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Local Arctic air pollution: A neglected but serious problem

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Key Points:

- Local Arctic air pollution is among the most severe world-wide
- Arctic meteorological conditions exacerbate air pollution and create unique pollution formation mechanisms
- Future economic activities in the Arctic are expected to increase local air pollution

This is the author manuscript accepted for publication and has undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the Version of Record. Please cite this article as doi: [10.1029/2018EF000952](https://doi.org/10.1029/2018EF000952)

Abstract

Air pollution in the Arctic caused by local emission sources is a challenge that is important, but often overlooked. Local Arctic air pollution can be severe and significantly exceed air quality standards, impairing public health and affecting ecosystems. Specifically in the wintertime, pollution can accumulate under inversion layers. However, neither the contributing emission sources are well identified and quantified nor are the relevant atmospheric mechanisms forming pollution well understood. In the summer, boreal forest fires cause high levels of atmospheric pollution. Despite the often high exposure to air pollution, there are neither specific epidemiological nor toxicological health impact studies in the Arctic. Hence, effects on the local population are difficult to estimate at present. Socio-economic development of the Arctic is already occurring and expected to be significant in the future. Arctic destination shipping is likely to increase with the development of natural resource extraction, and tourism might expand. Such development will not only lead to growth in the population living in the Arctic but will likely increase emission types and magnitudes. Present-day inventories show a large spread in the amount and location of emissions representing a significant source of uncertainty in model predictions that often deviate significantly from observations. This is a challenge for modeling studies that aim to assess the impacts of within Arctic air pollution. Prognoses for the future are hence even more difficult, given the additional uncertainty of estimating emissions based on future Arctic economic development scenarios.

1 Introduction

Air pollution in the Arctic is not a problem that has only recently surfaced. Arctic Haze - an accumulation of fine particulate matter and trace gases in the lower Arctic atmosphere in winter and early spring - has been observed since the 1950s (Mitchell, 1956). A significant number of studies have demonstrated long-range transport of air pollution to the Arctic (AMAP, 2015a and references therein; Law & Stohl, 2007; Law et al., 2014). Local sources of pollution (e.g. shipping, oil extraction, flaring or metal smelting) are also important, affecting atmospheric composition at regional and Arctic-wide scales (e.g. see Law et al., 2017 for further discussion). Yet, very few studies have examined local sources and their impacts on inhabited areas and their surroundings. There is poor understanding of atmospheric chemical reactions of air pollutants under the unique Arctic conditions, including extremely cold and dry air with little solar radiation in winter. Air pollution can have adverse effects on human health and ecosystems (Anenberg et al., 2012; Shindell et al., 2012), and induce climate-relevant change through direct scattering and absorption of radiation, altering cloud properties, precipitation (Maahn et al., 2017), and snow albedo (Flanner et al., 2007).

During winter, people living in Arctic cities and villages (Figure 1) are often exposed to high levels of pollution (Wang & Hopke, 2014; Ward et al., 2012), leading to concerns about health effects on residents. However, few chemically detailed observations exist since most research sites in the Arctic have historically focused on measurements of “background” atmospheric composition at remote sites for climate studies, often disregarding or not collecting data when influenced by local pollution (Dutkiewicz et al., 2014; Heintzenberg & Leck, 2012; Sharma et al., 2013; Uttal et al., 2016).

Sources of, and mechanisms for human exposure to pollution in the Arctic vary seasonally. During wintertime, cold temperatures and strong surface-based temperature inversions, which can trap locally emitted pollution, are common (Serreze et al., 1992; Tran & Mölders, 2011). This trapping of pollution is particularly acute when population centers are in valleys or bowls, where topography further decouples surface winds from airflow aloft. In this way, Arctic cities can be particularly vulnerable to pollution caused by the activities of the residents and local industry (Leelasakultum et al., 2012; Tran & Mölders, 2011). Additionally, some local sources of pollution are stronger in wintertime because cold temperatures require more intense domestic heating and power generation, and mobile sources may be operating poorly due to extreme cold leading e.g., to increased emissions from automotive cold start (Platt et al., 2017). In contrast, during summer, near-constant sunlight and subsequent atmospheric heating causes stronger vertical mixing, while also more clouds are present and precipitation can remove air pollutants. However, in the summer, large forest fires are common and can cause episodic extreme pollution events (Kelly et al., 2013). Overall, sources and impacts of current local Arctic air pollution remain poorly quantified.

Similarly, information on the potential future evolution of in-Arctic air pollution sources is scarce. Changes in local sources are expected due to sea ice loss that is progressing rapidly as the Arctic warms, and by 2050 the Arctic could be nearly sea ice free in summer (Overland & Wang, 2013). This sea ice reduction is catalyzing growth of Arctic industrialization, commercial shipping, fishing, and tourism (Allison & Bassett, 2015; Meier

et al., 2014). It is estimated that around 30 % of the world's undiscovered gas and 13 % of undiscovered oil resources are located in the Arctic, providing significant incentive for increasing fossil fuel exploration and extraction with sea ice loss (Gautier et al., 2009). All these activities are expected to be increasing in-Arctic emissions.

With the realization of such commercial opportunities, the number of residents in the Arctic is also set to increase. While the Arctic population (~4 million) slightly decreased between 2000 and 2010, an increase on the order of 30 % by 2030 is projected for the North American Arctic, much less increase or population stagnation in the European Arctic, and a decrease in Russia (Larsen & Fondahl, 2015). However, populations may increase in regions where there is significant resource extraction, as is already the case in northern Canada. Also, given that the world population is projected to grow beyond 9 billion by mid-century, with associated increases in demand for resources, putting pressure on those found in the Arctic, it can be expected that the current urbanization trend in the region accelerates in the future (Larsen & Fondahl, 2015). With this projected socio-economic development, local emissions of air pollutants and greenhouse gases are likely to increase and will mostly be emitted around industrial activities and human settlements.

The objectives of this study are (1) to identify gaps in knowledge about local Arctic air pollution sources and associated impacts on human health and ecosystems, and (2) to recommend a research agenda to address these gaps. Against the background of near- to mid-term socio-economic development of the Arctic, answering the most pertinent questions is essential for a timely design of sustainable development policy strategies. In this article, we summarize what is known of air pollutant emissions from Arctic sources in section 2. Section 3 highlights uncertainties in future Arctic pollution emission estimates. In section 4, we discuss specific aspects of Arctic meteorological conditions that can exacerbate air pollution, and in section 5 we provide a concrete example of severe local air pollution. Section 6 elaborates further on the present knowledge and open questions of atmospheric chemical processing of air pollution in Arctic specific conditions, i.e. in a cold and dark environment in winter and long sunshine hours in summer. Section 7 and 8 briefly summarize what is known about air pollution effects on human health and ecosystems in the Arctic, respectively. Finally, in section 9, we present recommendations for a research agenda that addresses questions that are key to mitigating effects of current local Arctic air pollution and to limiting these effects in the future.

2. Local Arctic Air Pollutant Emissions

Local Arctic emissions include combustion sources such as vehicle emissions from transportation, domestic heating by wood, oil, coal, or natural gas, and power generation by burning of diesel, coal, or natural gas (AMAP, 2015a). Industrial activities in the Arctic also generate emissions and are linked to oil and gas extraction, metal smelting, and mineral extraction (AMAP, 2015a). Shipping is another source of air pollutants in the Arctic along the coast and inland-rivers (Aliabadi et al., 2015; Eckhardt et al., 2013). Furthermore, waste burning and biomass burning produces emissions of reactive gases and aerosol particles (AMAP, 2015a). Boreal forest fires are an important natural source of pollutants (even if fires

can be triggered by human activity, AMAP, 2015a). Local emissions from vegetation (forests, tundra), oceans, and occasional volcanic eruptions are other sources of natural emissions. Re-suspended dust from volcanic ash sediments or glacial deposits also provides a source of natural aerosol particles.

Recent emission estimates for the Arctic have often focused on black carbon (BC) due to its contribution to global warming via direct radiative forcing and snow and ice albedo reduction, rather than its health impacts as an air pollutant. Other pollutants, especially those that have implications for public health or ecosystems, have received less attention: e.g. nitrogen oxides, NO_x ; volatile organic compounds, VOCs; sulfur dioxide, SO_2 ; carbon monoxide, CO; and particulate matter, PM. In addition, not all sectors that contribute to local emissions in the Arctic were considered in previous assessments, e.g. waste burning is not considered (AMAP, 2015a). There are also considerable uncertainties in current emissions which are reflected in the differences between inventories. Figure 2 provides an example of discrepancies between three recent BC emission inventories in the Eurasian Arctic: the Arctic Black Carbon dataset (Huang et al., 2015), ECLIPSEv5 emissions (Stohl et al., 2015), and the Hemispheric Transport of Air Pollution (HTAPv2) inventory (Janssens-Maenhout et al., 2015). The main differences are the overall magnitude and spatial distribution of emissions. Those are partly due to the inclusion of specific emission sources such as gas flaring or high emitting vehicles in the Arctic Black Carbon and ECLIPSEv5 inventories but not in the HTAPv2 emissions (see Sergent (2017) for further discussion). In chapter 5 of the AMAP assessment report on 'Black carbon and ozone as Arctic climate forcers' (AMAP, 2015a) emissions of BC, organic carbon, SO_2 , NO_x and CO from eleven global emission inventories are compared for several years in various latitudinal bands. For emissions north of 60°N the ratio between maximum and minimum emissions in 2010 (the most recent available year) is ~ 3.1 for BC, ~ 1.5 for NO_x , 1.6 for SO_2 , and exceeds 2.3 for all other pollutants (3.1 for BC). For comparison, globally ratios are around 1.2 (1.4 for CO) for all substances. This uncertainty in emissions emphasizes that improvements are needed, especially for the Arctic.

In the following sub-sections, we review the different emission sectors pertinent to the Arctic. However, there are very few Arctic-specific emission inventories, although recently, a number of emission inventories have been improved for both sector and national emissions in the Arctic region.

2.1 Resource extraction including flaring

Resource extraction is an important source of local emissions in the Arctic, especially in Russia, Alaska, and Norway. Emissions come from activities linked to metal smelting and oil and gas extraction. Mining activities may also generate local dust emissions but this source is poorly quantified at present. Studies investigating dust sources within the Arctic focus only on natural sources as opposed to industrial activities such as mining (Groot Zwaafink et al., 2016).

It has been recognized for some time that metal smelting activities in northern Russia at Norilsk and the Kola Peninsula in northwest Russia are a large source of SO_2 emissions in the Arctic (e.g., Stohl et al., 2013). SO_2 is oxidized to particulate sulfate. Prank et al. (2010) updated SO_2 and PM emissions for the Kola Peninsula after finding that emissions from one

of the largest point sources, the Nickel smelter plant on the border with Finland, were either missing, misplaced or wrongly attributed in major European inventories. These emissions were shown to make a significant contribution to air pollutant levels in neighboring northern Finland (Ruoho-Airola et al., 2015).

