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Unveiling the Implicit: Arctic Coastal Aerosol Processes

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Abstract: Arctic coasts cover more than 101,000 km and emulsify terrestrial, marine and socio-economic ecosystems. All three components produce specific emissions that contribute to the mix of atmospheric constituents, which are processed and dispersed in the coastal atmosphere to contribute to cloud formation through cloud condensation nuclei and ice nucleating particles. Clouds strongly influence the coastal energy balance. Importantly, Arctic coastal ecosystems are exposed to multiple pressures such as the warming atmosphere and ocean, the thawing cryosphere and the expanding anthropogenic activities. This means that coastal emissions and atmospheric processes are in constant evolution. Given the large area covered by coasts and the mix of emission sources, coastal aerosol processes deserve quantification to better understand their role in accelerated Arctic climate change.

Keywords: Aerosol · Arctic · Coast · Cloud condensation nuclei · Ice nucleating particles

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1. Introduction

Conceptionally, the Arctic has been experiencing two aerosol regimes: anthropogenic emission-dominated Arctic haze in winter, and natural emission-dominated low concentrations in summer.[1,2] During polar night, the Arctic atmosphere receives mid-latitude air masses and due to the strong stratification and absence of precipitation, anthropogenic aerosols then accumulate over the region between November and May forming the so-called Arctic haze.[3,4] During polar day, the energy from solar radiation leads to an unstable atmosphere with frequent cloud formation and precipitation.[3] This reduction of pollution from long-range transport reaching the Arctic favors local emissions, *e.g.* from the ocean, to determine Arctic aerosol composition.[5,6] Aerosol properties across the Arctic have been studied widely, because they influence Arctic surface temperature.[7] The Arctic is warming at an accelerated rate, up to four times faster than the global average, a phenomenon referred to as Arctic amplification.[8,9] Arctic haze generally contributes to cooling the Arctic through direct scattering of solar radiation. However, given the recent decrease in air pollution emissions in the northern hemisphere, the ensuing decline of Arctic haze has led to a net warming.[4,7] Aerosols also influence cloud radiative properties through acting as cloud condensation nuclei (CCN) and ice nucleating particles (INP) that are responsible for cloud droplet and ice crystal formation, respectively.[10] In the high Arctic, clouds are typically mixed-phased, *i.e.* containing both liquid droplets and ice crystals. Arctic low-level mixed-phase clouds have an overall warming effect, but they can have cooling effects over the open ocean and Arctic land.[11]

Characterizing the Arctic aerosol regime as 'haze' and 'non-haze' is useful for a simple classification of Arctic atmospheric composition regimes. There are however far more nuances to Arctic aerosol sources and processes that are related to regionality, seasonality and climate change.[12,13] Neither the winter nor the summer regime, can be understood as homogeneously present across the Arctic. The Arctic is a region of highly diverse environments, including (snow-covered) landmasses, islands and archipelagoes, open ocean, the marginal ice zone and the ice-covered ocean. Emissions of aerosols and their precursors can be very different in each area, as well as varying seasonally. Briefly, Arctic terrestrial emissions include volatile organic compounds (VOCs), which are aerosol precursors, from the tundra,[14] VOCs and particles from wild fires[15,16] and human activities, *e.g.* fossil fuel

combustion such as gas flaring.[17] The open ocean is a source of sea spray aerosol (SSA) that consists of sea salt and organic material, as well as primary biological aerosol particles.[18,19] The marginal ice zone is microbially very active, meaning that VOCs emitted from phytoplankton blooms, *e.g.* dimethyl sulfide, are released[20] and they constitute important precursors to secondary aerosols, *i.e.* aerosols that are formed through chemical reactions in the atmosphere. A more recently studied source originates from the pack ice which is covered with salty snow throughout much of the year, leading to the formation of airborne particles within blowing snow.[21]

