

MEMORANDUM

June 30, 2020

To: Molly Birnbaum, Alaska Department of Environmental Conservation

From: Jean Guo and Ralph Morris

Subject: Identification of Potential Emission Sources that Can Contribute to Sulfate at Alaska IMPROVE Sites

OVERVIEW

As part of a scoping study to determine the potential sources of sulfate at Alaska IMPROVE sites, Ramboll identified various types of reactive sulfur¹ emissions that could contribute to sulfate extinction at IMPROVE sites in Alaska.

Emissions inventories from the Harvard-NASA Emissions Component (HEMCO²) tool and Fire Inventory from NCAR (FINN³) can be used to analyze the amount of sulfur emissions for an Alaska domain to provide an estimate of the relative importance of different source sectors to regional sulfur emission loadings in the region. The CAMx oceanic pre-processor could also be used to identify sulfate emissions from sea salt and oceanic dimethyl sulfide (DMS) emissions that is converted to SO₂ and then sulfate. Ramboll conducted an Area of Influence (AOI) and Weighted Potential Analysis (WPA) analysis of Alaska anthropogenic emissions that could be extended to natural emissions to understand the relative importance of anthropogenic and natural sources. Finally, the use of photochemical transport models, such as the recent EPA Alaska CMAQ 2016 modeling platform, could be used to explicitly estimate the contributions of various emission sources to ammonium sulfate (AmSO₄) visibility extinction at Alaska IMPROVE sites using source apportionment or sensitivity (e.g., brute force) analysis.

DATA SOURCES FOR EMISSIONS

Sulfur emissions contributes to sulfate extinction in Alaska. Sources of sulfur emissions include, but are not limited to the following:

- Alaska anthropogenic emissions
- Commercial Marine Vessels (CMV)
- Wildfires, prescribed burns and agricultural burning
- Volcano degassing/leakage
- Volcano eruptions
- Sulfate in sea spray aerosols (SSA)
- Oceanic Dimethyl Sulfide (DMS)
- Long-range transport of anthropogenic and natural emissions from outside of the immediate vicinity of Alaska (e.g., Asia and Russia)

¹ We are using the term reactive sulfur emissions to refer to sulfur compounds that are eventually converted to aerosol sulfate in the atmosphere so does not include sulfur compounds like hydrogen sulfide (H₂S)

² http://wiki.seas.harvard.edu/geos-chem/index.php/The_HEMCO_User%27s_Guide

³ <https://www2.acom.ucar.edu/modeling/finn-fire-inventory-ncar>

The Harvard-NASA Emissions Component (HEMCO) tool, which provides emissions inventory information to the GEOS-Chem global chemical transport model, is one potential data source for sulfur emissions. HEMCO reads in various emissions inventories⁴ and provides sulfur (and other species) emissions rates from worldwide anthropogenic emissions, shipping emissions, biomass burning, volcanic degassing/leakage, and oceanic DMS, as shown in **Table 1**.

HEMCO global anthropogenic emissions, including emissions from shipping sources, come from the Community Emissions Data System (CEDs) inventory.⁵ Species in this inventory include SO₂, BC, OC, NO_x, NH₃, CH₄, CO, NMVOC, and CO₂. Biomass burning emissions come from the Global Fire Emissions Database, Version 4 (GFED4) inventory.⁶ Volcanic emissions of SO₂ come from the AeroCom inventory. Oceanic DMS emissions comes from the DMS ocean exchange inventory.

The contribution of sulfur emissions to Alaska and other regions were extracted from HEMCO inventories for the year 2014. SO₂ emissions (in Metric tons (Mt)/year) from anthropogenic emissions, shipping, biomass burning, volcanic degassing, and oceanic DMS are shown in **Table 2**. The emissions from the listed sources are provided for the following regions: (1) an Alaska region; (2) the contiguous United States (CONUS); and (3) global world-wide. **Figure 1** shows the Alaska (dark pink) and CONUS (lighter pink) emission extraction domains on the GEOS-Chem 2x2.5 degree domain, which is used by HEMCO. The Alaska emission extraction domain was defined to roughly correspond to the 9-km grid resolution domain used in EPA’s 2016 CMAQ chemical transport modeling.

