{Water\_Arctic}}\cdot 24$	mol/(d·Pa)
River[c]	$D_{\mathrm{Tran\_River\_SA}}$	$f_{\mathrm{Runoff\_SA}}k_{\mathrm{Soil\_Runoff\_Source}}V_{\mathrm{Soil\_Source}}Z_{\mathrm{Soil\_Source}}\cdot 24$	mol/(d·Pa)
Soil runoff[c]	D_{Runoff}	$k_{\mathrm{Soil\_Runoff}}V_{\mathrm{Soil}}Z_{\mathrm{Soil}}\cdot 24$	mol/(d·Pa)
Degradation and physical loss			
Degradation in each phase[d]	D_{Deg}	$k_{\mathrm{Deg}}VZ\cdot 24$	mol/(d·Pa)
Air transfer to the Stratophere[e]	$D_{\mathrm{Air\_Strat}}$	$A\ MTC_{\mathrm{Air\_Strat}}Z_{\mathrm{Air}}\cdot 24$	mol/(d·Pa)
Seawater mixing to deep ocean[f]	$D_{\mathrm{Seawater\_sink}}$	$Flow_{\mathrm{Sink}}Z_{\mathrm{Water}}\cdot 24$	mol/(d·Pa)
Gas deposition			
Gas exchange with ocean[g]	$D_{\mathrm{Air\_Water}}$	$\frac{f_{\mathrm{Seawater}}A_{\mathrm{Ocean}}}{1/(MTC_{\mathrm{AW}}Z_{\mathrm{Air}})+1/(MTC_{\mathrm{WA}}Z_{\mathrm{Water}})}\cdot 24$	mol/(d·Pa)
Gas exchange with soil[g]	$D_{\mathrm{Air\_Soil}}$	$\frac{A_{\mathrm{Land}}}{1/(MTC_{\mathrm{AS}}Z_{\mathrm{Air}})+1/(MTC_{\mathrm{SA1}}Z_{\mathrm{Air}}+MTC_{\mathrm{SA2}}Z_{\mathrm{Water}}+MTC_{\mathrm{SA3}}Z_{\mathrm{Soil}})}\cdot 24$	mol/(d·Pa)
Gas exchange with snowpack[g]	$D_{\mathrm{Air\_Snowpack}}$	$\frac{A_{\mathrm{Arctic\_Land}}}{1/\left(MTC_{\mathrm{Air\_Snow}}Z_{\mathrm{Air\_Arctic\_Cold}}\right)+1/\left(MTC_{\mathrm{Snow\_Air}}Z_{\mathrm{Air\_Arctic\_Cold}}\right)}\cdot 24$	mol/(d·Pa)

Gas exchange with sea ice[g]	$D_{\mathrm{Air\_Sea\_Ice}}$	$\frac{(1-f_{\mathrm{Seawater}})A_{\mathrm{Arctic\_Sea\_Ice}}}{1/(MTC_{\mathrm{Air\_Ice}}Z_{\mathrm{Air\_Arctic\_Cold}})+1/(MTC_{\mathrm{Sea\_Air}}Z_{\mathrm{Air\_Arctic\_Cold}})}\cdot 24$	mol/(d·Pa)
Rain scavenging	D_{Rain}	$MTC_{\mathrm{Rain}}A\ Z_{\mathrm{Droplets}}\cdot 24$	mol/(d·Pa)
Snow scavenging	D_{Snow}	$MTC_{\mathrm{Snow}}A\ Z_{\mathrm{Snowflake}}\cdot 24$	mol/(d·Pa)
Particle deposition			
Dry deposition[h]	D_{DD}	$\alpha MTC_{\mathrm{Qd}}\ A\ \varphi_{\mathrm{Particle\_Air}}\ Z_{\mathrm{Particle}}\cdot 24$	mol/(d·Pa)
Wet deposition with rain[h]	$D_{\mathrm{WD\_Rain}}$	$\alpha MTC_{\mathrm{Rain}}A\ Q_{\mathrm{Rain}}\varphi_{\mathrm{Particle\_Air}}Z_{\mathrm{Particle}}\cdot 24$	mol/(d·Pa)
Wet deposition with snow[h]	$D_{\mathrm{WD\_Snow}}$	$\alpha MTC_{\mathrm{Snow}}A\ Q_{\mathrm{Snow}}\varphi_{\mathrm{Particle\_Air}}Z_{\mathrm{Particle}}\cdot 24$	mol/(d·Pa)