From a seasonal perspective, aerosol emissions and processes within the Arctic change dramatically due to the drastically changing sea ice extent, temperatures, and rapid shift from constant day to constant night. In the absence of light, biological emissions are at a minimum and the terrestrial and marine biospheres are covered under snow and ice. In addition, the absence of solar radiation inhibits many atmospheric photochemical processes.[26] With an increase in temperature and solar radiation comes increasing melt of sea ice and glaciers and an increase in biological productivity meaning that in Arctic summer biogenic emissions are at their maximum. Marine phytoplankton blooms are driven up to one third by riverine discharge of nutrients which reach the Arctic Ocean in late spring.[27,28] In summer, the retreat of the sea ice is associated with emissions from human marine activities, *e.g.* shipping.[29] Moreover, the Arctic is home to several hundred million[30] migratory sea birds that are a large source of ammonia during polar day, a caustic gas that is critical to the formation and growth of new particles.[11] Another seasonal source of aerosols in the Arctic is high latitude dust. High latitude dust is generated, in part, due to the exposure of highly sedimented land regions during the melt season and is lofted by surface winds, contributing then to the airborne INP number concentration.[23,31]

Due to the Arctic amplification, climate change is particularly visible across the Arctic and bound to impact emissions and atmospheric processes relevant for aerosol formation and properties. Sea ice extent has declined by more than 40% in the last decades,[32] allowing for more increased marine VOC emissions.[20] With thawing permafrost, the Arctic coastline is eroded by roughly 0.5 m year^{-1} adding nutrients for marine productivity to the ocean.[33] Terrestrial VOC emissions are increasing[14] as well as emissions from more frequent and intense boreal wild fires.[34] In addition, there are changes in atmospheric dynamics that favor air mass transport along latitudinal transects allowing for more moisture to enter the Arctic,[35] with more precipitation expected in the future Arctic,[36] meaning an increase in wet deposition.[37] Finally, atmospheric composition of oxidants is changing, with ozone mixing ratios exhibiting a decreasing trend in the spring and increasing in summer.[38] Bromine monoxide concentrations are increasing. The formation of the gas is related to first year sea ice formation, and it is a significant sink of atmospheric oxidants, particularly ozone in spring.[39]

Summarizing the above description on a map (Fig. 1) highlights that many environmental changes as well as aerosol relevant emission sources are located around the Arctic coastline, which covers a distance of > 101,000 km.[33] More specifically, marine net primary production and marine VOC emissions are fueled by riverine outflow, high latitude dust sources are located along some coasts as are many of the bird colonies. Many anthropogenic activities, be it oil and gas extraction, shipping or urban life occur close to the coast. Here, we define 'coastal' loosely as 50 km in either direction from the coastline covering an area where emissions can mix and react at moderate windspeeds such as 2-3 m/s. In this review article, we focus on Arctic coastal emission sources and aerosol processes that have an impact on the cloud condensation nuclei and ice nucleating particle number concentrations, which constitute the vitally important link to aerosol climatic effects through cloud formation.

2. Coastal Aerosol Sources and Processes

The following sections discuss coastal Arctic aerosol processes by emission sources with a focus on four aspects: riverine influ-

