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Arctic air pollution: Challenges and opportunities for the next decade

S.R. Arnold[1*] • K.S. Law[2] • C.A. Brock[3] • J.L. Thomas[2] • S.M. Starkweather[3,4] • K. von Salzen[5] • A. Stohl[6] • S. Sharma[7] • M.T. Lund[8] • M.G. Flanner[9] • T. Petäjä[10] • H. Tanimoto[11] • J. Gamble[12] • J.E. Dibb[13] • M. Melamed[4,14] • N. Johnson[15] • M. Fidel[12,16] • V.-P. Tynkkynen[17] • A. Baklanov[18,19] • S. Eckhardt[6] • S.A. Monks[3,4] • J. Browse[1] • H. Bozem[20]

[1]Institute for Climate and Atmospheric Science, School of Earth and Environment, University of Leeds, Leeds, United Kingdom
[2]LATMOS/IPSL, UPMC Univ. Paris 06 Sorbonne Universités, UVSQ, CNRS, Paris, France
[3]Earth System Research Laboratory, National Oceanic and Atmospheric Administration, Boulder, Colorado, United States
[4]Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, Colorado, United States
[5]Canadian Centre for Climate Modelling and Analysis, Environment Canada, Victoria, British Columbia, Canada
[6]NILU-Norwegian Institute for Air Research, Kjeller, Norway
[7]Science and Technology Branch, Environment Canada, Toronto, Ontario, Canada
[8]Center for International Climate and Environmental Research, Oslo, Norway
[9]Climate and Space Sciences and Engineering, University of Michigan, Ann Arbor, Michigan, United States
[10]Department of Physics, University of Helsinki, Helsinki, Finland
[11]Center for Global Environmental Research, National Institute for Environmental Studies, Tsukuba-City, Ibaraki, Japan
[12]Aleut International Association, Anchorage, Alaska, United States
[13]Institute for the Study of Earth, Oceans, and Space and Department of Earth Sciences, University of New Hampshire, Durham, New Hampshire, United States
[14]International Global Atmospheric Chemistry Project, Boulder, Colorado, United States
[15]Institute for the Study of Environment and Society, Brown University, Providence, Rhode Island, United States
[16]Resilience and Adaptive Management Group, University of Alaska Anchorage, Alaska, United States
[17]Department of Social Research and Aleksanteri Institute, University of Helsinki, Helsinki, Finland
[18]World Meteorological Organization, Genève, Switzerland
[19]Danish Meteorological Institute, Copenhagen, Denmark
[20]Institute for Atmospheric Physics, University Mainz, Mainz, Germany

*s.arnold@leeds.ac.uk

Abstract

The Arctic is a sentinel of global change. This region is influenced by multiple physical and socio-economic drivers and feedbacks, impacting both the natural and human environment. Air pollution is one such driver that impacts Arctic climate change, ecosystems and health but significant uncertainties still surround quantification of these effects. Arctic air pollution includes harmful trace gases (e.g. tropospheric ozone) and particles (e.g. black carbon, sulphate) and toxic substances (e.g. polycyclic aromatic hydrocarbons) that can be transported to the Arctic from emission sources located far outside the region, or emitted within the Arctic from activities including shipping, power production, and other industrial activities. This paper qualitatively summarizes the complex science issues motivating the creation of a new international initiative, PACES (air Pollution in the Arctic: Climate, Environment and Societies). Approaches for coordinated, international and interdisciplinary research on this topic are described with the goal to improve predictive capability via new understanding about sources, processes, feedbacks and impacts of Arctic air pollution. Overarching research actions are outlined, in which we describe our recommendations for 1) the development of trans-disciplinary approaches combining social and economic research with investigation of the chemical and physical aspects

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of Arctic air pollution; 2) increasing the quality and quantity of observations in the Arctic using long-term monitoring and intensive field studies, both at the surface and throughout the troposphere; and 3) developing improved predictive capability across a range of spatial and temporal scales.

1. Introduction

Arctic ecosystems, climate, and societies are affected by air pollution from both remote and local sources. However, because both the environment and economy of the Arctic are rapidly changing, Arctic air pollution is influenced by a complex web of environmental and atmospheric feedbacks and socio-economic responses. Changes in atmospheric pollutants such as aerosol particles and tropospheric ozone affect the atmospheric radiation balance, and contribute to Arctic climate warming (Shindell and Faluvegi, 2009). The resulting sea ice loss may increase accessibility of the Arctic, leading to increases in air pollutant emissions within the Arctic from activities such as oil and gas extraction or shipping. It is thought that Northern Hemisphere mid-latitude emissions (from Europe, Asia, and North America) are currently the main source of air pollutants in the Arctic (Stohl, 2006; Sharma et al., 2013), including also toxic contaminants with important atmospheric pathways (e.g. mercury (Hg), certain persistent organic pollutants (POPs)) However, sources of air pollution from within the Arctic or nearby sub-Arctic (defined here as 'local') are already important in some regions (e.g., Stohl et al. 2013), and these and other sources may grow rapidly in the future (Corbett et al., 2010; Peters et al., 2011).

It is crucial to improve quantification of the relative contributions of different anthropogenic pollutant sources to provide a sound scientific basis for sustainable solutions and adaptive strategies. The rapid pace of Arctic environmental change puts a high priority on improving understanding of processes controlling sources and fate of Arctic air pollutants and their impacts on Arctic communities. Deficiencies in predictive capability and a lack of observations at high latitudes present major challenges to advancing this understanding, and to making credible near- and long-term projections of Arctic environmental change.

Here, we describe a new international initiative - air Pollution in the Arctic: Climate, Environment and Societies (PACES) (see http://www.igacprojects.org/PACES) which has been recently launched following recognition within the international community[1] for a need to improve our understanding of Arctic air pollution and its impacts. This paper outlines our views about how these issues could be tackled with future collaborative research efforts. The PACES initiative is being developed under the auspices of the International Global Atmospheric Chemistry project (IGAC) (under Future Earth) and the International Arctic Science Committee (IASC-Atmosphere Working Group (WG)). PACES will benefit the wider community by providing improved scientific knowledge on processes controlling air pollutants in the Arctic, in particular those linked to economic and climate change drivers, and subsequent impacts on human health and ecosystems. PACES aims to provide 1) motivation and coordination of research efforts at national and international level on Arctic air pollution and its impacts over the next decade, and 2) recommendations to guide new research on this topic. PACES is driven by an acknowledgement in the scientific community of key deficiencies in our understanding, which remain despite substantial progress made in recent years regarding processes such as the long-range transport of pollution to the Arctic (Law et al., 2014). These deficiencies are defined and discussed in Section 2. A main goal of PACES is to establish a framework for new and improved collaborative efforts, particularly those crossing traditional disciplinary boundaries that take into account societal perspectives and engage with Arctic communities. Such opportunities, together with the needs for tackling these issues, are outlined in Section 3, including capacity building for coordinated measurement programmes (both short-term campaigns and long-term observations) and modelling initiatives. Lastly, we recommend a roadmap for moving forward, taking into account synergies with other national and international initiatives and identifying several key focus areas (Section 4).