In the past years, it has been recognized that flaring linked to oil and gas extraction, particularly in northern Russia, is an important source of BC, suggested to be 66 % of the total Arctic BC emissions (Stohl et al., 2015; Stohl et al., 2013). More recent estimates suggest higher BC emissions from flaring activities in Russia (Huang & Fu, 2016; Huang et al., 2015) although Evans et al. (2017) note that estimates are very dependent on the emission factors used in different studies, e.g. 1.6 g m^{-3} of emitted gas in Stohl et al. (2013) and 2.27 g m^{-3} in Huang et al. (2015). Using measured isotope ratios, Winiger et al. (2017) showed that flaring BC emissions in Russia might have been overestimated, while emission distribution and source attribution were found faulty in available emission inventories. Estimates of flaring emissions based on VIIRS satellite data suggest that Russian emissions peaked in 2005 and may have declined in more recent years (Huang & Fu, 2016). However, analysis of 2004-2015 flaring NO_2 emissions of in Siberia suggested there has not been a trend (Li et al., 2016), but this study did not include all Russian production fields.

In Alaska (USA), the North Slope of Alaska (Prudhoe Bay) is the third largest oil and gas field in North America. While the United States accounted for 18% of Arctic oil production from 1990-2004, the Alaskan Arctic was estimated to be responsible for 34% of NO_x and 29% of CO emissions from Arctic oil and gas extraction (Peters et al., 2011). Elevated NO_x , NO_y , CH_4 , and CO_2 have been observed at Utqiagvik, downwind of the oil fields (Jaffe et al., 1991; Jaffe et al 1995). In addition, long-range transport and enhanced growth of ultrafine particles has been observed downwind of the oil fields, impacting nearby villages (Creamean et al., 2018; Gunsch et al., 2017; Kolesar et al., 2017). Even though these ultrafine particles might not contribute significantly to PM mass, their impact on human health could be significant (Franck et al., 2011, see also discussion in section 7). With low PM and semi-volatile trace gas concentrations, aging timescales of particles in the Arctic are much longer than elsewhere in the world (Fierce et al., 2017). It is therefore important to understand potential health impacts for existing industrial emission sources and potential future sources that might be located near local communities or cities in the Arctic.

Offshore oil and gas extraction can also impact local and regional air quality in coastal regions and may increase in the future. Some first indications about possible effects were examined based on aircraft data collected downwind of platforms in the southern Norwegian Sea in summer 2012. Data analysis and subsequent modeling show that these activities produce a range of emissions, not only linked to oil and gas production (flaring, venting) but also from stationary “mobile” sources, such as storage tankers and drilling rigs, as well as shuttle tankers, and power generation on the platforms (Roiger et al., 2015; Tuccella et al., 2017). In a model assessment, Tuccella et al. (2017) showed that background ozone and aerosol concentrations are influenced by emissions downwind of the platforms in this region. These studies, and further discussion of these results in Law et al. (2017), highlight significant shortcomings in current inventories and reported emissions such as missing mobile sources and key components like PM and volatile organic compounds, precursors to secondary

organic aerosol (SOA). One challenge is the characterization of emissions through measurements. For example, characterization of BC emissions from flaring is complicated by instrument limitations, such as missing the freshly emitted BC particles that are very small. Also, the highly intermittent nature of these emissions, which is not currently taken into account in inventories that provide annual means, affects emissions estimates (Roiger et al., 2015; Law et al. 2017).

2.2 Combustion

This emission category includes emissions from (a) domestic combustion such as wood, oil, coal, and natural gas burning for residential heating, (b) transport, and (c) power generation by oil and coal-fired power plants. A recent AMAP assessment (2015) reports that surface transportation in the Arctic Council nations contributes about 50 % of the BC and NO_x emissions. Emissions from domestic heating are difficult to determine as they are grouped with commercial emissions in many inventories. Similarly, power generation emissions are often reported together with energy extraction and conversion activities. However, the power generation sector contributes the largest share of SO₂ emissions (AMAP, 2015a)

Stohl et al. (2013) highlighted the importance of domestic heating emissions (as well as flaring) for the Arctic BC burden and showed that inventories need to take into account the seasonality in emissions for models to be able to reproduce observed BC burdens. In the European Arctic, emissions from domestic wood burning have been underestimated, as shown by Hienola et al. (2013) for Finland in a regional modelling study. However, the magnitude of the missing emissions is not quantified. Analysis of BC data, including new measurements of carbon isotopes in BC, collected at Tiksi on the northern coast of eastern Siberia, showed that emissions from transport and domestic heating contribute each more than one third to the atmospheric BC concentrations on average per year (Winiger et al., 2017). Only small contributions come from other sources (gas flaring, power plants and open fires). Measurements and model-based source apportionment of BC in the Kara Strait pointed towards the importance of gas flaring emissions from the Yamal-Khanty-Mansiys and Nenets-Komi regions (Popovicheva et al., 2017). In the White Sea, biomass burning from the mid-latitudes, surface transportation, and residential and commercial combustion in Central and Eastern Europe are important sources of BC (Popovicheva et al., 2017).

Evans et al. (2015) focused on BC emissions from diesel sources on the Kola Peninsula with special emphasis on Murmansk, the largest city north of 60°N. They found that 69% of BC emissions originate from diesel combustion in the mining industry in the Murmansk region due to high fuel consumption and absent control standards. On-road vehicles also made a significant contribution (13%) with a few old heavy-duty vehicles contributing the largest share. Locomotives and smaller diesel generators are also non-negligible sources. In addition, Murmansk operates a large port but emissions from shipping, particularly from fishing ships, are difficult to estimate since data on fuel consumption is highly uncertain. It is not known how much these different activities, and also metal smelting in the Kola Peninsula, contribute to urban air pollution in this region.

This limited number of examples shows that information on emissions from the transport, domestic and power generation sectors is scarce. Detailed knowledge is confined to

select study regions within the Arctic (e.g., Murmansk) that are not necessarily representative of Arctic emissions as a whole.

2.3 Shipping

Arctic shipping emissions have received attention due to the possibility of additional traffic through tourism and oil and gas extraction shipping, as well as traffic likely shifting from lower latitudes to transiting via the Northern Sea Route (NSR) along the northern Russian coast and the Northwest Passage (NWP) across northern Canada and near Alaska (Figure 1). Cross-polar shipping routes have also been proposed (Smith & Stephenson, 2013). In addition, growth in Arctic destination shipping is expected, associated with, for example, increasing tourism and development of resource extraction activities such as along the northern Russian coast e.g. the Yamal LNG facility (MOL, 2017). Despite projections of a continued decline in summer sea ice over coming decades, the future development of Arctic shipping will also be determined by factors affecting economic viability (e.g. fuel savings). Nevertheless, commercial shipping is already using the NSR (Winther et al., 2014), and cargo and cruise ships have already transited the NWP. A recent study showed that during the 2011–2014 period, Arctic shipping was already extensive, concentrated in the Norwegian and Barents Seas, and accessed mainly via NSR (Egufluz et al., 2016). Shipping emissions affect pollutant concentrations along coastal regions, particularly in and near port areas and also via deposition of pollutants to marine and coastal regions. For example, Aliabadi et al. (2015) found pollutant enhancements of 18% in ozone and 32% in PM_{2.5} (particulate matter with a diameter smaller than 2.5 µm), due to summer shipping activities along the NWP. Enhanced equivalent black carbon (EBC), by 45% on average and up to 72% in stagnant conditions compared to the baseline, were found in Ny Ålesund, due to the presence of tourist cruise ships (Eckhardt et al., 2013). However, this study reported lower (by 5%) ozone mixing ratios. Ozone depletion, or titration, is expected near the emission sources where the measurement site was located.

To date, several shipping emission inventories have been developed either for the entire Arctic (Corbett et al., 2010; Peters et al., 2011; Winther et al., 2014), and for regions like the Norwegian or Baltic Seas (Jalkanen et al., 2012). The datasets developed by Winther et al. (2014) and Jalkanen et al. (2012) made use of satellite automated identification system (AIS) ship position data. Jalkanen et al. (2012) used the STEAM2 model (Ship Traffic Emissions Assessment model, version 2) that includes individual ship data, such as position, ship speed, engine and fuel type, to build very high temporal (e.g. 30 minutes) and spatial (e.g. 5 km) resolution datasets. Marelle et al. (2016) found that STEAM2 emissions over the Norwegian coastal region in summer 2012 were comparable to Winther et al. (2014) but were higher than older estimates likely due to the inclusion of fishing ships, the use of AIS data and updated emission calculations. These emission inventories have recently been updated (Johansson et al., 2017; Winther et al., 2017) making use of additional years of AIS data with more complete information about vessel numbers, routes, technical information and more up to date information about emission factors but have yet to be extensively used in model assessments. The Arctic inventory from Winther et al. (2017) also provides new predictions for shipping emissions in 2020, 2030 and 2050; differences with older inventories include an

increased fraction of liquid natural gas vessels in the future, as well as more up to date assumptions about sulfur and nitrogen oxide emissions especially in areas where these emissions are subject to tighter controls (e.g. Baltic Sea).

Using the STEAM2 emissions, Marelle et al. (2016) simulated enhancements in ozone (5%), BC (40%), and PM_{2.5} (10%) for the Norwegian coast during July 2012. This suggests that Arctic air quality is already impacted by shipping emissions and that these effects are comparable to current urban impacts of shipping emissions in Europe (Viana et al., 2014). More recent Arctic-wide simulations using the Winther et al. (2014) emissions investigated the impacts of future shipping and found significant increases in ozone concentrations particularly along the northern Norwegian coast, the Barents Sea, and off the northern coast of Alaska (Law et al., 2017) in line with prior estimates (Dalsøren et al., 2007; Granier et al., 2006). A recent study focusing on the impacts of Canadian shipping along the NWP found limited present-day effects on aerosols and ozone but potentially large effects based on a business as usual scenario for 2030 with up to 5% increases in ozone, and 5 to 20% in PM_{2.5} along shipping corridors (Gong et al., 2018). Browse et al. (2013) investigated the deposition of BC from future Arctic shipping in 2050 based on the scenarios from Corbett et al. (2010). They estimate that shipping north of 60°N will not be an important source of deposited BC.

However, prediction of future Arctic shipping impacts is sensitive not only to emission scenarios but also to the way in which the diversion of ship traffic is considered as a function of changed sea ice conditions. In addition, changes in ozone concentrations depend strongly on dispersion of precursor emissions due to non-linearity in the relationship between ozone production rates and NO_x concentration (Marelle et al., 2016; Ryerson et al., 2001).