FINN is another source of wildfire emissions. The inventory is an optional addition in the HEMCO tool. It can also be downloaded directly from the FINN website.⁷ FINN uses satellite observations of active fires and other data to provide daily estimates of opening burning emissions for use in air quality models.

The CAMx regional photochemical transport model has an oceanic pre-processor. The processor can be used to generate emissions from sea salt, which includes primary sulfate emissions and oceanic DMS.

Table 1. Emissions Inventories from HEMCO.

Type	Emission Inventory
<i>Aerosol</i>	<ul style="list-style-type: none"> • AeroCom volcanic • DMS ocean exchange
<i>Anthropogenic, Biofuel</i>	<ul style="list-style-type: none"> • CEDs anthropogenic emissions
<i>Aircraft and ship</i>	<ul style="list-style-type: none"> • CEDs ship
<i>Biomass burning</i>	<ul style="list-style-type: none"> • GFED4 • FINNv1 – OPTIONAL

⁴ wiki.seas.harvard.edu/geos-chem/index.php/HEMCO_data_directories

⁵ http://wiki.seas.harvard.edu/geos-chem/index.php/CEDs_anthropogenic_emissions

⁶ https://daac.ornl.gov/VEGETATION/guides/fire_emissions_v4_R1.html

⁷ <https://www2.acom.ucar.edu/modeling/finn-fire-inventory-ncar>

Table 2. Emissions of SO₂ (Mt/year). Data comes from the inventories in the HECMO emissions tool.

Source	Alaska	CONUS	World-Wide
<i>Anthropogenic</i>	0.17	5.46	102.80
<i>Shipping</i>	0.18	0.92	8.90
<i>Biomass Burning</i>	0.076	0.031	2.25
<i>Volcano degassing</i>	0.58	0.062	18.48
<i>Oceanic DMS</i>	0.43	0.72	38.17