2. Major science issues

Progress in understanding processes controlling air pollution in the Arctic has benefited greatly from observations of pollutants and processes at the surface for many years. Such efforts have resulted in improved understanding of the sources of Arctic air pollution at the surface (e.g., Sharma et al., 2006), fluxes and interactions of chemical constituents between the snow, sea ice, ocean, biosphere and atmosphere (e.g., Dibb et al., 1998; Honrath et al., 2002; Levasseur, 2013), and chemical reactions within the snowpack and lowermost atmosphere (e.g., Simpson et al., 2007; Grannas et al., 2007). More recent progress in understanding the sources, processing, fate and impacts of air pollution in the Arctic throughout the depth of the troposphere has been enabled, in part, by extensive and unprecedented observations from aircraft, surface observatories and satellites during the International Polar Year (IPY). These included the POLARCAT (Polar Study using Aircraft, Remote Sensing, Surface Measurements and Models, Climate, Chemistry, Aerosols and Transport) campaigns in spring and summer 2008 (Law et al., 2014), as well as increased collaborative efforts making observations

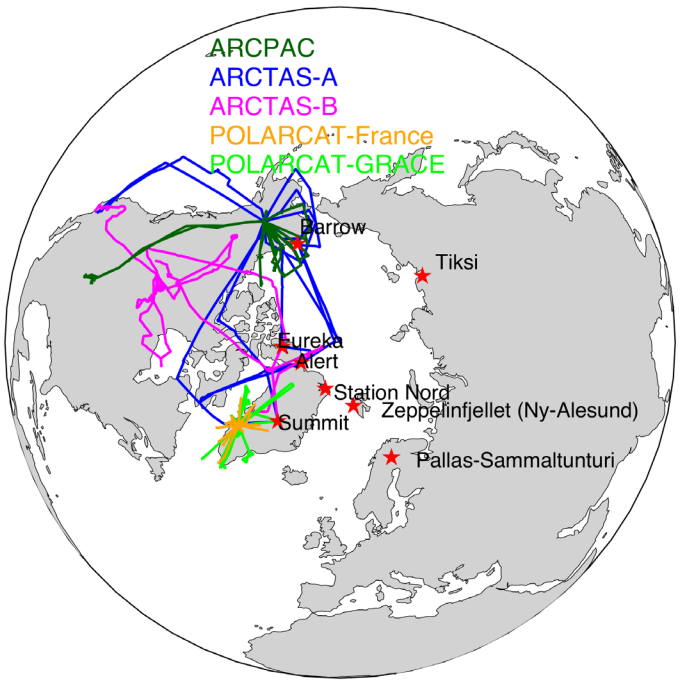

Figure 1

Arctic long-term surface observations and year 2008 research flights.

Locations of key long-term surface observing sites in the Arctic, and flight tracks from aircraft campaigns that took place during the International Polar Year 2008.

Date ranges of flights shown for each campaign are: ARCPAC (April 1–Apr 23, 2008); ARCTAS-A (April 1–April 19, 2008); ARCTAS-B (June 18–June 26, 2008); POLARCAT-France (June 30–July 14, 2008); POLARCAT-Grace (July 2–July 14, 2008).

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from the long-term surface sites, such as International Arctic Systems for Observing the Atmosphere (IASOA) and the Network for the Detection of Atmospheric Composition Change (NDACC). Locations of key long-term surface observing sites and aircraft campaigns taking place during the International Polar Year 2008 are shown in Figure 1. Aircraft observations in particular, have enabled new understanding on pollution distributions, and processes controlling them, away from the Arctic surface. Subsequent analysis of these (and other) data, and their use in evaluating chemical transport models and climate models (e.g. Fisher et al., 2010; Monks et al., 2015; Emmons et al., 2015), has highlighted remaining issues and new questions that require development of coordinated observational and modelling efforts, interdisciplinary research, and increased engagement of communities and societal perspectives. In this section, we provide a qualitative overview of the scientific understanding of the processes that govern Arctic air pollution and summarize areas where there are still large uncertainties in our understanding.

2.1 Arctic pollution sources – pollution imported to and emitted within the region

The Arctic region is a receptor of air pollution from diverse source regions at mid-latitudes. Pollution import from southerly latitudes is largely facilitated by transport in low pressure weather systems (Stohl, 2006). Recent studies based on both in-situ vertical profile measurements and modelling have demonstrated enhanced pollutant layers throughout the depth of the Arctic troposphere, with northern Eurasian sources dominating near the surface, and import from mid-latitude North America and Asia becoming more important in the mid and upper troposphere (e.g. Sharma et al., 2006, 2013; Fisher et al., 2010; Brock et al., 2011; Bourgeois and Bey, 2011; Wespes et al., 2012; Stohl et al., 2013; Law et al., 2014; Monks et al., 2015). These different source sensitivities in the vertical result from pseudo-isentropic transport pathways that bring mid-latitude emissions to the Arctic, and supressed vertical mixing of pollutants within the Arctic, due to the high vertical stability of the Arctic troposphere (Fig. 2; Stohl, 2006; Hirdman et al., 2010). Changes in mid-latitude emissions, and climate-driven changes in transport patterns, coupled to increasing local emissions are expected to shift the balance among pollutant source contributions in the coming decades. Short-lived air pollutants such as aerosol and precursors of ozone emitted outside and within the Arctic have the potential to impact Arctic climate (Shindell and Faluvegi, 2009; Sand et al., 2015). The sensitivity of Arctic climate to emissions of pollutants is not well characterised, particularly from local Arctic sources.

The Arctic lower troposphere is influenced by pollution from local sources and sources in high-latitude Eurasia, which are currently poorly quantified. These sources include emissions associated with resource extraction (e.g. flaring of gas associated with oil production; Stohl et al., 2013) and shipping (e.g. Corbett et al., 2010; Aliabadi et al., 2015). These local sources are already influencing atmospheric composition on local and regional scales (Roiger et al., 2015; Marelle et al., 2016). As Arctic sea-ice thins and retreats, it is expected that new shipping routes within the Arctic Ocean will seasonally open up, substantially decreasing transport distances between Asia and North America and Europe. This may result in up to 5% of global

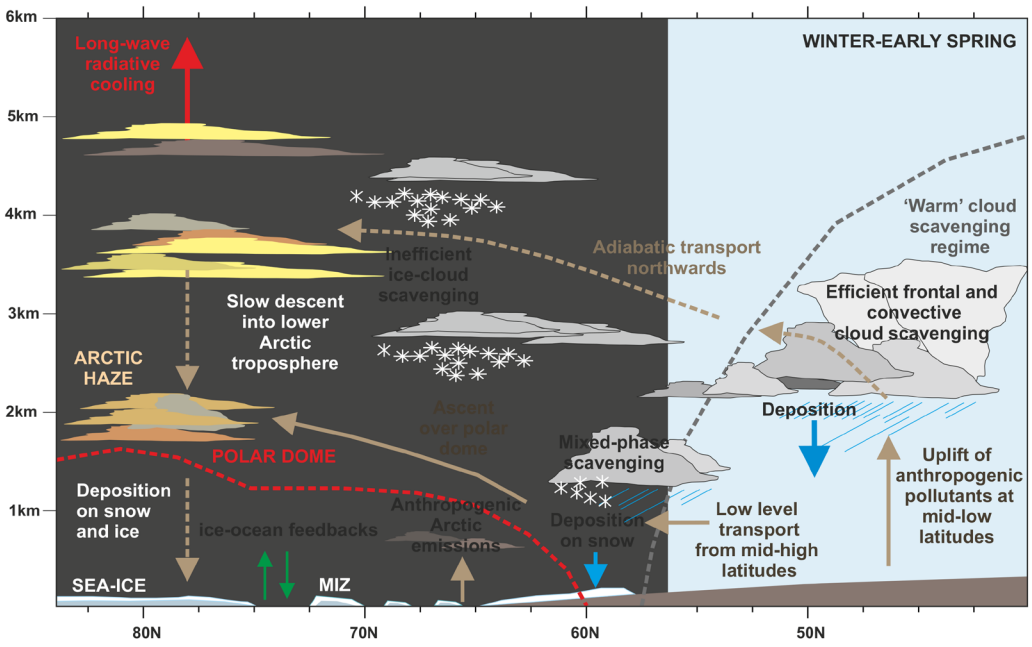

Figure 2

Import and processing of Arctic air pollution.

