Alaskan Layered Pollution And Chemical Analysis (ALPACA) White Paper

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Plain Language Summary:

Wintertime air pollution is a serious problem for urban areas and areas affected by industrial activities in the Arctic and sub-Arctic regions. The cause of this pollution is local emissions coupled with poor dispersion caused by strong temperature inversions, yet many uncertainties remain in understanding the physical and chemical processes driving this pollution. In the case of Fairbanks, Alaska, fine particulate matter ($PM_{2.5}$) concentrations exceed health-based pollution thresholds, requiring action to mitigate this problem. This particulate matter comes from a mix of sources and is likely enhanced by condensation of gaseous precursors, especially under these low temperature conditions and through chemical gas-to-particle conversion processes. Prior Fairbanks studies have shown that wood combustion is the largest single source, but other sources also contribute. Progress on this problem requires research into source apportionment and a better understanding of chemical processing of pollution during cold and dark conditions. The role of meteorology, particularly surface-based inversions also needs study. Therefore, we propose the ALPACA study, which is organized under the international PACES initiative and in coordination with the Pan Eurasian Experiment (PEEX). ALPACA investigates emissions and chemical and meteorological influences on pollution in Fairbanks. In addition to measurements, modeling, laboratory, and health effect studies, outreach efforts are planned. Involvement of citizens is highly desired. The intended outcomes of ALPACA are improved mechanistic understanding of pollution behavior under cold and dark conditions, improved public understanding of the problem, and better-informed pollution mitigation strategies. The ALPACA study will provide a benchmark to inform sister studies at high northern latitudes and assist in understanding air pollution impacts of increasing development in the rapidly changing Arctic. ALPACA will also provide a colder/darker comparison for mid-latitude wintertime pollution studies.

1 Introduction:

1.1 Introduction to high-latitude air pollution

The Arctic is undergoing unprecedented changes such as the highly visible decline in summer sea ice as well as rapidly increasing surface temperatures due to global warming. In addition to these climate changes, the Arctic is being developed for natural resources and transportation through the region is increasing in response to the more hospitable conditions. These coupled changes affect air pollution in the Arctic, which occurs both in the remote Arctic and on local scales in Arctic cities and near industrial activities. The broader Arctic regional changes are due to increasing emissions, especially in northern mid-latitudes of greenhouse gases and substances such as black carbon (BC) aerosols that warm the atmosphere. While Arctic warming is primarily due to increasing CO_2 and cryosphere-atmosphere positive feedback loops (Arctic amplification), there is also an important contribution from so-called short-lived climate pollutants (SCLPs): black carbon, ozone and methane (AMAP, 2015). Black carbon, and other absorbing aerosols, can also be deposited on snow and ice leading to reductions in surface albedo. Reductions in sulfate aerosols that cool the atmosphere are also responsible for additional warming (e.g. Breider et al., 2017). The Arctic has been impacted by long-range transport of aerosols, ozone and their precursors from mid-latitudes since the industrial revolution leading to the formation of "Arctic haze" each winter and early spring when loss processes such as wet deposition are slower (e.g. Barrie, 1986). Pollution from boreal forest fires may affect regional air quality, deposition to ecosystems in the summer and contribute to radiative effects even if faster loss processes at this time of year tend to reduce the impacts of such events. The recent review by Willis et al. (2018) describes the most up-to-date assessment of remote Arctic aerosol abundance and composition. Aerosols also affect cloud properties and radiative effects but these aerosol-cloud indirect effects remain highly uncertain (AMAP, 2015).

While transport of pollutants from Asia, Europe and North American emission regions as well as boreal fires are the main source of air pollutants in the remote Arctic, local emissions cause significant air quality issues in cities and near industrial facilities. Such emissions impact local and regional air quality (Schmale et al., 2018) and may also impact ecosystems, for example via nitrate deposition (Hodson et al., 2010) and regional climate (Marelle et al., 2018). These Arctic emissions can also be transported downwind to contribute to Arctic aerosol loadings. Other downwind influences are BC emissions from gas flaring in northern Russia, enhanced wintertime domestic combustion (Stohl et al., 2013), sulfur from metal smelting operations (Prank et al., 2010) or Arctic shipping (e.g. Stohl et al., 2013; Marelle et al., 2016). Favorable socio-economic drivers (Larsen and Fondahl, 2015), combined with the accelerated opening of the warming Arctic region, are expected to promote large-scale industrial developments including shipping, resource extraction, and are likely to worsen local pollution. Increasing impacts of human activities such as increasing pollutant emissions in the Arctic are highlighted as a major issue in the recent Arctic Science Ministerial summary (Arctic Science Ministerial, 2018).

1.2 The air Pollution in the Arctic: Climate, Environment, and Societies (PACES) initiative

The air Pollution in the Arctic: Climate, Environment and Societies (PACES, https://pacesproject.org) initiative has been developed as a bottom-up community activity to address deficiencies in our understanding of sources, processing and fate of Arctic air pollution. Despite much progress in developing knowledge from recent initiatives, such as the POLARCAT-International Polar Year campaigns of 2008 (Jacob et al., 2010; Brock et al., 2011; Law et al., 2014), there remain important knowledge gaps, which have implications for our confidence in predicting future Arctic climate response to remote and local emission changes, potential effects of increases in local Arctic pollution sources, knowledge of pollution interactions with natural cycles, and societal and ecosystem impacts of Arctic air pollution. The overarching aim of PACES is to create new collaborative efforts between observational and modeling groups, social and human science researchers and local Arctic communities to address these issues. PACES aims to provide international community leadership in the coordination of large international collaborative efforts, such as coordinated field experiments and model comparison and evaluation exercises. The major scientific foci and aims of PACES are given in Arnold et al. (2016). Two working groups (WGs) have been established under PACES. The first is tasked with developing approaches to reduce model uncertainties regarding the processing and impacts of remote and local Arctic pollution sources, with a focus on improved understanding of pollution transported from mid-latitudes to the Arctic (WG1). The second focuses on interactions between Arctic air pollution and Arctic societies, and local sources of Arctic air pollution (WG2). The ALPACA study has been developed under the umbrella of PACES WG2.

1.2.1 Arctic Pollution and Societies (PACES WG2)

PACES WG2 supports observational studies guided by community concerns, investigation of local air quality in Arctic communities, and feedbacks between economic development, air pollution and environmental change in the Arctic. For WG2 to conduct salient research for and with Arctic citizens, the use of a transdisciplinary study design is key (Schmale et al., 2014). Here, transdisciplinarity means that scientists (social, human and natural) together with local citizens, regulatory agencies and other stakeholders co-develop knowledge to create options for mitigation. This approach fosters common ownership of solutions and a higher likelihood of implementation. Fairbanks, Alaska, USA, has been identified as first city for a major international field study as described in this whitepaper. Lessons from Fairbanks are envisaged to be applied to a city in the Eastern Arctic via a “twin city approach”.

1.2.2 Northern Urbanization and other efforts complementary to ALPACA

Urbanization, a global phenomenon, is accelerating, resulting in a rapid increase in the number of cities and urban complexes, many of them in Northern latitudes. Such a trend causes transformation in the geosphere, biosphere, atmosphere, and hydrosphere, which affects the environment over both short and long timescales. Northern cities represent a broad and highly dynamic interface between Earth Components (atmosphere, land, water, etc.) and societal factors. High latitude cities are some of the most vulnerable to the climate change, which is the most pronounced in the Arctic, and have direct and indirect impacts on the local atmospheric composition, climate, hydrology and ecology. Most previous studies of urban pollution and climate have focused on lower-latitude cities, yet Northern cities have extra challenges with

pollution such as the effects of wintertime conditions like temperature inversions, meteorology, and urban heat islands. These complicating conditions require deeper and more focused study, particularly in a changing climate. To address this problem, there are several ongoing initiatives which can provide a good platform for complex northern urbanization studies for different Arctic cities. Some of these efforts are:

- Pan-Eurasian EXperiment (PEEX) WG: Northern Urbanization: Environmental challenges and their impact on urban societies (currently focusing on urban heat island and air pollution studies), https://www.atm.helsinki.fi/peex
- UHIARC: Urban heat island observation network in Arctic cities (Apatity, Vorkuta, Nadym, Urengoy, Murmansk, Norilsk) http://www.geogr.msu.ru/science/grants/
- HIARC: Anthropogenic Heat Islands in the Arctic - Windows to the Future of the Regional Climates, Ecosystems and Society – https://www.nersc.no/project/hiarc
- TRAnsferable Knowledge and Technologies for high-resolution environmental impact assessment and management (TRAKT-2018) https://www.nersc.no/project/trakt-2018
- WMO Global Atmosphere Watch (GAW) Urban Research Meteorology and Environment (GURME) project, http://www.wmo.int/pages/prog/arep/gaw/urban.html
- WMO Integrated Urban Weather, Water, Environment and Climate Services & Multi-Hazard Early Warning Systems, http://www.wmo.int/pages/prog/arep/gaw/documents/UrbanIntegratedServicesPart1aConceptandMethodologyEC-70.pdf
- Year of Polar Prediction (YOPP) and WMO WWRP Polar Prediction Project (PPP) – https://www.polarprediction.net

Under the umbrella of PACES, the ALPACA study will be linked to the above efforts through its study design, stakeholder involvement methodology and scientific findings. PACES is developing the "twin city approach" through which lessons learned from ALPACA and the North American Arctic can be used to provide insight into Arctic Eurasian urban air pollution. Potential focus cities in addition to Fairbanks include Tromsø, Murmansk and Norilsk. The ultimate objective is to develop an Arctic urban air pollution understanding to inform sustainable development policies for an Arctic as it undergoes rapid urbanization and economic development.

1.2.3 The Pan-Eurasian Experiment (PEEX) program

Pan-Eurasian Experiment (PEEX) program is a large scale, bottom-up research collaboration network in the Arctic-boreal context especially with European, Russian and Chinese organizations (Kulmala et al., 2015; Lappalainen et al., 2016), https://www.atm.helsinki.fi/peex. The PEEX science plan (https://www.atm.helsinki.fi/peex/images/PEEX_Science_Plan.pdf) is available and provides a multidisciplinary approach to resolve the interconnected grand challenges, such as climate change and air quality, in the Arctic and boreal environment. The tools include 1) joint science plan identifying the gaps in knowledge, 2) capacity building in observation infrastructures, 3) supporting science-based decision making and 3) capacity building via education and training. For ALPACA, the PEEX program provides connections to study air quality in urban environment in the different Arctic cities with the twin-cities approach.