2.4 Boreal and agricultural fires

Boreal wild fires and agricultural fires (crop burning) are another source of air pollutants in the Arctic (Warneke et al., 2009). Information on agricultural fires north of 60° N, a minor emission source, is extremely scarce. While fires lead to increases in PM concentrations in the Arctic (Warneke et al., 2009; Yttri et al., 2014), such events tend to result from fires occurring at lower latitudes in Eurasia, particularly in Eastern Europe and Siberia, with subsequent transport of pollutants northwards (Paris et al., 2009; Stohl et al., 2007). Regarding agricultural fires, Russia is the largest contributor with about 35% of global agricultural fires. Fire activities show two peaks: First in March and April in Eastern Europe and European Russia, and second in August in Central Asia and Asian Russia contributing most.

Emissions from boreal forest fires are the largest natural source of air pollutants in the Arctic. Emissions are highly seasonal, with large inter-annual and spatial variability (Giglio et al., 2013). Boreal fires have been shown to be the main driver of inter-annual variability in CO observed at several surface sites in the North American and European Arctic (Monks et al., 2012). They occur predominantly in spring and summer. Temperature, precipitation and humidity control variability in fire activity, with increased emissions occurring under drier conditions in the boreal regions in both Siberia (Macias-Fauria & Johnson, 2008) and North America (Bartsch et al., 2009). The strong climatic control on fire emissions means that boreal fire variability is linked to large-scale climatic modes, including the North Atlantic Oscillation, Arctic Oscillation, El Niño-Southern Oscillation, and Pacific Decadal Oscillation

(Balzter et al., 2007; Beverly et al., 2011; Monks et al., 2012). Modelling studies suggest that future increase in temperatures and dryer boreal forests will likely increase fire activity (Flannigan et al., 2009), with potential consequences for Arctic aerosol and trace gas burdens.

Further, it is important to understand how boreal fires burn. They tend to behave differently than mid-latitude fires. First, a major fraction (60-90%) of fuel consumption in boreal forest fires comes from carbon stored in boreal soils (Genet et al., 2018; Kasischke & Hoy, 2012; Kasischke et al., 2005; Mack et al., 2011; Randerson et al., 2006; Turetsky et al., 2010). Second, biomass burning in peatlands and the duff layer corresponds to smoldering rather than flaming (Akagi et al., 2011). Consequently, larger amounts of VOCs and organic aerosol particles are emitted from boreal forest fires compared to fires in other regions. Third, it has also been found that the BC content in peatland burning emissions in Alaska tends to be lower than that of peatland burning emission in the tropics (Chakrabarty et al., 2016), possibly caused by temperature and humidity differences (Turetsky et al., 2014).

The dominance of smoldering fires in the Arctic may pose a challenge for satellite detection of wildfires. For example, Waigl et al. (2017) find that MODIS failed to detect 45% of wildfires in Alaska in 2016, leading to a large bias in emission estimates for this region. In addition, fires burning low to the ground, as opposed to high crown fires, possibly emit into the boundary layer rather than further aloft which may lead to less dilution and hence higher pollutant concentrations at the surface. An investigation of the contribution of 12-year wildfire emissions on the BC mass deposited in the Arctic shows that fires in northern Eurasia contributed on average 85 % out of all biomass burning sources in the Northern Hemisphere (Evangelidou et al., 2016). The authors also estimate that Siberian fires contributed 46 % to the deposited BC from fires in northern Eurasia. Thomas et al. (2017) showed that deposition of BC to the Greenland ice sheet may be due to particularly large fire events in any one year. A multi-model study based on 8 global chemical transport models using FINN fire emissions for year 2008, suggested that 45-55% of all Arctic (> 60°N) CO is sourced from fire emissions during summer (Monks et al., 2015).

Future emissions of wild fires are highly uncertain (Knorr et al., 2016). Depending on the level of climate change that will modulate vegetation shifts (Kicklighter et al., 2014), some model results suggest that wildfire emissions will increase during the second half of the century. Regional forecasts identified north-eastern Europe as a potential area of increased fire activities (Knorr et al., 2016) which might affect air quality in the Arctic. While extensive studies on air pollutant emission factors from fires have been conducted in the Arctic (Kelly et al., 2013; Simpson et al., 2011; Vasileva et al., 2017), little is known about the extent to which fire emissions impair urban air quality. Wang and Hopke (2014) found that hourly PM_{2.5} mass concentrations that frequently exceed 60 µg m⁻³ can occur in Fairbanks, Alaska due to nearby forest fires leading to pollution exceeding EPA standards. While this is only one study reporting evidence of poor air quality caused by forest fires, it is highly likely that many more inhabited Arctic areas are affected. It is hence important to collect information across the Arctic to understand local impacts.

2.5 Natural emissions

While wildfires as described above can occur naturally and be triggered by human activity, terrestrial and marine trace gas and aerosol emissions occur in the Arctic without direct anthropogenic influence. Yet, these natural emissions interact with anthropogenic emissions to influence resulting air quality, both in terms of composition and pollutant levels (e.g. Hoyle et al., 2011).

Biogenic VOC (BVOC) emissions from vegetation are chiefly driven by temperature, radiation intensity, vegetation type, stress factors, and leaf area (Guenther et al., 2012). These emissions are important ozone precursors and can also lead to the production of SOA. However, BVOC emissions have been poorly characterized in the boreal regions and the Arctic. Measurements have generally focused on European boreal forests with a major emphasis on monoterpenes (Bäck et al., 2012; Jurán et al., 2017; Rantala et al., 2015; Rinne et al., 2000; Spirig et al., 2004; Zhou et al., 2017). Coniferous tree species characteristic of the boreal forests are efficient monoterpene emitters (Rinne et al., 2000), which leads to larger monoterpene/isoprene emission ratios in the boreal regions compared with forest ecosystems at lower latitudes (Guenther et al., 2012). However, some earlier measurements show abundant isoprene concentration over Alaskan boreal forests (Blake et al., 1992). Isoprene fluxes in tundra systems have also been measured in Greenland (Kramshoj et al., 2016; Lindwall et al., 2016; Schollert et al., 2014; Vedel-Petersen et al., 2015), northern Sweden (Faubert et al., 2010; Tang et al., 2016), and the Alaskan North Slope (Potosnak et al., 2013). All of these measurements show a very strong positive temperature dependence of isoprene fluxes, likely due to the higher emission potentials for isoprenoids in tundra vegetation than in temperate species (Rinnan et al., 2014). Schollert et al. (2014) also measured temperature-dependent emissions of monoterpenes and sesquiterpenes for four heath vegetation types in northeast Greenland, and also observed significant emissions of 'other reactive VOCs', including methylcyclohexane and 3-methylhexane. Thus, this high temperature sensitivity suggests an important role of climate warming for BVOC emissions that is compounded by increasing Arctic vegetation due to warming (Macias-Fauria et al., 2012).

Boreal biogenic emissions peak during the spring and summer growing season when they may make substantial perturbations to reactive atmospheric chemistry. Several studies have described the role of biogenic emissions, particularly monoterpenes, in driving increases in aerosol particle number and mass in the boreal regions (Lappalainen et al., 2016; Tunved et al., 2006), and others have quantified their role in climate feedbacks (Scott et al., 2018). Measurements conducted at the ZOTTO site in central Siberia have shown that secondary organic aerosols formed from BVOC emissions dominate aerosol composition during summer (Mikhailov et al., 2015). An understanding of the extent to which boreal BVOC emissions contribute to aerosol and tropospheric ozone loadings in Arctic settlements is not available. Bossioli et al. (2012) identified a role for interactions between BVOC sources and fire-emitted NO_x in controlling ozone production over Western Russia during spring 2006. Further, local NO_x emissions are expected to alter BVOC oxidation pathways, enhancing biogenic SOA formation, as described by Hoyle et al. (2011). The formation of sulfate from local SO_2 emissions also impacts biogenic SOA production by promoting acid-catalyzed

reactions, and formation of particle-phase organosulfates that have been observed in the Arctic (Hansen et al., 2014).

Mineral dust can be another natural source of PM. While long-range transported dust can dominate the mass burden in the atmospheric column, local dust sources dominate the surface dust concentration (~85 %) and dust deposition (~90 %) in the Arctic (Groot Zwaafink et al., 2016). Local mineral dust is mainly mobilized from erodible soils and becomes hence more important in the warm season with reduced snow cover. This also implies that higher contributions of mineral dust to PM can be expected in a warming climate. In addition, with increasing development in the Arctic region, mineral dust lofted from vehicle use on dirt roads is expected, but the contribution of locally-emitted dust to air quality in Arctic communities is not known. Further, the extent of atmospheric aging of local and transported mineral dust by anthropogenic trace gases (e.g. HNO₃) and impact on trace gas composition (as a NO_x sink) is an open question.

With rapidly declining sea ice extent, marine trace gas and aerosol emissions are expected to be increasing. Perhaps the most commonly observed interaction between natural and anthropogenic emissions in the Arctic is the formation of aged sea spray aerosol. During atmospheric transport, sea spray aerosol readily reacts with acidic anthropogenic gases (e.g. HNO₃, H₂SO₄), producing aged sea spray aerosol (Chi et al., 2015; Geng et al., 2010; Gunch et al., 2017; Kirpes et al., 2018). During both winter and summer, Kirpes et al. (2018) and Gunch et al. (2017), respectively, observed increased aging of sea spray aerosol at Utqiagvik, Alaska for air masses influenced by the Prudhoe Bay oil fields, in comparison to clean Arctic Ocean influence. Beyond sea spray aerosol, there is the potential for marine trace gas emissions, in the forms of dimethylsulfide and BVOCs, to react with anthropogenic pollutants, altering reaction pathways, trace gas composition, and aerosol formation. Yet, little is known about the interactions between marine emissions and anthropogenic pollutants in the Arctic.