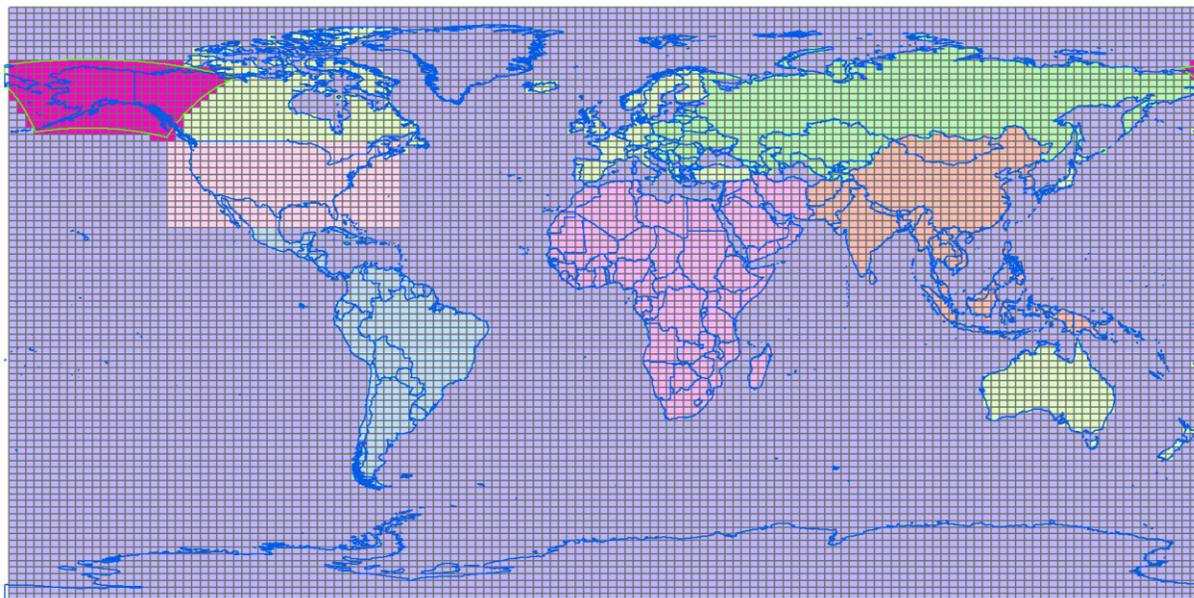


Figure 1. Map of the EPA Alaska 9-km CMAQ modeling domain (green) overlaid over the GEOS-Chem 2x2.5 degree grid Alaska emission extraction domain (dark pink) and CONUS emission extraction domain (lighter pink).

METHODS FOR DETERMINING IMPACT OF EMISSIONS ON SULFATE VISIBILITY EXTINCTION

In this section we discuss methods that can be used to and quantify the contributions of sources to sulfate at Alaska IMPROVE sites.

AOI/WEP Analysis

One method to assess and quantify the impacts of sulfur emissions on the visibility extinction of sulfate at Alaska IMPROVE monitors is to conduct an Area of Influence (AOI) and Weighted Emissions Potential (WEP) analysis. The AOI/WEP analysis uses HYSPLIT back trajectories from Alaska IMPROVE sites to get the frequency of occurrence, or residence time (RT), that air parcels originating from a particular location arrive at the IMPROVE site

on Specific days (e.g., the IMPROVE Most Impaired Days [MID] or highest sulfate extinction days). The RT or AOI analysis can then be weighted by sulfate extinction to generate Extinction Weighted Residence Times (EWRT). The HYSPLIT derived sulfate EWRT can be overlaid on gridded sulfur emission to determine the sources of emissions within and near Alaska that are most likely to contribute to visibility impairment at IMPROVE sites on high sulfate extinction or the MID. Ramboll has recently completed an AOI/WEP analysis for Alaska IMPROVE sites and the 2014-2018 MID. The Ramboll Alaska WEP analysis focused on the contributions from seven anthropogenic source sectors: (1) Electric Generation Unit (EGU) point sources; (2) non-EGU point; (3) oil and gas area and point source; (4) on-road mobile sources; (5) non-road mobile sources, which included rail, CMV and airports; (6) non-point (area) emissions; and (7) total anthropogenic emissions. The Ramboll Alaska AOI/WEP analysis could be extended to included natural sources in order to get the relative importance of these other source sectors.

Photochemical Grid Modeling

Another method to obtain source contributions to sulfate at Alaska IMPROVE sites is to run a photochemical grid model (PGM), such as CMAQ⁸ or CAMx⁹. A PGM can be used to explicitly estimate the contributions of various emissions sources to sulfate at IMPROVE sites. Long-range transport contributions to Alaska can be determined by applying global chemistry models (e.g., GEOS-Chem or Hemispheric CMAQ) that provide boundary condition (BC) inputs to the PGM. Zero-out modeling emissions modeling of different world-wide source regions or source sectors could be used to modify the boundary conditions (BC) of the PGM and the world-wide contributions could then be tracked by the PGM using the source apportionment or conducting Brute Force sensitivity modeling. CMAQ and CAMx have source apportionment tools that are able assess source attribution from different sources at a fine resolution and can be used to analyze sulfate visibility extinction at Alaska IMPROVE monitors. The modeling results can be compared with data from IMPROVE monitors to determine source contributions on the most impaired days.

Figure 2 displays the 27-km and 9-km CMAQ modeling domains used in EPA's Alaska regional haze modeling.¹⁰ The 9-km modeling domain roughly corresponds to the Alaska emissions extraction domain used to extract GEOS-Chem emissions from the HEMCO pre-processor as shown in **Figure 1** and used to generate the data in **Table 2**. Limitations of the EPA Alaska 2016 and 2028 CMAQ modeling include several missing source categories such as lightning NO_x, windblown dust and volcano emissions. We also do not think it includes DMS emissions, although sulfate from SSA should be included. As discussed in the next section, using the GEOS-Chem emissions inventory these missing sources could account for approximately 70 percent of the reactive sulfur emissions in the CMAQ 9-km Alaska modeling domain.