1.3 Prior wintertime air quality studies

Pollution in the Arctic is most problematic and preventable during the winter when pollution dispersion rates are low and local sources related to heating and industry are high. Summertime forest fires are also an issue for the Arctic, but these are difficult to control, so the ALPACA study focuses on wintertime air quality. There have been several previous research efforts aimed at understanding wintertime atmospheric chemistry within the last decade, including a number of medium- to large-scale campaigns in the U.S, as well as a limited number in Europe and China. Though the following list is not exhaustive, it may be representative of studies that have some scientific overlap with the goals of ALPACA. Nitrogen Aerosol Composition and Halogens on a Tall Tower (NACHTT) took place north of Denver, Colorado during January and February 2011 and featured measurements from a movable carriage on a 300 m research tower that allowed detailed investigation of the vertically resolved chemical composition in an urban influenced winter boundary layer (Brown et al., 2013; Gilman et al., 2013; Öztürk et al., 2013; Riedel et al., 2013; VandenBoer et al., 2013; Wagner et al., 2013; Kim et al., 2014; Jordan et al., 2015). The Uintah Basin Winter Ozone Studies (UBWOS) followed the discovery of unusual, severe winter (but not summer) ozone pollution in two sparsely populated oil and gas producing mountain basins of the western U.S. (Schnell et al., 2009). Ground based field intensives in the Uintah Basin of Northeast Utah in early 2012, 2013 and 2014 characterized the meteorology, emissions and atmospheric chemistry leading to winter ozone, demonstrating that photochemistry can be rapid in winter under specific levels of common pollutants, particularly volatile organic compounds (VOCs) (Edwards et al., 2013, 2014; Helmig et al., 2014; Oltmans et al., 2014; Warneke et al., 2014; Ahmadov et al., 2015; Lee et al., 2015; Veres et al., 2015; Wild et al., 2016; Yuan et al., 2016; Schnell et al., 2016; Koss et al., 2017). The Clean Air for London (ClearfLo) project (Bohnenstengel et al., 2015) established new understanding regarding the influence of urban heat fluxes and wintertime boundary layer stability, as well as the importance of interactions between low levels of photochemical activity and diurnal cycles in mixed layer height and emissions, in controlling primary pollutant concentrations in wintertime London. Measurements during ClearfLo showed that vehicle emissions, solid fuel combustion, and cooking dominate organic aerosol in urban central London in winter (Liu et al., 2015; Xu et al., 2016). Similarly, observations have shown that solid fuel (wood) burning makes important contributions to organic aerosol during winter months in many regions of Europe, including the Po Valley region of northern Italy (Pietrogrande et al., 2015), a residential district of a city in the Czech Republic (Hovorka et al., 2015), and Helsinki, Finland (Pirjola et al., 2017). There have been a number of field experiments studying wintertime urban pollution in China. Measurements in wintertime Beijing have included detailed measurements of aerosol size distribution (Wu et al., 2017; Du et al., 2018), and gas-phase chemistry, with a focus on processes controlling HONO (Wang et al., 2017) and enhanced sulfate formation during severe pollution episodes (Wang et al., 2014). Observations from the North China Plain under a severe pollution episode have shown that wintertime PM_1 mass is dominated by organics, with large sources from coal and biomass combustion (Li et al., 2017).

Airborne observations have also been used to understand processes controlling regional-scale wintertime atmospheric composition over the US. The Wintertime INvestigation of Transport, Emissions and Reactivity (WINTER) surveyed the Eastern U.S. and offshore areas during February and March of 2015 using the NSF C-130 aircraft. This was the first study in the U.S. to bring detailed chemical composition information using an instrumented aircraft to the winter

season. Flights characterized regions that include the Northeast U.S urban corridor, the Ohio River Valley, urban areas of the Southeast U.S., and transport of polluted airmasses over the Atlantic Ocean. Scientific analyses to date have included emissions and wintertime chemistry from coal fired power plants (Fibiger et al., 2018; Lee et al., 2018) factors governing winter aerosol composition, chemistry, pH and trends (Guo et al., 2016; Sullivan et al., 2017; Schroder et al., 2018; Shah et al., 2018), evaluation of winter nitrogen oxide chemistry and emissions (Fibiger et al., 2018; Jaeglé et al., 2018; Kenagy et al., 2018; McDuffie et al., 2018; Salmon et al., 2018) and understanding of halogen partitioning and sources (Haskins et al., 2018). The recent Utah Winter Fine Particulate Study (UWFPS) utilized a light aircraft, the NOAA Twin Otter, to investigate the chemical and meteorological structure of polluted winter boundary layers in the region in and around Salt Lake City, Utah, which is subject to some of the most severe winter particulate matter pollution in the U.S. Results from this 2017 study are currently being prepared for publication.

In addition to in-situ chemical and aerosol analysis studies, a number of studies have used meteorological reanalysis, in-situ or remote sensing datasets to demonstrate a key role for synoptic forcing on surface-based inversion layers and on pollutant distributions in winter. The ERA-Interim dataset has been used to demonstrate forcing of Eastern European inversions in winter by the Russian high pressure system (Palarz et al., 2018). Relationships between meteorological variability and both satellite and in-situ observations of air pollutants have also been used to reveal strong synoptic scale control on wintertime pollutant variability in northern European urban regions (Pope et al., 2014; Grundstrom et al., 2015). The influences of regional and local circulation patterns on severe wintertime pollution episodes in China have also been the subject of several recent studies (Bei et al., 2016, 2017; Han et al., 2018). In addition, the formation of Arctic haze depends on the transport of trace gases and aerosols and their precursors from mid-latitudes and local within or near Arctic sources such as domestic wood combustion (Stohl et al., 2013) or metal smelting (Prank et al., 2010). The contribution of local sources to Arctic haze remains uncertain but a recent study by Yang et al. (2018) estimated that more than 80% of surface sulfate originates from within or near Arctic emissions.

1.4 Air pollution under dark, cold winter conditions: The example of Fairbanks, Alaska

Northern cities and industrial facilities can have serious air quality problems due to local emissions and poor dispersion of this pollution that is caused by severe meteorological inversions. Fairbanks, Alaska is an example of a small city with severe wintertime air quality problems. Fairbanks has ~32,000 residents within the city limits and ~100,000 including surrounding residential and remote areas. Major emission sources are domestic heating (typically using oil, wood, coal, gas) power generation plants (fueled by coal and oil), transportation, and light industry. The climate is continental with very cold winters (-40°C air temperatures occur most winters), light winds, and strong meteorological inversions that can exceed 0.5°C / m in the lowest ~10m AGL (Benson, 1969; Mayfield et al., 2013; Fochesatto et al., 2015). At cold temperatures, domestic heating demands are large leading to a large pollution source near ground level. These factors combine to cause the highest $PM_{2.5}$ concentrations in the United States, exceeding the US EPA regulatory limit of 35 microgram m^{-3} on a 24-hour basis for weeks of the winter.

A number of studies have examined the relationship between meteorological conditions and these pollution episodes (Tran and Mölders, 2011, 2012; Leelasakultum et al., 2012; Mölders et al., 2012). These strong inversions also caused trapping of carbon monoxide (CO), and Fairbanks used to violate US EPA CO standards (National Research Council, 2002) before automotive emission controls improved to the point where CO violations stopped happening in the early 2000s.

Fine particle ($PM_{2.5}$) chemical composition has been measured in Fairbanks and the nearby residential community of North Pole, Alaska. Figure 1 shows that organic carbon (OC) species dominate the particulate composition, with lesser amounts of inorganic ions (sulfate, ammonium, nitrate) and elemental carbon (EC). The composition differs between Fairbanks and North Pole, with higher OC and lower inorganics at the residential community of North Pole (Ward et al., 2012; Nattinger, 2016).

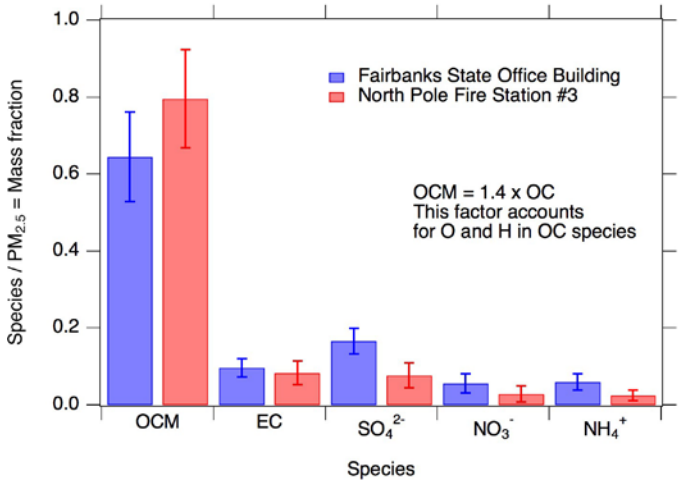

Figure 1: Mass fractional composition of $PM_{2.5}$ in Fairbank. The species are OCM = organic carbon mass, including a 1.4x multiplier to scale up organic carbon (OC) mass to total organic species mass, EC = elemental carbon. Data are from 2012-2014.

Source apportionment analysis points to domestic wood combustion as the largest single source of the pollution, estimated at 60-80% (higher at North Pole than Fairbanks) by chemical mass balance methods (Ward et al., 2012) and 40% in Fairbanks by positive matrix factorization, PMF, (Wang and Hopke, 2014) and 52% in a more recent PMF analysis that includes a secondary wood smoke component (Kotchenruther, 2016). Although sulfate is a significant component of $PM_{2.5}$, it is not clear what chemical mechanisms, if any, are responsible for atmospheric sulfur oxidation. Therefore, a number of modeling and observational studies have examined sulfur chemistry in Fairbanks (Leelasakultum et al., 2012; Shakya and Peltier, 2013; Joyce et al., 2014) with the general result that models cannot reproduce $PM_{2.5}$ sulfate observations, which can exceed 10 microgram $SO_4^{=}$ m^{-3}. Nocturnal nitrogen chemistry that could contribute to aerosol nitrate and deposition of nitrate to snowpack has also been studied (Ayers and Simpson, 2006; Huff et al., 2011). A few studies have connected air quality with health effects (Kossover, 2010) and investigated the effectiveness of technologies in remediating residential air quality (Ward et al., 2017). Although Fairbanks is a particularly extreme example of wintertime pollution, other Arctic cities suffer similar poor air quality (Mölders and Kramm, 2018), likely as a result of similar meteorology, emissions, and chemistry.

1.5 Atmospheric pollution problems at other high latitude locations

Future efforts under PACES intend to study other high latitude locations, so it is useful to consider pollution in these regions for the context of the ALPACA study. In general, the atmospheric environment over remote areas of Siberia, Northern Asia, the Barents region, and the Kola Peninsula are relatively unpolluted outside of hotspots compared with other surrounding regions of Asia and Eastern Europe (Baklanov et al., 2013). However, air pollution from industrial centers in these regions pose significant environmental threats. Siberian ecosystems have begun to show stress from the accumulation of deposited pollution that came from cities and industrial plants. Urban air quality in several Siberian cities (*e.g.*, Norilsk, Barnaul, Novokuznetsk) is some of the worst among the Russian and European cities. Stable atmospheric stratification and temperature inversions are predominant weather conditions for more than half a year. This contributes to accumulation of pollutants at the surface, where ecosystems function and people live. In addition to the severe climatic conditions, man-made impacts on the environment in industrial areas and large cities have intensified. The impacts manifest themselves in pollution of environment, change of the land use, hydrology, and hydrodynamic regimes of the atmosphere. Ultimately, these impacts feedback to people, affecting their health and well-being.

Sources of atmospheric pollution

Atmospheric pollution sources at high latitudes can be grouped into anthropogenic (industrial, transportation, combustion, etc.) and natural (biogenic emissions, wildfires, dust storms, sea salt aerosol particles, volcanic eruptions, pollen, etc.) emission sources. Among the natural emission sources, biomass, soils, and peatlands of Siberia contain one of the largest pools of terrestrial carbon (Groisman and Gutman, 2013). In Siberia under current climate change scenarios, some of the largest temperature increases have occurred and are expected to occur, the stored carbon has the potential to be released. Permafrost and fires can be natural sources of gases like carbon dioxide (CO_2), methane (CH_4), sulfur dioxide (SO_2), nitrogen oxides (NO_x), and ammonia (NH_3). These gases are locked inside frozen soil and can be released to the atmosphere gradually (*e.g.,* during the permafrost thaw with warming) or abruptly (*e.g.*, during wildfires). The frequency, duration, and severity of forest fires in the boreal forest zone, which is a source of SO_2 and sulfur aerosols in Siberia, appear to have increased in the last decades (Derome and Lukina, 2011), and there are no reasons to assume that this increase will not continue in the near future.

The Siberian environment also has sources of atmospheric particulates and their gaseous precursors such as SO_2, dimethylsulfide (DMS), and VOCs. Natural aerosol particles contain sulfates, which can be formed through gas-to-particle conversion of SO_2 derived from oxidation of DMS, and sea-salt aerosols, both originating from ice-free ocean and sea-water (Sokolik et al., 2011). Organic aerosols formed from biogenic emission of VOCs are also common. Local sources also include volcanoes (outside Siberia) and wildfires, either natural or human-related. Open biomass fires produce several important types of aerosols, including organic carbon (OC) and black carbon (BC).

Ammonia and NO_x emissions, derived from diffuse sources such as agriculture and vehicular traffic, are of more local concern in Siberia. Sulfur dioxide emissions are mainly associated with point sources such as power plants, the pulp and paper industry, non-ferrous metal smelters, and

oil and gas processing. The latter involves the emission of exhaust gases containing CO_2, NO_x, SO_x and volatile organic compounds (VOC).