Schematic diagram showing key processes associated with emission, transport, and deposition of pollution to the Arctic in late winter and early spring when Arctic haze is at a maximum. Brown arrows show atmospheric transport pathways, with dashed lines indicating less efficient routes. The dashed black line shows the approximate position of the -15°C isotherm, which is the approximate temperature threshold for ice clouds. The "polar dome", shown by the red dashed line, presents a barrier to adiabatic atmospheric transport from the southerly warmer mid-latitudes to the Arctic surface. Transport from northern Europe, within the polar dome, to the Arctic surface occurs much more efficiently. The coupling between different patterns of vertical uplift and northerly transport into the Arctic has implications for the locations of wet and dry deposition (shown by blue arrows) from air masses imported from the different mid-latitude regions. "MIZ" is the marginal ice zone. For clarity, chemical transformations and biogeochemical processes are not shown. Adapted from Browse et al. (2012).

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shipping traffic moving to high Arctic routes (Corbett et al., 2010). In addition, a large portion of the Earth's undiscovered oil and gas is predicted to be under the Arctic Ocean floor and will be increasingly utilized in the future (Peters et al., 2011; Gautier et al., 2009). Population numbers, associated urbanization, infrastructure development, and mining activities are also expected to increase in the Arctic (Andrew, 2014), adding to local emissions of air pollutants, unless compensated by tighter pollutant emission controls. A detectable increase in particulate matter concentrations at Resolute Bay on remote Lord Cornwallis Island in Canada has been associated with ship traffic in the newly accessible Northwest Passage (Aliabadi et al., 2015). Similar increases have been associated with cruise ship traffic near Svalbard in the Norwegian Arctic (Eckhardt et al., 2013). Flaring of excess natural gas during oil extraction in the Russian Arctic has been estimated to be the dominant contributor to black carbon concentrations in the atmosphere and surface snow in this portion of the Arctic (Stohl et al., 2013) but these emissions are highly uncertain. For example, the officially stated volumes of flared gas in Russia, reported by oil and gas companies, differ greatly from satellite-based data (e.g. Vasileva et al, 2015). There is evidence that use of wood for household heating could be an important source of absorbing and non-absorbing particles as well as toxic compounds such as polycyclic aromatic hydrocarbons (PAHs). Indications are that such domestic combustion has increased during the last ten to fifteen years (Pearson et al., 2013) but these estimates are only partially quantified. In Finland, for example, this source accounts for 40% of $PM_{2.5}$ (mass of airborne particles of diameter less than 2.5μm), more than half of all national black carbon emissions, and 80% of PAHs (Ahtoniemi et al, 2010). We note that AMAP (2015) recently recommended mitigation, together with improved quantification, of this (and other) pollutant source.

Long-term observations at the surface provide the main source of information on seasonal cycles and long-term trends in Arctic pollutants. Such sites will be critical for understanding how Arctic environmental change and changes in local sources affect future Arctic air pollutant abundances in the coming years and decades. Surface sites are most sensitive to local and Eurasian emissions (Baklanov et al., 2013), and so provide much less information on transport of pollution from North American and Asian sources. Models display diverse and often poor skill in simulating Arctic pollution enhancements both at the surface and throughout the depth of the Arctic troposphere (Emmons et al., 2015; Monks et al., 2015; Eckhardt et al., 2015; Figs. 3, 4), suggesting potential deficiencies in their ability to diagnose pollutant contributions from both local and remote sources and their impacts.

2.2 Processing, fate and impacts of Arctic pollution on climate and ecosystems

The environmental fate of airborne pollutants in the Arctic is largely determined by transfer from the atmosphere to the surface, and is thus influenced by the stratified vertical structure of the Arctic troposphere (see 3.1). Understanding vertical transport within the Arctic is one of the key uncertainties in evaluating the impacts of extra-Arctic pollutants on Arctic biogeochemical systems. Early studies, mainly on the Greenland ice sheet, measured in-situ dry deposition rates of aerosol constituents, and samples from deposited and precipitating snow (e.g. Davidson et al., 1987; Bergin et al., 1994, 1995). Despite these early studies, rates of dry deposition to the large-scale Arctic surface in models, and wet scavenging within and underneath liquid-phase, ice-phase and mixed-phase clouds remain poorly constrained. Lack of understanding of pollutant

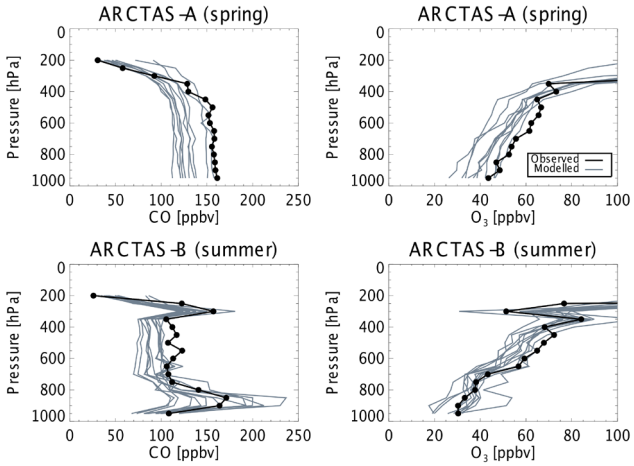

Figure 3

Evaluation of model carbon monoxide and ozone vertical profiles.

Comparison of measured (black line) and modelled (gray lines) carbon monoxide (CO, panels a and c) and ozone (O_3, panels b and d). Measurements are from the NASA ARCTAS campaign in the Arctic and high northern mid-latitudes in spring (panels a and b) and summer (panels c and d) 2008. Model output is from 10 different chemical transport models participating in the POLMIP model comparison exercise (Emmons et al., 2015). CO is produced from combustion and oxidation of volatile organic compounds and has a lifetime of months at high latitudes. Tropospheric ozone is formed in-situ in the atmosphere from emissions of NOx from fossil fuel and biomass combustion, soils and lightning, and volatile organic compounds sourced from anthropogenic and natural sources. Ozone in the upper troposphere has an additional source from the stratosphere. Models display varied skill and large inter-model diversity for both species, indicating errors in emissions, transport (especially for CO), and/or chemical processes. Adapted from Monks et al. (2015).

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deposition rates may be a key driver for the poor skill of many models in simulating the seasonal cycle and magnitude of aerosol pollutants when compared to Arctic measurements (AMAP, 2015; Eckhardt et al., 2015). In particular, disagreement with observations has been shown to be sensitive to the representation of wet scavenging and aerosol microphysical (e.g., black carbon aging) processes in models (e.g., Browse et al., 2012; Garrett et al., 2011; Liu et al., 2011; Lund and Berntsen, 2012). More recently, a model simulation using observed meteorology to reasonably simulate the seasonal cycle and vertical distribution of pollutants in the Arctic in the year 2008 found that aerosol wet scavenging was ~3 times larger than dry scavenging in the Arctic (Breider et al., 2014).

