Impacts of Sea Ice Leads on Sea Salt Aerosols and Atmospheric Chemistry in the Arctic

Supplemental Information

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Equation S.1. Wind speed and sea surface temperature SSA number source function in GEOS-Chem from Jaegle et al. (2011).

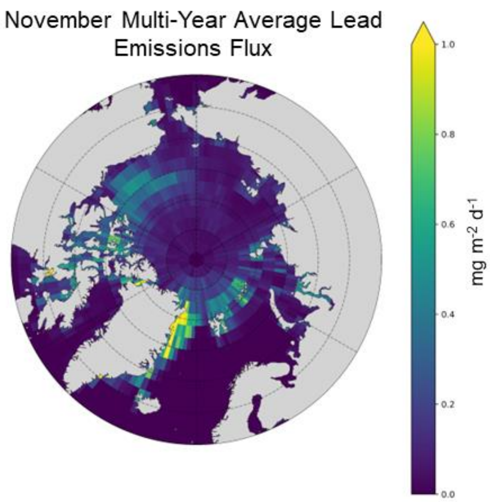
$$\frac{dF}{dr_{80}} = (0.3 + 0.1 \times T - 0.0076 \times T^2 + 0.00021 \times T^3)1.373u_{10\,m}^{3.41}r_{80}^{-A}\left(1 + 0.057r_{80}^{3.45}\right) \times 10^{1.607e^{-B^2}}$$

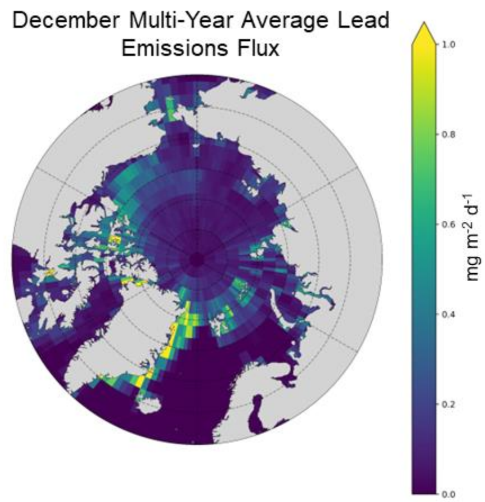
Where $\frac{dF}{dr_{80}}$ expresses a density function in m^{-2} s^{-1} μm^{-1}; T is the sea surface temperature (SST) expressed in ºC; $A = 4.7(1 + \Theta r_{80})^{-0.017r_{80}^{-1.44}}$; $B = [0.433 - \ log_{10}(r_{80})]/0.433$; r_{80} is the particle radius at RH= 80% ($r_{80} \sim 2r_{dry}$); $u_{10\,m}$ 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).

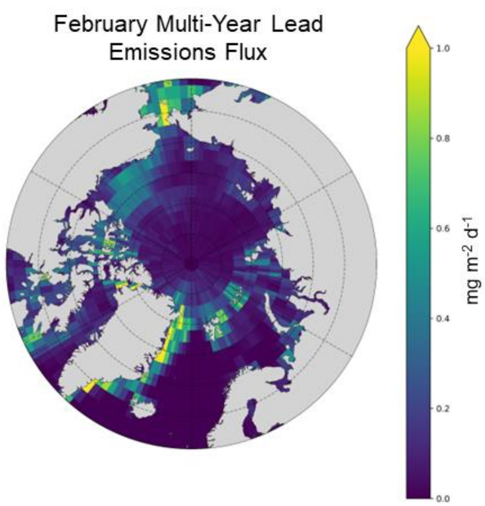
Equation S.2. SSA number source flux equation derived in Nilsson et al. (2001)

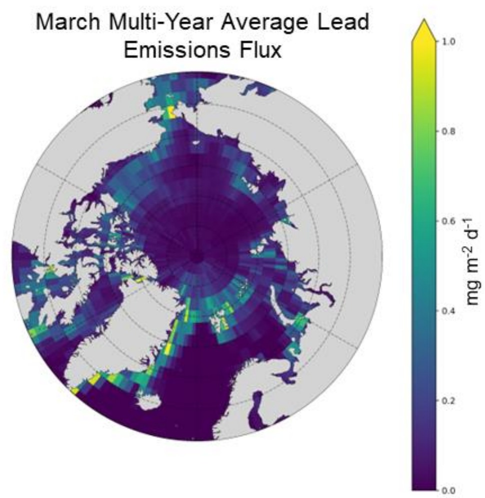
$$\log(F) = 0.20\overline{U} - 1.71$$

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
mg m^{-2} d^{-1}
1.0
0.8
0.6
0.4
0.2
0.0