⁸ <https://www.epa.gov/cmaq>

⁹ www.camx.com

¹⁰ "Technical Support Document for EPA's Updated 2028 Regional Haze Modeling for Hawaii, Virgin Islands, and Alaska" prepared by Office of Air Quality Planning and Standards, United States Environmental Protection Agency. June 2020.

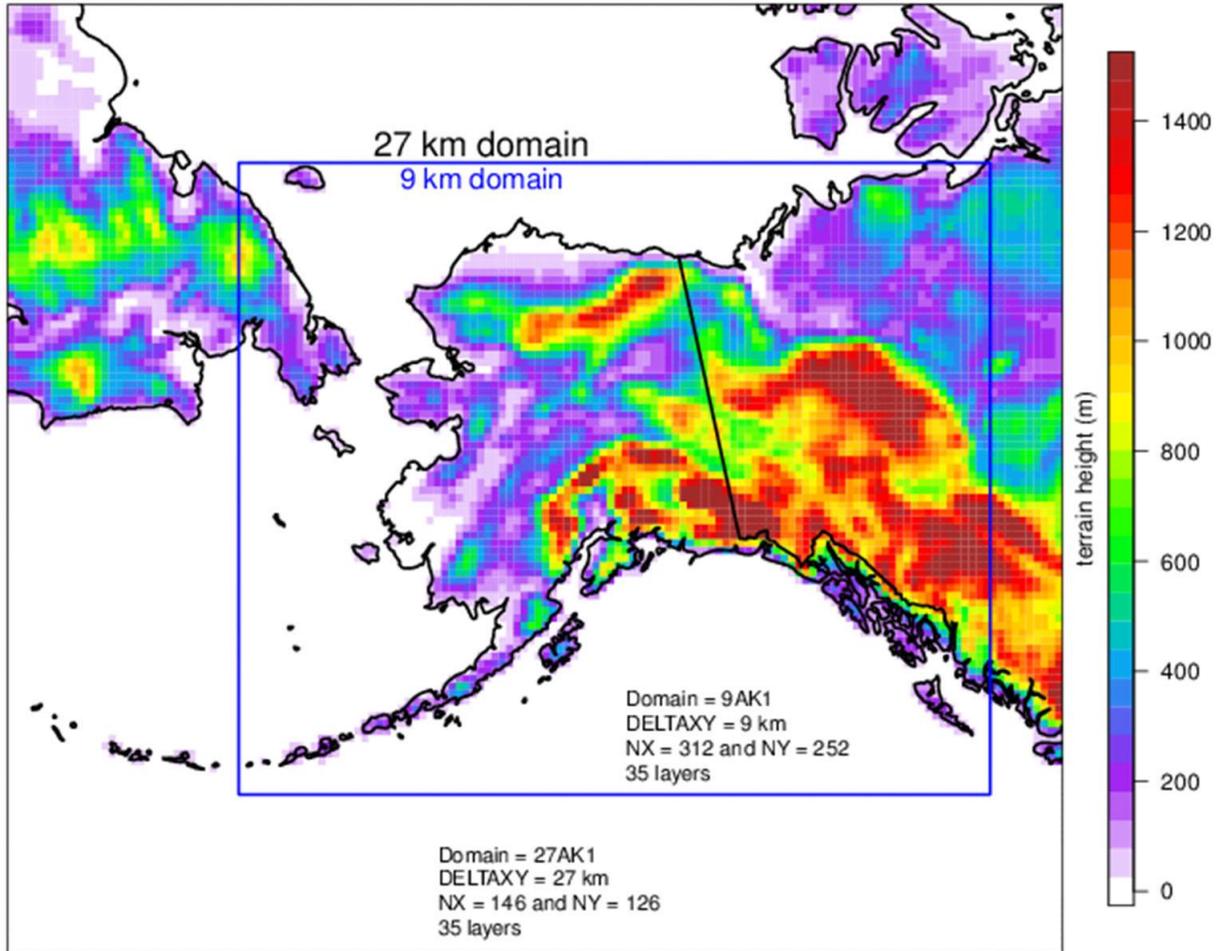


Figure 2. 27-km and 9-km grid resolution modeling domains used in EPA’s Alaska CMAQ 2016 PGM modeling platform and 2028 regional haze modeling.