A better understanding of the processing, fate and impacts of Arctic pollution on climate and ecosystems can be realized only if a number of research challenges are surmounted. Much better representations of current and future emissions of pollutants including black carbon, nitrogen oxides, sulphur species, methane, and other volatile organic compounds (VOCs) from Arctic sources are needed. The environmental consequences of potentially large quantities of VOCs, including alkanes, alkenes, and aromatic species, emitted during oil and gas extraction activities in the Arctic have not been evaluated.

As warming of the Arctic continues, uptake of carbon by the biosphere will increase. At the same time biogenic emissions of volatile organic gases increase. Based on long-term observational data, these two processes lead to elevated concentrations of secondary aerosol particles and possible cooling of the climate in the region (Paasonen et al. 2013), by increasing the number of cloud condensation nuclei, affecting cloud properties (e.g. Spracklen et al. 2008). Furthermore, the biogenic vapors condense onto the aerosol particles, increasing the amount of diffuse radiation that is more efficiently used in photosynthesis increasing the carbon uptake further (Kulmala et al. 2004, 2014). These two negative feedback mechanisms affecting the Arctic environment are just examples of many feedbacks associated with the system (Arneth et al. 2010). As another example, thawing permafrost is associated with elevated emissions of methane, which is a potent long-lived greenhouse gas as well as an important species that affects the tropospheric OH budget (e.g., Wuebles and Hayhoe, 2000). All of these emissions alter atmospheric chemical processing in the Arctic atmosphere, where photochemical oxidation has large temporal variability due to strong variation of solar insolation between polar night and polar day.

Earth system and chemistry-transport models, combined with simplified climate models and metrics (e.g. UNEP-WMO 2011), form the basis for climate policy development. The fact that most models currently cannot adequately replicate observations, particularly the vertical profile (Figs. 3, 4), therefore has important implications for estimates of the climate impacts from Arctic air pollutants. The deposition and vertical transport processes themselves need to be studied in the context of pollutant deposition and re-emission (e.g. nitrate or contaminants such as mercury and persistent organic pollutants (POPs) from snow surfaces). The Arctic is a key receptor region for long-range transport of both POPs and mercury (Hg), which can accumulate particularly in the Arctic environment due to low temperatures, snow covered surfaces and extended conditions of darkness (AMAP, 2004, 2011; Ariya et al., 2004). In addition to primary emissions and import into the region, such contaminants are also re-mobilised to the atmosphere from the surface. Many POPs were banned by the Stockholm Convention (UNEP, 2001) due to their high levels of toxicity. However, their long environmental lifetimes mean that they may have impacts in the Arctic for many years and decades into the future. Production of reactive trace species such as nitrogen oxides and formaldehyde (CH_2O) within snow (Sumner and Shepson, 1999; Honrath et al., 1999) and emission to the atmosphere can dominate surface atmospheric photochemistry in polar regions remote from anthropogenic influence (Grannas et al., 2007). Finally, the indirect effects of aerosols on cloud radiative properties have been examined in several studies, but the results have been varied, and no consistent understanding of cloud-aerosol interactions has evolved

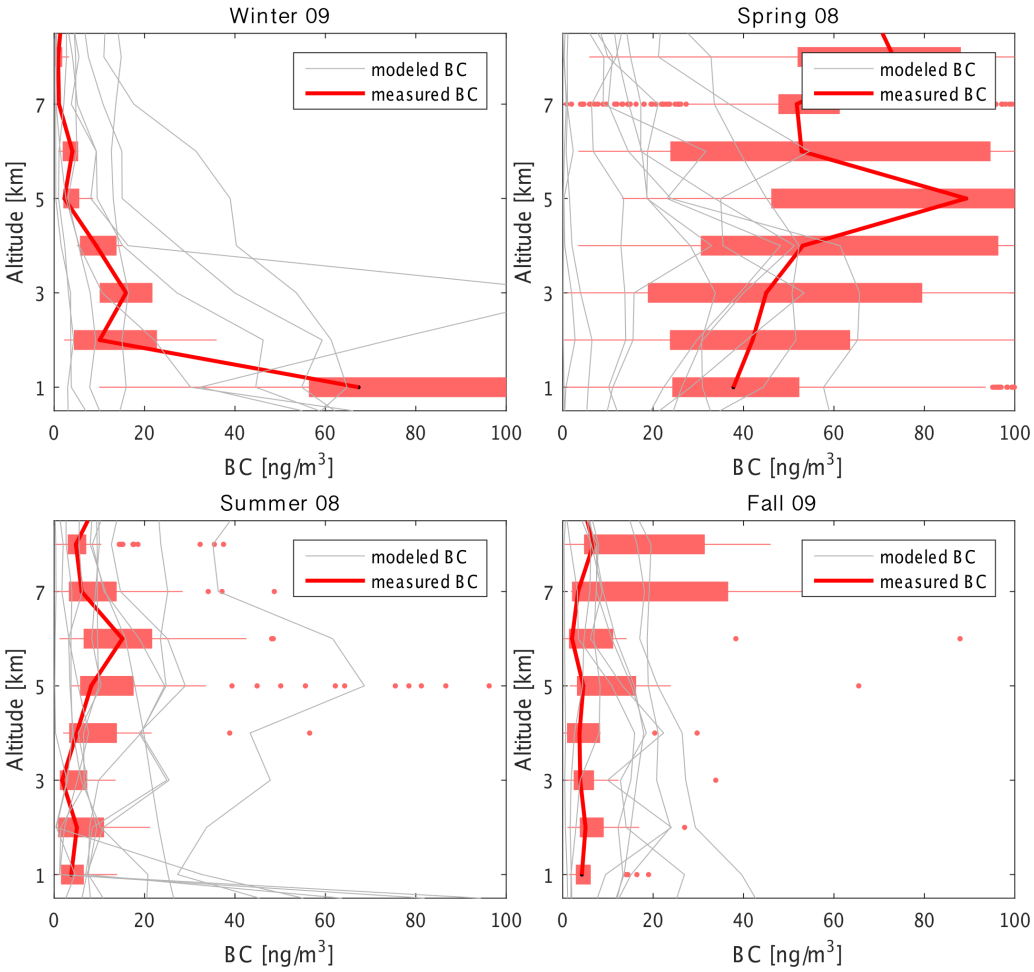

Figure 4

Evaluation of model black carbon aerosol vertical profiles.

Comparison of measured (red line, bars, points) and modelled (gray lines) black carbon (BC) based on measurements north of 70° made in a) winter 2009, b) spring 2008, c) summer 2008, and d) fall 2009 during the HIPPO, ARCTAS, ARCPAC, and PAMARCMiP campaigns. Each gray line indicates output from a different model. Most of the models display particularly poor skill in spring, and considerable inter-model diversity throughout the year. Adapted from Eckhardt et al. (2015).

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(e.g., Garrett and Zhao, 2006; Gayet et al., 2009; Earle et al., 2011; Lance et al., 2011). Thus cloud-aerosol interactions, which affect the radiation balance, the hydrological cycle, and the sea-ice state, remain one of the primary uncertainties regarding the effect of pollutants on the Arctic climate (Browse et al., 2014; Fig. 2).

