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Impacts of sea ice leads on sea salt aerosols and atmospheric chemistry in the Arctic

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Equation S1. Wind speed and sea surface temperature SSA number source function in GEOS-Chem from Jaegle et al. (2011).

$$\frac{dF}{dr_{80}} = (0.3 + 0.1 \times T - 0.0076 \times T^2 + 0.00021 \times T^3)1.373 u_{10\,m}^{3.41} r_{80}^{-A}\left(1 + 0.057 r_{80}^{3.45}\right) \times 10^{1.607 e^{-B^2}}$$

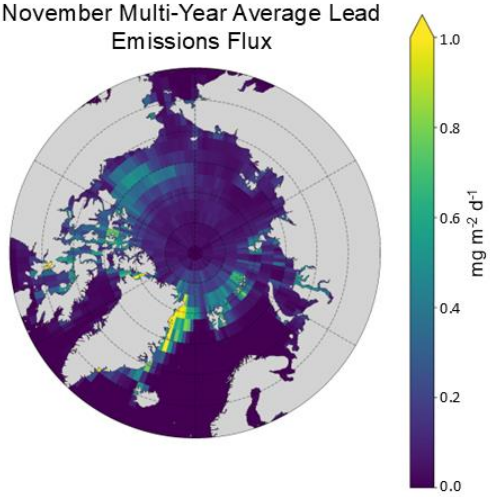
Where $\frac{dF}{dr_{80}}$ expresses a density function in $m^{-2}\ s^{-1}\ \mu m^{-1}$; T is the sea surface temperature (SST) expressed in ºC; $A = 4.7(1 + \Theta r_{80})^{-0.017 r_{80}^{-1.44}}$; $B = [0.433 - log_{10}(r_{80})]/0.433$; r_{80} is the particle radius at RH= 80% ($r_{80} \sim 2r_{dry}$); $u_{10\,m}$ is the 10-meter wind speed; and Θ is an adjustable parameter controlling the shape of the size distribution of submicron (recommended value of Θ = 30, which is used in our study).

Equation S2. SSA number source flux equation derived in Nilsson et al. (2001)

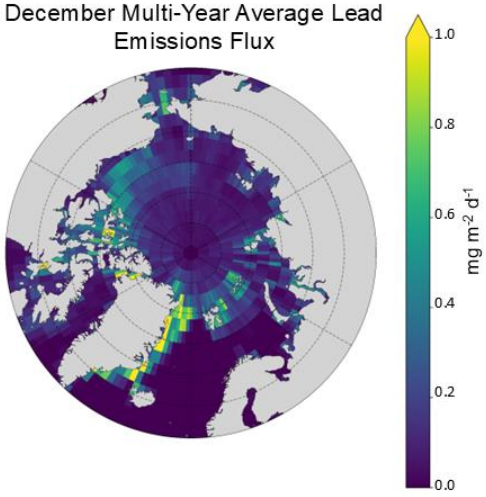
$$\log(F) = 0.20\overline{U} - 1.71$$

Where F is the number source flux in $10^6\ m^{-2}s^{-1}$ and $\overline{U}$ is the local wind speed.

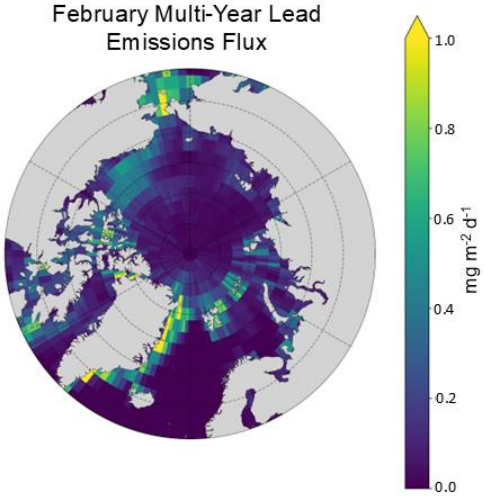
November Multi-Year Average Lead Emissions Flux