December Multi-Year Average Lead Emissions Flux
mg m^{-2} d^{-1}
1.0
0.8
0.6
0.4
0.2
0.0


February Multi-Year Lead Emissions Flux
mg m^{-2} d^{-1}
1.0
0.8
0.6
0.4
0.2
0.0


March Multi-Year Average Lead Emissions Flux
mg m^{-2} d^{-1}
1.0
0.8
0.6
0.4
0.2
0.0

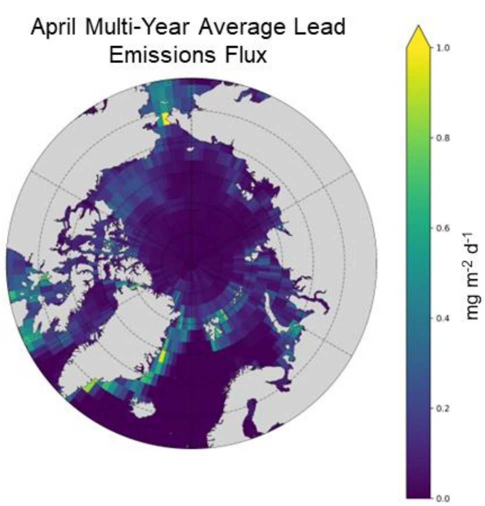

April Multi-Year Average Lead Emissions Flux
mg m^{-2} d^{-1}
1.0
0.8
0.6
0.4
0.2
0.0

Figure S.1-Lead emissions totaled for months during the cold season.

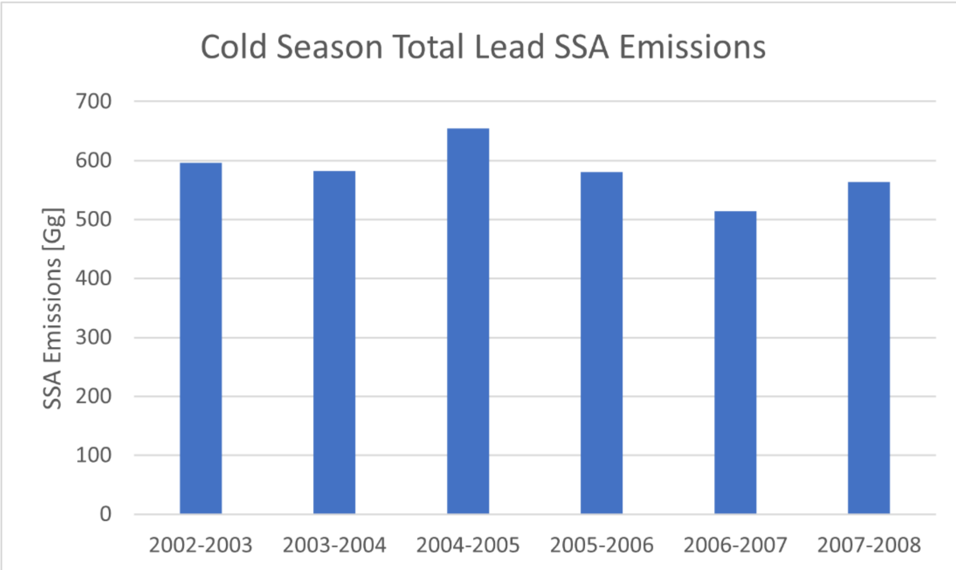


Figure S.2- Cold Season Total Standard + Lead SSA Emissions.