Hotspots and urban air pollution

Observations show that atmospheric concentrations tend to be the highest around the major industrial centers, with their highest levels in the south-central part of Siberia (Irkutsk, Krasnoyarsk, and Novosibirsk regions). Two regions are most polluted: Tyumen oblast (West Siberia) and Krasnoyarsk Krai (East Siberia). In these areas, anthropogenic activities have contributed over two times the amount of pollution as compared to any other area in Siberia. The next most polluted areas are Irkutsk and Kemerovo oblasts located in East and West Siberia, respectively. The city of Norilsk is an extreme example of Arctic pollution. The nearby "Norilsk Nickel" metallurgical complex emitted about 1.9 million tons of SO_2 in 2015 (GGO, 2016). With the nickel factory located in the southern part of the city of Norilsk, the copper factory just to its north, and the Nadezhda metallurgical plant 12 km to the east, Norilsk was caught between heavy industry no matter which way the wind blew. After a 2007 visit, the Blacksmith Institute declared Norilsk one of the top 10 worst-polluted places in the world.

According to a Russian urban emission statistical analysis for 1998–2007 (GGO, 2009), the highest atmospheric emissions of particulate matter (PM) are observed in Siberian and Ural cities. For Barnaul, Krasnoyarsk, Novokuznetsk, Omsk and Chelyabinsk, they have exceeded 30 thousand tons yr^{-1}. For gaseous compounds, the maximum emissions included: SO_2 is emitted at a 30 tons yr^{-1} in Novokuznetsk, Novosibirsk, Omsk and Ufa, and nitrogen dioxide is emitted at more than 35 thousand tons yr^{-1} in Krasnoyarsk and Omsk. The density of PM emissions per capita is maximal in Novokuznetsk, and per unit area in Krasnoyarsk and Novokuznetsk. The highest densities of sulfur dioxide emissions per capita and per unit area are in Novokuznetsk and Omsk. One of the highest densities of nitrogen dioxide emissions per capita is observed in Novokuznetsk. In most of the above-mentioned Siberian cities, annual concentrations of several chemical compounds can exceed the national permissible threshold values. The integrated air pollution index (API) calculated for 5 major pollutants (CO, NO_2, NO, O_3, and formaldehyde), showed a high or very high level of air pollution in more than 71% of Siberian cities in 2007. Additionally, during the 1998-2007 period this pollution index increased by 26.5% (GGO, 2009).

Contamination of the surface air layer to a large extent depends on meteorological conditions. On average, the air pollution potential in Siberian cities is higher than in European cities of Russia due to a lower dispersion potential (Bezuglaya, 1999). In certain periods, when meteorological conditions trigger an accumulation of harmful substances in the surface layer, the pollution concentrations may drastically increase – leading to high pollution / smog episodes. Both summer and winter episodes with high concentrations occur often in most of industrial Siberian cities.

Although the anthropogenic emissions and environmental pollution in the Barents Euro-Arctic region are relatively low in comparison with other industrially developed areas of Europe, there are severe 'hot spots' mostly in the Russian part of the Barents region. The Kola Peninsula, despite the presence of areas with undisturbed nature in the eastern part of the region, is the most industrially developed and urbanized region in the Russian Arctic. The main polluters are the 'Severonikel' smelter in Monchegorsk in the central Kola Peninsula and the 'Pechenganickel' smelter in Nickel and Zapolyarnyi near the Russian-Norwegian border. For comparison, the

emissions of SO_2 only from the Nickel smelter are 5-6 times larger than the total Norwegian emissions (Sandanger et al., 2013).

The main impact from the smelters is exerted by the emissions, dominated by SO_2 and heavy metals, which result in high air concentrations and deposition to the surface (Baklanov et al., 2012). The impact is manifested as deterioration of forest ecosystems, acidification of soils and surface waters (Derome and Lukina, 2011) even at considerable distances from the smelters. Heavy metals and alkaline pollutants contaminate areas around the sources of pollution within a few hundred kilometers, while acid sulfates can be spread to long distances (Mahura et al., 2018). The main heavy metal pollutants in the region are nickel and copper, but other metals are also emitted. Mercury is the latest known threat to the Arctic environment. The group of persistent organic pollutants (POPs), including a large number of organic chemicals, is also potential environmental hazard. Examples of POP chemicals are PCB and DDT (AMAP, 2006).

The Murmansk county (Kola Peninsula) is one of the most urbanized and industrially developed regions of the Russian Arctic. Although the area of the region occupied by cities, settlements and industrial enterprises covers only 0.5% of the whole peninsula, its environmental influence is known to be very high, particularly with regard to airborne chemical pollution from mining and metallurgical industry and remains to be a concern also for the neighboring Fenno-Scandinavian countries.

It should be noted that cities are not isolated systems. They may distribute as much pollution in surrounding areas as they receive from the outside. Thus, the problem of mutual risk assessment for the cities and their surroundings should be considered, especially for Siberian conditions. The influence of the transboundary atmospheric transport between Russia, China, Kazakhstan, and Mongolia should not be ignored as well. All these problems are connected with the high-latitude ecological safety and quality of life. Moreover, there are indications (Jaffe et al., 2004) that the long-range transport of Siberian biomass burning emissions could produce pollution episodes and impact on surface ozone as far as in western North America.

For more details, see the overview by Baklanov et al. (2013), which considers specific atmospheric pollution problems in Siberia including the current state and projections, peculiarities of Siberian environmental protection problems, risk assessment, and tendencies in atmospheric pollution, including health-affecting pollutants, greenhouse gases (GHGs), and aerosols.

1.6 Wood smoke pollution remediation efforts

As described in Sections 1.3-1.5, wood smoke is often a major contributor to local ground-level air pollution. Wood burning is not only a traditional practice for domestic heating and cooking in many areas of the world, but also an opportunity for industrialized countries to transition towards renewable energy sources. However, wood burning in stoves and boilers emits large amounts of compounds harmful to human health and biomass burning emissions may be more harmful than other types of organic aerosols (Tuet et al., 2017). It has become clear, therefore, that renewables based on burning wood (and other biomass types) are an option if only technical measures are introduced to make emission factors for particles and other pollutants comparable with those already achieved for appliances employing gas/liquid fossil fuels. Nevertheless, many governments, implementing strategies to reach their national greenhouse gas emissions reductions, have subsidized wood burning. As a consequence, wood burning consumption has

increased in many European countries (Levander and Bodin, 2014). The studies conducted in Nordic (Scandinavian) and Alpine countries, where fuels are obtained from vegetation adapted to cold climates, can provide potentially useful information to address wood burning pollution in Arctic countries. Moreover, a recent review of the existing residential wood burning techniques was supported by the Arctic Council, as a contribution to the assessment of the transport of short-lived climate forcers (primarily black carbon) towards the glaciated regions in the Arctic (Levander and Bodin, 2014).

The scientific literature on this subject, summarized in Table 1, and the official governmental and EU reports agree in showing that emissions factors of small-scale wood burners (boilers, stoves, fireplaces, etc.) vary of several orders of magnitude between appliance types (Nussbaumer, 2010; EEA, 2016b). Traditional woodstoves can emit more than thirty times more particulate matter than an automated pellet stove and almost one hundred times more than an oil boiler (McDonald, 2009). In open fireplaces employing wood logs as fuel, the excess of combustion air is essentially un-adjustable making combustion very inefficient and the emissions from unburned fuel very high. Even in modern appliances, emission factors strongly depend on practices, and, since combustion efficiency is improved when frequently adding small logs, such stoves are often non-ideally operated and emissions are high. All studies indicate that the emissions of small-scale facilities depend very much on the user behavior (up to a 100 factor of difference). Nevertheless, significant improvements in terms of both energy efficiency and emission reduction were achieved by introducing new technical standards on certain log-fueled appliances, as in the case of modern two-stage combustion boilers equipped with a heat storage tank (Nussbaumer, 2010).

Table 1. Typical PM emission factors in milligrams PM per megajoule heat output for various residential appliance types using wood and other fuels.

Appliance Type	**Typical* PM emission factors today (mg/MJ)**	**Achievable* PM emission levels (mg/MJ)**
Open fireplaces	50 to >1 000	50-100
Wood stoves and closed inset appliances	20 to >1 000	15-25
Log wood boilers without heat storage tank	20 (at full load) to >1 000	Not recommended
Log wood boilers (with heat storage tank)	20 to > 100	10-20
Pellet stoves & boilers	10-50	10-20
Automatic wood combustion plants … with cyclone	50-300	50-100
… with simple ESP	25-50	15-35
… with advanced ESP	5-15	5-15
… with fabric filter	<5	<5
ASTM #2 oil boiler**		1.3
ULSD oil boiler**		0.025
Natural gas boiler**		0.016

*The left column shows typical operation today, and the right column shows laboratory measurements or best achievable PM emission levels under ideal conditions. Adapted from Nussbaumer (2010), with the exception of the lines marked with **, which are from McDonald (2009). ASTM means American Society for Testing and Materials and #2 is a common grade of heating oil, ULSD is ultra-low sulfur diesel, which is heating oil having <15 ppm S by mass.

Post-combustion control technologies such as electrostatic precipitators and catalytic converters have been investigated in a number of European studies (see the recent review: Vicente and Alves, 2018). A number of these studies have shown that these technologies can be effective at reducing $PM_{2.5}$ emissions (Fine et al., 2004; Hukkanen et al., 2012; Kaivosoja et al., 2013),

although there are concerns about the economics of small-scale ESPs (Karvosenoja et al., 2007). Beyond the question of reducing $PM_{2.5}$ mass concentrations, ESPs preferrentially remove particles of differing chemical composition (Migliavaca et al. 2014, Nussbaumer and Lauber, 2010), potentially altering the toxicity of emitted particles (Kaivosoja et al., 2013). Catalytic converters oxidize condensable organic vapors and soot particles resulting in PM emissions reductions during the high temperature part of the burn cycle (Hukkanen et al., 2012). Catalysts are typically only engaged when the woodstove emissions reach appropriate temperature, so startup emissions still occur and burn-cycle averaged emissions are not reduced as much as the main burn phase emission reduction may indicate (Fine et al., 2004; Hukkanen et al., 2012). Both ESPs and catalytic converters also need maintenance to perform properly, which could affect their overall effectiveness in pollution mitigation (Fine et al., 2004; Migliavaccaa et al., 2014; Reichert et al., 2017). In addition to pollution reductions for outdoor air, McNamara et al. (2017) and Ward et al. (2017) studied the effectiveness of indoor air filtration units in reducing exposure to wood smoke in homes heated with woodstoves.

In alternative to manual wood combustion devices, pellet stoves and boilers can be employed. The use of pellets guarantees automatic feeding with small-size solid fuel, allowing to reach improved combustion conditions in practical operations (Table 1). For instance, Shen et al. (2012) estimated that 95, 98, 98, 88, and 71% reductions in the total emissions of CO, OC, EC, PM, and PAHs could be achieved by replacing the raw biomass fuels combusted in traditional cooking stoves with pellets burned in modern pellet burners.

These studies indicate that replacement of old biomass-burning appliances with modern, more efficient ones, the emission of air pollutants can be considerably reduced (EEA, 2016a). At the same time, the evaluation of dirty versus clean technologies must account for the diversity of solid fuels available in the national/local markets. Even among processed wood products, such as pellets, significant differences can be found in the quality of the fuel (in terms of emission factors of specific pollutants). When wood briquettes or pellets are stored for long periods before they are sold, they may contain preservatives against fungi and insects, but such chemicals can increase the amount of specific pollutants (e.g., copper) in the exhaust (AIRUSE, 2016). For these reasons, the introduction of a certification for pellets was recommended (EEA, 2016a). The evaluation of appropriate policies (eco-labelling, economic incentives) to promote the changeover of old stoves and boilers is a critical issue, because wood is a very cheap, abundant fuel in many rural regions, including large areas in the Arctic. Incentives should target also building renovation to improve thermal insulation, not only the changeover of the old appliances. Finally, local information campaigns must complement regulatory measures and incentives, to improve maintenance of the stoves and to achieve better practices for combustion.