3 Potential future Arctic emissions

While it is already difficult to quantify present Arctic emissions, there are significant challenges to produce realistic emission estimates for the future. This is true for both anthropogenic and natural emissions. Focusing on the former, there is a lack of socio-economic development scenarios specific to the Arctic which makes estimating future activity data difficult. Controlling factors are industrial development in terms of e.g. oil and gas exploration, mining activities, shipping diversion, tourism, agriculture and lifestyle change, and urbanization (AMAP, 2017a; Peters et al., 2011). For Arctic shipping, previously mentioned emission inventories (Corbett et al., 2010; Peters et al., 2011; Winther et al., 2017; Winther et al., 2014) provide future scenarios. However, scenarios in Winther et al. (2014) are based on Corbett et al. (2010) only assuming different ship type activity data. The new Winther et al. (2017) predictions update the ship activity data and consider regions of emission control. Furthermore not all studies consider a variety of Arctic shipping routes but limit the scenarios to the Europe-Asia passage. Peters et al. (2011) also provide estimates for future marine oil and gas exploration and associated shipping, considering both current and best practice emission factors for future estimates. Future scenarios for both types of

activities, however, depend strongly on sea ice evolution, and prediction skills of sea ice models are still low (AMAP, 2011, 2017b). Also, prognoses of future oil and gas activities diverge widely. Land-based emissions of flaring related to oil/gas extraction were not predicted to greatly increase in the ECLIPSE emission dataset (Stohl et al., 2015). In 2016, large-scale project plans were stopped and concessions sold due to low oil and gas prices (Hug, 2016). Recently, the US government initiated a process for drilling activities in the Arctic National Wildlife Refuge in Alaska (NYT, 2017), and despite a joint US and Canada ban on offshore drilling in 2016, offshore drilling in the Alaskan Arctic resumed in 2017. Without relevant international policies being in place, constraining the scenarios is difficult.

For anthropogenic BC emissions in particular, however, the Arctic Council has recently established policy guidance. At the 10th Arctic Council Ministerial Meeting in 2017, the member states set a target in the Fairbanks Declaration (AC, 2017) to reduce their BC emissions by 25 – 33 % below the 2013 levels by 2025. Among the measures recommended by the Arctic Councils Expert Group on Black Carbon and Methane are emission reductions from diesel use, e.g. by using up-to-date vehicle exhaust emission standards and ensuring the availability of clean fuels as well as voluntary emission control in the shipping industry (EGBCM, 2017). Regarding residential emissions, old biomass burning appliances should be replaced with cleaner and more efficient devices. Also, home heating efficiency and/or insulation should be enhanced for reduced fuel use. Moreover, the Arctic States with relevant oil and gas production (Canada, Norway, Russia, United States) have endorsed the World Bank's 'Zero-Routine Flaring by 2030' initiative. While this progress is promising for the reduction of BC emissions, from an air quality perspective it is insufficient. The supported mitigation measures are targeted towards the reduction of BC only within the context of achieving co-benefits for climate change (BC warms the atmosphere) and air quality. However, emissions sources such as shipping, power plants, and metal smelters that emit mainly non-absorbing particulate matter or precursors forming e.g. particulate sulfate and nitrate and organic aerosol particles, or reactive trace gases (e.g. nitrogen oxides, sulfur oxides) are not targeted, despite the fact that they deteriorate air quality.

While pure anthropogenic emissions can be controlled through policy measures, there is only limited control over 'semi-natural' emissions. Forest and agricultural fires are partly initiated by humans and future changes in Eurasian agricultural practices may be important considerations in the development of Arctic pollution scenarios over the coming decades. The Climate and Clean Air Coalition, for example, makes efforts to reduce emissions from agricultural open burning practices. However, with regard to purely natural emissions, i.e. naturally occurring wildfires, soil, and vegetation emissions, policy has no leverage. Nevertheless, potentially large changes must be expected in this respect. Forest fires in boreal North America have already been identified to have exceeded the envelope of the last 10,000 years emission history (Kelly et al., 2013), and generally fire emissions are estimated to increase in the Arctic due to lengthening of the boreal fire season (Flannigan et al., 2009). Significant increases in methane and BVOCs from terrestrial and marine Arctic ecosystems are expected as the currently frozen reservoirs will likely thaw with global warming (AMAP, 2015c; Commane et al., 2017; Anthony et al., 2016) and increased temperatures spur vegetation emissions (Holst et al., 2010; Lindwall et al., 2015).

3.1 Future anthropogenic emission scenarios

Despite the challenges for predicting Arctic air pollutant emissions, a limited number of emission inventories exist that provide future emission scenarios for the region. While these have not been developed specifically for the Arctic, they include emissions in the region north of 60°N, which we consider here to represent the Arctic region. We compare emissions from the ECLIPSE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants, Klimont et al., 2017, Stohl et al., 2015) and CMIP5 RCP (Representative Concentration Pathways, van Vuuren et al., 2011) emissions datasets for this region, going forward to the year 2050. These are two key inventories used widely in studies of future atmospheric composition and climate change (AMAP, 2015; IPCC, 2013). Key aspects of the emission datasets are summarized in Table 1, and Figure 3 shows the projected emissions of SO₂, NO_x, and BC from 2010 to 2050 north of 60 °N under different scenarios from each of these datasets.

The ECLIPSE and RCP emissions datasets make different assumptions regarding the future evolution of air pollutant emissions. The RCPs project greenhouse gas and air pollution emissions forward to the year 2100, based on 4 different scenarios of varying assumptions regarding the trajectories of population growth, economic and technological development, and air quality and climate policies. The projections are focused on achieving different amounts of net anthropogenic radiative forcing in the year 2100. These range from the RCP2.6 mitigation scenario of lowest forcing, to a high baseline emissions high forcing RCP8.5 scenario, with two intermediate stabilization scenarios (RCP4.5 and RCP6.0). The focus of the RCP scenarios is on development of net forcing scenarios driven mainly by changes in emissions of long-lived greenhouse gases. All four RCP scenarios assume development of increasingly stringent legislative and technological control on air pollution emissions linked to economic development, with similar assumptions among the scenarios, leading to overall reductions in air pollution emissions going forward in each scenario. ECLIPSE emissions data were produced using the GAINS (Greenhouse gas–Air pollution Interactions and Synergies) model (Markus Amann et al., 2011). These emissions were designed using updates to population data and activity location data used in the RCP emissions, while also specifying monthly temporally distributed emissions for all sources (Klimont et al., 2017; Stohl et al., 2015). A key addition in the ECLIPSE dataset of particular relevance to the Arctic region is the inclusion of emissions from gas flaring associated with oil production. This source contributes approximately one third of total BC emissions north of 60 °N, and two thirds of those north of 66 °N, and is estimated to account for more than 40% of all Arctic BC near the surface (Stohl et al. 2013). Future projections within the ECLIPSE datasets include a Current Legislation (CLE) scenario (which includes current and planned environmental laws and policies), a SLCP mitigation scenario (including measures of co-benefit to air quality and climate), a No Further Control (NFC) scenario (no further legislation introduced after 2015), a Maximum Technologically Feasible Reduction scenario (MTFR), as well as a baseline control scenario.

Present day (2010) Arctic emission totals, and the trajectories of future emission changes out to 2050 between the RCP and ECLIPSE scenarios are similar for NO_x, but are substantially different for SO₂ and BC (Figure 3). Flaring emissions contribute to the substantially larger BC emissions in ECLIPSE (see above). Energy sector sources contribute

less than 1% of BC 2010 Arctic emissions in each of the RCP scenarios, with the largest sources being transport, industry, and domestic fuel combustion. Despite similar global SO₂ emission estimates from ECLIPSE and the RCP scenarios for the year 2010, ECLIPSE emissions are almost a factor of 3 in the Arctic region (Figure 3c). Energy sector sources dominate SO₂ emissions in the RCP 2010 emissions, accounting for between 63 and 80% of Arctic emissions across the different RCP scenarios. Energy sector SO₂ emissions from ECLIPSE are of the same magnitude, but account for only 19% of total emissions. Industry sector SO₂ emissions are a factor of 10 larger in ECLIPSE, accounting for around 80% of the 2010 total, and producing the large offset between the two datasets in the Arctic.

Estimates of future Arctic anthropogenic emissions vary considerably with projected policy and development scenarios, and between the two datasets. The spread among the different ECLIPSE scenarios is larger than between the different RCP scenarios, particularly for BC and SO₂ (Fig 3b,c). This is a consequence of air pollutant emissions following similar pathways between the different RCP scenarios, with an assumption that they would decline to 2050 in all scenarios, linked to economic growth (Amann et al., 2013). Uncertainties regarding the development and degree of adoption of air quality control policies are also specifically accounted for in the ECLIPSE projections. This leads to a large degree of spread in potential future emissions, even though each ECLIPSE scenario follows a single pathway of future energy demand (Stohl et al., 2015). In addition to the spread in pollutant concentrations, the spatial pattern of emissions also changes among the scenarios. A more detailed discussion can be found in the supplementary material.

The spread in future Arctic emission projections can be interpreted to reflect uncertainty in our current knowledge of Arctic emissions sectors and activities (e.g. gas flaring), as well as the future development of emission control policies. The possibility of specific policy or legislation being introduced to protect the unique Arctic environment needs to be considered alongside general or lower latitude policy drivers when considering future Arctic emission changes. It would be misleading to interpret narrower spread among the RCP scenarios as an indication of greater confidence in future Arctic emission trajectories, since assumptions regarding development of pollution control policy and technologies is rather simplistic (see above). Policies aimed at mitigating climate warming, in addition to reducing air pollution, can result in large reductions in Arctic emissions of BC and NO_x, demonstrated by the ECLIPSE SLCP scenario. Emission mitigation measures under this scenario are targeted to mitigating air quality impacts in addition to their 20-year time horizon climate impact (Stohl et al., 2015). Due to their warming impacts (NO_x being a precursor for tropospheric ozone), large reductions in Arctic BC and NO_x emissions are achieved in this scenario, with little noticeable impact on SO₂ emissions. The ECLIPSE MTRF scenario gives a lower limit to emissions across the three species, based on implementation of theoretically achievable control technologies. However, these measures do not necessarily map to realistic policy options.

4 Implications of Arctic specific meteorological conditions for air pollution

There is not only a shortfall in quantifying Arctic air pollutant emissions, as discussed in the previous sections, but also little has been done to investigate the interplay between specific

Arctic meteorological conditions and air pollution. At cold temperatures inversion layers trap pollutants, low temperatures also likely increase condensation of semi-volatile gases to the particle phase, fog and cloud formation influence aqueous-phase reactions, and reduced solar insolation in wintertime limits photochemical reactions. From mid-latitude research, it is known that temperature inversion layers trap air pollution and contribute significantly to increased pollutant concentrations (e.g. Sandradewi et al., 2008). At high latitudes, inversion layers play an even more crucial role due to their frequent occurrence, not only in winter, whereby they exacerbate local air pollution (Tran & Mölders, 2011). For the Fairbanks area, Alaska, a study based on 10-year meteorological and vertical sounding data found that in 64 % of the cases surface-based inversions occurred (Mayfield & Fochesatto, 2013). Those were accompanied by additional elevated inversion layers > 80 % of the time. Generally, inversion layer formation can be controlled either locally by surface-based radiative cooling or synoptically, the most prominent synoptic influence being anticyclonic conditions followed by warm air advection (Mayfield & Fochesatto, 2013). An earlier study (Serreze et al., 1992) used data from up to twelve years of data from 31 stations across the Eurasian Arctic and six drifting stations near the North Pole. They observed that the frequency of low level inversion layers increases towards the east from Norway to eastern Siberia. Occurrences east of Novaya Zemlya in winter are > 95 % of the time. In summer, inversion occurrence is much lower, however, still > 50 % in all locations. Eastern Siberia is especially prone to surface based inversions due to its topography, similar to some regions in Alaska, with even > 85 % inversion occurrence in summer. Note that inversion layers are significantly lower in altitude in winter than in summer. With regards to the interplay of inversion layers and air pollution, Asmi et al. (2016) found strongly elevated particle concentrations at Tiksi, East Siberia, on very cold winter days with little air movement. The relatively small diameter (50 nm) of the dominant particle mode and high black carbon (BC) concentrations pointed towards local combustion sources, which were, however, not specified.