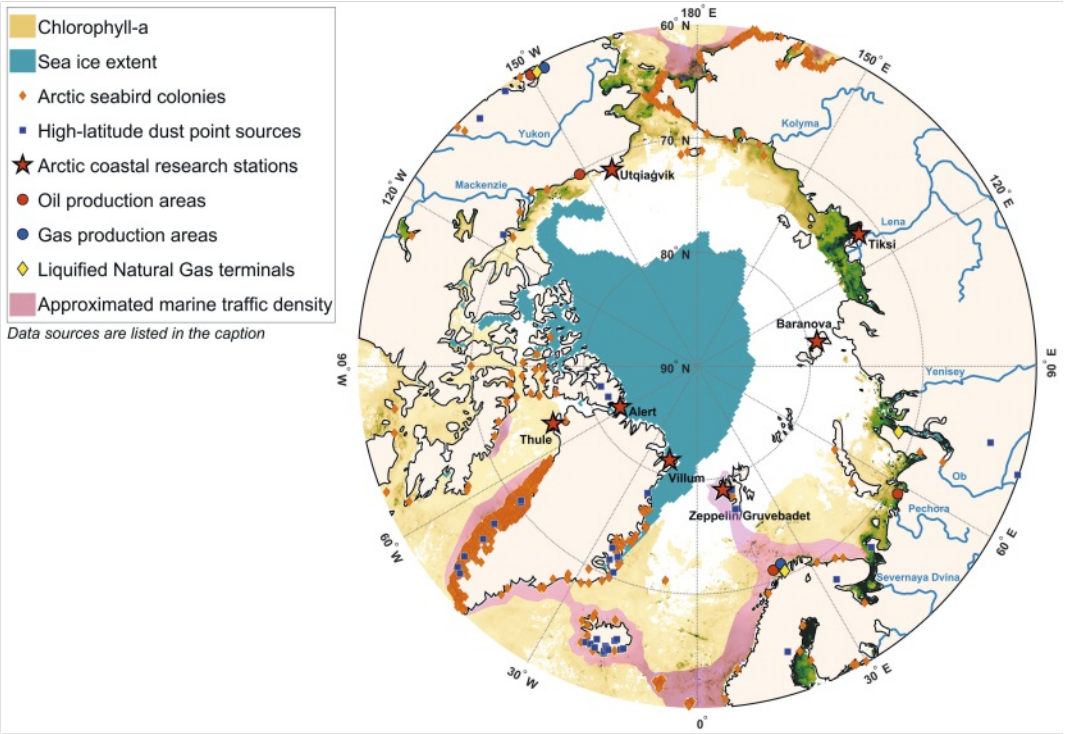

Fig. 1. Map of Arctic coastal processes impacting aerosol populations. Sea ice extent is at the 2020 minimum (Sep. 15th), from Nimbus-7 SMMR and DMSP SSM/I-SSMIS passive microwave data (National Snow and Ice Data Center). Chlorophyll-a concentrations, from the MODIS sensor on the NASA Aqua satellite, are also provided for that same date along with the tracks of the main Eurasian and North American rivers discharging nutrients in the Arctic Ocean. The location of the main Arctic seabird colonies, retrieved from the Riddick *et al.*,[22] global ammonia emissions from seabird inventory, is indicated with orange diamonds. High-latitude dust point sources, as identified by Meinander *et al.*,[23] are indicated with blue squares. Examples of anthropogenic activities related to energy production (oil and gas production[24]) and approximated marine traffic density,[25] are indicated on the map. Finally, the non-exhaustive location of Arctic coastal research stations is indicated with red stars.

ence, animal colonies and coastal ecosystems, high latitude dust and anthropogenic activities. Processes discussed in the text are illustrated in Fig. 2 and referred to by the numbers in square brackets.

2.1 Riverine Influence and Coastal Erosion

On a volume basis and compared to other oceans, the Arctic receives the highest terrestrial input of freshwater, organic matter and nutrients through riverine outflow, mainly in the Eastern sector.[40] A similarly significant source of sediments to the ocean is coastal erosion.[41] This influx of nutrients leads to enhanced phytoplankton blooms near the river deltas[27] with three main implications for aerosols. First, the presence of riverine organic matter has been shown to increase the number concentration of SSA and thereby the number concentration of CCN that were equally efficient in forming droplets as SSA from ocean water not influenced by riverine outflow.[42] Second, waters with riverine sediment off the Russian coast were reported to contain high INP concentrations under strong wind conditions (Fig. 2, [1]).[43] This suggests that these coastal waters are rich in ice-nucleating material, potentially related to the strong riverine input or coastal erosion due to thawing permafrost, with the latter having been demonstrated to provide effective INP.[44] In contrast, the pack ice, open leads and the MIZ appear to be weak sources of INP. Third, the enhanced phytoplankton activity in areas with riverine influence has driven an increase in dimethyl sulfide (DMS) emissions (Fig. 2, [2]) in 2011 – 2016 versus 1998 – 2003 north of 70°N, and in particular north of 75°N where previously no riverine influence was detected.[20] It is also postulated that additional sulfur input from riverine outflow could mitigate marine microbial sulfur limitation further enhancing DMS production.[45] In the atmosphere, DMS is converted to sulfuric acid, an important condensable gas active in new particle formation,[46] and methanesulfonic acid (MSA) which was found to be important for the growth of aerosol particles in the Arctic.[47]