1.7 Stakeholder issues and questions from Fairbanks stakeholders

Because Fairbanks, Alaska is in serious violation of the EPA $PM_{2.5}$ National Ambient Air Quality Standards (https://www3.epa.gov/airquality/greenbook/rnca.html#PM-2.5.2006.Fairbanks), the community has concerns about breathing healthy air and about how to heat homes and run businesses with affordable energy. As a part of the process of developing this whitepaper, we compiled questions that stakeholders have expressed. Many of these questions motivate our study and we also wish to formulate the study in such a way that we can address them and make an outreach plan to bring results back to the community.

Some members of the public ask: "Is it possible to reach attainment? If so, what needs to be done?" Some members of the public feel that they are doing their part by using EPA-certified device and operating it properly with dry wood, but still worry that they will be restricted further if these measures are not deemed effective.

Members of the public have concerns about the accuracy of the prior source apportionments, which identify wood smoke as the largest cause of ground-level air quality reductions (see Section 1.4). Each of these studies gets a slightly different apportionment of sources, and this variability is used as evidence to not trust the results at all. There is also a concern about the contribution of the power plants to ground-level air quality reductions. Members of the public point out that the Fairbanks emissions inventory shows power plants emit a large fraction of the total $PM_{2.5}$ on an annual basis, yet the remediation strategy focuses on wood smoke. The confusion arises because much of the power plant emissions are emitted aloft from smoke stacks, so only a fraction of their emissions actually impacts the ground-level air quality on the strong inversion days when air quality is worst. Modeling has been used to try to quantify this effect, but there is a general mistrust of modeling. The point sources who emit pollutants from high stacks feel that their contribution to ground-level air quality reductions are minimal, yet they are being asked to add expensive retrofit control technologies to reduce pollution. Therefore, the point source operators would like a better quantification of their actual contribution to ground-level air quality reductions. Models are also used to predict when to call burn bans (curtailments), but the public doesn't understand the basis for identifying these time periods and if a curtailment is called and no exceedance occurs, some feel it may have been unnecessary.

With respect to monitoring, there are public questions about the representativeness of the monitoring sites. Some members feel that the monitors are sensing localized hotspots and are not representative of the general air quality, while some others think some monitors are in non-residential areas and thus are not representing un-detected hotspots.

Many in the public are concerned about health effect of fine particulate matter. Some wonder if Fairbanks pollution is less or more harmful than pollution in the lower 48 states. People ask: "How many people are dying as a result of $PM_{2.5}$ pollution?" Some feel that EPA health studies are biased and/or that the health standards are set too low. Some even say that strong inversions make the current health based standard impossible to achieve, so Fairbanks should have a different standard. There is also confusion about summertime wildfires, which are often excluded from the monitoring data as a uncontrollable "exceptional" events. The public has concerns that if $PM_{2.5}$ is harmful, why isn't EPA addressing wildfire.

Fairbanks recently completed a "Stakeholders" process to identify appropriate control measures to address air quality. Information from the group is freely available on the web: http://fnsb.us/transportation/Pages/stakeholders.aspx. Members of the ALPACA steering committee (Simpson and Mao) and local ALPACA workshop participants worked with this group to provide the current state of the science. Although the currently meeting stakeholders group has been completed before the ALPACA field studies could be carried out, the process has started the dialog and presents an opportunity for the ALPACA project to hear concerns and eventually for ALPACA to deliver results back to the community and stakeholders.

2 Open questions:

2.1 Which aerosol emissions and chemical processes are most important for ground-level wintertime air quality problems?

2.1.1 What are appropriate source characterizations for cold climates?

People living in cold climates and at high latitudes often use more energy and have more emissions than residents in lower latitudes. The need to heat homes and power them with electricity, which is typically done by combustion of wood, oil, coal, or natural gas, is a large source of these emissions. Mineral and oil/gas extraction industries are common reasons for people to be living in the North, leading to industrial emissions. Finally, transportation of those materials also contributes to pollution. Many of these sources also occur at lower latitudes and these sources have been characterized for primary particulate and gaseous emissions. However, these emissions, upon exit from the smokestack or tailpipe cool to much colder temperatures than are typical of source tests that have been developed for the more populated mid-latitude regions. How these colder temperatures affect prompt particle formation is not well known at this time. Wood combustion is a common source of heat in northern homes. Source tests on wood appliances show that different appliances can have drastically different emissions, and that the emissions depend upon wood moisture, wood species, and the skill of the operator (Fine et al., 2004; Vicente et al., 2015; Fachinger et al., 2017). Additionally, emissions vary over the burn cycle of fire ignition and appliance warm up, burn phase, and eventual burnout of the fuel load. In some places, operators may also burn fuels other than the appliance was designed for, leading to uncertain emissions. Therefore, it is challenging to build appropriate emissions inventories relevant to cold locations.

To address these issues, a Fairbanks-specific emission inventory was produced (Alaska DEC, 2014a). This emission inventory includes all major source classes for point sources, mobile sources, and residential heating. Many of the emission profiles were generated by tests of Fairbanks fuels in relevant devices, although those tests were carried out at OMNI-test laboratories in Portland Oregon, so emissions were not cooled as much as they would cool in Fairbanks during winter. The inventory also places emissions into appropriate altitude layers, which is important for modeling the impact of elevated stack emissions on ground-level air quality (see Section 2.3). The ALPACA study will improve and test this inventory. These considerations lead to these questions:

- How do cold temperatures affect formation of particulate matter?
- What are appropriate burn-cycle-averaged wood smoke profiles?
- How can operator skill, fuel moisture, and other factors be included in emission inventories?

2.1.2 Do cold conditions and particle-phase chemistry produce non-traditional SOA?

Atmospheric organic aerosol (OA) composition, transport, and transformation has been studied in a wide range of environments during summer months at low- to mid-latitudes (e.g. Zhang et al., 2007). Traditional SOA sources include OH, ozone, or nitrate radical initiated reaction of VOCs to form low-volatility material that partitions reversibly to the particle-phase. Much of the

low-temperature sampling of ambient OA has occurred at high elevations (mountain or aircraft), representative of remote air that has undergone significant aging and dilution since the point of emission.

Fairbanks, AK represents an understudied high-latitude environment that traps high concentrations of fresh wintertime emissions in a shallow boundary layer that is largely void of traditional photochemical activity (low-angle wintertime sun that approaches only 3 hours of daylight). It is of interest to explore possible phase-partitioning of an analytically-challenging range of semi-volatile organic compounds (SVOCs) and intermediate-volatile organic compounds (IVOCs) that tend toward the gas-phase at low-mid latitudes, but likely enter the particle-phase at the low temperatures realized in Fairbanks. Such species can exist from direct combustion emissions of biomass and other fuels, but the variety and concentration of these species can be amplified by any potential chemical reactions in the atmosphere.

Non-traditional wintertime chemistry, while less studied, has been reported in a range of polluted environments. Deviations from equilibrium, diffusivity limitations (see Section 2.1.3), enhanced oxidants, temperature modifications of volatility, or heterogeneous reactions could lead to non-traditional SOA. Regions with large combustion sources that produce high concentrations of organic gases and nitrogen oxides have observed enhanced SOA production as a result of several possible formation pathways. Nocturnal oxidation of precursor organic gases by NO_3 radicals have been shown to produce significant SOA mass through the production of organic nitrates (Ng et al., 2017), a process that is included in most modern models. Aerosol (e.g., soot) or other surface heterogeneous formation of nitrous acid (HONO, from NO_2 uptake) and proceeding reactions can generate OH and O_3 that in turn react with VOCs/IVOCs/SVOCs to produce SOA, as has been observed in other wintertime environments (e.g. Xing et al., 2018). The reaction of O_3 with common alkenes can lead to additional OH formation which perpetuates further SOA production (Paulson and Orlando, 1996). Like HONO, nitryl chloride ($ClNO_2$) can form through nighttime heterogeneous reactions in the presence of aerosol-phase chloride (Thornton et al., 2010). While Fairbanks is removed from major marine sources of NaCl particles, $ClNO_2$ chemistry has been observed at mid-continent locations as a result of anthropogenic sources (Thornton et al., 2010), and we expect significant wood-burning-derived particle-phase KCl to be present in the Fairbanks wintertime airshed. Another example of non-traditional SOA formation potential occurs in regions with very high VOC emissions (such as the oil and gas fields of the Western U.S.), where wintertime photolysis of carbonyls has led to very high concentrations of O_3 (Edwards et al., 2014), which can enhance SOA formation. This chemistry is notably dependent on snow cover (increasing overall actinic flux) and cold temperatures to form a shallow boundary layer, conditions common in Fairbanks (see Sections 2.3 and 2.2.3). Aerosol acidity can contribute to enhanced SOA production through acid-catalyzed heterogeneous reactions (Jang et al., 2002). These considerations lead to the following open questions:

- What oxidants are present under cold and dark conditions and what is their role in SOA formation (also see Sections 2.2.1 and 2.2.2)?
- What is the phase partitioning of semi-volatile organic compounds under these cold and dark conditions (also see Section 2.1.3)?
- Are heterogeneous reactions (e.g. Jang et al., 2002) important in forming SOA under these conditions?

2.1.3 What is the phase of organic aerosol (liquid, solid, semi-solid) in cold and dark conditions?

Models of organic aerosol, a major component of wood smoke particulate matter, typically use equilibrium phase partitioning theory to describe the partitioning of semi and low volatility organic compounds between the gas and particle phases (Pankow, 1994). Partitioning theory is implemented to account for evaporative losses of primary (directly emitted) organic aerosol upon dilution from the point of emission and for the formation of secondary organic aerosol via oxidative conversion of organic vapors, either separately emitted or the result of primary organic aerosol evaporation, into semi and low volatility products (Robinson et al., 2007). Equilibrium partitioning of organics has been validated in early studies of urban photochemical smog in experiments forming organic aerosol from fuel vapor oxidation under warm summertime conditions (Pankow, 1994). However, few studies have adequately tested its validity under cold wintertime conditions.

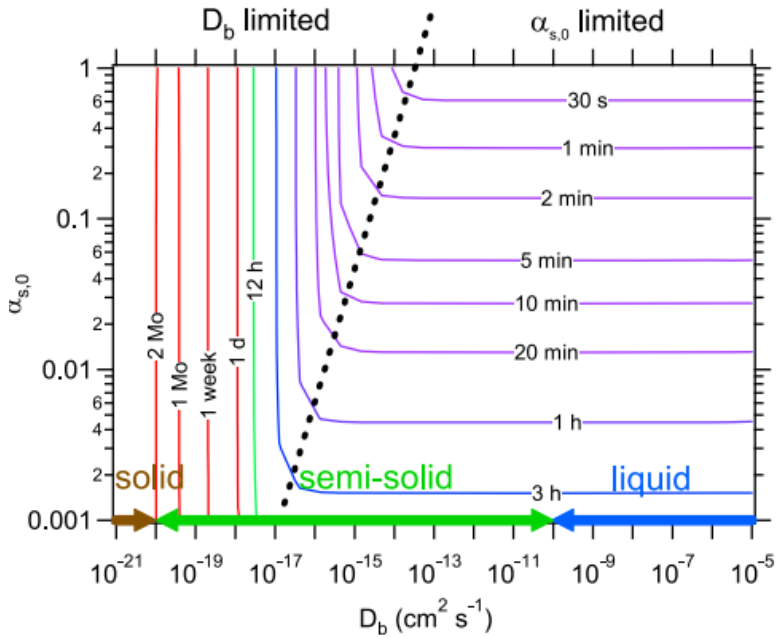

Figure 2. Time scales to achieve gas-particle equilibrium (contours) as a function of particle phase diffusivity (D_b) and surface accommodation efficiency (a). D_b reflect the viscosity and thus phase state of the aerosol (solid, semi-solid, or liquid). Adapted from Shiraiwa and Seinfeld 2012

Recent studies have found that the physical phase state of atmospheric organic aerosol can range from liquid-like to semi-solid (e.g. wax-like) or even solid depending upon temperature, humidity, and the chemical composition of the organic aerosol (Virtanen et al., 2010; Koop et al., 2011; Perraud et al., 2012; Renbaum-Wolff et al., 2013; DeRieux et al., 2018). The distinction of aerosol phase is important to predicting the atmospheric evolution of organic aerosol in that phase state controls the diffusivity of components in the particle phase, and thus the timescale required for vapors and particle phase components to achieve equilibrium. The more viscous (solid-like) a medium, the lower the diffusivity of components through the medium. Slower diffusion leads to longer timescales to achieve equilibrium as components arriving from the gas-phase will require longer time to fully mix throughout the particle or for a component in the particle to reach the surface and enter the vapor phase; both requirements for achieving equilibrium between the phases. For liquid-like atmospheric particles, the timescale to achieve

equilibrium between vapors and sub-micron particle components is minutes or less (Shiraiwa and Seinfeld, 2012). However, for semi-solid or solid particles, equilibration could require hours to weeks or longer as shown in Figure 2 (Shiraiwa and Seinfeld, 2012). Thus, the assumption of nearly instantaneous phase equilibrium, typical of gas-particle partitioning in models of atmospheric organic aerosol could lead to significant errors in the predictions of organic aerosol mass evolution in the atmosphere if conditions favor semi-solid or solid-like phase states. Slow diffusion of water, oxidants and organic molecules could promote ice nucleation (Knopf et al., 2018) and facilitate long-range transport of reactive and toxic organic pollutants embedded in SOA (Shrivastava et al., 2017; Mu et al., 2018).