An additional aspect of near-ground atmospheric inversion structures is that they favor the formation of radiation fog and/or ice fog (Kumai, 1973; Przybylak, 2003; Serreze & Barry, 2005). Very early Arctic work already established that anthropogenic emissions of water vapor can lead to (ice) fog formation (Benson, 1969; Robinson et al., 1957). A more recent study confirmed that 10 to 100 m thick near-surface ice fog forms from water vapor emitted at very low temperatures (Kim et al., 2014). In addition to being harmful to health, as found for Siberian fog (Romanov, 2001), it also reduces visibility and can be a hazard for aviation (Gultepe et al., 2014, 2017; Willet, 1929). Scientific observations of fog formation in the Arctic are limited and show that conditions that favor its formation can vary widely by region. For example, Rae (1954) found that fog formation on Canadian islands increases with temperatures < -34 °C. Siberian winter fog was found to occur between -45°C and -5°C (Lydolph, 1977). During the International Arctic Ocean Expedition winter fogs were found to have a maximum probability at temperatures between -10°C and -5°C (Ye 2009). These studies highlight that the probability of fog formation can vary considerably by location, especially between Arctic continental and coastal regions. Near the coast, fog can occur due to the advection of cold air. Regardless of the formation mechanism, temperature is an important determining factor. Ye (2009) shows for the Siberian Arctic that at some stations fog can be present as many as 60 days per winter at temperatures < -30 °C, increasing with

decreasing temperature. Conversely, at other locations fog days increase with increasing temperature, reaching e.g. 50 days at -5°C .

How fog and low-level clouds interact with air pollution in the Arctic has not been a focal area of scientific research. Work from Bowling (1986) suggested that low level ice fogs in Alaska may deepen the mixing layer leading to the potential entrainment of elevated pollution plumes. Such pollution plumes can originate from high stacks. At the same time, mixing can also induce dilution of air pollution. Generally, fog formation will lead to scavenging of the activated aerosol particles and will thus have a cleansing effect, but only if it precipitates. Without precipitation the evaporated fog will leave aerosol particles behind. In fact, recent investigations of the toxicology of PM - in form of reactive oxygen species (ROS) present in the aerosol particles - have shown that ROS can be produced in fog droplets. Hence, the fog-processed PM can be more harmful to health (Decesari et al., 2017).

These previous studies imply that for understanding Arctic air pollution and its impacts, an accurate representation of (sometimes multiple) inversion layers and cloud/fog formation in models is essential in order to predict realistic ambient concentrations, and their transport and transformation in the atmosphere, as well as their wet and dry deposition. Accurately simulating observed inversion layers is challenging for the current generation meteorology models (Mölders et al., 2012), meaning that the potential exceedance of regulatory air quality limits might not be predicted correctly. In addition, simulating the chemical evolution of emissions in high-latitude locations requires taking into account the local temperature, humidity, and solar radiation. For example, under winter inversion conditions processing of secondary aerosol species such as sulfate and organic matter can differ from mid-latitude conditions (see section 5). The ability of our current state-of-the-art chemical mechanisms to simulate observed concentrations of criteria air pollutants under high emission scenarios at very cold temperatures has so far not been rigorously tested. The following sections 5 and 6 review a case study of urban Arctic air pollution in Alaska and open questions regarding chemical processing under these specific conditions.

5 An urban example of Arctic air pollution: Fairbanks, Alaska

Here, we illustrate the magnitude that urban Arctic air pollution can reach using the example of Fairbanks, Alaska, USA. The city of Fairbanks exemplifies many of the problems of pollution in the Arctic and sub-Arctic regions. Urban pollution in Fairbanks was first examined by Benson (1969) who identified meteorological inversions as an important factor affecting pollution in Fairbanks, leading to ice fog. Carbon monoxide pollution was identified as a problem in Fairbanks during the early 1970s with CO concentrations commonly exceeding the US Environmental Protection Agency (EPA) standards (National Research Council, 2002). Automobile CO emissions, often exacerbated by cold start emissions, were identified as the primary source. Improving automotive technology began dramatically reducing wintertime CO pollution events during the 1990s, and Fairbanks was removed from the non-attainment list in the 2000s. This early success story clearly shows how technological solutions can be effective at addressing pollution problems, even in the face of extreme meteorological trapping conditions.

However, Fairbanks has had challenges with fine particulate pollution standards since the EPA established the standards in 1997. Figure 4 shows the annual cycle of exceedances of the 24-hour $PM_{2.5}$ standard ($35 \mu\text{g m}^{-3}$) with a wintertime “violation season” and a summertime peak (June, July, August) from forest fires (see section 2.4). Because forest fires are not generally preventable in Alaska, EPA often waives exceedances that are from forest fires during summertime, but counts wintertime exceedances, which are considered preventable through regulation. These wintertime events caused Fairbanks to become a non-attainment area in 2009, and lack of progress on this problem has led Fairbanks to be re-classified as a “serious” non-attainment area in 2017. Note that $PM_{2.5}$ mass concentration measurements are made every third day, so the peak of approximately six measured exceedances per month actually represents two measurements per month in both December and January.

Given the poor air quality in Fairbanks, a number of studies were carried out to understand causes for this problem and the sources of the particulate matter. Tran and Mölders (2011) found that strong temperature inversions and low wind speeds were the most common conditions during $PM_{2.5}$ violations. An important aspect of the finding that poor vertical mixing was associated with air quality violations is that emissions from point sources (e.g. power plants) that have high smokestacks often inject pollution above the surface mixed layer and thus have a reduced effect on breathing-level air quality. This effect also complicates use of emission inventories to apportion sources of air quality degradation at breathing level because some pollution from major emitters such as power plants may remain aloft and decoupled from the surface layer sufficiently long to transport out of the air shed. Therefore, receptor-based analyses have to be carried out to determine which sources actually impact urban residents.

In Fairbanks, the chemical composition of breathing-level $PM_{2.5}$ has been quantified for source apportionment. Figure 5 shows the fractional composition, by mass, of wintertime $PM_{2.5}$ in Fairbanks and the nearby residential community of North Pole, Alaska. These measurements show that organic carbon species, either from direct emission (primary) or through organic aerosol formation in the atmosphere (secondary), make up the greatest fraction of the $PM_{2.5}$ mass ($> 60\%$) with smaller amounts of inorganic ions and elemental carbon (EC). Ward et al. (2012) applied chemical mass balance (CMB) modelling to apportion these receptor-based observations and found wood smoke was the dominant component and that the fraction of wood smoke varies between measurement sites in the region. Wang and Hopke (2014) used positive matrix factorization (PMF) modelling to analyze the same data and found again that wood smoke was the leading single factor. Kotchenruther (2016) recently used PMF modelling for source apportionment of wood smoke at multiple cities in the US Pacific Northwest, including Fairbanks. They found that wood smoke makes up 50% of the considered particulate mass. They also showed that smaller, more residential cities with little other industry producing $PM_{2.5}$ had increasing fractions of their $PM_{2.5}$ problem arising from wood smoke. This urban/residential difference is also observed between Fairbanks, which shows more inorganic pollution, and North Pole Alaska, which shows a greater fraction of organic pollution largely produced by wood combustion.

Tran and Mölders (2012) modelled the effect of replacing woodstoves with newer, lower emitting stoves and found a small air quality benefit.

This example demonstrates that Arctic air pollution can be so severe that the city of Fairbanks has been labelled a “serious” non-attainment area by the US EPA. While it is unclear whether anthropogenic emissions in Fairbanks are actually higher compared to other US non-attainment cities, it is evident that the specific Arctic meteorological conditions exacerbate the air pollution problem, because the pollutants are trapped under the shallow inversion layers. What is not clear, however, is how significant a role atmospheric chemical reactions specific to the dark and cold season play a role in this wintertime air pollution formation. Potential mechanisms and knowledge gaps are discussed in the following section.

6 Air pollution chemistry under Arctic conditions

The studies of pollution in northern cities, discussed earlier, show that wintertime pollution is typically caused by local emissions coupled with poor dispersion conditions, while summertime pollution is caused by large episodic forest fire events. A critical question for all pollution studies is how much of the pollution is directly emitted (primary) versus produced through chemical transformations in the atmosphere (secondary). This question is critical because pollution control strategies typically differ between primary and secondary pollution. Secondary pollutants can have complex chemical interactions leading some well-intentioned control strategies to fail or even be counterproductive.

Studies at lower latitudes show that SOA is often a major component of fine particulate pollution (Jimenez et al., 2009). However, typical SOA formation chemistry is thought to be photochemical. This suggests that SOA should be a small fraction of organic aerosol particulate matter under wintertime Arctic pollution conditions with the main organic aerosol contribution being primary. On the other hand, low temperatures favor partitioning of semi-volatile species to partition from the gas phase to the particle phase, adding to $PM_{2.5}$ mass. For SOA formed from the ozonolysis of α -pinene, Kristensen et al. (2017) observed suppressed formation of dimer esters, relative to carboxylic acids, at 258 K, compared to 293 K, suggesting reduced oligomerization at lower temperatures. However, at an even lower temperature of 223 K, Huang et al. (2018) observed increased mass fractions of oligomers in the SOA formed from α -pinene ozonolysis, which was attributed to increased hydrogen-bonding in the higher viscosity SOA (compared to SOA formed at 296 K). The studies of Fairbanks pollution, for example, have only used standard OC/EC analysis (Chow et al., 2004), which has little information about the actual chemical structures of the organic carbon species. The recent work of Kotchenruther (2016) used fractions of OC derived from the thermal evolution of OC during analysis to increase chemical speciation within OC. The PMF analysis showed two wood smoke profiles, whereby one was identified with “aged” wood smoke or SOA, and the other with primary wood smoke. During wintertime, this SOA fraction was relatively small (~20 % of OA), but in summertime, most wildfire OA was identified as SOA. However, this analysis still lacks detail on the chemical nature of the OA. Therefore, studies of molecular composition, and the relationship to particle phase, are needed for wintertime conditions.