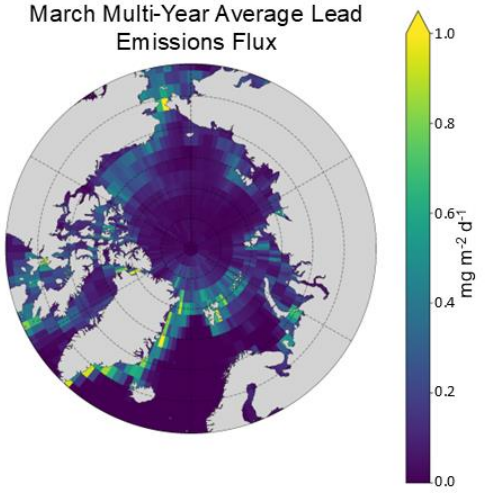
December Multi-Year Average Lead Emissions Flux



February Multi-Year Lead Emissions Flux



March Multi-Year Average Lead Emissions Flux



April Multi-Year Average Lead Emissions Flux

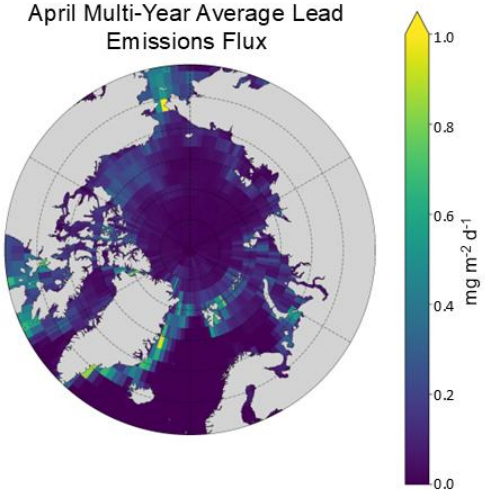


Figure S1-Lead emissions totaled for months during the cold season.

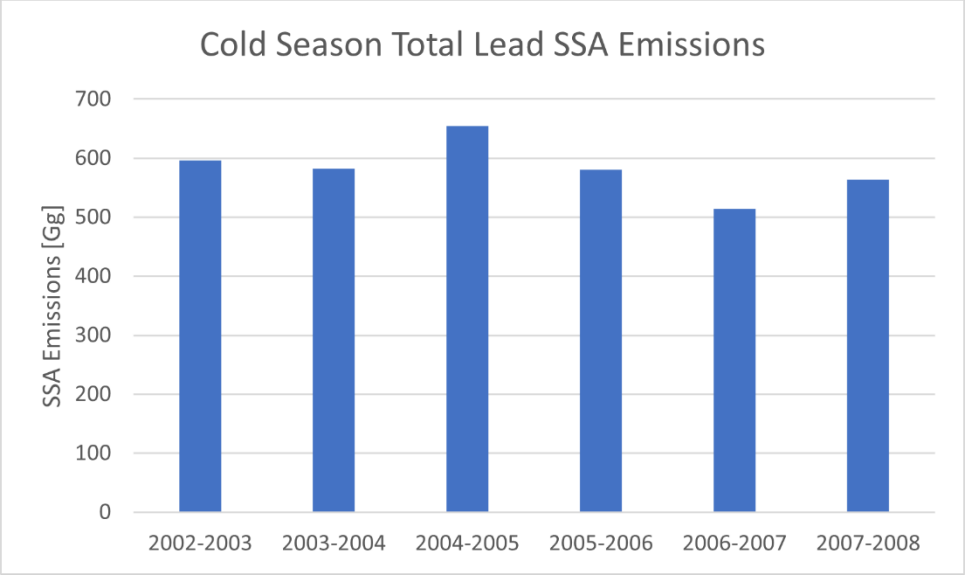
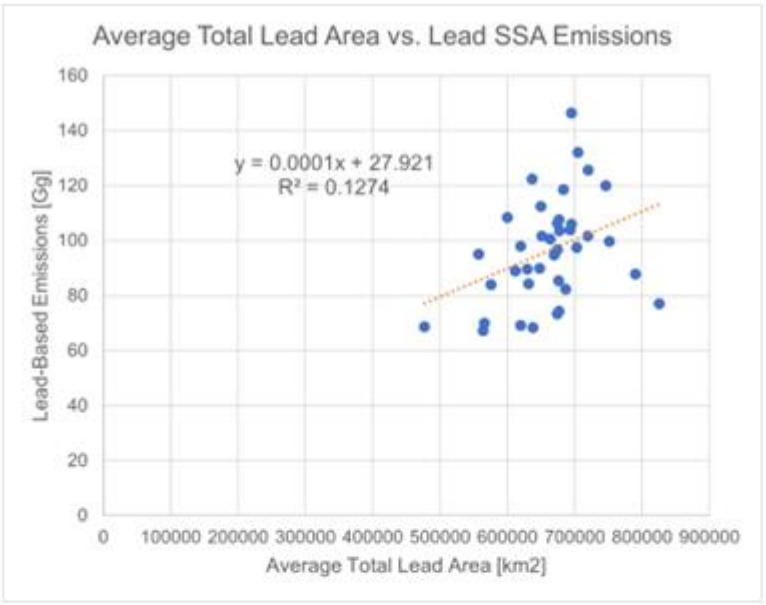