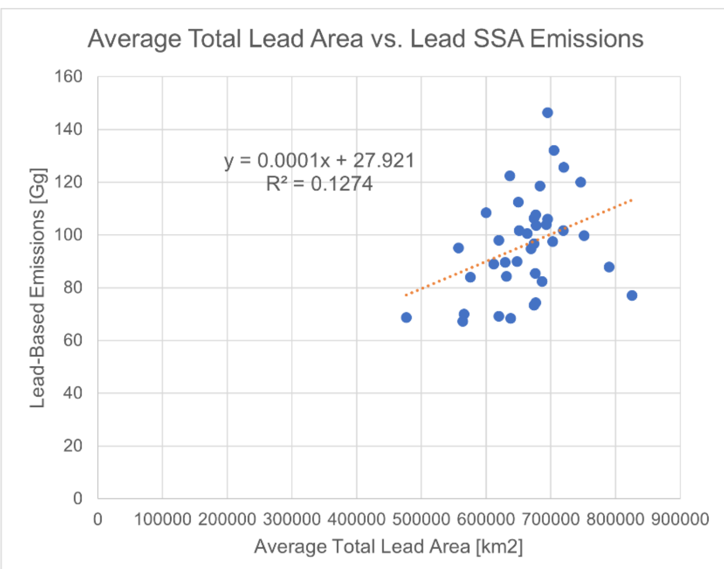


Fig S.3 - Average total lead area (km^2) vs. monthly lead emissions (Gg).

Text S.1

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).