2.2 Arctic Wildlife and Coastal Ecosystems

The Arctic coastline is a key habitat to migratory seabirds[48] and seals.[49] Both species are strong emitters of ammonia,[50] while much more evidence for ammonia emissions of seabirds in the Arctic currently exists.[11,51] Ammonia is an important ingredient to both new particle formation (NPF) and particle growth.[3] Direct measurements of NPF in Ny-Ålesund, Svalbard, elucidated that ternary nucleation from sulfuric acid, ammonia and water vapor is the major process[46] with subsequent growth of particles to CCN sizes by sulfuric acid and MSA in spring. While this study only inferred that ammonia originated from sea bird colonies, Croft *et al.*[11] conducted a modeling study to investigate specifically the impact of ammonia emission from Arctic seabird colonies on NPF, CCN and cloud properties. Using above-mentioned ternary NPF scheme, the pan-Arctic seabird influence was shown to be responsible for high number concentrations of particles > 10 nm. The seabirds' ammonia emissions were also shown to be important for growth of particles to CCN-relevant sizes, in line with observational evidence of high neutralization levels of submicron aerosol in the marine boundary over the Canadian archipelago.[51] Across the Arctic, according to this modeling study, ammonia emissions increase the cloud droplet number concentration resulting in local cooling effects due to enhanced solar radiation scattering by higher cloud albedo (Fig. 2, [4]).

In addition to ammonia-emitting animal colonies, coastal ecosystems also contain highly active marine microbial areas and diverse species of terrestrial vegetation. As mentioned, springtime particle growth around Svalbard is driven by sulfuric acid and MSA, which originates from phytoplankton activity.[46] In summer, an additional driver of growth was found, *i.e.* condensable organic vapors (Fig. 2, [5]) both around Svalbard as well as in the Canadian archipelago creating CCN-sized particles.[46,47,52] The exact source of the organic vapors is not known in either location, but the fact that they emerged after snow melt on Svalbard suggests a terrestrial source, whereas in the coastal regions of the Canadian archipelago both terrestrial and marine ecosystems are possible sources. Generally, nucleating and condensable vapors such as sulfuric acid and organics may also be transported from further away and lead to NPF and growth at Arctic coasts,[53] where they encounter precursor gases such as ammonia and further favorable conditions for the observed processes to occur. Coastal NPF is a world-wide phenomenon[54] and recent observations confirm that the Arctic is no exception.

2.3 High Latitude Dust Sources

Dust is one of the most important (mass-wise) components of Arctic aerosols with diameters < 10 µm, *e.g.* second only to sea