Figure S2- Cold Season Total Standard + Lead SSA Emissions.

(a)



(b)

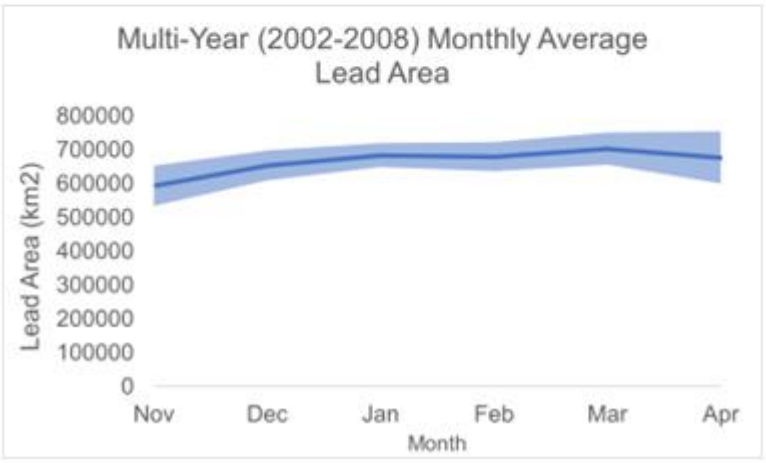


Fig S3 - (a) Average total lead area (km^2) vs. monthly lead emissions (Gg) and (b) multi-year monthly average lead area (km^2), with shading representing the standard deviation margin.

Text S1

The total monthly average lead area is calculated for each month separately by multiplying the monthly average lead fraction (from the daily AMSR-E files) in each grid cell by the grid cell area and summing all values. We correlate the monthly average total lead area and monthly lead emissions and find low correlation (R^2= 0.1274).