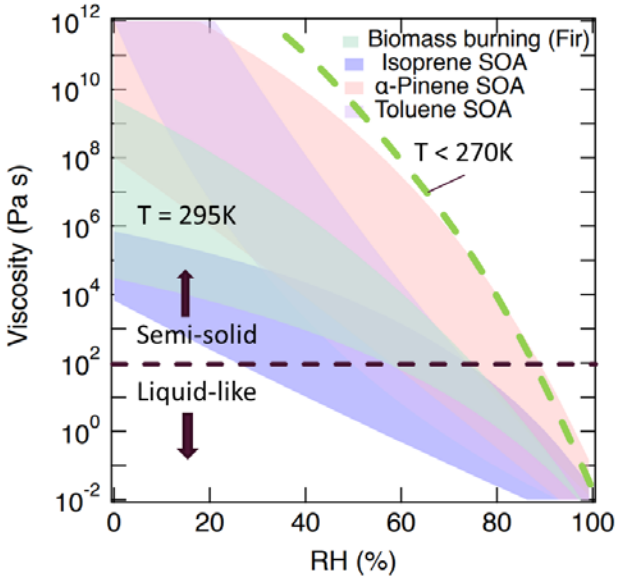

Figure 3. Viscosity estimates of different organic aerosol types as a function of ambient relative humidity (RH) at T = 295 K (shaded regions) adapted from DeRieux et al. 2018. Viscosity relates inversely to diffusivity, and a typical transition from liquid-like to semi-solid behavior occurs for a viscosity of ~10^2 Pa s (horizontal dashed line). As the ambient temperature approaches the material glass transition temperature (T_g see text) viscosity will increase substantially. The green dashed line indicates a possible viscosity response or biomass burning organic aerosol for wintertime temperatures.

Recent global model simulations have predicted that the annual average of SOA phase state in Alaska would be liquid-like reflecting high relative humidity (Shiraiwa et al., 2017). However, strong diel and seasonal variations of the SOA phase state is expected and oxygenated organic aerosol is more likely to exist as a semi-solid or amorphous solid phase under colder and/or lower relative humidity (RH) conditions. The glass transition temperature (T_g) characterizes the transition between liquids and amorphous solids for different organic substances. The closer ambient temperature is to a substance's T_g, the more viscous and thus solid-like the substance becomes (Koop et al., 2011). For constant temperature, a higher ambient relative humidity (RH) will lead to penetration of water into the organic matrix, lowering viscosity and moving the medium towards a more liquid-like phase state (DeRieux et al., 2018). Figure 3 illustrates the estimated viscosity at 295K for different RH of different organic aerosol types, including biomass burning organic aerosol which would presumably be similar to residential wood smoke organic aerosol. Under typical RH conditions, biomass burning organic aerosol is expected to have a viscosity similar to viscous semi-solids, even at 295K. The T_g of biomass burning organic aerosol from lodgepole pine and subalpine fir combustion are likely in the range of 270 – 280 K (DeRieux et al., 2018). Thus, under wintertime conditions where $T \sim 250 – 270$ K, the

expectation is that wood smoke organic aerosol (and secondary organic aerosol generally) would have even higher viscosity, potentially by factors of 10^3 or 10^4, than indicated in Figure 3 (DeRieux et al., 2018). Furthermore, the cold temperatures may cause condensation of higher volatility species, like intermediate volatility compounds, as their vapor pressure would be lower than at standard temperature and pressure.

Questions that stem from these considerations include:

- What is the phase state of particulate organic matter under typical wintertime conditions?
- To what extent is equilibrium gas-particle partitioning of organics achieved under cold winter conditions within urban areas near direct emissions of POA?
- How should models that utilize dynamic gas-particle partitioning simulate aging of wood smoke aerosol and SOA formation from VOC under wintertime conditions?
- Do the cold temperatures modify vapor pressures enough to condense intermediate volatility compounds?
- How does the phase state affect chemical transformation and cloud formation properties of organic aerosols under wintertime conditions in Alaska?

A wintertime study could provide key insights into these questions. A combination of existing particle physical state measurements, such as inertial bounce fraction and hygroscopicity, together with simultaneous measurements of semi-volatile organic vapors in the gas and particle phases could provide the observational constraints needed to test theories of organic aerosol phase state and gas-particle partitioning thermodynamics.

2.2 Which precursor gases interact to form/grow aerosol particles?

2.2.1 How is sulfur oxidized under cold and dark conditions?

Sulfate is a ubiquitous component of global aerosol with implications for human health, visibility, and climate. Sulfate is also the dominant source of acidity in clouds and aerosols with implications for acid rain and the formation and abundance of other inorganic and organic aerosol particles. For example, sulfate abundance influences the pH-dependent gas-aerosol partitioning of ammonia and nitrate, two other important sources of inorganic aerosol. The largest source of sulfur to the atmosphere today is from coal and oil combustion (Smith et al., 2011). Most sulfur from human activities is emitted as sulfur dioxide (SO_2) gas, with a small fraction (< 10%) emitted directly as sulfate. Sulfate is also a dominant component of Arctic aerosol known as Arctic haze, which typically peaks in winter and early spring (Sirois and Barrie, 1999). Remote Arctic sulfate abundance has been decreasing at surface sites since about 1980 due to reductions in sulfur emissions in Europe and North America resulting from air quality regulations (Geng et al., 2015; Breider et al., 2017) even though emissions over Asia were increasing over the same time period. However, this reduction has not occurred in urban Arctic areas such as Fairbanks where local sources of sulfur dominate sulfate and $PM_{2.5}$ abundance (Tran and Mölders, 2011; Wang and Hopke, 2014; Nattinger, 2016).

Sulfate emitted directly to the atmosphere is referred to as "primary" sulfate, while sulfate formed through the oxidation of SO_2 gas is referred to as "secondary" sulfate. Secondary sulfate formation occurs through gas-phase reactions of SO_2, through aqueous-phase reactions in clouds, and through reactions on the liquid-layer surface of existing aerosol. Globally, in-cloud SO_2 oxidation is the dominant form of secondary sulfate (~60%) followed by gas-phase oxidation by

OH (~30%). In-cloud sulfate formation is dominated by oxidation by hydrogen peroxide (H_2O_2) (Alexander et al., 2012), while gas-phase oxidation is dominated by the hydroxyl radical (OH). Both OH and H_2O_2 require sunlight for formation, making SO_2 oxidation and sulfate formation rates generally higher in summer than in winter. Wintertime oxidation pathways include oxidation by ozone, in-cloud/fog formation of hydroxymethane sulfonate (Moch et al., 2018), and transition metal-catalyzed oxidation by O_2 in clouds. The latter is thought to be the dominant formation pathway of secondary sulfate in the Arctic winter (McCabe et al., 2006; Alexander et al., 2009).

In wintertime in Fairbanks (at the downtown NCORE site), sulfate typically represents 1/5 of the mass of $PM_{2.5}$ (Figure 1; Nattinger, 2016). The origin of this sulfate is unclear. Although coal and heating oil burned in Fairbanks contain relatively large amounts of sulfur, most of the sulfur emissions are in the form of SO_2, not sulfate (US-EPA, 2010). The degree of oxidation of sulfur can be quantified by comparing the relative molar abundance of sulfate to total sulfur (SO_2 + sulfate), a metric often referred to as the sulfur oxidation ratio (SOR). The SOR of primary sources is not well known. If the actual SOR is large enough, primary emissions could explain the particulate sulfate at the ambient monitoring stations. It is also possible that SO_2 is being oxidized in plume as emissions equilibrate to ambient conditions, forming a pseudo-primary sulfate emission source. Whatever sulfate is not formed by prompt emissions would arise from secondary atmospheric oxidation of SO_2. However, It is unclear how SO_2 would be oxidized to sulfate the cold, dark and dry conditions of wintertime in Fairbanks when OH and H_2O_2 concentrations are expected to below, and clouds are likely to be ice rather than liquid water, slowing the rate of in-cloud sulfate formation. Higher SOR values are indicative of higher rates of SO_2 oxidation and secondary sulfate formation. Paradoxically, SOR values are higher in winter than in summer in Fairbanks, the opposite of expectations. Although typical wintertime SOR values are only 5-6% (Nattinger, 2016), this is still on the high end of that expected from primary emissions, suggesting some secondary sulfate formation. Air quality models underestimate the sulfate abundance and the mass fraction of sulfate in $PM_{2.5}$ in wintertime in Fairbanks (Mölders and Leelasakultum, 2012). It is possible that oxidation on pre-existing aerosol may be an important form of secondary sulfate formation in this unique environment, as has been suggested to be the case in the extreme pollution episodes in wintertime in China (Wang et al., 2014; He et al., 2018). This oxidation pathway is typically not included in air quality models because its contribution to secondary sulfate formation is usually negligible. However, this may not be the case in the unique conditions of a Fairbanks winter. Understanding the degree, rate and mechanism of secondary sulfate formation is critical for determining if reducing sulfur emissions in Fairbanks would significantly alleviate the contribution of sulfate aerosol to poor wintertime air quality. As sulfate abundance also influences the formation of other inorganic and organic aerosols, understanding the origin of sulfate is central for determining the most effective air-quality mitigation strategies. These ideas lead to the following open questions:

- What are sources of atmospheric sulfur and what is the SOR from these sources?
- How does vertical mixing of precursors and oxidants affect processing of sulfur to particulate sulfate?
- What is the phase dependence of sulfur oxidation processes?
- Which oxidants are responsible for sulfur oxidation under cold and dark conditions?

- Do SO_2 levels exceed National Ambient Air Quality Standards (75 ppb in 1 hour, which would then trigger health concerns) in areas around Fairbanks?
- What are the processes governing background sulfate over the Fairbanks region?