The measurements in Fairbanks show that inorganic components make up a significant fraction of $PM_{2.5}$ mass, with sulfate being the largest component followed by nitrate and ammonium. However, sulfate formation mechanisms in polluted Arctic cities in winter remain to be elucidated. Traditional pathways for sulfate formation include SO_2 -oxidation by the hydroxyl radical (OH) in the gas-phase, and hydrogen peroxide (H_2O_2) and O_3 in the aqueous phase. These standard pathways appear to be insufficient to explain the formation of wintertime sulfate aerosol particles in China, for example, under meteorological conditions comparable to Fairbanks (Wang et al., 2014). In China, observations show that particulate sulfate mass increases as a function of relative humidity (Elser et al., 2016). A few hypotheses have been developed to explain the high level of wintertime sulfate observed in China. One is the oxidation of HSO_3^- or SO_3^{2-} by NO_2 in aqueous aerosol particles, initiated by the formation of the $NO_2-SO_3^{2-}$ complex (Clifton et al., 1988). This pathway, however, requires sufficient neutralization in aerosol particles to be effective, as the solubility of SO_2 increases rapidly with pH in aerosol water. Two studies find from their calculations that pH in fine particles of Chinese haze can be as high as 6-7, and thus they concluded that aqueous oxidation by NO_2 is responsible for the high sulfate formation (Cheng et al., 2016; Wang et al., 2016). However, more recent studies suggest that wintertime fine aerosol particles in urban China have a pH range of 3.0–5.0, despite the very high levels of gaseous NH_3 (Guo et al., 2017; Liu et al., 2017). If pH of ambient aerosol particles is indeed in the range of 3.0-5.0, the pathway of aqueous oxidation of SO_2 by NO_2 is unlikely to be effective (Guo et al., 2017). Another hypothesis is the aqueous oxidation of SO_2 by O_2 catalyzed by transition metals, as they are ubiquitous in combustion and crustal aerosol particles (Jacob, 2000). Several studies have suggested that transition metal ions can catalyze sulfate formation in cloud droplets (Alexander et al., 2009; Harris et al., 2013; Huss et al., 1982), although the importance of these reactions in aerosol particles remains unknown. The catalytic mechanism by iron ions has been described by Deguillaume et al. (2005). It has been suggested that the catalytic mechanism is particularly important when photochemical activity is low (Pandis et al., 1992; Warneck, 1991), favoring wintertime sulfate formation in cities. Transition metals identified to catalyze SO_2 -oxidation include iron (Warneck, 1999), manganese (Berglund & Elding, 1995), and copper (Conklin & Hoffmann, 1988a, 1988b). At Alert, Canada, McCabe et al. (2006) used oxygen isotopes of non-sea salt sulfate to show a 10-18% contribution from a non-photochemical sulfur oxidation pathway, likely Fe^{3+}/Mn^{2+} -catalyzed O_2 oxidation, during the Arctic winter. However, the number fraction of Arctic aerosols containing these transition metals is not known to further evaluate this oxidation pathway.

Heterogeneous nocturnal chemistry involving dinitrogen pentoxide (N_2O_5) is known to produce particulate nitrate (Brown & Stutz, 2012), but relies on the presence of ozone to form nitrate. Poor dispersion conditions, such as those observed in Fairbanks, coupled with emissions of automotive-sourced NO lead to near complete removal of O_3 and thus little N_2O_5 is present for much of the winter (Ayers & Simpson, 2006). As urban-polluted air transports away from the city and mixes with ozone-rich air, nocturnal nitrogen oxide chemistry becomes active (Apodaca et al., 2008; Huff et al., 2011) leading to deposition of nitric acid and particulate nitrate. Joyce et al. (2014) considered this chemical-meteorological coupling for Fairbanks wintertime conditions through use of a 1-D chemical transport model and found that N_2O_5 chemistry was suppressed in the urban airshed, which was consistent with the small

mass fraction of nitrate in urban Fairbanks particulate matter, but occurred aloft and downwind and was a strong function of available ammonia, affecting the regional fate of the pollution. In regions where more vertical mixing occurs, such as in the mid-latitude city of Salt Lake City, Utah, this mixing may bring ammonium nitrate formed aloft back down to impact breathing-level air quality, which is consistent with the large fraction of ammonium nitrate in Utah wintertime particulate matter. Sources of ammonia in urban areas are not well understood in the Arctic, but $\text{PM}_{2.5}$ in Fairbanks is typically observed to have about two moles of ammonium per mole of sulfate, indicating sufficient ammonia is available to neutralize the particles. N_2O_5 can also react with aerosol chloride to form nitryl chloride, ClNO_2 , a photolabile source of reactive chlorine (Osthoff et al., 2008). This chlorine-activation chemistry can happen quite far inland, as demonstrated by (Thornton et al. (2010) and presumably happens at Arctic coastal and even inland locations. Heterogeneous reactions can also occur on ice particles and at the snow surface, leading to deposition of N_2O_5 (Apodaca et al., 2008; Huff et al., 2011). While the chemical reactions underlying nocturnal nitrogen chemistry appear to be relatively well understood, the way in which vertical mixing brings ozone and NO_x together exerts strong controls on the rate of nitrogen oxidation and formation of nitric acid.

With regards to secondary gaseous air pollutants, ozone could potentially become a problem in the Arctic in the future with the development of intensive oil and gas extraction facilities. At southerly latitudes, recent observations from regions of oil and natural gas extraction in northern Utah showed very large episodic wintertime enhancements in surface ozone. These events, driven mainly by photochemical processes (available sunlight), are also a result of reduced dry deposition and high albedo due to snow cover, coupled with shallow boundary layer heights and calm conditions (Ahmadov et al., 2015; Edwards et al., 2014; Schnell et al., 2016; Wild et al., 2016). Emissions of VOCs from extraction activities have been shown to be key drivers of ozone production during such events, with large contributions from aromatic VOCs (Ahmadov et al., 2015). Photochemical production of ozone during these wintertime events occurs at larger VOC concentrations and lower NO_x concentrations than found during common summertime pollution episodes, with photolysis of carbonyl compounds being the main oxidant source (Edwards et al., 2014). However, estimating how future Arctic ozone concentrations may respond to new precursor sources is partly hindered by larger uncertainties in knowledge of gas-phase chemical reaction kinetics at low temperatures (compared with those at the laboratory standard temperature of 298 K). This produces a substantial spread in predicted oxidant (ozone) concentrations in not only anthropogenic but also biomass burning pollution plumes (Ridley et al., 2017) and is due to particularly large uncertainties in a small number of key reactions (most notably the $\text{NO} + \text{O}_3$ reaction) at low temperatures.

The above discussion highlights that the chemical processing of air pollutants in cold and dark conditions, in particular, are not yet well understood. Being able to model these reactions to predict air pollution episodes is crucial for the effective design of air pollution mitigation measures to reduce health impacts.

7 Arctic specific health impacts from air pollution

While many epidemiological studies examining associations between air pollutants and health outcomes have been conducted around the world, primarily in the United States, Europe, and now Asia, few short-term and no long-term studies have been conducted among Arctic communities. Hence our current understanding of relationships between air pollution exposure and health outcomes among Arctic communities is limited. Previous work on environmental health in the Arctic focused mainly on persistent organic pollutants (POPs) and metals, and has not addressed criteria air pollutants to the same extent (AMAP, 2015b). The Arctic Monitoring and Assessment Programme report on 'Acidifying Pollutants, Arctic Haze, and Acidification in the Arctic' (AMAP, 2006), which summarized air pollution health studies in the Arctic, concludes, debatably, that no evidence was found of health implications from emissions of the metallurgical plants on the Kola Peninsula or Norilsk, which are among the areas of highest air pollution in the Arctic. Since then no outdoor air pollution health impact studies in the Arctic have been published. A few studies on the health impacts of indoor air pollution are available, though most either have poorly defined exposure variables (e.g. the general use of wood or coal stove for heating versus personal exposure measurements) or a small study population (Bulkow et al., 2012; Guggisberg et al., 2003; Nieminen et al., 2013; Singleton et al., 2018; Ware et al., 2014). Therefore, at the moment effects of neither indoor nor outdoor air pollution are well quantified.

To overcome this limitation in availability of epidemiological concentration-response relationships in other parts of the world, the Global Burden of Disease study developed Integrated Exposure Response (IER) curves, which draw information from epidemiological studies globally on exposure to ambient air pollution, household air pollution, and smoking (Burnett et al., 2014; Cohen et al., 2017). These curves provide a continuous function relating PM_{2.5} exposure levels with stroke, ischemic heart disease, chronic obstructive pulmonary disease, lung cancer, and lower respiratory infections. They are frequently used to extrapolate concentration-response relationships from places where epidemiology studies have been conducted to populations where no epidemiological studies have been carried out. Applying these IERs to the global population, the Global Burden of Disease study found that ambient air pollution and household air pollution are associated with 4.2 million and 2.9 million premature deaths in 2015, respectively (Forouzanfar et al., 2016). In the western Arctic countries, premature mortality increased by a factor of 6 from 1990 to 2015, mostly due to population aging; while in Russia and Finland premature deaths decreased by up to 26.6 % (Cohen et al., 2017).

In the absence of Arctic-specific air pollution epidemiology, it would be a reasonable extrapolation to apply the IERs to assess health impacts of air pollution in Arctic populations. Indeed, these curves have been used to assess air pollution health impacts on a global scale covering all populations (Anenberg et al., 2017; Apte et al., 2015; Cohen et al., 2017), and in individual countries where no air pollution epidemiology has been carried out (e.g., Anenberg et al., 2017; Pillarisetti et al., 2016). Using similar extrapolations of epidemiologically-derived concentration-response functions, several studies have estimated the health impacts of emissions from particular sources among Arctic nations, including shipping (Jonson et al., 2015), solid fuel heating (World Bank & ICCI, 2013.; Chafe et al., 2015; Sigsgaard et al.,

2015), and transportation (Anenberg et al., 2017; Crippa et al., 2016). The World Bank's and International Climate and Cryosphere Initiative's "On Thin Ice" report estimated that expansion of short-lived climate-forcing pollutants mitigation measures could avoid 48,000 (i.e. > 1%) premature deaths in Arctic nations in 2030. The greatest health benefits from black carbon mitigation measures were replacing biomass and coal heating stoves, and reducing Eurasian open burning by 90% (see discussion on Fairbanks stoves in section 5 and agricultural burning in section 2.4).