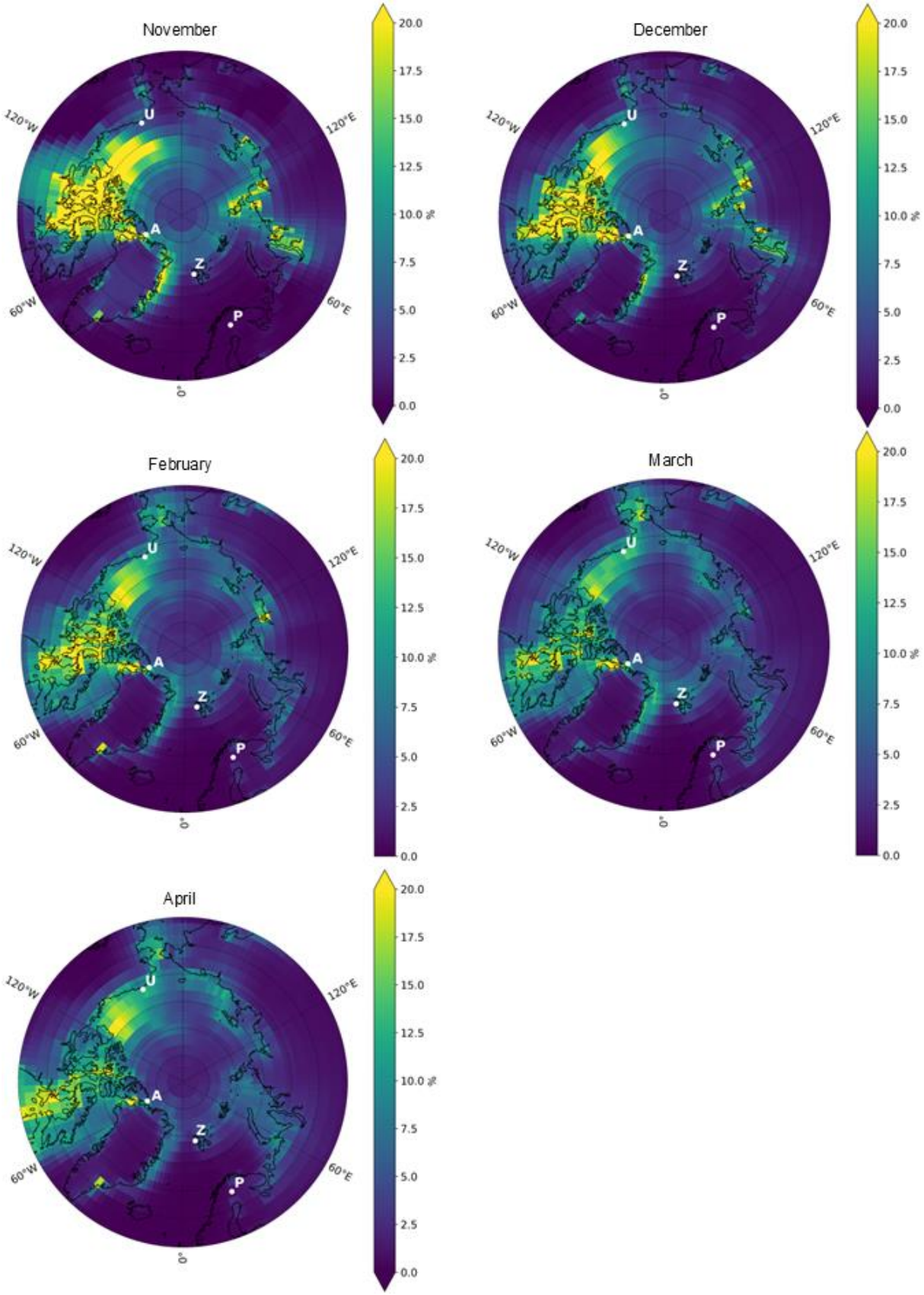

November
December
February
March
April
120°W
120°E
60°W
60°E
0°
U
A
Z
P
20.0
17.5
15.0
12.5
10.0 %
7.5
5.0
2.5
0.0

Figure S4- Multi-year average percent increase in SSA concentration due to leads (calculated with Eq. (1)) for other months during the cold season.