Note:

a. According to the residence time calculation equation of air used in the Simplebox 4.0 model [17], the D values are calculated. A_{Arctic} and A_{Source} are area of the Arctic and the source area, $A_{\mathrm{Arctic}}= 2.13\times10^{13}$ m^2, $A_{\mathrm{Source}}= 2.40\times10^{14}$ m^2. $V_{\mathrm{Arctic\_Air}}$ and $V_{\mathrm{Source\_Air}}$ are air volumn in the Arctic and source zone, where the height of the atmopshere is set to 6000 m. v is the wind speed of atmospheric transport, set to 8 m/s [8].

b. Ocean currents' transport has been discussed by Macdonald et al. [18]. The inflow discharge of surface ocean is 4.86×10^{13} km^3/y, outflow flux is 2.86×10^{13} km^3/y. The rest sinks to deeper water.

c. Rivers' budget comes from soil runoff in source area and the Arctic area, respectively. $D_{\mathrm{Tran\_River\_MA}}$ refers to river inflow from the source area. $k_{\mathrm{Soil\_Runoff\_Source}}$ is soil runoff rate in the source, which was set to 2×10^{-5} h^{-1} [17]. $f_{\mathrm{Runoff\_MA}}$ is the fraction of soil runoff in source to the Arctic. According to Li et al. [19], we assumed 3 ‰ of soil runoff in the source finally enter the AO and the rest flows into oceans in the source zone. However, runoff process only happened in the warm season in the Arctic, as in the cold season, soil is reckoned to be frozen. And $k_{\mathrm{Soil\_Runoff\_Arctic}}$ is much smaller, which was modified by rain rate in regions ($MTC_{\mathrm{Rain\_Arctic}}$ and $MTC_{\mathrm{Rain\_Source}}$) as:

$$k_{\mathrm{Soil\_Runoff\_Arctic}} = k_{\mathrm{Soil\_Runoff\_Arctic}} \cdot \frac{MTC_{\mathrm{Rain\_Arctic}}}{MTC_{\mathrm{Rain\_Source}}}$$

Rain rate in the source zone and the Arctic zone are 9.7×10^{-5} m/h [9] and 1.2×10^{-5} m/h [10].

d. Besides atmospheric circulation and ocean current, chemicals are mostly removed through degradation in phases, air's exchange with the stratophere, and ocean's exchange with deep ocean. Degradation rate (k_{Deg}) was calculated from half-lives in different phases and modefied by temperature.

$$k_{\mathrm{Deg}} = \frac{ln\ 2}{t_{1/2}} e^{E\left(\frac{1}{298.15\ R} - \frac{1}{T}\right)}$$

$t_{1/2}$ is half-live in different media at 298.15K, given in Table S1, E is activation energy for degradation process in different phases. Wöhrnschimmel et al [4] estimated E value of α-HCH and β-HCH and accepted in this work ($E_{\mathrm{Air}} = 1.12\times10^4$ J, $E_{\mathrm{Ocean}} = 1.073\times10^5$ J, $E_{\mathrm{Soil}} = 3\times10^4$ J).

e. Air will transport to upper air above 6000 m. By habit, it was named transfer to stratophere.

f. Surface sea water also can mix with deep water. According to the Macdonald et al.[18], the sinking flux is 2×10^{13} km^3/y.

g. Gas exchange between air with soil, sea water, snowpack, and sea ice are calculated by the two-film theory widely used in fugacity method[9]. $f_{Seawater}$ is the bare sea water area fraction in the AO and in source there is no sea ice. $f_{Seawater}$ = 0.6 in Arctic warm season and $f_{Seawater}$ = 0.7 in Arctic cold season. *MTC* value are given in Table S2.