2.2.2 How active is nocturnal nitrogen chemistry in this poorly mixed environment?

Nitrogen oxides (NO_x = NO + NO_2) play a crucial role in urban air quality. NO_2 is a federally regulated pollutant and the chemistry of various reactive nitrogen species drive a number of important atmospheric chemical cycles, including the formation of other regulated pollutants such as ozone and secondary aerosol. Quantifying the rates of NO_x emissions and their oxidation cycles are therefore critical to understanding their impacts on air quality within any given region. While the role of nitrogen oxides has been well established for summertime air pollution, the situation in Fairbanks, Alaska, is unusual as pollution events occur in wintertime, when large amounts of NO_x are emitted into a cold, dark, and stable boundary layer, which in addition has high concentrations of a unique mix of aerosols. Understanding NO_x chemistry and its impacts on local air quality there is challenging due to the extremity of the environment (very cold temperatures, very limited sunlight) and the relatively small number of previous studies characterizing these rates in under these conditions (Ayers and Simpson, 2006; Apodaca et al., 2008; Huff et al., 2011; Joyce et al., 2014). Studying reactive nitrogen chemistry in Fairbanks would extend our understanding of NO_x chemistry across a range of environments, and provide contrast to other urban areas at lower latitudes that also experience severe winter air pollution, e.g., Salt Lake City, Utah (Baasandorj et al., 2017), cities in the San Joaquin Valley, California (Brown et al., 2006a; Chow et al., 2006), and Beijing, China (Huang et al., 2014; Wang et al., 2018). Fairbanks is also a model for similar urban areas across the Arctic and sub-Arctic regions of the world.

Broadly speaking, NO_x oxidation cycles fall into two categories: photochemical and dark. Photochemical NO_x oxidation cycles occur via OH reaction with NO_2, organic peroxy radical (RO_2) reaction with NO and formation of peroxy acetyl nitrates (PANs), reactions 1-3 below.

Photochemical NO_x oxidation

$$NO_2 + OH \rightarrow HNO_3 \quad (1)$$

$$RO_2 + NO \rightarrow RONO_2 \text{ (organic nitrate)} \quad (2)$$

$$R(O)O_2 + NO_2 \leftrightarrows PAN \quad (3)$$

Dark oxidation process via the reaction of O_3 with NO_2 to produce NO_3, which is photochemically unstable such that the mechanism does not contribute substantially during sunlit conditions (Brown and Stutz, 2012).

Dark NO_x oxidation

$$NO_2 + O_3 \rightarrow NO_3 \quad (4)$$

$$NO_3 + NO_2 \leftrightarrows N_2O_5 \quad (5)$$

$$N_2O_5 + (H_2O \text{ or } Cl^-) \rightarrow (2\text{-}\phi)HNO_3 + \phi ClNO_2 \quad (6)$$

The equilibrium in reaction (5) strongly favors N_2O_5 at high NO_x and / or low temperature ; reaction (6) is heterogeneous, leading to the production of HNO_3 or $ClNO_2$ according to the $ClNO_2$ yield, ϕ, which is dependent on the ratio of aerosol liquid water to chloride . Nitric acid produced in this reaction may partition to the gas or particulate phase depending on temperature and the availability of ammonia.

Photochemical NOx oxidation requires sunlight as a source of the radical species (OH, RO_2, $R(O)O_2$). In Fairbanks, at a latitude of 64.8 °N and a day duration on January 1 of approximately four hours (4 hours 3 minutes), photochemical radical sources are expected to be weak, and photochemical NO_x oxidation cycles quite slow. For example, the midday clear sky NO_2 photolysis rate is 3.8×10^{-4} s^{-1}, approximately 15 times slower than at midlatitudes (40°N). Major sources of the precursors of the photochemical radicals driving photochemistry include the photolysis of ozone itself, reactions of ozone with alkenes, and photolysis of species typically associated with air pollution, such as formaldehyde (CH_2O) and other carbonyls, nitryl chloride ($ClNO_2$) and nitrous acid (HONO). The degree to which each of these contributes to photochemistry in Fairbanks is an important scientific question. Of the potential photochemical radical sources, HONO is the shortest-lived and fastest photolyzing in the low light environment of Fairbanks, with a midday lifetime on the order of 4-5 hours, compared to 20 hours for $ClNO_2$ and more than 500 hours for CH_2O. HONO is produced chemically from the heterogeneous uptake of NO_2 on surfaces, particularly the ground or building surfaces (Stutz et al., 2002; Young et al., 2012 also see Section 2.2.3). This chemical source of HONO does not require ozone to be present and will be prevalent under high NO_x conditions characteristics of Fairbanks winter pollution events. HONO also arises from direct emissions from combustion sources such as gasoline and diesel engines (Kurtenbach et al., 2001a) and residential wood combustion (Kurtenbach et al., 2001b; Akagi et al., 2011). Since residential wood combustion is implicated as a source of the organic aerosol that constitutes the majority of the Fairbanks particulate matter, characterizing the HONO from this source will be important as well. The sources and amount of HONO is thus of particular scientific interest.

Dark NO_x oxidation requires ozone, which is typically absent at surface level within urban areas at night, due to the efficient titration of O_3 by excess NO_x, which is emitted primarily in the form of NO.

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (7)$$

Average O_3 mixing ratios at the main monitoring site in Fairbanks are 5 ppbv during the winter months, and are frequently zero at high NO_x, presumably during stagnant conditions associated with high air pollution events. However, the atmosphere above the surface layer and outside of urban Fairbanks likely has sufficient ozone to drive NO_x chemistry in the dark. The extent to which these air masses contribute to trace gas levels at the urban surface will depend on the stability of the lower atmosphere, efficiency of entrainment of free tropospheric air into the boundary layer, vertical mixing and advection processes.

Neither photochemical nor dark oxidation cycles ought to be rapid at surface level in the immediate vicinity of Fairbanks during winter pollution events. Nevertheless, there is potential for NO_x chemistry within the winter Fairbanks environment. This potential depends on several factors. For photochemical cycles, it depends on the magnitude and sources of photolytic radical precursors. Nighttime chemistry will depend on the horizontal and vertical extent of O_3 titration and the degree to which horizontal or vertical mixing within the Arctic boundary layer and into/out of the Arctic boundary layer, leads to regions above or around Fairbanks with substantial NO_x mixing ratios but without ozone titration.

Mixing of NO_x from the ozone titrated region of the urban boundary layer into regions with excess ozone is likely to be an important process governing the dark oxidation of NO_x, as it is in other, lower latitude polluted winter boundary layers (see references above). A recent modeling study of the evolution of NO_x in Fairbanks suggests that this process is an important source of

soluble nitrate (HNO_3 or pNO_3^-) (Joyce et al., 2014), but there is little data to constrain such models on either a vertical or horizontal scale. Measurements of NO_x, O_3 and intermediates such as N_2O_5 and $ClNO_2$ as a function of height or in plumes transported downwind from the Fairbanks region are required.

The open scientific questions regarding nitrogen chemistry and cycling in Fairbanks winter are summarized as follows.

- What are the major sources of NO_x emission in the Fairbanks boundary layer? Is residential wood combustion an important source of NO_x, as well as radical precursors such as CH_2O, other carbonyl species and HONO? How much does power-plant emitted NO_x affect the boundary layer or chemistry aloft?
- Do NO_2 levels in Fairbanks approach national (NAAQS = 100 ppbv in 1 hour) standards that would trigger concerns regarding its health effects?
- What is the partitioning between gas and particulate phase soluble nitrate (HNO_3 and pNO_3^-)? How is this coupled with sulfate, ammonia, and pH?
- What are the major transport patterns in the Fairbanks region, including exchange between the Arctic boundary layer and free troposphere, and is there evidence for significant dark reactive nitrogen chemistry downwind?
- What are the reactive uptake coefficients for N_2O_5, $\gamma(N_2O_5)$ and yield of $ClNO_2$, $\phi(ClNO_2)$, what factors govern these parameters, and how do they compare to nighttime chemistry in other regions?
- What role does HONO play as a daytime radical precursor?

2.2.3 How does the presence of snow affect processing of wintertime pollution?

In addition to being dark and cold, snow is continually present on the ground during winter in Fairbanks. Snow impacts atmospheric stability, as discussed in Section 2.3.1, due to its high visible albedo and high IR emissivity. The presence of snow, though its unique albedo, directly impacts photochemistry, by increasing actinic fluxes compared to summer conditions at similar solar zenith angles (Lefer et al., 2001). Snow on the ground and suspended in the atmosphere as well as ice particles provide reactive surfaces for heterogenous chemistry, which can influence atmospheric composition though the uptake of trace gases and aerosols and the emission of various trace gases to the atmosphere. Dry deposition on snow is a complicated process that involves both air-snow gas exchange as well as the uptake on snow grains. Parameterizations of dry deposition on snow are implemented in air quality models, but, considering the well-known complexity of physical and chemical processes in snow, their accuracy merits validation. Reactions on surface fog particles may be faster than dry deposition because there is less transport limitation to bring gases into contact with fog particles than are required to bring gases to the ground surface. However, little is known about the role surface fog plays as a sink for trace gases and aerosols in wintertime Fairbanks.

Over the past decades it has also become clear that heterogeneous/multi-phase chemistry on ice surfaces can impact the overlying atmosphere (e.g. Domine and Shepson, 2002; Grannas et al., 2007). A prime example of such chemistry is the recycling of snow/ice nitrate into NO, NO_2, and HONO in the atmosphere, which has been observed at polar locations (e.g. Honrath et al., 2000, 2002; Grannas et al., 2007). Release of nitrogen oxides from snow impacts the atmospheric reactive nitrogen budget, which may be potentially contributing to elevated pollution levels. The

release of HONO has been implied as an explanation for elevated OH levels observed in the Arctic and Antarctica (Chen et al., 2001). Heterogeneous chemistry of NO_2 to produce HONO may in fact be enhanced in the highly NO_x-polluted Fairbanks environment. The photolysis of H_2O_2 in snow is also know to produce gas-phase OH (France et al., 2007). There is ample literature showing the release of reactive halogens from ice surfaces (Abbatt et al., 2012 and references therein) in remote regions, which may also occur under dark conditions (Simpson et al., 2018). While bromine release is likely not important for Fairbanks, release of chlorine compounds could provide an additional radical source in winter. Salts, used in road de-icing, are present in snow in urban regions and may also be present through other industrial processes (e.g. halides present in coal or added for scrubbing purposes). It is well known that species present under dark conditions, such as N_2O_5, can convert salts on surfaces into atmospheric species providing a source of radicals to drive atmospheric chemistry (Osthoff et al., 2008; Roberts et al., 2008; Thornton et al., 2010). Apodaca et al. (2008), for example, show that the lifetime of N_2O_5 is three times shorter under conditions that favor ice particle formation, providing strong evidence for its heterogeneous uptake in the wintertime environment in Fairbanks. In a follow-up study, Huff et al. (2011), derived a N_2O_5 deposition velocity of 0.59 cm s^{-1} over snow in Fairbanks. In addition to impacting the reactive nitrogen budget, N_2O_5 undergoes efficient chemistry with halogens on ice surfaces (Lopez-Hilfiker et al., 2012), which could provide another pathway of reactive chlorine formation. There is also evidence of active organic chemistry in snow leading to the formation and release of formaldehyde and other light aldehydes (Sumner and Shepson, 1999; Grannas et al., 2002; Hutterli et al., 2004).

While most of these studies have been performed in environments much cleaner than Fairbanks, a few studies have shown evidence for snow chemistry urban areas. Nevertheless, our understanding of snow chemistry under polluted conditions is incomplete. The ALPACA experiment offers a unique opportunity to investigate the influence snow/ice can have on the composition of the overlying atmosphere. ALPACA will thus address the following questions related to snow/ice chemistry:

- How does the reactive uptake of trace gases and multiphase chemistry on snow impact air quality in Fairbanks?
- What parallels exist between snowpack chemistry and chemistry on surface fog during wintertime pollution episodes?

These questions will be answered in close collaboration with the participants in the IGAC Cryosphere and Atmospheric Chemistry (CATCH) initiative, which has been endorsed by SOLAS.

2.2.4 What is the aerosol acidity and how does it affect other processes?

Particle acidity is a fundamental aerosol property that modulates many aerosol processes resulting in varied environmental impacts. Fine particle pH affects PM mass through controlling the gas-particle partitioning of semi-volatile acidic and basic species and by influencing rates of important heterogeneous reactions, such as catalyzing formation of certain secondary organic aerosol (Jang et al., 2002; Surratt et al., 2007). The proportion of acidic and basic species divided between gas and particle phases also affects both nitrogen and acid deposition patterns due to large differences in gas versus particle deposition velocities (Duyzer, 1994; Schrader and Brümmer, 2014), which leads to adverse effects on ecosystems from eutrophication to acidification (Erisman et al., 2007). Particle acidity also affects the solubility of important trace

metals, including iron, copper and manganese, which have been linked to aerosol toxicity (Ghio et al., 2012), see Section 2.4.2. Reactions with soluble Fe(III) and Mn(II) can also be an important route for SO_2 oxidation to sulfate, especially in locations where other oxidants (OH, H_2O_2, O_3) are expected to be low, such as the winter conditions in Fairbanks during pollution events (see Section 2.2.1).