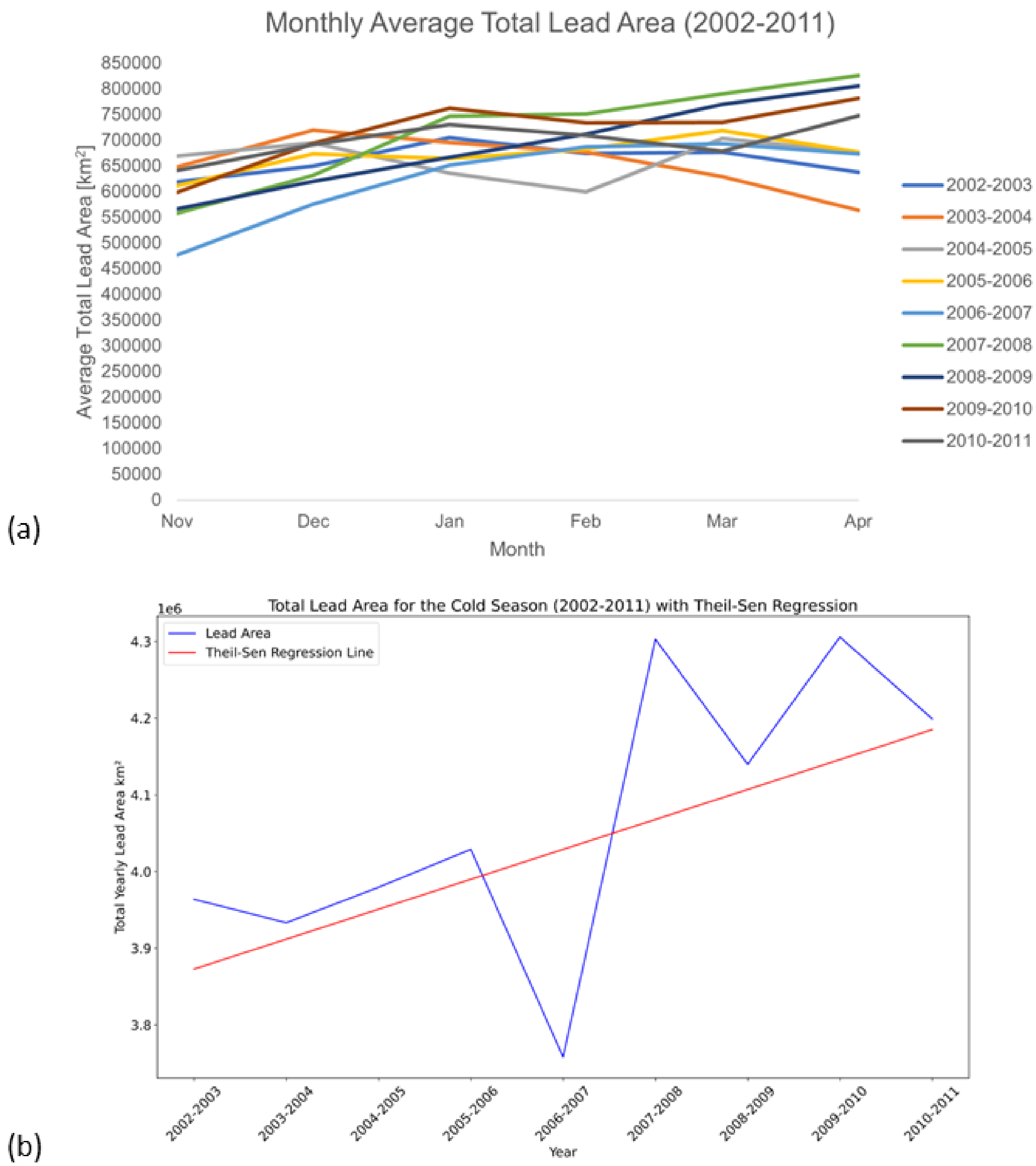


Figure S.4 - (a) Average monthly total lead area and (b) average lead area totaled over the cold season (November – April) by year, for 2002-2011.

Text S.2

To assess the trend in the total lead area for each cold season from 2002-2011 (blue line in Figure S.4 above) we employ a Theil-Sen regression method (red line in Figure S.4 above). This analysis reveals a statistically significant positive trend, with a slope of +39,018.5 km^2/year (95% Confidence Interval: 1,385 to 69,217.5 km^2/year).