h. Li et al. proposed the steady-state equation for G/P partition calculation [20]. This method has been proved to be more accurate for OCPs' stimulation [21] and successfully used in models [22, 23]. Based on the equilibrium gas/particle partition theory, a parameter α was added to correct environmental process about the particle in the air. α is calculated as [20]:

$$\alpha = 1/\left(1 + 4.18 \times 10^{-11} f_{\mathrm{OM\_Particle\_Air}} K_{\mathrm{OA}}\right)$$

$f_{OM\_Particle}$ is the fraction of organic matter in particle, which was set to 0.2 [24]. $\varphi_{Particle\_Air}$ is the volumn fraction of particle in the air. It can be calculated by total suspend particle (TSP, μg/m^3).

$$TSP = 10^{-9} \frac{TSP}{\rho_{\mathrm{Particle}}}$$

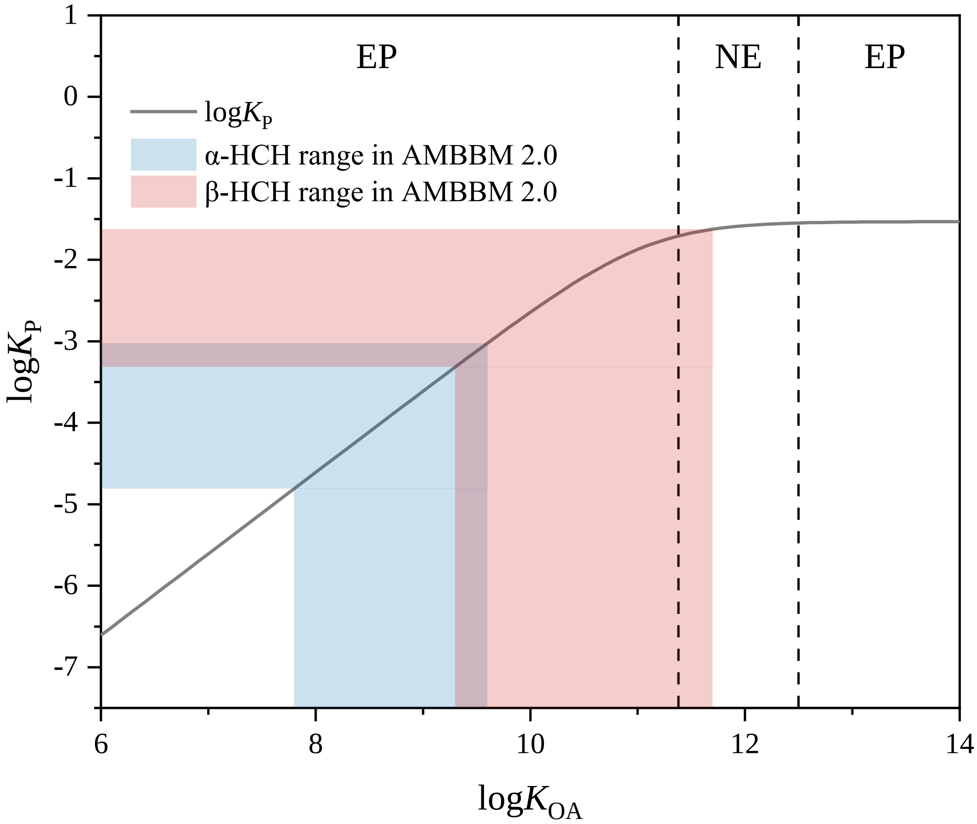
The atmophere in the Arcitc is clear, the total suspend particle in the Arctic (TSP_{Arctic}) is set to 1 μg/m [25]. Correspondingly, TSP_{Source} is set to 30 μg/m^3. Q_{Rain} and Q_{Snow} are scavenging ratio of rain and snow, Q_{Rain} = 200000 [9] and Q_{Snow} = 320000 [26].