2.3 Interactions and feedbacks between anthropogenic pollution and natural processes

Trace gas and aerosol budgets in the Arctic are heavily influenced by natural emissions in the region, including species emitted from sea-ice, snow, open water, and land. For example, DMS (dimethyl sulphide) emissions from the open ocean/ice-edge are oxidized to sulphate in the atmosphere, forming new particles and contributing to growth of existing aerosols (Charlson et al., 1987; Sharma et al., 2012; Leaitch et al., 2013, Kulmala et al., 2007; Gross and Baklanov, 2004). Similarly, sea salt emissions may change with a reduction in Arctic sea-ice extent. Halogens, especially bromine-containing compounds, present in seawater, snow, sea-ice, and aerosols are activated in the Arctic and cause ozone depletion events during spring and summer (Simpson et al., 2007; Abbatt et al., 2012). In addition, it has been recently discovered that there is active chlorine chemistry occurring in the Arctic boundary layer, likely arising from activation of chloride present in seawater (Liao et al., 2014).

As warming of the Arctic continues, it is expected that biogenic emissions of volatile organic gases from vegetation will increase. This may lead to increased biogenic aerosol concentrations (Paasonen et al., 2013). New particles formed from gas-to-particle conversion in the boreal forest environment may increase the numbers of cloud condensation nuclei, affecting cloud properties (e.g., Spracklen et al., 2008). Natural and human-influenced open fires also emit large quantities of trace gases and aerosols within and near the Arctic (e.g., Warneke et al., 2009, 2010). Both the frequency and geographic distribution of these fires is expected to increase in a warming climate (de Groot et al., 2013). The climate impacts of such emissions may result in further warming of the high latitudes, leading to increases in natural fire activity, and a potential but poorly understood climate feedback. Ecosystems are impacted by the deposition of pollutants, such as nitrogen and

sulphur compounds. Model studies suggest that critical loads of acidifying deposition due to S and N were exceeded in the 1990s across large regions of N Europe and the Norilsk region of western Siberia (Forsius et al., 2010). Total deposition of S to the Arctic was found to decrease by around 50% by 2000. However, future increases in local industrial activities within the Arctic could lead to some offsetting of such reductions.

In light of the potential for further development of human activities in the Arctic region, studies to understand natural chemical and aerosol processes and how they are modified by anthropogenic pollution are essential. For example, nitrogen emissions from snow and sea-ice occur after deposition of nitrogen that was originally partially anthropogenic in origin (Grannas et al., 2007; Bartels-Rausch et al., 2014). Activation of chlorine in the Arctic, through release and photolysis of nityrl chloride ($ClNO_2$) by reaction of chloride (Cl^-) with nitrogen pentoxide (N_2O_5) in aerosol, will likely be enhanced as nitrogen emissions increase with new anthropogenic activities, such as shipping and petrochemical extraction and processing. The reduction of Arctic sea-ice, caused by environmental change, represents both a transition of the Arctic ice-edge progressively moving north and a transition from snow/ice to open ocean in summer and fall. This change has implications for atmosphere-ocean exchanges of reactive species because the ice cover limits exchange and also inhibits the biogeochemical cycles that produce DMS (Clarke and Ackley, 1984). Atmospheric chemistry and formation of new aerosols depends on the rate of atmospheric oxidation, which will be influenced by natural emissions, anthropogenic inputs, and the nature of the surface in the Arctic (sea-ice, snow, land, ocean), which controls surface albedo and the efficiency of dry deposition. The effects of climate change on snow cover, soils, vegetation, and water bodies, in addition to changes in surface temperature, ocean and atmospheric circulations, will likely have impacts on the cycling of POPs and Hg through different environmental media at high latitudes, and their atmospheric mobilisation (Wöhrnschimmel et al., 2013; Pacyna et al, 2015).

2.4 Arctic climate response to radiative forcing within and outside the Arctic

A large portion of the rapid warming seen in the Arctic has been attributed to increases in well-mixed greenhouse gas concentrations in the atmosphere (Gillett et al., 2008; Fyfe et al., 2013; Najafi et al., 2015). However, short-lived air pollutants such as tropospheric ozone and aerosol also substantially affect Arctic climate (Shindell and Faluvegi, 2009). Many of the physical and chemical processes that control short-lived air pollutants and their influence on Arctic climate are poorly understood. Such short-lived pollutants have heterogeneous distributions in the atmosphere, leading to complicated radiative forcing patterns, and uncertain regional climate response. In general, changes in Arctic climate can arise from changes in radiative forcings and feedbacks within the Arctic, changes in forcing outside the Arctic and subsequent changes in the transport of heat to the Arctic, and unforced internal variability in the Arctic atmosphere, ocean, snow, and ice. For instance, positive radiative forcings exerted by pollutants outside the Arctic warm the Arctic via enhanced heat transport to the Arctic (Shindell and Faluvegi, 2009; Flanner, 2013; Sand et al., 2013a). The radiative forcing of black carbon occurring at mid-latitudes may even have a greater impact on Arctic air temperatures than black carbon forcing within the Arctic (e.g. Flanner, 2013; Sand et al., 2013b). However, emissions of black carbon within the Arctic may produce much stronger Arctic temperature response per mass emitted (Sand et al., 2013a). In the Arctic, the absorption of solar radiation by black carbon, organic carbon, and mineral dust (Breider et al., 2014) causes the air to warm locally (Quinn et al., 2007; Brock et al., 2011). However, vertical mixing of heat is inefficient owing to high static stability of the Arctic atmosphere. Consequently, radiative forcing by elevated layers of black carbon within the Arctic can cool the near-surface air owing to a lack of mixing of sensible heat and dimming of solar radiation at the surface. Warming aloft from the black carbon, together with reduced solar radiation reaching the ground, can increase stability, initiating a positive feedback mechanism that further reduces vertical mixing (Petäjä et al., 2016). These surface-cooling processes occur despite net positive radiative forcing of black carbon at the top of atmosphere (Quinn et al., 2007; Brock et al., 2011; Flanner, 2013; Sand et al., 2013b; Lund et al., 2014). In contrast, black carbon that deposits on snow and sea-ice or resides in the lower Arctic troposphere can cause strong surface atmospheric warming (e.g. Flanner et al., 2007). Changes in temperatures caused by black carbon radiative forcings are amplified by powerful snow and sea-ice albedo feedbacks in the Arctic (Flanner, 2013).

Recent results from model simulations indicate that black carbon has very likely contributed to the observed warming of near-surface air in the Arctic since pre-industrial times. In contrast, negative radiative forcing by sulphate aerosol has caused a reduction in the net rate of Arctic warming (UNEP-WMO, 2011; AMAP, 2015; Sand et al., 2015). The magnitude and spatial distribution of the net combined radiative forcing of these aerosol changes is presently unknown, with responses of clouds to changes in aerosol concentrations (i.e. via so-called indirect effects) particularly uncertain. Regionally, emissions of black carbon from East and South Asian domestic sources, and fires in Siberian and tropical forests are particularly important for Arctic temperatures (Sand et al., 2015). Emissions of black carbon from current oil/gas production within the Arctic may be efficient in warming the Arctic locally (Ødemark et al., 2012; Sand et al., 2013a; AMAP, 2015) whereas local shipping emissions are likely causing a small cooling due to sulphate formation (Ødemark et al., 2012; Marelle et al., 2016).

Tropospheric ozone also leads to Arctic warming (Shindell et al., 2006) with most of the ozone resulting from methane oxidation at mid- and northerly latitudes. (UNEP-WMO, 2011; AMAP, 2015; Sand et al., 2015). Release of methane due to thawing of permafrost and gas hydrates in deep-sea sediments in the Arctic in a warming climate may lead to increased radiative forcing, as well as enhanced ozone production and further amplification of Arctic warming trends (Isaksen et al., 2014).