Particle acidity is largely driven by non-volatile species, such as sulfate and certain cations (e.g., Na^+, Ca^{2+}, Mg^{2+}, K^+), and semi-volatile nitrate, organic acids and ammonia (Guo et al., 2015, 2016, 2017a; Murphy et al., 2017; Nah et al., 2018). Fairbanks aerosol during winter pollution events is dominated by wood-burning emissions, which is also a source for species that can influence particle pH, such as nitrate (from NO_x) (Liu et al., 2016), ammonia (Reis et al., 2009) and K^+ (Peltier et al., 2007; Pachon et al., 2013; Fourtziou et al., 2017). These species, along with sulfate, which comprises roughly 15 to 20% of the Fairbanks $PM_{2.5}$ mass (Figure 1), could largely drive the pH of fine particles in Fairbanks. Figure 4 shows that $PM_{2.5}$ sulfate, nitrate and ammonium are likely the major inorganic anionic and cationic species, but that the aerosol is moving toward a decreasing trend in ammonium relative to nitrate and sulfate, possibly due to an increasing role of other cations, such as K^+ or change in ammonia concentrations from changing ammonia emissions from other sources.

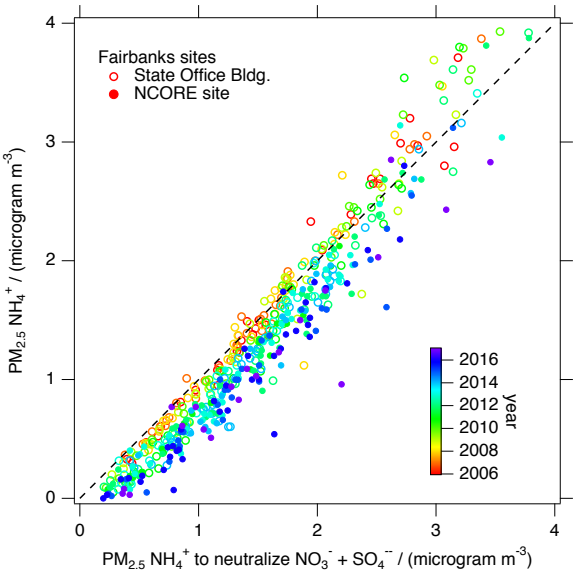

Figure 4: A partial ion balance showing observed ammonium in particles in Fairbanks, Alaska versus amount of ammonium that would be needed to neutralize strong acids observed in these particles. The colors show a trend towards less neutralized particles in time.

The sources of NH_3 in the winter atmosphere in Fairbanks are not well understood. The NEI 2014 inventory for NH_3 emissions for Alaska indicates that wildfires (34130 tons) and prescribed fires (192 tons) dominate the annual emissions reported for the state (35236 tons). Sources that are more relevant to the winter urban atmosphere include fuel combustion by industrial boilers (411 tons), residential oil and natural gas combustion (200 tons), residential wood combustion (34 tons), waste disposal (23 tons), on-road gas and diesel vehicles (176 tons). Typical emission factors for NH_3 from residential wood burning are 0.9 – 1.8 lbs of NH_3 per ton of fuel burned (CARB, 2015). Emission factors of NH_3 from gasoline vehicles are higher than from diesel vehicles, though this may change as selective catalytic reduction (SCR) technology is more widely adopted by diesel vehicles. Suarez-Bertoa et al. (2017) reported on-road emission factors

of 5-8 mg NH_3 per km driven for gasoline vehicles and 2-7 mg NH_3 per km driven for diesel vehicles, but during cold starts the emissions from the gasoline vehicles could be 10-20 times higher. Emissions of NH_3 into the cold winter atmosphere are likely to partition into the condensed phase, for example, clouds, fog, and particles. Paulot et al. (2017) found that the abundance of ammonia in the winter will strongly influence the pH of the condensed phase, with consequences for the rate and mechanism of sulfur oxidation (see Section 2.2.1).

Modeling studies have indicated that the pH in remote Arctic regions can be very low (Fisher et al., 2011). Determining particle pH under extremely cold conditions could be challenging, both in terms of measurements and the application of thermodynamic models. Unbiased measurements of the gases and particles would require maintaining samples at very low temperatures, up to the time of analysis. When ambient air is drawn indoors through an inlet and the temperature and relative humidity of the air change, the partitioning between the gas and particle phases may be dramatically impacted. For thermodynamic calculations, both limited data on thermodynamic properties at very low temperatures, and issues with particle phase state, such as ice or super-cooled liquid water and separate organic phases, will pose challenges in predicting pH and would have to be investigated. The low temperatures may also compromise the assumption that the gas and particles phases have fully equilibrated (see Section 2.1.3). The ALPACA study provides an ideal venue to study particle pH under extreme cold conditions give the large suite of supporting measurements proposed.

- What are the major sources of reduced nitrogen (NH_3 or amines) in Fairbanks, such as agriculture, emissions from mobile combustion sources (Nowak et al., 2012) or biomass burning (Perrino et al., 2002; Akagi et al., 2011; Behera et al., 2013) or long range transport (Fisher et al., 2011) from lower latitudes?
- How well understood is the cold temperature thermodynamics of the inorganic aerosol components in Fairbanks? What aerosol pH range is characteristic of this region? How well do cold Fairbanks conditions help to constrain processes at similarly cold high-altitude conditions?
- Does acidity affect the concentrations of transition metal ions, and in turn influence SO_2 oxidation and/or SOA formation?

2.2.5 What is the mixing state of aerosols and how that can impact chemical transformation?

Aerosol mixing state, the distribution of chemical species within an aerosol population and between individual aerosol particles, affects aerosol toxicity, acidity, hygroscopicity, gas-particle partitioning, and particle-phase and multiphase reactions (Ault and Axson, 2017), as shown in Figure 5. Aerosol mixing state reflects both the aerosol source, as well as atmospheric aging, during which internal mixing of chemical components occurs via coagulation, multiphase reactions, and gas-particle partitioning (Fierce et al., 2017). Estimated errors in simulations comparing assumptions of fully internally mixed aerosol (bulk aerosol assumption) versus size- and type-resolved aerosol (single-particle) show increased biases for freshly emitted (un-aged aerosol) compared to aged aerosol, particularly for aerosol absorption simulations (Fierce et al., 2017). Despite its importance, there is little knowledge of Arctic aerosol mixing state (Kirpes et al., 2018), particularly for the aerosol population impacting northern cities and villages (Schmale et al., 2018).

The identification and quantitation of aerosol sources using bulk chemical composition measurements can be challenging, and these measurements do not provide aerosol mixing state. In contrast, chemical measurements of individual particles directly measure aerosol mixing state and provide insights into atmospheric aging mechanisms (Prather et al., 2008); however, few measurements of individual particles have been conducted in the Arctic (Kirpes et al., 2018). Using secondary sulfate as an example, models may assume sulfate to be externally mixed (Sand et al., 2017) or completely internally mixed (e.g. CMAQ, https://www.epa.gov/cmaq, WRF-Chem SAPRC-MOSAIC (e.g. Marelle et al., 2018)), yet Kirpes et al. (2018) observed sulfate to be internally mixed with sea spray aerosol and organic aerosol during the winter at Utqiaġvik, Alaska. In Fairbanks, Alaska, source apportionment efforts have separated sulfate from contributions from diesel combustion and wood smoke (Wang and Hopke, 2014), leaving questions about the sources, mixing states, and fractions of primary and secondary sulfate, particularly during air quality violation episodes. During the winter in Alert, Canada, McCabe et al. (2006) found a significant contribution to non-sea salt sulfate formation from a non-photochemical oxidation pathway, proposed to be Fe^{3+}/Mn^{2+}-catalyzed O_2 oxidation. The potential for metal-catalyzed sulfate formation within individual particles can be probed by individual particle measurements (Li et al., 2013). This further illustrates the need for single-particle mixing state measurements, combined with isotopic measurements, to inform modeling and regulations for northern cities and villages. Example questions to be probed related to aerosol mixing state include:

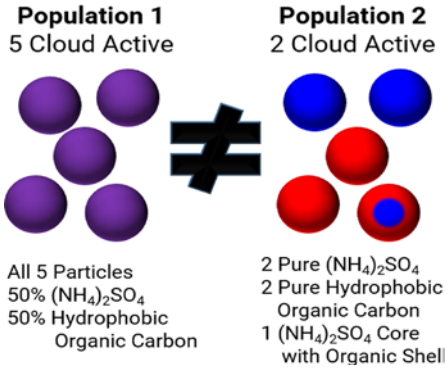

Figure 5. Illustration of an example of the importance of single-particle measurements in the cloud droplet activation (Ault and Axson 2017). This shows that while two populations can have the same average chemical composition, the individual particle properties and resulting CCN activities can be significantly different.

- What is the relationship between aerosol mixing state and secondary aerosol formation (including e.g., uptake of precursors, such as HNO_3, N_2O_5)?
- Are metals (particularly Fe, Mn) internally mixed within particles with enhanced sulfate?
- What is the mixing state of aerosol containing elevated sulfate levels (nitrate, oligomers, etc), and what formation mechanisms are implicated from this knowledge?
- What are the pathways for wood burning aerosol aging in the dark?

Investigating the mixing state of particles in wintertime Fairbanks is important to understand and ultimately to predict their atmospheric behavior in terms of a) how far particles can be transported before they are deposited, b) whether and which fractions of particles contributes to liquid and ice fog formation, and c) understanding of secondary aerosol formation, including wood burning aerosol aging and sulfate formation processes. Such studies would also provide insights into the mechanisms governing wintertime Arctic haze production from local sources and the extent to which such mechanisms are influencing haze aerosols transported from remote emission regions.

2.3 How do poor dispersion conditions exacerbate these pollution problems?

2.3.1 How does boundary layer meteorology play a role in wintertime urban air pollution?

During the cold and dark winters of the Interior of Alaska, the absence of diurnal cycle combined with ice- and snow- covered surfaces creates a unique meteorological condition in the lower troposphere leading to the formation of shallow temperature inversions layer. In such cases, and under low-pressure-gradient forces at regional level, the topographic configuration and mountain orientation in the area constrain the low-level tropospheric circulation and the airflow in the planetary boundary layer (PBL) becomes locally quasi-laminar and regionally stagnant dominated mainly by local-scale circulation and radiative cooling (Fochesatto et al., 2015). Under the mentioned conditions, when a stagnant airflow makes contact with ice- and snow-covered surfaces, the air layers immediately above the surface cool down by infrared radiation loss forming a temperature inversion with the base at the surface. Figure 6 shows near-surface temperature gradient measurements for an annual cycle in Fairbanks, Alaska. During wintertime, long periods having air at 30m aloft more than 5°C warmer than air at 2 m are observed, demonstrating the ubiquity of strong surface-based inversions in Fairbanks.