Table S6 Dispersion factors of properties

Properties	**dispersion factors**
K_{AW}	**5**
K_{OA}	**5**
HL_{Air}	**10**
HL_{Water}	**10**
HL_{Soil}	**10**

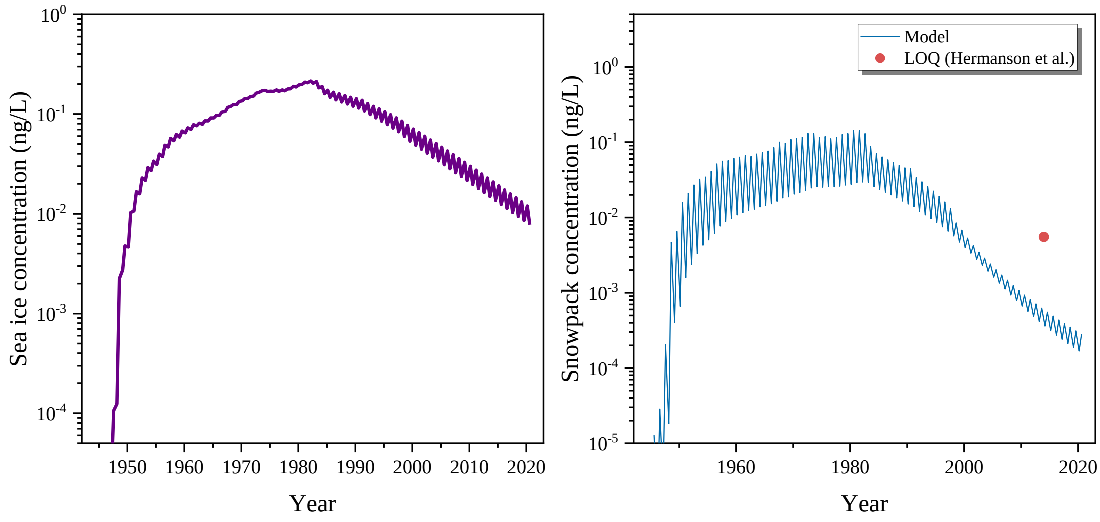
Figure

Figure S1 Gas/partition of α-HCH and β-HCH in AMBBM 2.0



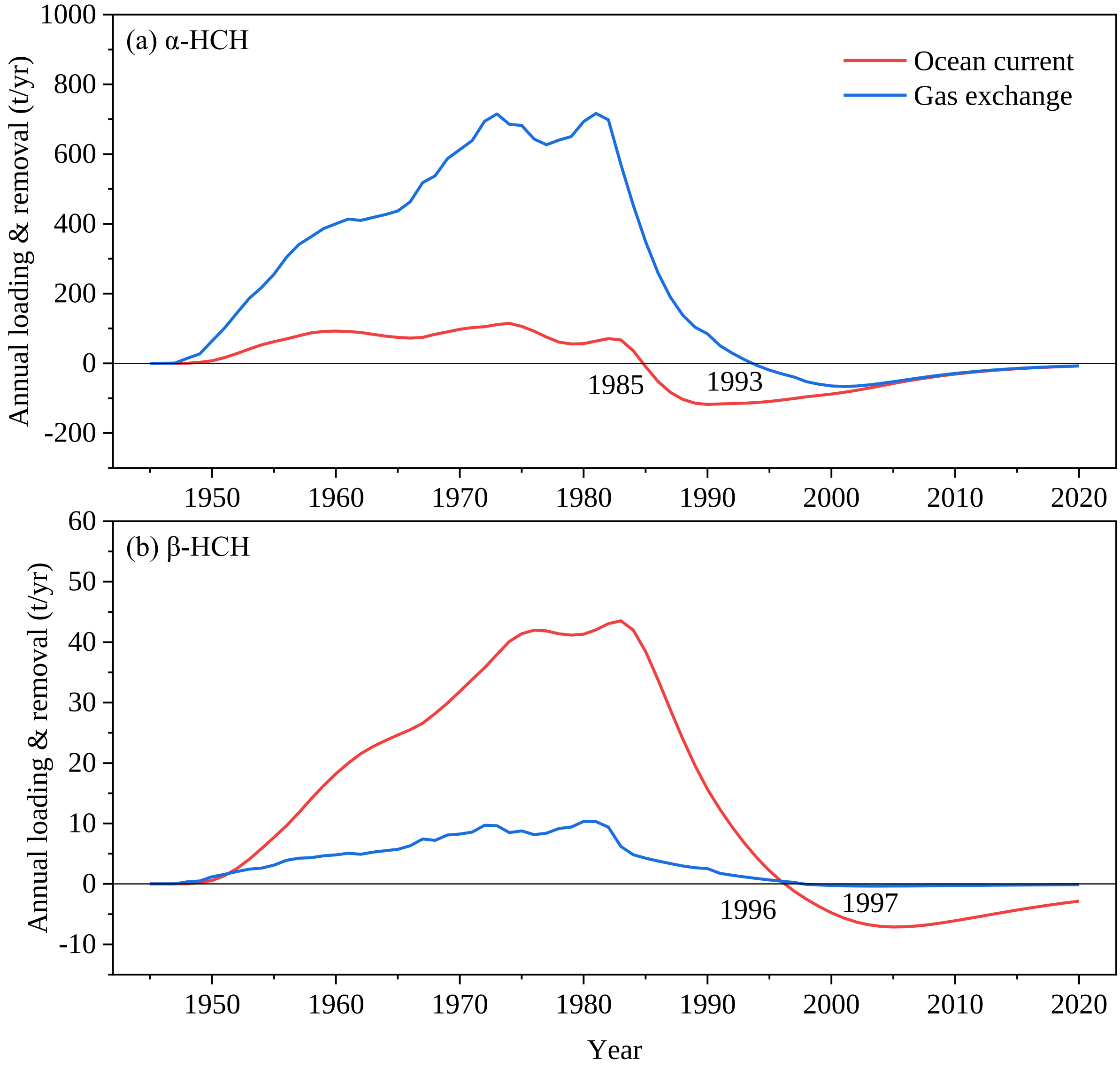
This figure depicts the possible logK_{OA} and logK_P range of α-HCH and β-HCH in the model, which indicates that the gas/particle partition of α-HCH and β-HCH are basicly both falled in the EP domain [24]. Thus, whether equilibrium or steady state assumption used in the model won't raise a huge influence on the result.