In the future, reductions in global emissions of aerosol and precursors may result in positive net aerosol radiative forcing, due largely to reduced sulphur emissions, and an increase in the rate of Arctic warming (Gillett and von Salzen, 2013; Gagné et al., 2015). Emission reductions may also lead to a strengthening of the meridional atmospheric temperature gradient at high latitudes and thereby changes in large-scale circulation (Rotstayn et al., 2014). At present, it is unknown whether pollutant-induced radiative forcings contribute to changes in atmospheric circulation and extreme weather at mid-latitudes, which have been linked to observed changes in Arctic climate (Francis and Vavrus, 2012; Coumou et al., 2014).

Recent research on climate impacts of Arctic air pollutants has mainly focused on present-day radiative forcings and impacts of changes in emissions of pollutants on Arctic temperature and precipitation trends are less well understood. There is low scientific confidence in the relationship between long-term changes in Arctic climate and emissions within and outside the Arctic. Relative contributions of individual long (CO_2, chlorofluorocarbons) and short-lived climate forcers (black carbon, sulphate, organic carbon, CH_4, O_3) to observed temperature trends in the Arctic are highly uncertain, which limits confidence in projections of future changes in Arctic climate (AMAP, 2015).

2.5 Societal perspectives for Arctic air pollution

Projections of Arctic air pollution must account for ever-changing human activities and evolving governance and socio-economic responses to these activities. Arctic air pollution is both an extra-Arctic and intra-Arctic phenomenon, since its sources are both local and from import from lower latitudes. Therefore, understanding these activities and responses must be addressed at global and regional scales, and also take into account variations on year-by-year timescales. For example, in Russia, flaring of petroleum gas decreased from 40–50 billion cubic meters (bcm) in 2007 (accounting one third of global flaring volume) to about 30 bcm in 2011 (e.g. Røland, 2010; EBRD, 2013). This relatively successful, but unintended action, was caused by Russian electricity reform in 2008 that encouraged oil companies to utilize unburned gas in small-scale electricity power plants for the needs of oil drilling (EBRD, 2013). Further success in flaring reduction lies within deliberative Russian energy and environmental policies. Emission scenarios provide a means to estimate how likely socio-economic factors might drive future growth (Peters et al., 2011; Stephenson et al., 2013). These critical tools require interdisciplinary efforts to address how different scenarios will impact Arctic air quality, and which governance/regulatory structures are available to limit negative anthropogenic impacts on Arctic air quality, whether they are adequate and whether they have scientific basis.

There is substantial opportunity for partnership between Northern communities and atmospheric scientists to expand and exchange knowledge about Arctic air pollution. Arctic residents are important observers of changing air quality and other impacts of Arctic industrialization. Community-based observations of air quality could improve the seasonal and geographic coverage of sampling, identify factors leading to extreme events and increase the salience of scientific evidence about air pollution risks in northern communities. Such practices, including the development of small user friendly sensors, require carefully developed knowledge exchange frameworks to bridge differences between communities of practice (Fazey et al., 2013). New tools are emerging to support citizen inquiry into air quality, such as the US Environmental Protection Agency's Citizen Science Toolbox (http://www.epa.gov/heasd/airsensortoolbox/). Topics that could be explored using community-based monitoring and/or citizen science approaches include identifying the concerns of Arctic residents regarding air pollution and the observations that are meaningful to them and what they can learn from them. It is also important to investigate how residents can best make use of scientific information and work with scientists to co-produce knowledge on Arctic air pollution for greater societal relevance.

Understanding the multiple interactions between physical and social science issues related to Arctic air pollution requires social science expertise ranging from sustainability and adaptation studies to economics and political science. Arctic air pollution affects Arctic residents both directly in terms of their health, but also via ecosystems impacts, which affect their cultural continuation and food security (AMAP, 1998). Perceptions of risk due to air pollution may lead to complex adaptation responses, even when risks are not well defined by scientific evidence. Scientific assessments of risks compete with many other types of information in forming adaptation responses. Value demands by stakeholders for information fall into three broad categories: salience, credibility, and legitimacy (Cash and Buizer, 2005; Cash and Clark, 2001). It has been recognized that the provision of salient, credible and legitimate information relies upon engaging the perspectives of relevant stakeholders, ideally in sustained settings (Dilling and Lemos, 2011; Fazey et al., 2013).

Interdisciplinary research questions on the regional and local social impacts of Arctic air pollution include: 1) What are the impacts of diminishing air quality on the lives of those at northern high latitudes? 2) How do Arctic residents, decision-makers, and scientists perceive and weigh air pollution risks relative to other

risks, and how does this evaluation process shape decision-making? 3) Do northern community decision makers, as well as external actors making decisions that have an impact in the region (e.g. representatives of oil and gas companies, and state ministries), have the required information about impacts of air pollution on human and ecosystem health? 4) How might they respond to such information? Answering these questions will require involvement from anthropologists, sociologists, and political scientists who employ qualitative and quantitative methods to develop new understanding of these issues.

3. Cross-cutting perspectives and opportunities

A major PACES goal is to foster and drive new international research efforts where progress will not be possible without a collaborative interdisciplinary approach. It aims to capitalise on existing capabilities, covering topical specialities and extensive geographies with multiple groups contributing unique expertise, tools and capacities. PACES also aims to foster and build links with Arctic programmes that do not currently have a strong focus on air pollution and its impacts. This section highlights opportunities for collaboration, which PACES seeks to exploit and drive forward.

3.1 Developing a trans-disciplinary approach

Understanding of the mechanisms and processes controlling atmospheric pollution in the Arctic also requires more coordinated observations of Arctic atmosphere dynamical processes, ocean-ice-atmosphere interactions, atmospheric boundary layer processes, in addition to observations of atmospheric composition. PACES will facilitate coordination of atmospheric pollution observations and modelling studies with other international Arctic meteorology, climate and oceanography research programmes. These include the WMO World Weather Research Programme (WWRP) Polar Prediction Project (PPP). The PPP (Polar Prediction Project, 2013; http://polarprediction.net) promotes cooperative international research in order to enable the development of improved weather and environmental prediction services for the Polar Regions on timescales from hours to seasonal. The forthcoming Year of Polar Prediction, 2017–19 (YOPP; Year of Polar Prediction, 2014), organized through PPP, will cover an extended period of coordinated intensive observational and modelling activities in order to improve polar prediction capabilities on a wide range of time scales in the Arctic. PACES will encourage the coordination of new observational capabilities and field campaigns with YOPP-related activities (e.g. Multidisciplinary drifting Observatory for the Study of Arctic Climate; MOSAiC) and also via participation in Arctic Observing Summits (AOS) led by International Study of Arctic Change (ISAC) and contributing to the Sustaining Arctic Observing Networks (SAON) initiative of the Arctic Council (and co-led by the IASC).