Along with epidemiological concentration-response relationships, air pollution health impact assessments require information about the size, age, and health status of the exposed population. Disease incidence and prevalence rates have now been estimated for every country (GBD 2015 Disease and Injury Incidence and Prevalence Collaborators, Vos et al., 2016). However, disease incidence rates can vary dramatically within an individual country, and such subnational variability can influence results of air pollution health impact assessment (Chowdhury & Dey, 2016). In the case of Alaska, Alaska Natives have significantly higher rates of cancer, heart disease, and chronic obstructive pulmonary disease, among other health outcomes (AMAP Consortium, 2016). Such population differences should be taken into account when estimating the health impacts of air pollution.

This brief summary of the state of knowledge regarding Arctic air pollution health impacts demonstrates that information gaps are large. Given that higher emissions and population growth are expected for the future, air pollution health effects need to be investigated more thoroughly. While health impact estimates can be conducted based on existing IERs combined with Arctic-specific disease incident and other factors, several important questions remain including whether Arctic particulate matter might have toxicological properties different from other regions, and whether populations in Arctic communities respond differently to air pollution exposure levels given differences in health care systems and lifestyles. These questions are more difficult to address but deserve attention.

8 Ecosystem impacts

Beyond health impacts, air pollution can also have adverse effects on ecosystems. Emissions of SO_x and NO_x can lead to freshwater and ocean acidification. They can also cause acidification and eutrophication via the production of nitrate and sulfate aerosol particles, which are wet or dry deposited onto the surface (AMAP, 2006). A special region of concern is, for example, the Kola Peninsula and northern Finland, where industrial emissions are transported from Russia towards Finland (Ruoho-Airola et al., 2015). This particular Arctic region, together with the area around Norilsk, the location of the nickel smelter facility which is the single largest contributor to sulfur emissions in the Arctic, have been known for their high emissions and nearby ecosystem damage due to acidification for decades (AMAP, 2006). In addition to industrial facilities, Arctic shipping emissions may also result in deposition of acidic nitrate and sulfate compounds in particular to marine ecosystems. Modest increases in PM_{2.5} are predicted in studies focusing on European ship emissions with 5-10% enhancements due to present-day shipping along the southern Norwegian coast (Aksoyoglu et al., 2016; Jonson et al., 2015). Dalsøren et al. (2007) examined shipping impacts for the year 2015 and

found that deposition on land also contributes to acidification and eutrophication. In summer, the main impacted regions are Norway, Iceland and Greenland and the western part of northern Russia. Hassellöv et al. (2013) find that ocean acidification from shipping is of the same order of acidification from CO₂ uptake. Even though the authors conducted a global study that barely considers shipping north of 66 °N, it is evident that shipping near the coast of Norway has important local effects.

As discussed earlier, future shipping may also lead to significant increases in ozone levels (Granier et al., 2006; Marelle et al., 2016) thus introducing a potential extra burden on the already stressed Arctic ecosystems. Ozone is harmful to vegetation because it impairs photosynthesis via uptake through plant stomata (Sitch et al., 2007). A recent study showed only negligible effects from ozone sourced from boreal wildfires on forest productivity in boreal North America (Yue et al., 2017). But a study examining potential ecosystem damage due to ozone at the country scale found that ozone formed through NO_x has the highest impact in the high northern latitudes (van Zelm et al., 2016). In addition to ozone, also aerosol particles can have an effect on ecosystems. Changes in diffuse/direct radiation fraction induced by changes in atmospheric aerosol loading affect the efficiency of plant productivity (Rap et al., 2015). Fire-produced aerosol particles were found to produce an increase in net primary productivity via diffuse radiation effects, although decreases in productivity were found in a future mid-century scenario due to drought induced by fire aerosol impacts on atmospheric stability and moisture (Yue et al., 2017).

No study to date has assessed the combined aerosol, ozone, and nitrogen / sulfur deposition effects from different sources on the productivity of high latitude terrestrial ecosystems. The combined effect of several pollutants might be particularly relevant near industrial sites and urban areas. Joyce et al. (2014) point out that NO_x emissions are processed to particulate nitrate during night downwind of emission locations. This implies that nitrate deposition would take place outside urban or industrial areas. Given the scarcity of studies, understanding of current and potential future impacts of air pollutants on Arctic ecosystems is very poor. It is worth to note that a number of studies have focused on the albedo reducing effects of BC deposited on snow (e.g., Flanner et al., 2007), however, it has not been considered as a source of carbon in melt water.

9 Summary and recommendations

Understanding current and preventing potential future local Arctic air pollution is key to making the Arctic a sustainable living space. The above discussions of the uncertainty of current and future emission sources, local air pollution challenges, and unidentified and poorly characterized chemical mechanisms highlight that we are still a long way from having a comprehensive knowledge of the overall impact of Arctic air pollution on human health and ecosystems. For the scientific community to be able to generate policy relevant knowledge in the near future to mitigate current problems and to prevent future Arctic air pollution impacts, we present recommendations in the following on where to place research efforts.

Current emissions. Current Arctic air pollutant emissions are not well quantified although discrepancies between observed emissions and inventories have led to some inventory updates and improvements. This is especially the case for the metallurgical

activities and BC emissions on the Kola Peninsula (Evans et al., 2015; Hienola et al., 2013; Prank et al., 2010) and gas flaring, even though the magnitude of the contribution of flaring is still unclear (Huang et al., 2015; Stohl et al., 2013; Winiger et al., 2017). However, challenges with emissions are not limited to improving estimates for Arctic activities already included in the inventories, but future research needs to also include missing sources. Among those missing are local and irregular anthropogenic emissions, e.g. from open waste burning due to the lack of waste management in difficult to access regions (Aliabadi et al., 2015). Such sources could be particularly important for human health effects. Furthermore, current inventories focus on outdoor emissions. However, indoor air pollution exposure, especially in winter, when people tend to spend a significant amount of time inside, is highly relevant for health. In many remote areas heating and cooking happens on stoves that do not have very efficient burning processes and thus produce a significant amount of emissions (EGBCM, 2017).

Future emissions. Except for a limited number of datasets on shipping and some fossil fuel extraction emissions (Corbett et al., 2010, Peters et al., 2011, Winther et al., 2014, Winther et al., 2014), no Arctic-specific future emission scenarios exist that encompass all of the significant emission sectors for local air pollution. Global scenarios that include the Arctic exist, but these do not necessarily focus on air pollutant emissions. As a consequence, for example, the RCP scenarios, designed to achieve different strengths of net anthropogenic radiative forcing in the year 2100, show only a small spread in Arctic emissions of BC, NO_x, and SO₂ among the four different forcing scenarios. Other scenarios, for example from the ECLIPSE project, have been designed specifically around future emissions of short-lived climate forcing pollutants and include monthly temporally distributed emissions for all sources and additional emission sources such as flaring in the Arctic. Here, the spread in future emissions as a function of the scenario is significant (Figure 3). This highlights that without any international policies in place, which would put boundaries on the amount of emissions to expect within the Arctic, the creation of future Arctic air pollution emission scenarios is extremely challenging. One example of such challenges is the dependency of emissions on difficult-to-predict political decisions specific to the Arctic. In 2016, the US and Canada banned offshore drilling in the Arctic. In 2017, offshore drilling in Alaska resumed. In order to improve predictions and reduce uncertainty, socio-economic development scenarios, specifically designed for the Arctic, are needed. Scenario development with a range of stakeholders (e.g. local residents, policy-makers, international corporations, insurance industry) is needed to describe possible Arctic futures and to develop policy measures on time.

Winter air pollution formation mechanisms. Very few studies have addressed the impact of the specific Arctic environmental conditions for the formation of air pollution, particularly under wintertime conditions (Tran & Mölders, 2011). In the summer, air pollution can be dominated by forest fires, and chemical mechanisms are better understood. For the wintertime, however, many open questions remain regarding the degree to which chemical transformations affect wintertime air quality, a time of year when Arctic Haze due to remote sources can also be important. The most urgent questions are related to the balance between

direct emissions of pollution and chemical transformations that may be occurring under cold and dark conditions. Among the most urgent science questions are:

- What are the key wintertime emission sources, and how do these emissions impact processing of pollution?
- What is the balance of primary versus secondary pollutants under cold and dark conditions?
- To what extent do temperature inversions both affect trapping of ground-level emissions and facilitate export of pollution from high stack emissions?
- What are the health and ecosystem impacts of pollution from Northern cities?

Answers to these questions will allow improved predictions of air pollution and apportionment of emission sources. This information is necessary to design mitigation measures.

Air pollution control strategies. A sound scientific basis for understanding the chemistry, sources, and meteorological controls on wintertime pollution should underlie new control strategies. High latitude cities with poor air quality have various strategies for improving air quality and health. Because heating and local industries are sources of pollutants, reducing pollution often causes economic challenges for citizens and small businesses. On the other hand, poor air quality affects health and quality of life leading to negative economic consequences. Therefore, understanding sources of pollution, dispersion, and chemical transformation mechanisms is critical to finding air quality solutions. Because air pollution episodes are associated with poor dispersion conditions, it can be possible to predict these meteorological conditions and limit emissions on these days. For example, woodstove burn bans, with exceptions for homes that only have wood heat, have been effective in avoiding peak pollution events in some cities (LRAPA, 2016). In cities where woodstoves are a common form of domestic heating, changing heating sources to cleaner burning stoves or alternative fuels (e.g. natural gas) can assist in reducing pollution (Tran & Mölders, 2012).

Air quality impacts. Impacts of air pollution on human health and ecosystems in the Arctic are even less explored than its sources and atmospheric transformation mechanisms. There are no Arctic-specific epidemiological studies and it is important to notice that the Arctic holds a larger indigenous population with disparate health effects than other regions. Hence, impacts of air pollution are also an environmental justice questions. To estimate health impacts, in a first step, globally derived IERs can be applied to the Arctic population taking specific illness factors into account. The toxicity of Arctic particulate matter is not well known either. Health effects research has primarily focused on contaminants such as polycyclic aromatic hydrocarbons, persistent organic pollutants, heavy metals, and mercury, rather than the most abundant air pollutants. Further, there is evidence that fog-processes PM contains more reactive oxygen species (Decesari et al., 2017) and is hence more toxic. There is also a clear bias towards air pollution information in larger Arctic cities. In small communities, where household emissions and indoor pollution exposure might be most important, monitoring facilities are normally lacking. For both types of inhabited areas,

improved monitoring and personal exposure studies, for example through co-production with citizen science and traditional knowledge, are necessary. With regards to ecosystem impacts, information availability is even less. But it seems important to understand the effects of urban and industrial emissions with the perspective of socio-economic growth in the region.