INITIAL DISCUSSION OF POTENTIAL SOURCE CONTRIBUTIONS TO SULFATE AT ALASKA IMPROVE SITES

The WRAP 2014 GEOS-Chem emissions data were analyzed to provide an initial summary of the source of sulfur emissions in an around Alaska as well as a CONUS domain and across the globe. **Table 2** shows the contributions of reactive sulfur emissions by anthropogenic and natural emissions for an Alaska and CONUS emission extraction domain. It is recognized that sulfate impacts at Alaska IMPROVE sites are a combination of sources within and in the immediate vicinity of Alaska (e.g., the Alaska emissions extraction domain in **Figure 1** and EPA 9-km modeling domain in **Figure 2**) as well as long-range transport from other continents (e.g., Russia and China). Thus, analysis of emissions within and near Alaska is only part of the sulfate contribution.

Figure 3 displays the relative importance of reactive sulfur emissions within the Alaska emission extraction domain (**Figure 1**) and the five major source categories analyzed from the GEOS-Chem emissions inventory. The Volcano degassing SO₂ emissions account for 41% of the reactive sulfur emissions in the Alaska domain, with DMS accounting for another 30%. Anthropogenic and shipping emissions account for 12% each. Biomass burning (e.g., wildfires) account for 5% of the reactive sulfur emissions, but fire emissions have a lot of year-to-year variability and 2014 was not a high wildfire year in Alaska (**Figure 4**).

In contrast to the Alaska domain, where only 24% of the reactive sulfur emissions were from anthropogenic sources and shipping, almost 90% of the reactive emissions are anthropogenic and shipping in the CONUS domain (**Figure 3**). And world-wide, 65% of the reactive sulfur emissions are anthropogenic and shipping.