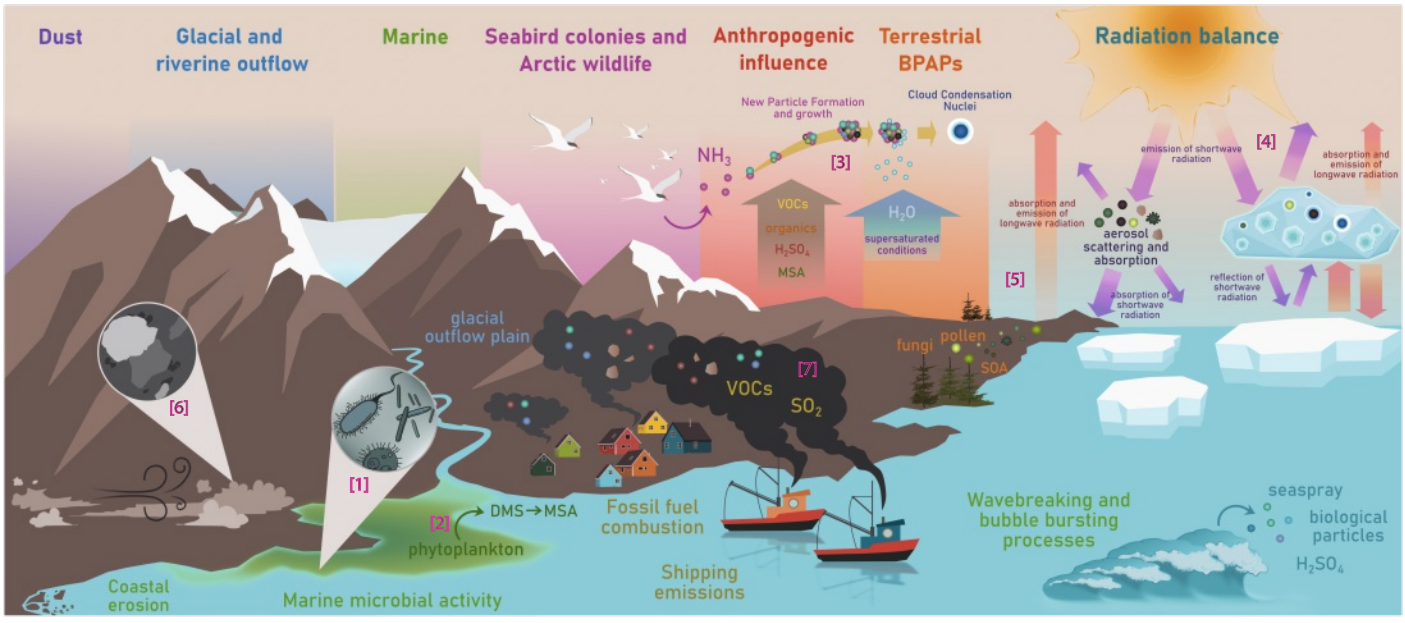

Fig. 2. Coastal Arctic aerosol processes. The cartoon illustrates a hypothetical landscape with elements discussed in the text as referred to by the numbers in square brackets. DMS is dimethylsulfide, MSA methanesulfonic acid, PBAPs primary biological aerosol particles, SOA secondary organic aerosols, VOCs volatile organic compounds.

salt on Svalbard.[15] Dust in the Arctic can be long-range transported from *e.g.* the Sahara desert,[55] but also sourced locally.[56] Dust sourced north of 60°N contributes around one third of total dust in the lower Arctic atmosphere.[23,57] Aerosolization of glacial outwash plain sediments,[6] located along the Arctic coasts, is one of the main contributing processes[58,59] and an important source of INPs across the Arctic.[31,60] Tobo *et al.*[31] collected samples from vegetation-free active outwash sediments (< 5 μm) in Svalbard that nucleated at warm temperatures up to –5°C.

The high ice nucleation ability was hypothesized to originate from the presence of small amounts of organic matter. In addition to the sediment samples, aerosol samples were collected at the Zeppelin observatory and INP concentrations exceeded previously reported values for summertime marine boundary layer at high latitudes. Dust from Svalbard is hypothesized to partly explain these higher concentrations: 0.1 μg/m^3 of outwash sediments from Svalbard would suffice to reproduce INPs observations from the samples (for reference: the average contribution of mineral dust in summer on Svalbard is 0.6 μg/m^3, ref. [15]), with one order of magnitude higher INP concentrations observed in July 2016 compared to March 2017, when outwash plains where still covered by snow. In addition, dust particles can act as carriers of biological particles,[61] which are very effective INP, freezing at higher temperatures, potentially making high latitude dust a versatile source of INP.

Arctic-wide model simulations have been run to estimate the importance of high latitude dust on the Arctic INP number concentration and results suggest that the concentration is 100 times higher compared to simulations neglecting high latitude dust.[57] Other modeling studies demonstrate the importance of high latitude dust in the lower troposphere and their impact on mixed-phase clouds.[62] By changing the cloud phase through contributing INP, high latitude dust induces a net cooling effect (Fig. 2., [4]). Also, glacial outwash plains are typically connected to the ocean and fjords *via* rivers that transport the sediments to marine ecosystems thereby providing nutrients, which are also provided through atmospheric transport of dust and subsequent wet and dry deposition, fostering marine microbial growth and their atmospheric emissions which contribute to the atmospheric aerosol population.[59]

2.4 Anthropogenic Emissions

Though sparsely populated, with the entire Arctic being home to approximately 4 million people,[63] anthropogenic activities have an important impact on Arctic climate.[7] Many of the Arctic anthropogenic activities producing emissions occur along the coast, such as fishing, shipping, oil and gas production and tourism.[24] Generally, these activities rely on fossil fuel combustion, producing emissions of aerosols and precursors comparable to other parts of the world. For example, emissions of semi-volatile hydrocarbons from the oil and gas extraction facilities at Prudhoe Bay, Alaska, were estimated to induce particle growth events[7] on approximatively 30% of days, excluding polar winter, following photochemical oxidation.[64] This influence of anthropogenic emissions was inferred to be greater than the influence of marine, terrestrial, and other coastal emissions combined for particle growth events. The growth events lead to particles large enough to act as CCN that are transported upwards[65] and influence cloud radiative properties.[4] It was found that enhanced CCN number concentrations from Prudhoe Bay emissions reduced cloud droplet effective radii by up to 1 μm thereby increasing cloud albedo and inducing cooling at the surface.[66] In addition, it was observed that up to 10% of the < 1 μm particles emitted from Prudhoe Bay are black carbon.[67]