Figure S2 Concentration of β-HCH in snowpack and sea ice



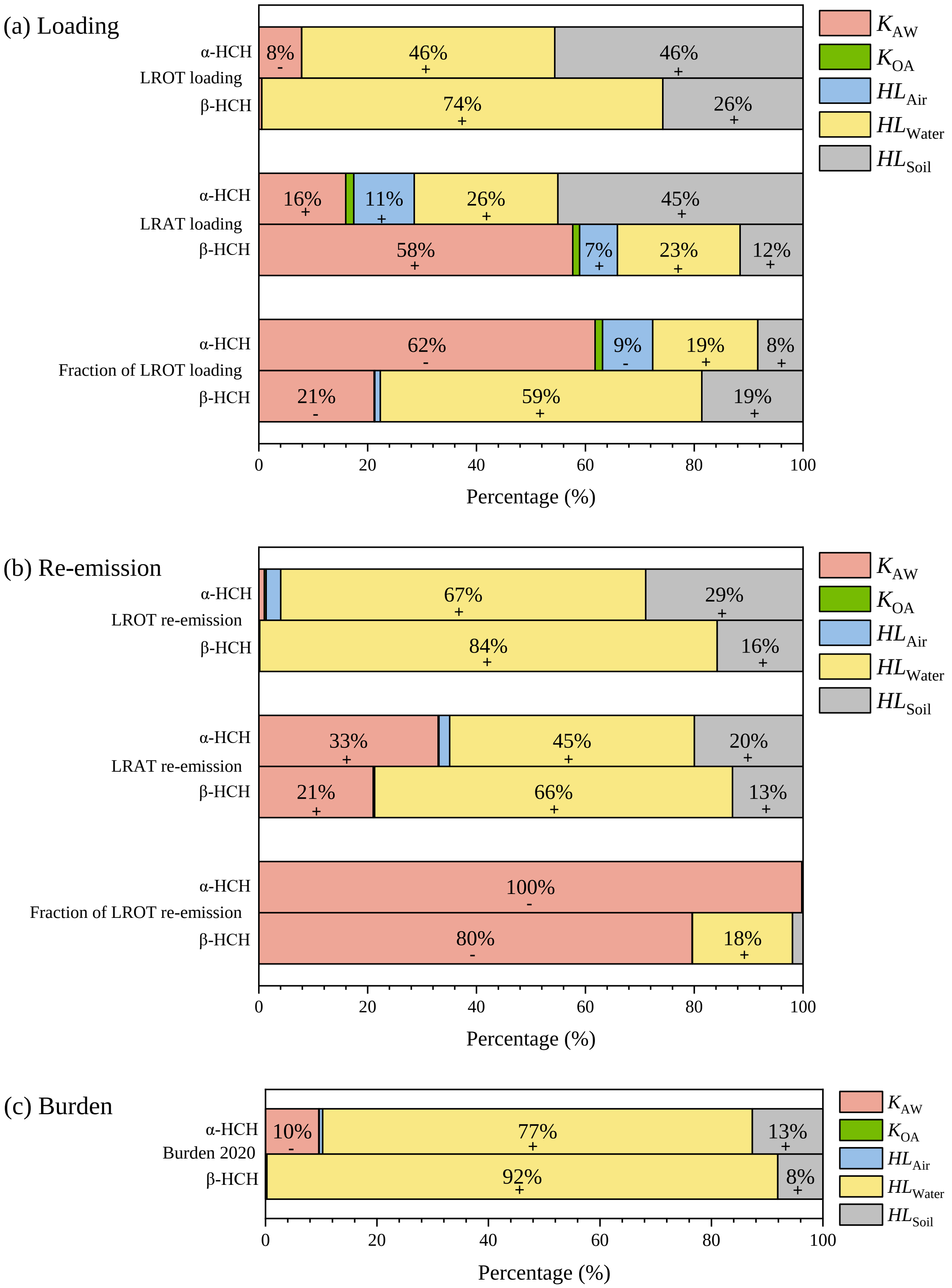
This figure depicts modeling results of sea ice and snowpack. The content of the figure is discussed in the section 4.1 in the manuscript.

Figure S3 "Switching point" of α-HCH and β-HCH



This figure depicts the yearly flux of ocean current and gas exchange. The top figure is α-HCH and the bottom figure is β-HCH. The red lines represent ocean current and the blue lines represent gas exchange. α-HCH's "switching point" of ocean current and gas-water exchange happened in 1985 and 1993. But β-HCH's "switching point" of them happened happened in 1996 and 1997.

Figure S4 Result of sensitivity analysis



This figure depicts the variance contribution of physical-chemical properties to HCHs' behaviors. The bars are depicted paired. The upper bar is α-HCH and the lower bar is β-HCH.

Positive or negative correlation is also marked under the percentage value as “+” or “-”. But properties whose contribution less than 5% won’t be marked.

(a) Loading

From top to bottom are the sum of HCHs entering the Arctic Ocean through LROT, LRAT, and the fraction of direct pathway.

(b) Re-emission

From top to bottom are the sum of HCHs’ re-emission in the Arctic Ocean through LROT, LRAT, and the fraction of direct pathway.

(c) Burden

From top to bottem are chemical’s burden in 2020.

Text

Text S1 Mass balance equations of this model

Source Zone

$$\frac{dM_{\text{Air\_S}}}{dt} = -\begin{pmatrix} D_{\text{Tran\_Air\_SA}} + D_{\text{Deg\_Air\_S}} + D_{\text{Air\_Seawater\_S}} \\ +D_{\text{Air\_Soil\_S}} + D_{\text{Rain\_S}} + D_{\text{DD\_S}} + D_{\text{WD\_S}} + D_{\text{Air\_Strat\_S}} \end{pmatrix} \frac{M_{\text{Air\_S}}}{V_{\text{Air\_S}} Z_{\text{Air\_S}}} + D_{\text{Air\_Water}} \frac{M_{\text{Seawater\_S}}}{V_{\text{Seawater\_S}} Z_{\text{Seawater\_S}}} + D_{\text{Air\_Soil}} \frac{M_{\text{Soil\_S}}}{V_{\text{Soil\_S}} Z_{\text{Soil\_S}}} + E_{\text{Air}}$$