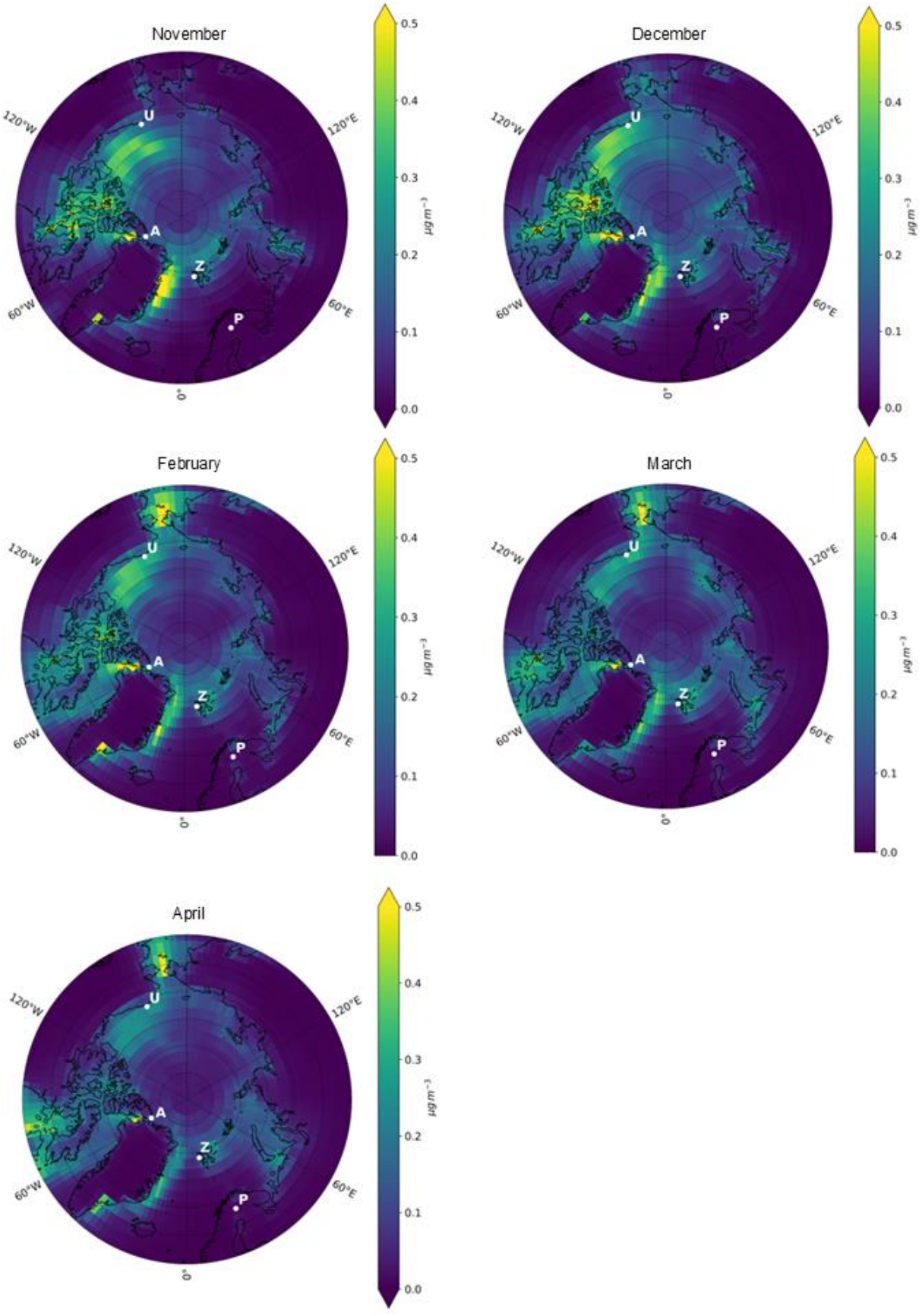
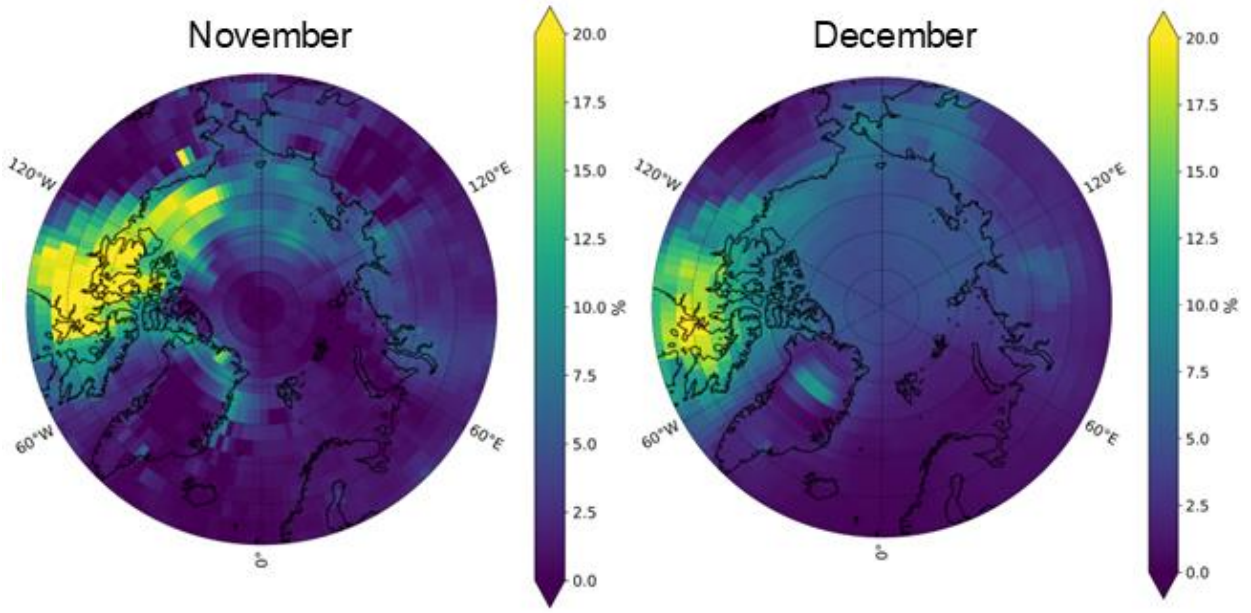

November
December
February
March
April
120°W
120°E
60°W
60°E
0°
U
A
Z
P
0.5
0.4
0.3
0.2
0.1
0.0
μg m⁻³

Figure S5- Multi-year average absolute difference of SSA concentration between the standard + leads and standard simulations for other months during the cold season.