At present, most of the fuel used in ships in the Arctic is still crude oil, producing a large variety of emissions of aerosols and precursor gases[68] that affect atmospheric composition significantly along major shipping routes, *e.g.* coastal Norway, or main touristic destinations, *e.g.* Svalbard.[29,69] Much of the research has focused on the impact of ship emissions on aerosol mass concentrations and ozone mixing ratios, rather than explicitly investigating changes in CCN, INP and cloud properties, while modeling studies predict generally a cooling effect due to cloud alteration over dark surfaces.[70,71] As regards black carbon, shipping off the northern coast of Norway increases its coastal concentration by ~40%.[70] Laboratory studies mimicking the ongoing change in fuel composition towards low-sulfur content and deployment of wet scrubbers show that scrubbers increase the CCN activity but not the total number of CCN when using high sulfur content fuel. Usage of low-sulfur fuel results in highly hydrophobic particles, while CCN concentrations decrease by one order of magnitude.[72]

3. Discussion and Outlook

Arctic coasts can be hotspots of aerosol emissions (*e.g.* primary particles such as dust and biological aerosols) and their precursors. In addition to being emission hotspots, they can also provide the right mix of ingredients to foster secondary aerosol production through new particle formation, *e.g.* when sulfuric acid meets ammonia, and growth, *e.g.* by methanesulfonic acid or organic vapors. Moreover, coastal aerosol processes can be triggered by natural or anthropogenic emissions or a mix of both. What distinguishes the coasts from other Arctic regions is that they bring together a particularly large variety of natural and socio-economic ecosystems on land and in the ocean that all impact atmospheric composition. Currently, quantification of Arctic coastal emissions and their climatic impacts remains elusive. What is understood however is that they are important and subject to drastic change through climate warming. In a relatively small area, coasts incubate climate forcing and impacts through the atmosphere, ocean, cryosphere, land, and biosphere as well as socio-economic changes.

Many open questions remain: With regards to riverine outflow and coastal erosion into the ocean, neither the source strength nor the INP efficiency of sea spray emissions has been assessed and relevant studies for CCN are scarce. In particular accelerating coastal erosion[41] could be an overlooked marine and aerosol source, as well as a source of VOCs,[73] *i.e.* aerosol precursors. Coastal ecosystems emit a rich mix of aerosol precursor gases, where in particular emissions of VOCs from terrestrial vegetation are not well constrained but likely important due to the ongoing large-scale vegetation shifts as a response to atmospheric and marine warming.[74] At the same time changes are expected in the ecosystems of coastal oceans,[75] where changes in VOC emissions are conceivable based on comparative studies across high latitudes[46] but not yet generally understood. While ammonia emissions from birds have attracted some attention, there is a near absence of knowledge on emissions from large seal colonies, which are also strong emitters of ammonia.[50] Moreover, some of the currently migratory sea bird species might shift to year-round residency in the Arctic, changing the seasonal cycle of inner-Arctic ammonia emissions.[30] Focusing on high latitude dust, early modeling attempts demonstrate the potential importance of this coastal aerosol source for Arctic clouds and warrant more investigations that assess specifically dust emission fluxes and INP ability for many Arctic sites. For example, Greenlandic outwash plains appear to be a strong dust source,[76] albeit not quantified and characterized for INP. There are also indications of wind speed intensification in some locations in the future Arctic,[77] which might increase the dust emission flux. From a socio-economic perspective, a strong interest in development of the Arctic exists and new initiatives such as the Arctic Economic Council established by the Arctic States provide evidence.[24] The Arctic is thought to hold 30% of the world's gas and 13% of the world's oil reserves,[78] and shipping is thought to increase in the future with more ice free ocean.[79] It is unclear

in how far Arctic development will be driven by fossil or post-fossil ambitions.

Altogether, the natural and socio-economic Arctic coastal ecosystems are an important and dynamic source of aerosols and their precursors within the Arctic that might be responsible for a significant part of the Arctic aerosol population. The coastal emission sources should be benchmarked against long-range transport, as well as emissions from the open and ice-covered Arctic Ocean.

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