$$\frac{dM_{\text{Seawater\_S}}}{dt} = -\begin{pmatrix} D_{\text{Tran\_Seawater\_SA}} + D_{\text{Deg\_Seawater\_S}} \\ +D_{\text{Seawater\_Sink\_S}} + D_{\text{Air\_Water\_S}} \end{pmatrix} \frac{M_{\text{Seawater\_S}}}{V_{\text{Seawater\_S}} Z_{\text{Seawater\_S}}} + \begin{pmatrix} D_{\text{Rain\_Seawater\_S}} + D_{\text{DD\_Seawater\_S}} \\ +D_{\text{Air\_Water\_S}} + D_{\text{WD\_Seawater\_S}} \end{pmatrix} \frac{M_{\text{Air\_S}}}{V_{\text{Air\_S}} Z_{\text{Air\_S}}} + k_{\text{Runoff}} D_{\text{Runoff\_S}} \frac{M_{\text{Soil\_S}}}{V_{\text{Soil\_S}} Z_{\text{Soil\_S}}} + E_{\text{Seawater}}$$

$$\frac{dM_{\text{Soil\_S}}}{dt} = -(D_{\text{Deg\_Soil\_S}} + D_{\text{Runoff\_S}} + D_{\text{Air\_Soil\_S}}) \frac{M_{\text{Soil\_S}}}{V_{\text{Soil\_S}} Z_{\text{Soil\_S}}} + (D_{\text{Rain\_Soil\_S}} + D_{\text{DD\_Soil\_S}} + D_{\text{Air\_Soil\_S}} + D_{\text{WD\_Soil\_S}}) \frac{M_{\text{Air\_S}}}{V_{\text{Air\_S}} Z_{\text{Air\_S}}} + E_{\text{Soil}}$$

Arctic Zone

$$\frac{dM_{\text{Air\_A}}}{dt} = -\begin{pmatrix} D_{\text{Tran\_Air\_AS}} + D_{\text{Deg\_Air\_A}} + D_{\text{Air\_Strat\_A}} + D_{\text{Air\_Soil\_A}} \\ +D_{\text{Air\_Snowpack\_A}} + D_{\text{Air\_Sea\_Ice\_A}} + D_{\text{Rain\_A}} \\ +D_{\text{Snow\_A}} + D_{\text{DD\_A}} + D_{\text{WD\_Rain\_A}} + D_{\text{WD\_Snow\_A}} \end{pmatrix} \frac{M_{\text{Air\_A}}}{V_{\text{Air\_A}} Z_{\text{Air\_A}}} + D_{\text{Tran\_Air\_SA}} \frac{M_{\text{Air\_S}}}{V_{\text{Air\_S}} Z_{\text{Air\_S}}} + D_{\text{Air\_Water}} \frac{M_{\text{Seawater\_A}}}{V_{\text{Seawater}} Z_{\text{Seawater}}} + D_{\text{Air\_Soil}} \frac{M_{\text{Soil\_A}}}{V_{\text{Soil}} Z_{\text{Soil}}} + D_{\text{Air\_Snowpack}} \frac{M_{\text{Snowpack\_A}}}{V_{\text{Snowpack\_A}} Z_{\text{Snowpack\_A}}} + D_{\text{Air\_Sea\_Ice}} \frac{M_{\text{Sea\_Ice\_A}}}{V_{\text{Sea\_Ice}} Z_{\text{Sea\_Ice}}}$$