The way forward. In order to provide salient science for future Arctic sustainable development from an air pollution perspective, we encourage the interdisciplinary science community to:

- Conduct source apportionment studies to identify emission sources.
- Improve and validate current Arctic emission inventories.
- Develop Arctic-specific future emission scenarios together with Arctic stakeholders.
- Undertake intensive field campaigns in different seasons to study air pollution formation and transformation mechanisms under Arctic-specific meteorological conditions, complemented by winter-relevant chamber and modelling studies.
- Perform personal exposure measurements to provide data for health studies and conduct epidemiological studies.
- Model the ecosystem impacts of air pollution.
- Perform an analysis of current legal mechanisms to mitigate air pollution in the present and to prevent its increase in the future.

To be successful in predicting future Arctic pollution, and more generally, designing future Arctic sustainable development, the actions recommended above need to be embedded in a broader coupled human-environment system approach (HES-approach, Seidl et al., 2013). The HES approach is based on the understanding of two-way interactions between the environment and humans: Human activities influence the environment, e.g. greenhouse gas emissions cause the sea ice to retreat. Subsequently, a changed environment changes human behavior, e.g. more shipping through the Arctic is followed by Arctic port enlargements and hence more local air pollution emissions. Retreating sea ice is only one example of such a coupling. To better understand the possible futures of the Arctic resulting from the two-way interaction, Earth system scientists need to collaborate with social scientists, humanities researchers and stakeholders. We encourage the science community to pursue such transdisciplinary research methods. For the mitigation of local Arctic air pollution, this requires integrating stakeholders, especially from communities subject to high air pollution, into the research design and execution (Arnold et al., 2016). Involvement of stakeholders early on will establish trust and a sense of ownership which create opportunities for the co-development of mitigation strategies (Schmale et al., 2015).

Acknowledgements

J. Schmale acknowledges funding from the European FP7 project BACCCHUS (grant agreement no. 49603445). K.S. Law acknowledges support from the French Arctic Initiative PARCS (Pollution in the Arctic System) project funded by CNRS-INSU. K.A. Pratt acknowledges support through an Early-Career Research Fellowship from the Gulf Research Program of the National Academies of Sciences, Engineering, and Medicine. The content is solely the responsibility of the authors and does not necessarily represent the official views of the Gulf Research Program of the National Academies of Sciences, Engineering, and Medicine. W.R. Simpson and J. Mao acknowledge support from the U.S. National Science Foundation under grant AGS-1818552.

We also acknowledge discussions with colleagues at meetings and workshops organized by the international air Pollution in the Arctic: Climate, Environment and Societies (PACES) initiative, under the auspices of Future Earth/International Global Atmospheric Chemistry (IGAC) and the International Arctic Science Committee (IASC).

ECLIPSE: We thank IIASA for the ECLIPSE emissions (available at

http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html)

HTAPv2: We thank the EDGAR team for compiling the HTAPv2 emissions (available at

http://edgar.jrc.ec.europa.eu/htap_v2/).

Arctic Black Carbon: We thank the US Department of Energy supported Arctic Black Carbon Initiative for Russian BC emissions (see

<http://acs.engr.utk.edu/Data.php#Arctic%20Black%20Carbon%20Initiative>)

Author contributions

J.S., S.R.A., and K.S.L. conceived the study. All authors contributed to writing the manuscript.

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Table 1. *Inventories Describing Future Emissions in the Arctic*

Inventory	Sectors	Considered time horizon	Scenario types
ECLIPSE	Agriculture (waste burning), domestic, energy, industry, surface transport and waste	until 2050	Baseline, current legislation (CLE), maximum technically feasible reduction (MTFR), no further control (NFC), short-lived climate-forcing pollutants (SLCP)
RCP	Energy, domestic, agricultural waste burning, industry, transport, waste, solvent production and use	until 2050	Pathways to different global net radiative forcing scenarios at year 2100 (range 2.6-8.5 Wm^{-2}). Pollution emissions follow assumption of enhanced pollution control technology with increasing economic growth across all scenarios.

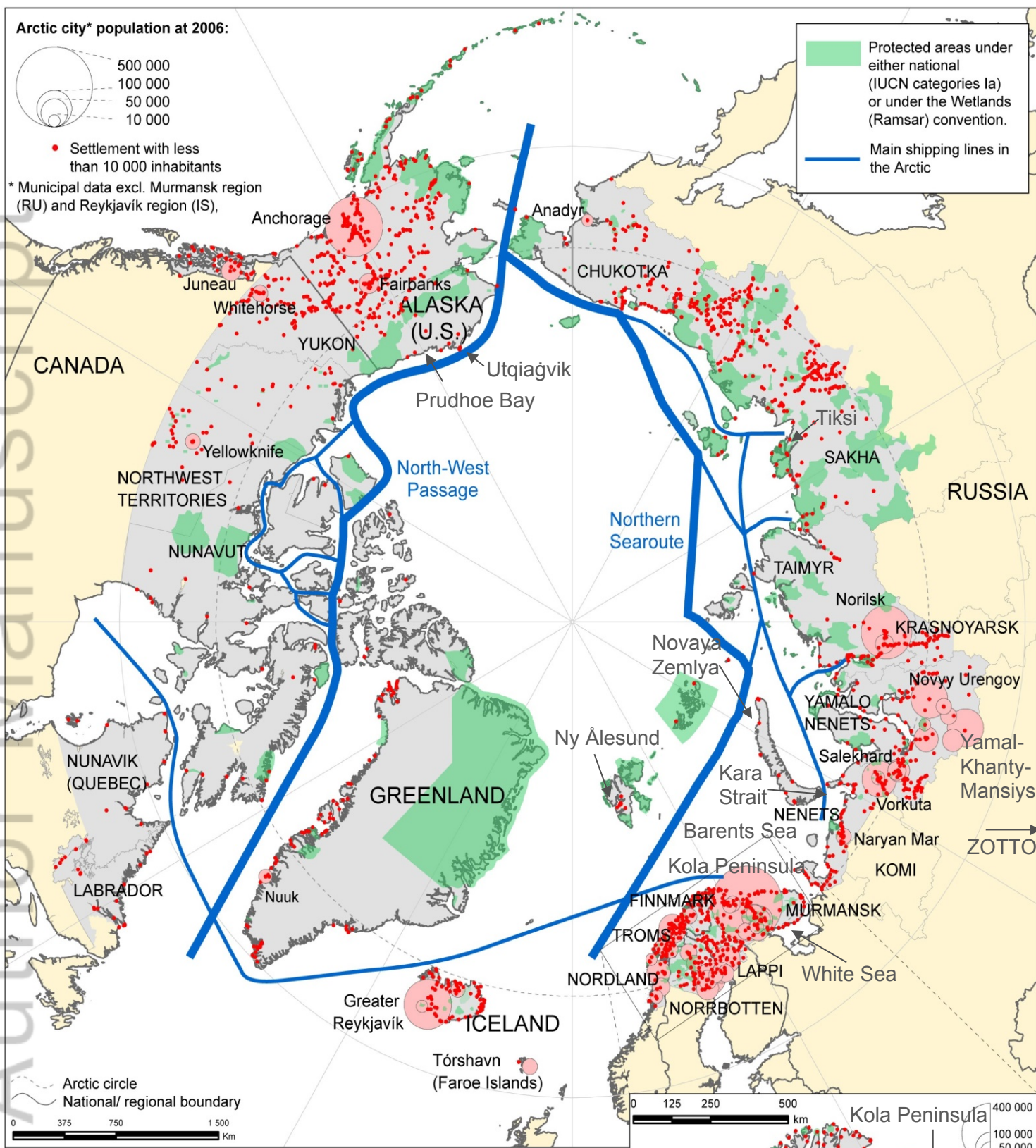
Figure 1: Population distribution in the Arctic. The map shows population per settlement in the Arctic region as of 2006. Murmansk, Anchorage, Reykjavik and Norilsk are the largest settlements in terms of size of population in the region while a number of small settlements with less than 10 000 inhabitants are visible in all regions and along the coasts of Iceland and western Greenland. The map also highlights protected areas under either national (IUCN categories IA) or under the Wetlands (Ramsar) Convention. With permission from Nordregio. (<http://www.nordregio.se/en/Maps/05-Environment-and-energy/Settlements-and-protected-areas-in-the-Arctic/>). Grey place names have been added to the map to highlight additional locations discussed in this work.

Figure 2: Comparison of different Eurasian BC emission inventories from (a) the Arctic Black Carbon dataset (ABC, Huang et al., 2015), (b) ECLIPSEv5 (Stohl et al., 2015), and (c) HTAPv2 (Janssens-Maenhout et al., 2015) for the year 2010, from Sergent (2017). Note not all inventories cover the entire Arctic, hence the focus on Eurasia.

Figure 3. Future emission scenarios. Scenarios are for (a) NO_x , (b) BC, and (c) SO_2 , based on the ECLIPSE v5a and RCP emissions datasets for regions north of 60°N (Stohl et al., 2015, van Vuuren et al., 2011). The colored lines show different future scenarios. Solid: ECLIPSE scenarios with Baseline, current legislation (CLE), maximum technologically feasible reduction (MTR), no further control (NFC), and short-lived climate-forcing pollutants (SLCP) mitigation. Dotted: RCP scenarios. Note that the ECLIPSE NFC scenario is from the v5 dataset (not v5a). See main text for description of scenarios.

Figure 4. Air quality exceedances in Fairbanks, Alaska, USA. Mean number of measurement days in exceedance of the EPA $\text{PM}_{2.5}$ standard ($35 \mu\text{g m}^{-3}$) per month 2006-2014 (from EPA air quality database). Measurements are made only every third day, hence a factor of three has been applied to the data shown in this figure.

Figure 5. Fractional mass composition of $\text{PM}_{2.5}$ in Fairbanks and North Pole, Alaska. The species shown are: Organic carbonaceous materials (OCM), which is calculated by multiplying organic carbon (OC) by 1.4 to account for the mass of O and H in these species. Elemental carbon (EC), and inorganic ions (SO_4^{2-} , NO_3^- , NH_4^+) are also shown. Data represent the average from three winter seasons between November 2011 and February 2014.



Settlements and protected areas in the Arctic 2006

Arctic region defined as in Arctic Human Development Report

Alaska (US); CA - Yukon, Northwest Territories, Nunavut, Nunavik (Quebec), Labrador
 Greenland, Iceland, the Faroe Islands; NO - Nordland, Troms, Finnmark; SE - Norrbotten;
 FI - Lappi; RUS - Murmansk, Nenets, Vorkuta (Komi), Yamalo-Nenets, Norilsk & Igrska
 (Krasnoyarsky Krai), Taimyr, Sakha (13 northernmost subregions), Chukotka.

Data source: World protected areas database, UNEP-WCMC, 2005 (Russian data digitized by WWF in 2005 by official sources); Population: National statistical institutes.
 Analysis & design: J. Roto / J. Sterling