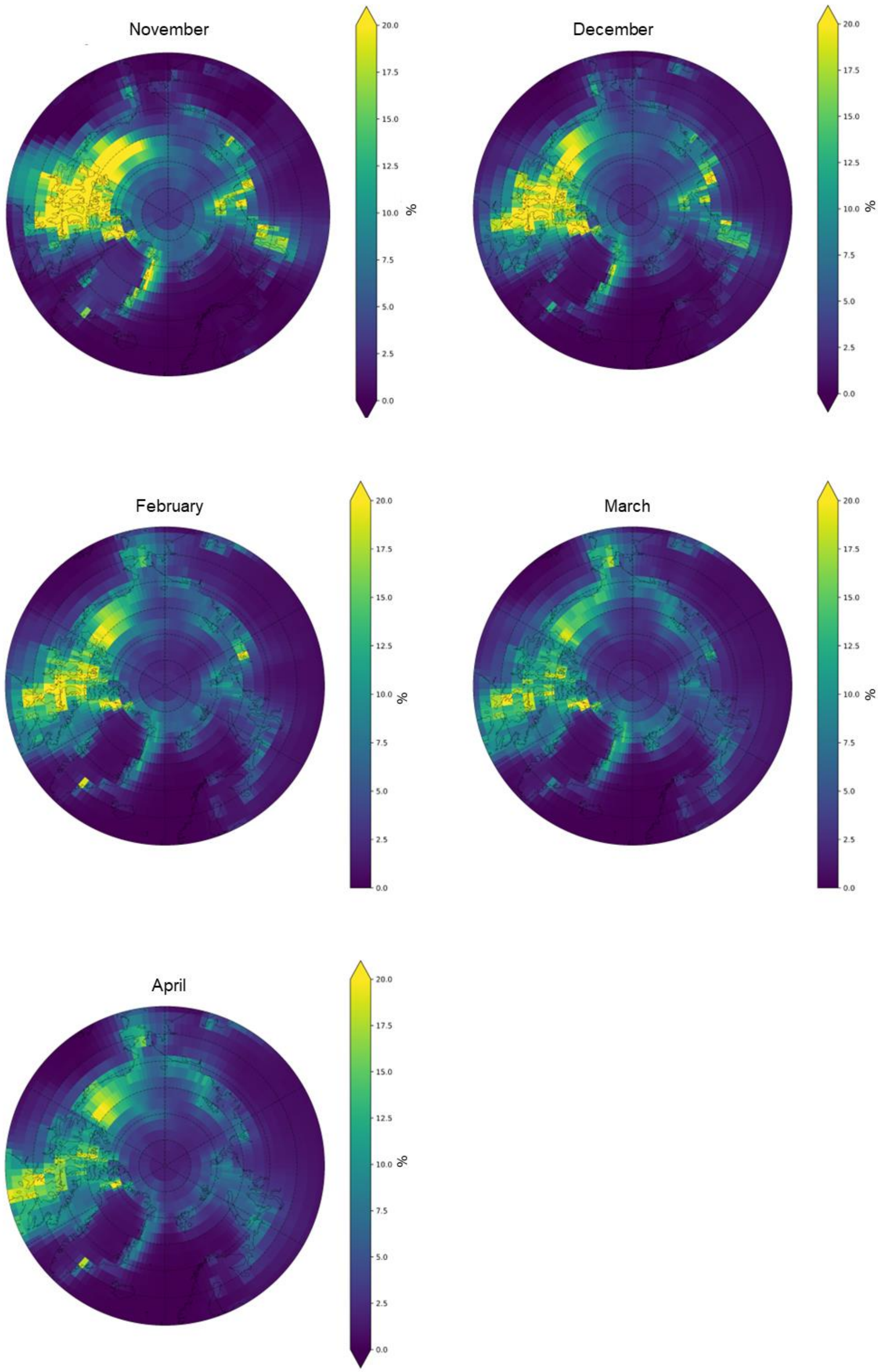

November
December
February
March
April
20.0
17.5
15.0
12.5
10.0
7.5
5.0
2.5
0.0
%

Figure S.5- Multi-year average percent increase in SSA concentration due to leads (calculated with Eq. (1)) for other months during the cold season.