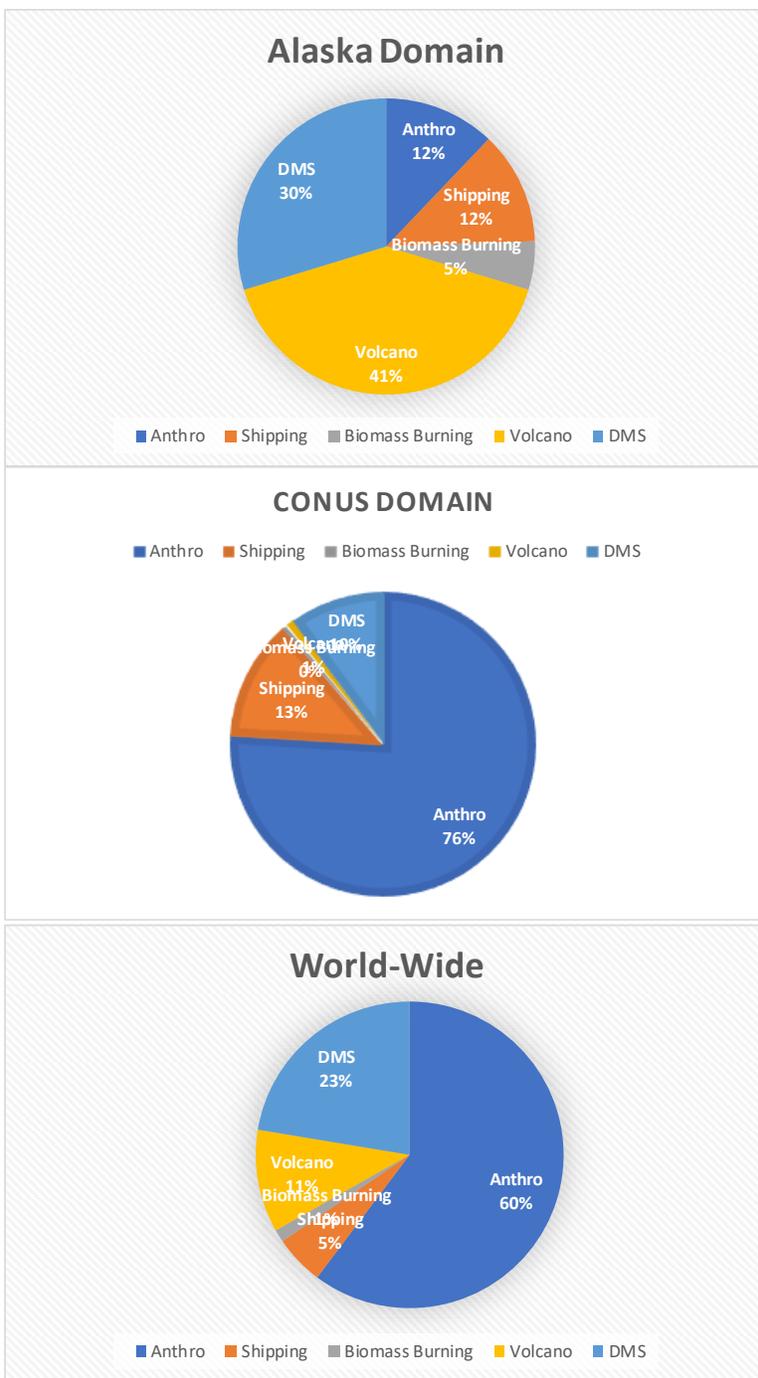


Figure 3. Relative importance of reactive sulfur emissions from the GEOS-Chem HEMCO emissions processor for 2014 of four emission extraction domains: Alaska, CONUS, and World-Wide.

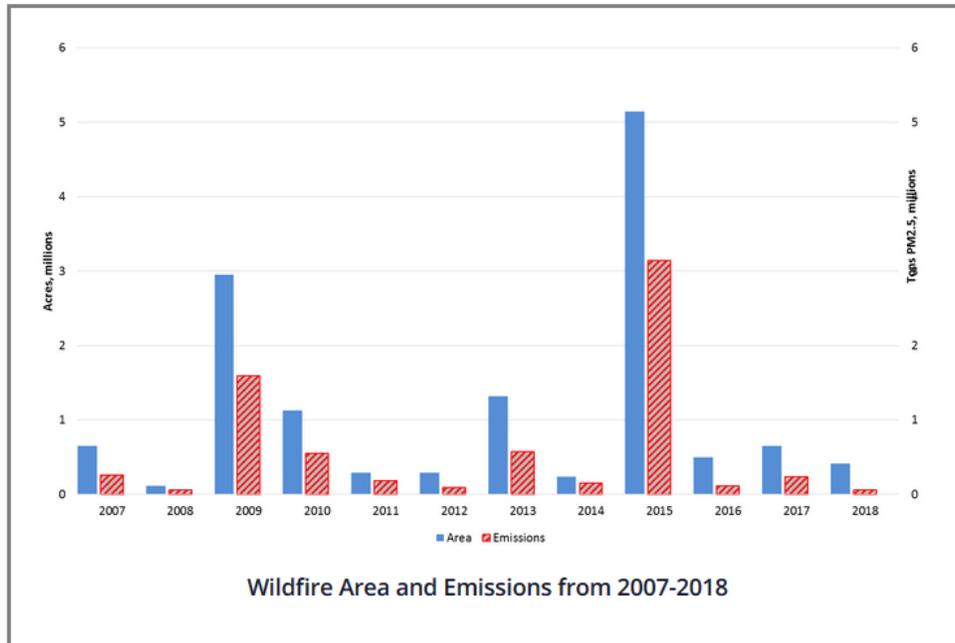


Figure 4. Annual acres burned (millions) and wildfire PM2.5 emissions (million tons per year) in Alaska 2007 – 2018 (Source: <https://dec.alaska.gov/air/anpms/projects-reports/fire-emission-inventory/>).