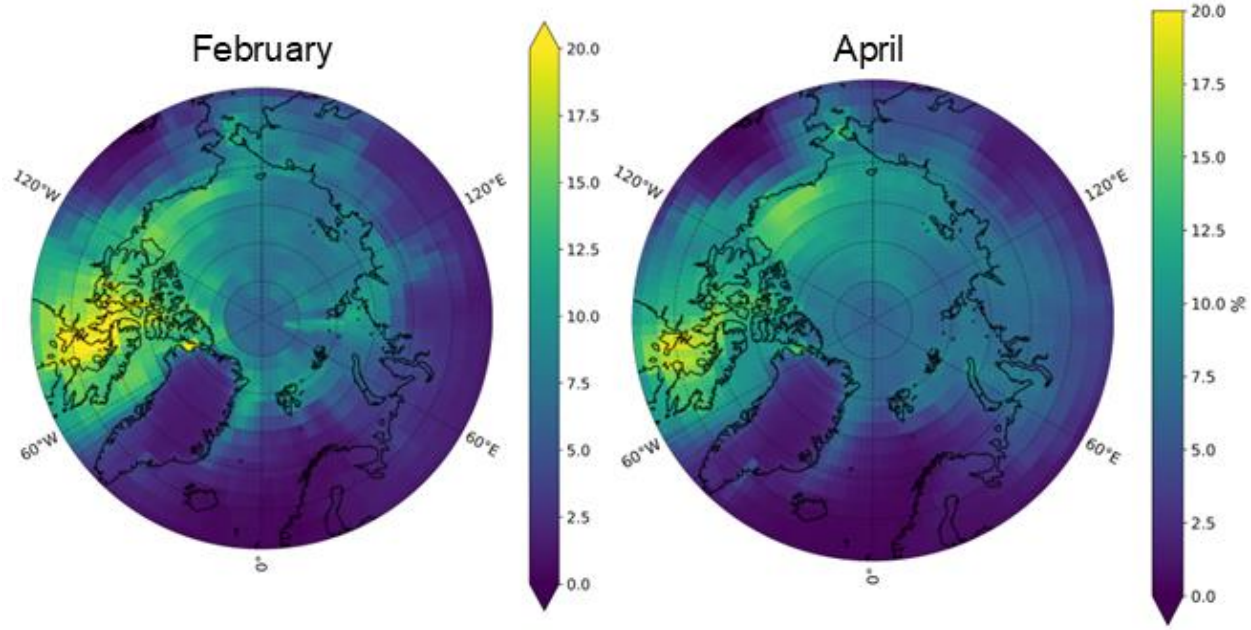


Figure S6- Multi-year average percent increase in Br concentration due to leads (calculated with Eq. (1)) for other months during the cold season.