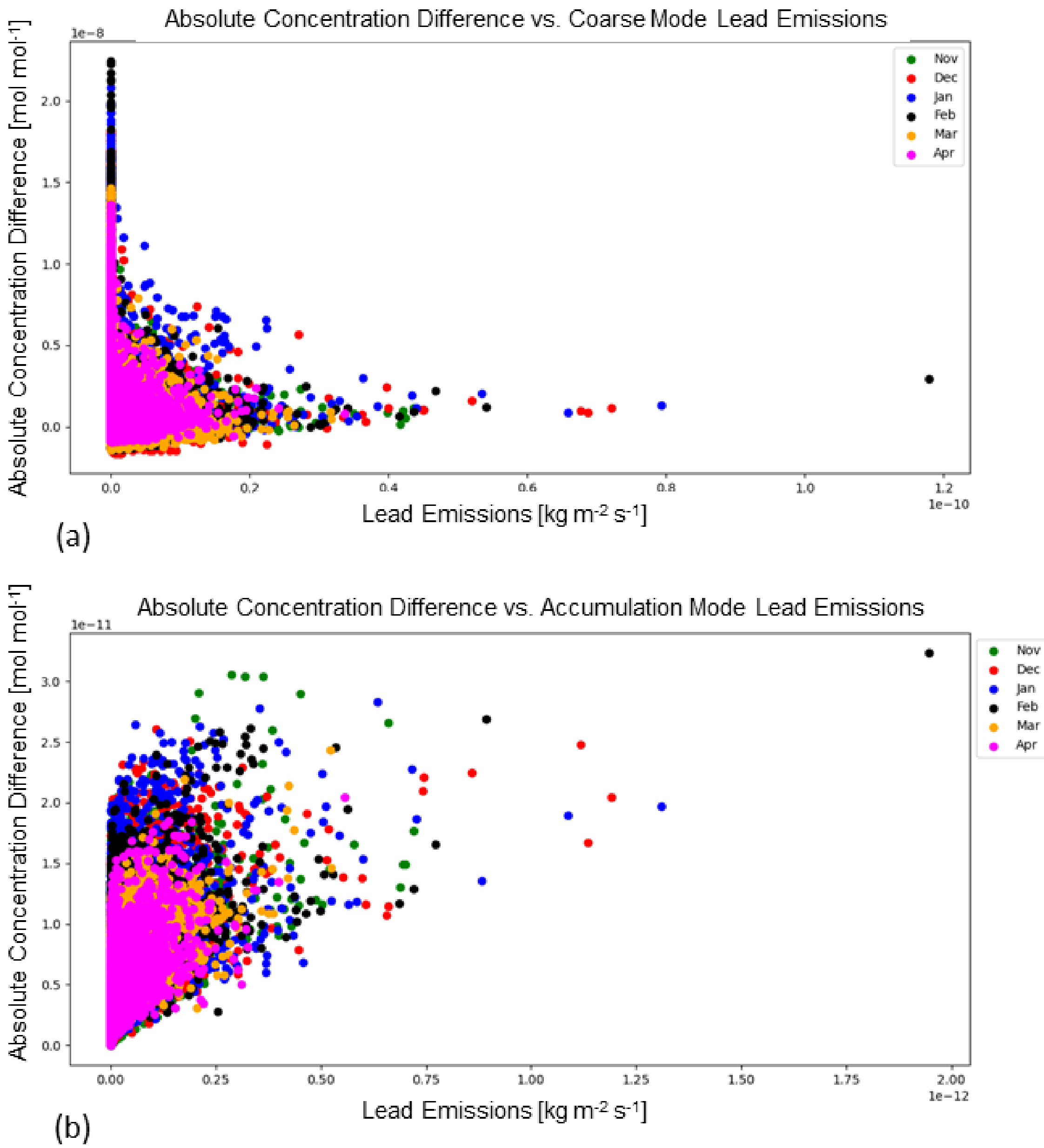


Figure S.6- Lead Emissions vs. absolute difference in SSA concentration between the standard+leads and standard models for (a) coarse mode and (b) accumulation mode.

Text S.3

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 S.6). 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 S.6a where the absolute concentration difference is larger than 0 when lead emissions are 0). This also occurs for accumulation mode particles (Figure S.6b).