$$\frac{dM_{\text{Seawater\_A}}}{dt} = -\begin{pmatrix} D_{\text{Tran\_Seawater\_AS}} + D_{\text{Deg\_Seawater\_A}} \\ +D_{\text{Seawater\_sink\_A}} + D_{\text{Air\_Water\_A}} \end{pmatrix} \frac{M_{\text{Seawater\_A}}}{V_{\text{Seawater\_A}} Z_{\text{Seawater\_A}}} + \begin{pmatrix} D_{\text{Rain\_Seawater\_A}} + D_{\text{Snow\_Seawater\_A}} \\ +D_{\text{DD\_Seawater\_A}} + D_{\text{Air\_Water\_A}} + D_{\text{WD\_Seawater\_A}} \end{pmatrix} \frac{M_{\text{Air\_A}}}{V_{\text{Air\_A}} Z_{\text{Air\_A}}} + (1 - k_{\text{Runoff}}) D_{\text{Runoff\_S}} \frac{M_{\text{Soil\_S}}}{V_{\text{Soil\_S}} Z_{\text{Soil\_S}}} + D_{\text{Tran\_Seawater\_SA}} \frac{M_{\text{Seawater\_S}}}{V_{\text{Seawater\_S}} Z_{\text{Seawater\_S}}} + D_{\text{Runoff\_A}} \frac{M_{\text{Soil\_A}}}{V_{\text{Soil\_S}} Z_{\text{Soil\_A}}}$$

$$\frac{dM_{\text{Soil\_A}}}{dt} = -(D_{\text{Deg\_Soil\_A}} + D_{\text{Runoff\_A}} + D_{\text{Air\_Soil\_A}}) \frac{M_{\text{Soil\_A}}}{V_{\text{Soil\_A}} Z_{\text{Soil\_A}}} + \begin{pmatrix} D_{\text{Rain\_Soil\_A}} + D_{\text{Snow\_Soil\_A}} \\ D_{\text{Air\_Soil\_A}} + D_{\text{WD\_Soil\_A}} + D_{\text{DD\_Soil\_A}} \end{pmatrix} \frac{M_{\text{Air\_A}}}{V_{\text{Air\_A}} Z_{\text{Air\_A}}}$$

$$\frac{dM_{\text{Sea\_Ice\_A}}}{dt} = -(D_{\text{Deg\_Sea\_Ice\_A}} + D_{\text{Air\_Sea\_Ice\_A}}) \frac{M_{\text{Sea\_Ice\_A}}}{V_{\text{Sea\_Ice\_A}} Z_{\text{Sea\_Ice\_A}}} + \begin{pmatrix} D_{\text{Rain\_Sea\_Ice\_A}} + D_{\text{Sea\_Ice\_A}} \\ D_{\text{Air\_Sea\_Ice\_A}} + D_{\text{WD\_Sea\_Ice\_A}} + D_{\text{DD\_Sea\_Ice\_A}} \end{pmatrix} \frac{M_{\text{Air\_A}}}{V_{\text{Air\_A}} Z_{\text{Air\_A}}}$$

$$\frac{dM_{\text{Snowpack\_A}}}{dt} = -(D_{\text{Deg\_Snowpack\_A}} + D_{\text{Air\_Snowpack\_A}}) \frac{M_{\text{Snowpack\_A}}}{V_{\text{Snowpack\_A}} Z_{\text{Snowpack\_A}}} + \begin{pmatrix} D_{\text{Rain\_Snowpack\_A}} + D_{\text{Snow\_Snowpack\_A}} \\ D_{\text{Air\_Snowpack\_A}} + D_{\text{WD\_Snowpack\_A}} + D_{\text{DD\_Snowpack\_A}} \end{pmatrix} \frac{M_{\text{Air\_A}}}{V_{\text{Air\_A}} Z_{\text{Air\_A}}}$$

This model is calculated by ordinary differential equations printed here. Each equation correspond to a compartment in a environment combines fugacity method and advection method. As mentioned in the section 2.2 of the manuscript, there are 3 compartments in the Source Zone and 5 compartments the Arctic. From top to bottom are equations about air in the Source Zone, seawater in Source Zone, soil in the Source Zone, air in the Arctic Zone, seawater in Arctic Zone, soil in the Arctic Zone, sea ice in the Arctic Zone, and snowpack in Arctic Zone.

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