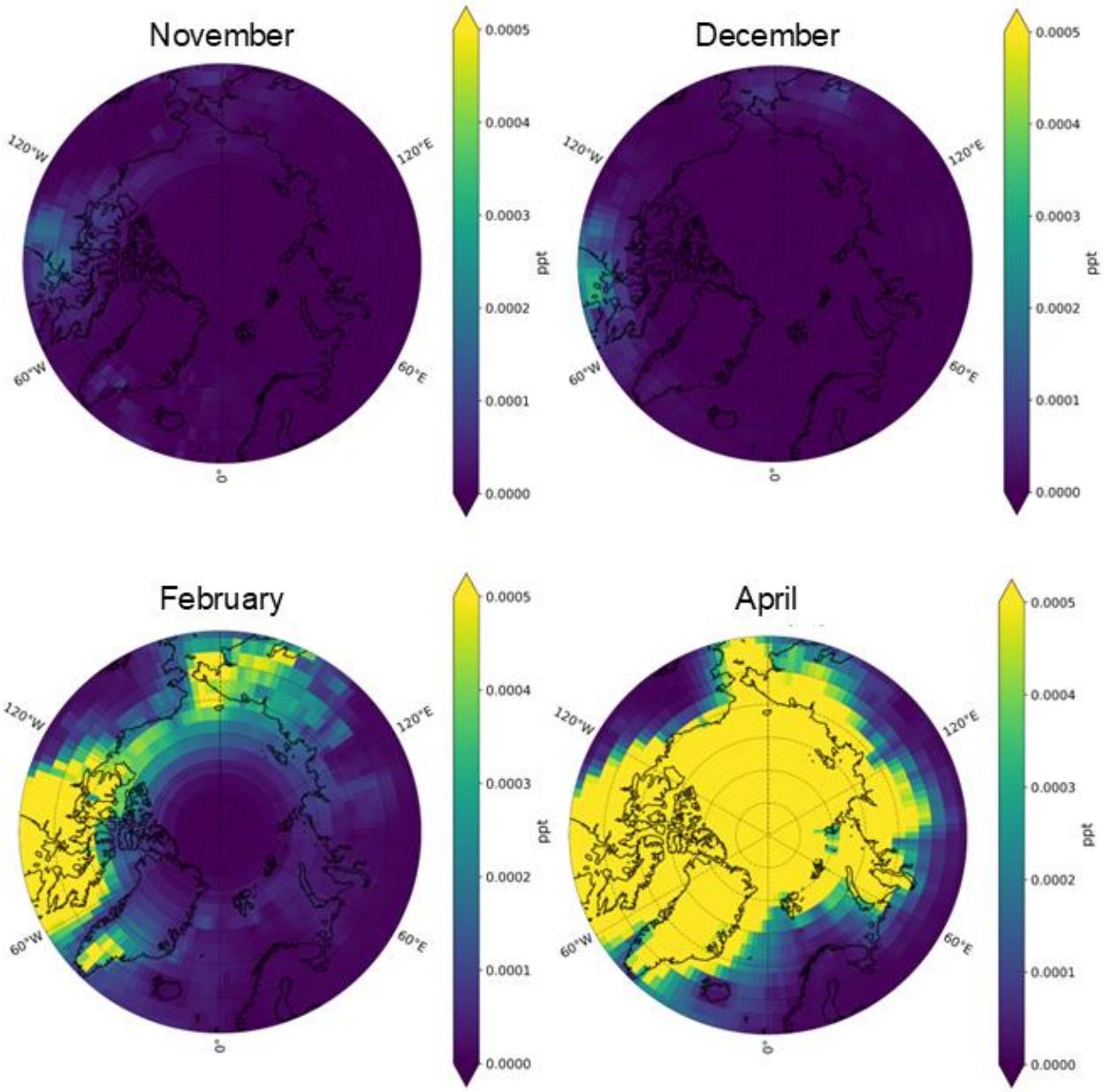


Figure S7- Multi-year average absolute difference in Br concentration due to leads for other months during the cold season.