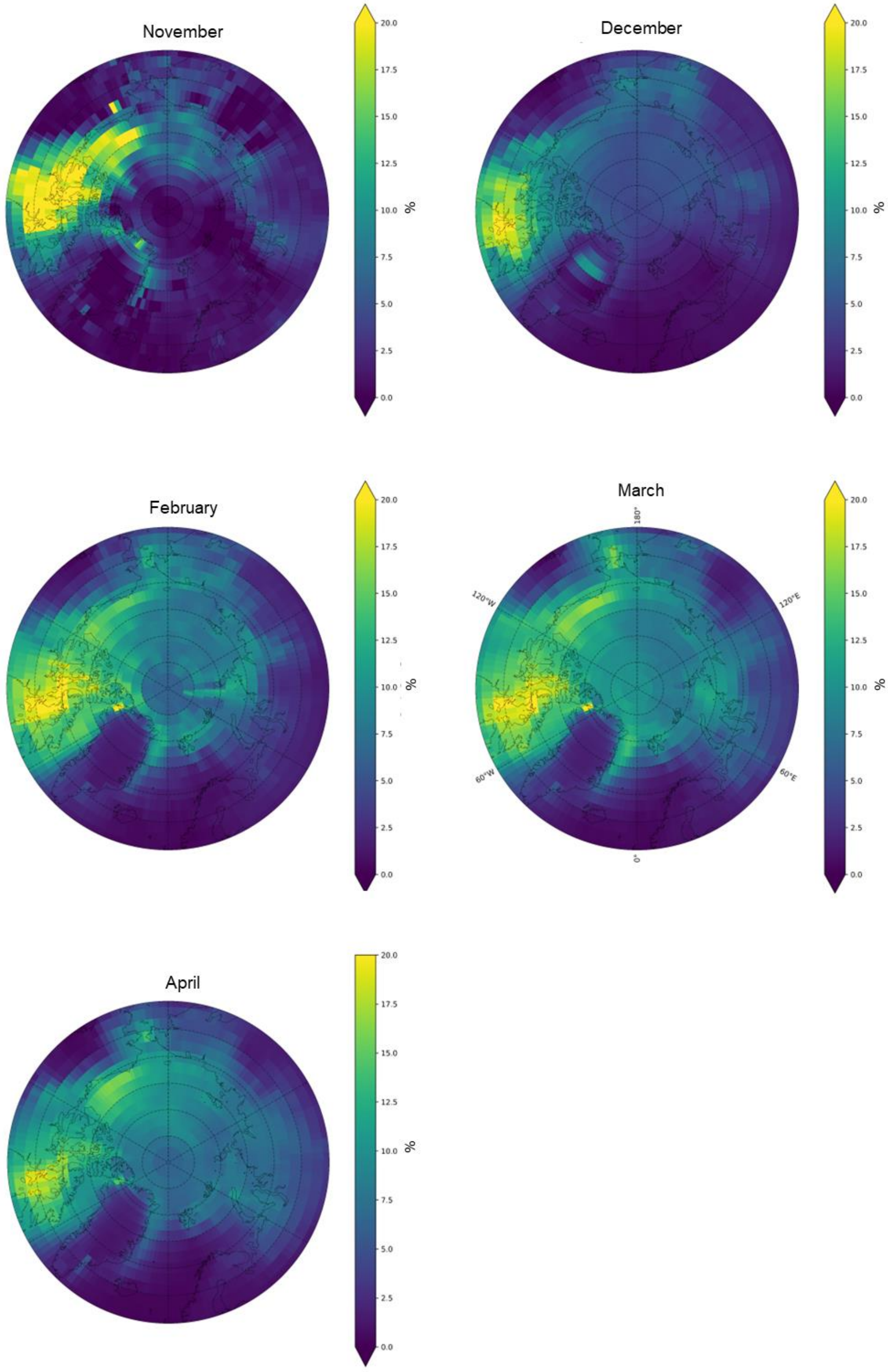

November
December
February
March
April
180°
120°W
120°E
60°W
60°E
0°
20.0
17.5
15.0
12.5
10.0
7.5
5.0
2.5
0.0
%

Figure S.7- Multi-year average percent increase in Br concentration due to leads (calculated with Eq. (1)) for other months during the cold season.

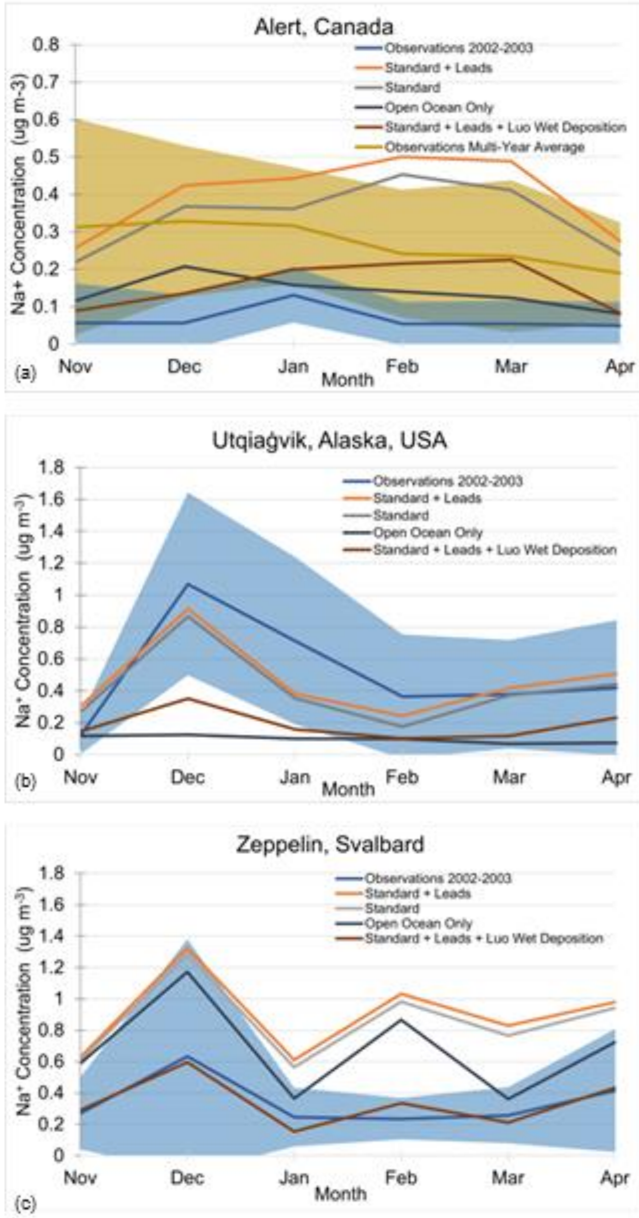


Figure S.8- Model evaluation for the cold season 2002-2003. Observed (blue + standard deviation margin) Na$^+$ concentrations are monthly averages for 2002-2003, and we add the multi-year average observed monthly concentration (gold + standard deviation margin) due to the low observed monthly concentrations in 2002-2003. We show monthly average modeled Na$^+$ concentrations for 2002-2003 for the standard + leads (orange) and standard (blue) with two additional sensitivity studies: open ocean only emissions contributing to Na$^+$ concentrations (dark navy blue) and the standard + leads emissions with Luo Wet Deposition applied to the GEOS-Chem full-chemistry run (red). Note the different axis for Alert (a), as concentrations are much lower at this site.

Text S.4

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 updated 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 run 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.