An important aspect of PACES will be to work with scientists focused on natural biogeochemistry in the Arctic, and to establish efforts to explore links between natural cycles and pollution, building on past work carried out as part of the Air-Ice Chemical Interactions (AICI) and Ocean-Atmosphere-Sea Ice-Snowpack (OASIS) activities under IGAC. PACES actions will also be developed in close cooperation with other international activities such as the Arctic Council Arctic Monitoring and Assessment Programme (AMAP) and its several relevant expert groups, and the Task Force on Hemispheric Transport of Air Pollution (TF-HTAP), which carry out assessments of pollution impacts on climate, ecosystems and health. On a more regional level, synergies will be developed with national and regional initiatives. For example, PACES is developing links via focused actions involving Russian researchers in collaboration with the PEEX (Pan Eurasian Experiment; https://www.atm.helsinki.fi/peex/; Lappalainen et al., 2014; Kulmala et al., 2015) research infrastructure program since this is a region where large gaps still exist in our knowledge about pollutant sources and their impacts (Kulmala et al., 2011). Additional coordination is possible between PACES and ongoing measurement efforts by the U.S. Department of Energy in Barrow, Alaska, the U.S. National Oceanic and Atmospheric Administration in Tiksi, Russia, and the Max Planck Institute in Zotino and Tomsk, Russia.

Existing long-term observatories also offer an opportunity for interdisciplinary questions to be addressed through already sustained and potentially expanded observations of the chemical and physical characteristics of the atmosphere as well as the cryosphere and terrestrial ecosystems. Collaborative platforms such as the World Meteorological Organization (WMO) WMO Global Atmosphere Watch (GAW) programme and IASOA are already exploring such Earth system science questions at long-term Arctic observatories through a coordinated atmospheric composition and surface flux observational network that could be leveraged for dry deposition studies, for example. GAW is a partnership involving the members of WMO, contributing networks and collaborating organizations and bodies (GAW, 2014). GAW focal areas are aerosols, greenhouse gases, selected reactive gases, ozone, UV radiation and atmospheric deposition. Having been established for 26 years, GAW provides the longest time-series of high quality observations of atmospheric composition and dynamics in the Arctic.

Community based monitoring (CBM) and Arctic citizen science are highly promising means by which observational "networks" can expand into under-sampled regions and seasons, and communities can become engaged in knowledge exchange frameworks with atmospheric scientists. CBM programmes exist across the

circumpolar region, although formal monitoring programs are most developed in North America (Johnson et al., 2015). While few CBM programmes currently engage community members in air pollution monitoring, several emerging programs have significant potential to contribute to PACES. Aleut International Association (AIA) is one initiative that is making progress on a circum-Arctic community-based monitoring initiative for black carbon. This emerging initiative presents a strong opportunity for collaboration that should be developed further. The Atlas of Community-Based Monitoring in a Changing Arctic (http://www.arcticcbm.org), a meta-database led by the Inuit Circumpolar Council (ICC), is another effort to support network building among CBM programs. Additionally, regional organizations in Canada and Alaska have focused on developing CBM programs that could be expanded to incorporate air pollution monitoring aspects. The Local Environmental Observer (LEO) program run by the Alaska Native Tribal Health Consortium and the Inuvialuit Settlement Region Community-Based Monitoring Program (ISR-CBMP) are two examples. On a more international level PACES may also contribute to the cross-disciplinary ArcticStar initiative being developed under Future Earth which aims to bring together groups working on natural and social sciences in order to develop a plan for solution-oriented, trans-disciplinary approaches to Arctic research.

3.2 Field missions and long-term monitoring

The PACES initiative will work to develop coordinated international field missions that address the science challenges detailed in Section 2. The remoteness of the Arctic and the particular need for vertical information on pollutants mean that airborne and ground-based *in-situ* measurements will necessarily play an important role. The goals of these missions will be 1) to challenge model performance regarding long-range transport, vertical distribution, deposition, and impacts on climate 2) to improve understanding of chemical and physical processes, especially deposition, 3) to better evaluate local sources, both natural and anthropogenic, and their impacts on Arctic air quality and ecosystems, and 4) to improve understanding of the social and economic interactions between Arctic air pollution and local populations and ecosystems. Any future measurement programs will need to collaborate closely with local residents, not only to understand the societal interactions with air pollution, but also to engage these important stakeholders in understanding the science and participating in policy development. PACES will develop and exploit new understanding of model process deficiencies, and use these to target new observational mission planning from the outset.

Improved understanding of relevant processes affecting pollutants and their impacts will require different tailored approaches to experimental design. In particular, a future field program is likely to involve a more sustained effort comprised of repeated flight profiles in multiple seasons and years. This could build on past activities such as the pan-Arctic survey flight series undertaken by the Alfred Wegener Institute from 2009 to 2015 (Herber et al., 2012). Vertically-resolved measurements over extended time periods are needed to better constrain model simulations of long-range transport to the Arctic. Such observations would help determine if there are model deficiencies associated with emissions, advective transport, and removal processes, or with vertical transport and deposition within the Arctic. The arrival of scientifically useful small unmanned aerial vehicles (UAVs) may herald new approaches to making long-term, vertically resolved in situ measurements of select parameters and improving understanding of the importance of both local and transported pollution to Arctic air quality.

Commercial aircraft serving Arctic communities are often small, so there is limited opportunity for installing comprehensive scientific payloads. However, with the miniaturization of sensors designed for UAVs or ultra-light aircraft (ULA) platforms, there is a potential for adding a small complement of select measurements to commercial aircraft that fly regularly to Arctic communities. Additionally, with the growth of the Arctic's political and military importance, there are potential opportunities to make regular measurements on military aircraft. For example, NOAA in the U.S. has a program to measure greenhouse gases on a Coast Guard aircraft that makes regular survey flights over the Alaskan Arctic (Karion et al., 2013). Equipping the commercial transport fleet with pollutant monitoring capabilities also offers excellent opportunity for regular sampling. Cargo ships are used for long-term monitoring of long-lived greenhouse gases in the atmosphere over global oceans by making use of the Ship of Opportunity Program (http://www.aoml.noaa.gov/phod/soop/index.php), a component of the Global Ocean Observing System.

In addition to regular flights, focused airborne campaigns, are also needed to better characterize local pollution sources especially in Eurasia (Russia) where knowledge is poor. Lagrangian experiments, where air masses are sampled multiple times during transport, have previously been very useful for the study of pollutant processing (e.g. at mid-latitudes, Fehsenfeld et al., 2006) and could be applied to the study of pollutant processing and deposition processes during transport to the Arctic, or processing of local pollutants under Arctic conditions. Continued observations of multi-decadal trends in pollutant concentrations in the atmosphere and in ice cores greatly improve our understanding of changes in Arctic composition and the origins of Arctic pollutants and need to be extended to include new compounds and sites. It will be especially important to engage and collaborate with Russian scientists and stakeholders to ensure that measurements are made in this data-poor region of the Arctic. Such efforts could build on the on-going series of flight

campaigns in Russia conducted by the French-Russian YAK-AEROSIB program (Paris et al., 2010) or the recently established Norwegian-Russian project (SLICFONIA) to study black carbon and methane in high-latitude Russia.

Satellite data provide very useful information about the spatial and temporal coverage of a limited number of trace gases, as well as certain aerosol properties, in regions where in-situ sampling is limited (e.g. see Jacob et al., 2010, Pommier et al., 2010). These data can be used to evaluate models, often in conjunction with in-situ data that provide much needed vertical information (e.g. Sodemann et al., 2011). CALIOP has provided such vertical profile data on aerosol properties such as backscatter up to 80N over a number of years allowing examination of seasonal and inter-annual aerosol variability in the Arctic (Di Pierro et al., 2013). Emmons et al., (2015) used satellite observations to demonstrate consistent bias in simulated NO_2 among several models in high latitude regions dominated by fire emissions, where in-situ sampling was not available. Satellite observations of CO can also be used to probe large-scale pollution outflow from continental regions and latitudinal gradients (e.g. Monks et al., 2015).