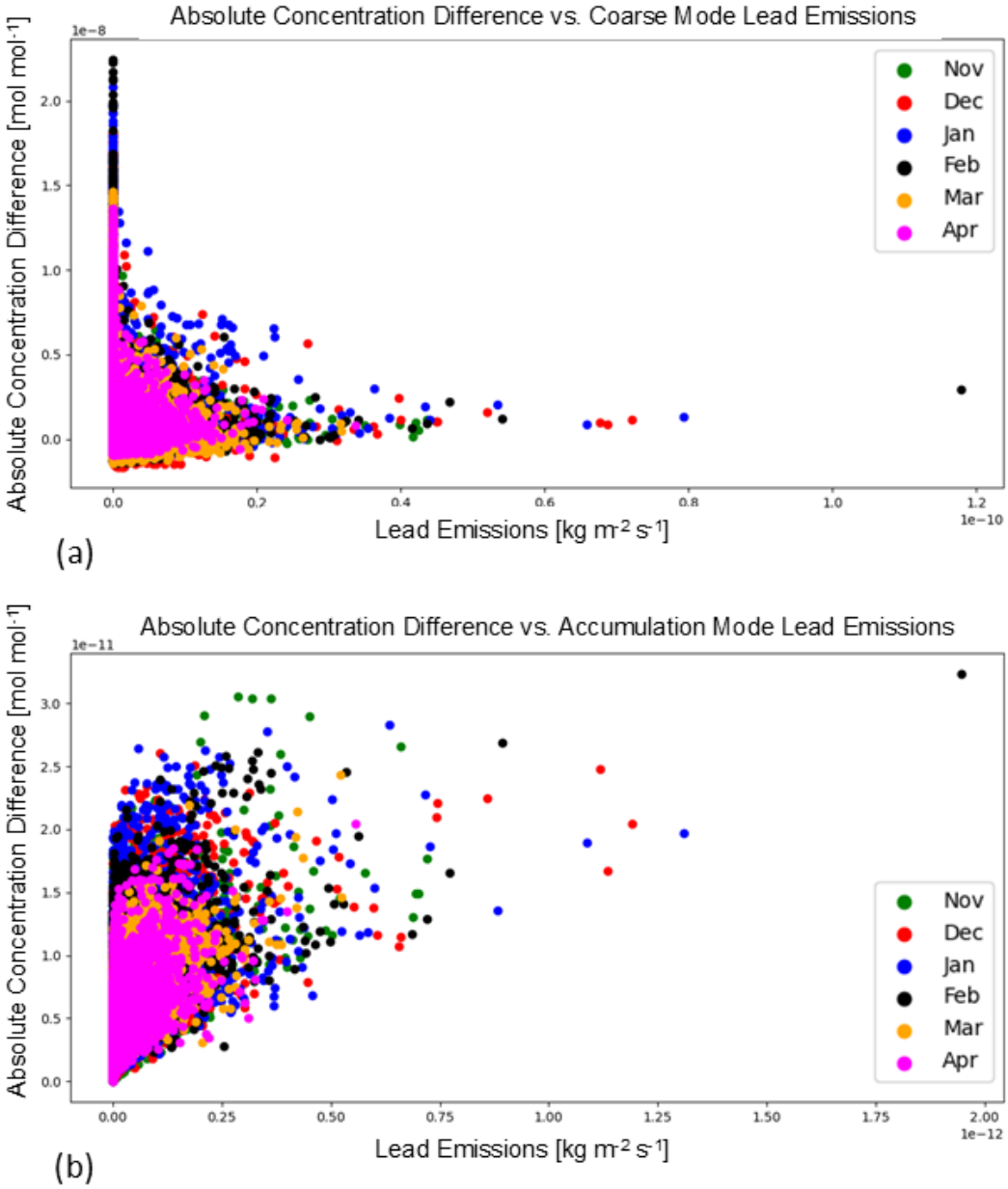


Figure S8- Lead Emissions vs. absolute difference in SSA concentration between the standard+leads and standard models for (a) coarse mode and (b) accumulation mode.

Text S2

To better understand the deposition and lifetime of the coarse and accumulation mode lead-based SSA, we correlate the lead-based emissions with the absolute difference between the standard+leads and standard SSA concentrations for each size bin (Figure S8). Coarse mode SSA dominates the total mass of SSA (note the different y-axes for both figures). We find evidence that the coarse mode SSA emitted from leads have long enough lifetimes in the atmosphere to be transported to regions of the Arctic where lead emissions are zero (many points on Figure S8a where the absolute concentration difference is larger than 0 when lead emissions are 0). This

also occurs for accumulation mode particles (Figure S8b).

Text S3

We run two additional sensitivity simulations to test the possible sources of overestimation in the model. For the first ("standard + leads + Luo Wet Deposition"), we use the calculated emissions of the standard leads from HEMCO and apply the Luo Wet Deposition scheme to the full-chemistry GEOS-Chem run. The Luo wet deposition scheme includes updates to pH calculations for cloud, rain, and wet surfaces; the fraction of cloud available for aqueous-phase chemistry; the rainout efficiencies for various cloud types; empirical washout by rain and snow; and wet surface uptake during dry deposition. We utilize the same approach of spin-up as the full standard + leads case, by running one year (November 1, 2002-November 1, 2003) and then running the simulation for analysis from November 2002 to April 2003, with the spun-up November 1, 2003, initial conditions. For the second sensitivity simulation ("open ocean only") we run HEMCO to calculate the open ocean only emissions by turning off blowing snow emissions. We calculate emissions starting November 1, 2001, which we use to spin-up the full chemistry GEOS-Chem run. We spin-up the GEOS-Chem simulation from November 1, 2001, to November 1, 2002, and run the simulation for analysis from November 2002 to April 2003.