While collaborations between modellers and groups making campaign observations are traditionally well established, opportunities for collaboration between modellers and long-term monitoring observations are not always fully exploited due, in some cases, to data products which are not readily usable or have poorly documented limitations on their use (Starkweather et al., 2012; Starkweather and Uttal, 2015). PACES aims to promote closer collaboration between modelling groups and the new WMO GAW science advisory group of the GAW near real-time data modelling applications. Investigator-driven working groups attached to global networks like NDACC and regional "networks of networks" like IASOA already provide a platform for cross-site collaboration and data product development among observational experts. Introducing more modellers into these discussions provides an opportunity to increase the relevance and accessibility of long-term observations to model evaluation and development.

3.3 Towards increased model complexity – what opportunities are offered by new generation Earth system models?

Scientific assessments of the impacts of air quality and climate on communities and economic development in the Arctic depend on accurate modelling capabilities for the region. Currently, global and hemispheric models are the primary tools available for investigating responses of Arctic air quality and climate to natural and anthropogenic influences. Chemistry-Climate Models (CCMs), and similar models, are based on physical principles and they reproduce important aspects of observed global and regional air pollution (Dameris and Jöckel, 2013; AMAP, 2015). In recent years, many global climate models have been extended into Earth System Models (ESMs), by including interactions between the physical climate system and ecosystems. ESMs were successfully used in scientific assessments of the past and future response of the global climate system to external forcing, in which biogeochemical feedbacks play an important role (Flato et al., 2013). It is anticipated that research on Arctic pollution and climate will also increasingly rely on simulations with ESMs owing to the need to account for emissions from the marine and terrestrial biosphere, and feedbacks driven by the effects of climate change on the biosphere, such as climate-fire feedbacks (de Groot et al., 2013). However, ESMs are currently limited by their large internal variability when it comes to e.g. assessment of contributions of specific emission sources to climate impacts and the evaluation of the effectiveness of mitigation strategies. In addition, an increasing focus on the need for information at the regional scale will require regional modelling and downscaled climate scenarios.

In the Arctic, models display varied skill in simulating enhancements in pollutants at the surface (Shindell et al., 2008; Breider et al., 2014; Eckhardt et al., 2015; Monks et al., 2015) and through the vertical profile, particularly of aerosol components (AMAP, 2015; Eckhardt et al., 2015), and ozone precursors, such as reactive nitrogen compounds (Emmons et al., 2015; Arnold et al., 2015). Poor model skill in the Arctic implies a lack of confidence in prediction of high latitude air quality and climate response to northern hemisphere emission changes. This lack of skill results from poorly-constrained and inter-dependent processes outlined in Section 2. In the case of aerosols, this includes uncertainties in emissions, continental export at mid-latitude source regions, wet deposition during uplift near source regions or at the Arctic front (Browse et al., 2012; Fig. 2), details of model chemistry and aerosol-cloud interactions.

Continued model development activities by climate and air pollution science communities can be expected to lead to improved global modelling capabilities that will also benefit Arctic pollution and climate research, especially with regard to a necessary integration of pollutant processes with biogeochemical cycles in ESMs and increased resolution of regional climate processes. A suite of modelling tools of varying complexity is likely required to address the diverse issues discussed in Section 2. New observations in the Arctic are of paramount importance for simulations of Arctic pollutant processes that are either missing or not adequately represented in models, including the transport of pollutants to the Arctic and atmospheric processes governing pollutant vertical profiles. In addition, successful model application to studies of pollution impacts on Arctic communities and economic activities requires that the development of physical models is informed by social sciences.

4. Moving forward and conclusions

This paper has outlined how Arctic air pollution plays a central role in a complex set of environmental and societal issues, driven by interactions with regional and global climate, and socio-economic responses to Arctic warming. Important goals of PACES are the provision of robust scientific knowledge to policy-makers, and engagement with local communities, to present findings and explore risks and benefits to Arctic communities while at the same time examining sustainable pathways in a changing environment. The PACES initiative aims to tackle key gaps in our understanding across the range of issues outlined in Section 2 by developing new focused actions over the next 10 years (2016–2026), and creating new collaborative efforts between observational and modelling groups, social science researchers, and local Arctic communities.

In particular, we make the following key overarching recommendations:

1. Advancement in Arctic air pollution research should take a trans-disciplinary approach. This should exploit collaborative platforms for observations across linked aspects of the Earth system (atmosphere, cryosphere, ocean, land surface, society), enable community-based monitoring approaches, and take account of societal and economic drivers and responses to Arctic change impacting climate, air quality and ecosystems. It should be carried in a collaborative framework linking to existing and planned programs.
2. Improved process understanding requires development of further capacity in terms of regular long-term monitoring and intensive field observations, both at the surface and throughout the troposphere (in-situ and satellite). A Lagrangian observational framework from source (mid-latitude) to receptor (Arctic) regions would allow characterization of pollutant processing during transport both en-route to the Arctic and within the Arctic leading to improved attribution of sources of Arctic air pollution and their effects. Commercial transport platforms (shipping and aircraft) and new technologies (e.g. unmanned aerial vehicles) should be exploited to improve sampling coverage and frequency especially in regions where data are sparse or completely lacking. Community-based monitoring would enhance spatial and temporal coverage of surface observations while engaging local populations in science and decision-making. Russian Siberia is a region where new observations and collaborative efforts are needed to improve data coverage.
3. Improved predictive capability is needed across a range of scales to diagnose wider impacts of Arctic air pollution on regional and global climate and the Earth system, and on local air quality and ecosystems. Results from current state-of-the-art models should be used to target uncertain processes (e.g. wet deposition during transport to and within the Arctic) and regions to be probed by observations, and observation activities should be collaborative between traditional disciplinary groups and with local communities. Modelling efforts should also exploit new capabilities in Earth system modelling, particularly with regard to interactions between atmospheric composition/climate and the biosphere/ocean. Improved regional-scale modelling is required to understand how changes in local sources of air pollutants will modify human and ecosystem exposure to harmful pollutant levels. New observations should be used to critically evaluate model processes across a range of scales to improve predictive capability. Quantification of impacts on societies and economic response requires that social science and economic drivers inform the development of physical models.

These recommended efforts should build on, and link to, existing programmes, ranging from networks making highly valuable observations to initiatives tackling closely related issues in different regions of the Arctic.

The broad recommendations highlighted above should lead to a significant international programme, modelled on the successful 2008 IPY/POLARCAT effort, in the 2018–2020 time frame. Longer-term support for enhanced monitoring efforts, as well as for sustained, two-way engagement with Arctic communities and corporate stakeholders, must be part of this programme. Following initial workshops, PACES working groups are now being formed to take forward the key recommendations and to develop a PACES implementation plan. For more information on PACES see http://www.igacproject.org/PACES.

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Notes

1. Arctic air pollution workshop: Boulder, United States, 2-4 February 2015 (see http://www.esrl.noaa.gov/psd/iasoa/node/131)

Contributions

- Contributed to conception and design: SRA, KSL, CAB, JLT, SMS, KvS, SS, AS, MM.
- Contributed to review of issues and summary of findings: All authors.
- Wrote the manuscript: All authors.
- Contributed revisions to the manuscript: SRA, KSL, CAB, JLT, JED.
- Approved the submitted version for publication: All authors.

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Competing interests

The authors declare no competing interests.