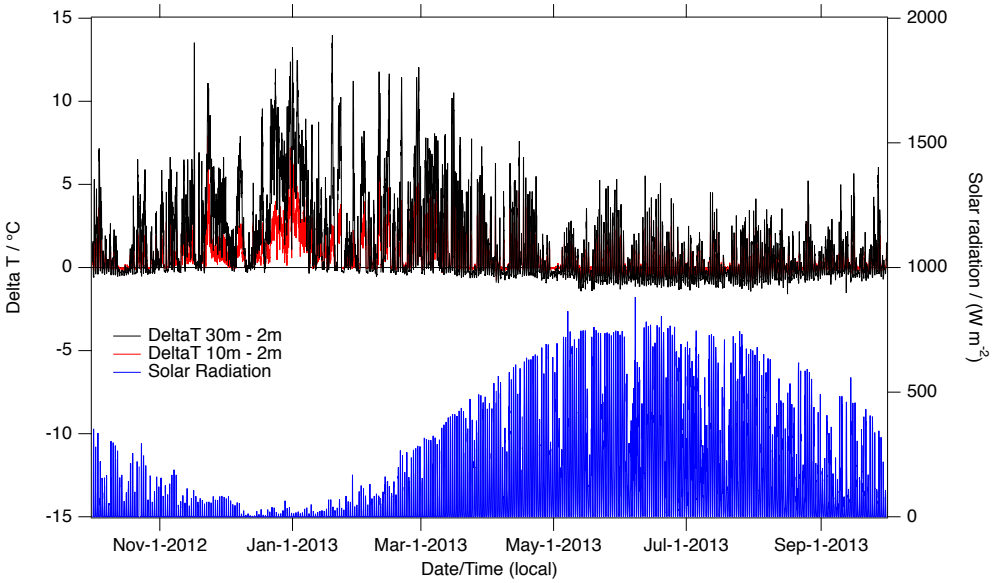

Figure 6. Temperatures at 2, 10, and 30meters above ground level and incoming solar radiation measured on a meteorological tower measured from the valley floor in Fairbanks, AK. Strong surface-based inversions are seen in winter months.

This layer is known as surface-based inversion (SBI) described earlier by Wexler (1936) and Billelo (1966) and by Bowling et al. (1968) in connection to the formation of anti-cyclonic (high pressure) meteorological conditions in Interior of Alaska. In the lowest layers of the atmosphere, this radiative cooling is manifested by a positive upward temperature gradient that extends to a height at which the surface cooling effect vanishes and the air mass "recaptures" the negative environmental temperature gradient. Changes induced by the radiative cooling process in the PBL (i.e., density gradients) promote the vertical stratification and therefore divergence of infrared fluxes (Fuggle and Oke, 1976; Estournel et al., 1983) instigating changes in the turbulent flux regime (Estournel and Guédalia, 1985). In some cases, a radiative equilibrium between the surface longwave radiation loss and incoming longwave radiation have been experimentally recorded (Malingowski et al., 2014). However, this radiative equilibrium is

fragile and therefore it can be disrupted by either the presence of low-level clouds (Wendler and Jayaweera, 1972; Jayaweera et al., 1975; Stone, 1997), the development of surface winds (Wendler and Nicpon, 1975; Hartmann and Wendler, 2005) or by the presence of shallow cold flows (i.e., drainage flows) consequently destroying the stratification (Mayfield et al., 2013; Fochesatto et al., 2015). The depth of the SBI is regulated by a number of factors, including local flows, quasi-total absence of shear-stress turbulence in the surface layer, and energy balance equilibrium between surface net radiation and atmospheric downwelling infrared (Overland and Guest, 1991). The presence of a SBI is important in regulating turbulent flux regimes, surface energy balance and deposition of chemical species and particulate matter (Cohen et al., 2007; Huff et al., 2011). Accurate representation of the SBI is central in the description of high-latitude air pollution problem specially when a connection between pollution sources and chemical receptors (Mölders and Kramm, 2010). Specifically, when there is a strong SBI, elevated plumes, such as from powerplants, may have reduced impact on ground-level air quality and conversely, ground-level sources are likely to dominate breathing air quality. Climatological and long-term studies of the Arctic environment reveal a consistent presence of SBIs in the North American Arctic (Maxwell, 1982; Kahl, 1990; Bradley et al., 1992; Walden et al., 1996) and in Eurasia (Serreze et al., 1992). The presence of the SBI was evaluated across the Alaskan climate regions, and its time variability was linked to large scale climate fluctuations (Bourne et al., 2010). In a similar way, studies covering Arctic and Antarctic stations by Zhang et al. (2011) evaluated the SBI space–time variability and its possible linkage to climatic fluctuations. Stable episodes are inter-dispersed with periods influenced by cyclonic activity advecting air masses from outside the region over Fairbanks and cleaning out the locally produced pollution.

Depending upon the synoptic condition, the flow above the SBI may also develop thermal inversions called elevated inversion layers. The elevated inversion layers occur between the SBI and the free troposphere, mainly as a function of specific synoptic large-scale flow. It has been hypothesized that elevated inversion layers may trap pollution regionally that can later recycle into the area, exacerbating pollution events caused by surface-based inversions. The occurrence of these elevated inversion layers promotes downward mixing of pollutants during ABL nocturnal-to-diurnal transition (Mayfield et al., 2013; Malingowski et al., 2014; Fochesatto et al., 2015).

This multilayered PBL structure is more likely to occur in Arctic and sub-Arctic locations during winter given the prominent synoptic large-scale dependence on surface and local thermodynamic parameters (Cassano et al., 2011). Fairbanks has been identified by the U.S. Environmental Protection Agency as an air-quality nonattainment area because of regular occurrences of severe wintertime pollution episodes, which are influenced by the SBI. The occurrence of an SBI combined with EIs can initiate two mechanisms of potential importance to the Fairbanks air pollution problem: 1) The presence of the SBI combined with an elevated inversions imposes an additional upper-level capping inversion for hindering the dispersion of pollutants at the regional level in a semi-enclosed basin where Fairbanks is located. 2) infrared radiative fluxes from the warm air aloft warming and eroding the SBI, enabling mixing between the SBI and the elevated inversions and changing consequently surface turbulent fluxes.

Overall, we summarize the open questions regarding boundary-layer meteorology:

- What are the horizontal and vertical scales of turbulence in this strongly stable environment?
- What dynamical and physical processes (e.g. synoptic forcing, down-slope flows,

radiative cooling, cloud radiative effects) affect atmospheric stability and turbulent mixing?
- What feedbacks are present and how do they affect stability and turbulence?
- How does boundary-layer meteorology affect atmospheric chemistry within the polluted surface layer, within lofted layers, and in the free troposphere?
- Do strong inversions prevent down mixing of lofted pollution plumes (e.g. powerplant plumes) and reduce that pollution's influence on ground-level air quality?

2.3.2 Can regional air quality models sufficiently resolve meteorology, emissions and impacts on atmospheric composition in urban areas?

Fairbanks, Alaska is classified as a "Serious nonattainment area" for the 24-Hour $PM_{2.5}$ National Ambient Air Quality Standard. Emissions from home heating and other sources, combined with stagnant meteorological conditions during cold winter episodes (-30 to -40 °C) result in daily-average concentrations of $PM_{2.5}$ that are hazardous to human health (> 35 μg m^{-3}). To reach attainment, there is a need to understand to what degree emissions, chemical processing, and meteorological conditions create high $PM_{2.5}$ levels. In addition, given the role of $PM_{2.5}$ in radiation, cloud, and fog processes, there is an additional question of how the high $PM_{2.5}$ concentration feeds back on the characteristics of the stagnant air mass. Motivating questions for Fairbanks in terms of air quality policy include:

- Can our state-of-the-art modeling tools recreate the pollution conditions experienced in Fairbanks?
- How much air pollution benefit is achieved from current and proposed pollution control measures?
- What is the role of atmospheric chemistry, snow surface chemistry, cloud and fog chemistry, and mixing with the free troposphere in producing or removing pollutants?
- How do these processes affect the effectiveness of pollution control measures?
- What role does the high concentration of $PM_{2.5}$ play in maintaining or destroying the stable, stagnant air mass?

A stagnant air mass combined with a stable stratified boundary layer limits the ability of air pollutants from different sources to interact chemically and results in spatially heterogeneous concentrations. In addition to meteorology and poor dispersion as factors contributing to pollution, poor air quality may influence the meteorological environment through modifications of cloud and fog properties and of the atmospheric and surface energy balance. The extent to which emissions of heat, water vapor and pollutants affect the structure of a stagnant stable atmosphere is unknown. Understanding these pollution and stable boundary layer interactions and potentially relevant feedback mechanisms is needed to make progress in terms of development of more process-based model representations of air pollution-boundary layer interactions. Such model representations are required in order to examine the benefits of pollution control measures and to improve air quality forecasts for cold wintertime conditions.

In a broader sense, high ambient concentrations in Fairbanks, as with other Arctic urban areas, contribute to increased levels of chemicals in the Arctic environment. After an episode, high concentrations pollutants are dispersed to the greater Arctic environment. Fairbanks is a convenient location to use as a case study of how a region with significant local sources, in this

case a polluted Arctic city, affects the broader Arctic environment. While other locations have their own unique air pollution sources and boundary layer dynamics, the information from Fairbanks will help answer fundamental questions about formation and fate of aerosols and their precursors including interactions with boundary layer dynamics and exchange with the free troposphere. An air pollution/stable boundary layer study in Fairbanks will ultimately help answer the science question: What are the implications of local (urban) Arctic emissions for the broader Arctic environment (in terms of haze, ecosystems, and climate)? During the periods between local pollution episodes, it will also be possible to study processes in cleaner background air masses during wintertime, including Arctic haze originating from more remote sources of pollution.

2.3.3 How do clouds/fog/aerosols and anthropogenic heat/water affect boundary layer structures in urban areas? Are there feedbacks?

During cold conditions typical of high-latitude winters, air may become saturated with respect to water or ice, resulting in the formation of liquid or ice fogs/low stratiform clouds, respectively. Such clouds change the particulate mass distribution with height, thereby impacting the vertical structure of radiative cooling. Specifically, the strong gradient at cloud top results in significant cooling there, creating a thermodynamically unstable situation and resulting in "upside-down" convection. Such convection begins to mix the atmosphere, further supporting cloud growth and development, and reducing the stable stratification of the lower atmosphere. This impact of clouds can be altered by the chemical and physical properties of aerosol particles present during cloud formation. Specific to nighttime conditions, a general reduction of droplet sizes associated with an increase in cloud condensation nuclei (CCN) concentrations increases the longwave emissivity of graybody clouds, generally with liquid water paths below 50 g m^{-2} or so (Garrett and Zhao, 2006). Such an increase in emissivity can increase the longwave cooling at cloud top, further enhancing the mixing effect of the cloud. The number of aerosol particles present can also alter the precipitation associated with cold clouds, with an increase in ice nucleating particles (INPs) enhancing snowfall and reducing the amount of water vapor available to form liquid droplets through the Wegener-Bergeron-Findeisen (WBF) mechanism (Wegener, 1911; Bergeron, 1935; Findeisen, 1938), thereby impacting cloud lifetime. Additionally, elevated CCN concentrations can change the riming efficiency of atmospheric ice crystals (Borys et al., 2003), changing the amount of mass associated with frozen precipitation. Finally, the coating of atmospheric aerosol particles by sulfates or organics may reduce their ice nucleation efficiency via deposition freezing, changing the phase partitioning of the atmosphere and thereby altering cloud microphysics and lifetime (Cziczo et al., 2009; Girard and Sokhandan, 2014), though the extent of this effect is still under debate (Sullivan et al., 2010). Through these mechanisms and the associated chemical processing of particles, clouds can have a significant impact on the properties of atmospheric particles, and vice-versa.

In addition to cloud effects, there are potential impacts of the surface on lower atmospheric mixing. Specifically, the presence of people can alter the sensible heat flux through enhanced surface heating over populated areas, a phenomenon known as the "urban heat island" effect (see also Section 2.4.5). Often such heating is a direct result of replacing trees with dark, absorbing construction materials (e.g. roofs, asphalt), however such effects are minimal during the Arctic night because they require incoming solar irradiance of the surface. In addition to the contribution of dark surfaces to urban heat islands, the heat introduced through heating of

buildings, vehicles and other entities may also play a small role. In Helsinki, the anthropogenic heat emissions have been estimated to reach 50 W m^{-2} in below freezing air temperatures (Järvi et al., 2014) thus being the primary energy input to the surface energy balance with substantial effect on the surface mixing conditions (Karsisto et al., 2016). Under dark conditions the role of human induced emissions can be expected to be even more pronounced. Overall these discussions lead to the following questions:

- What is the relationship between boundary-layer structure and clouds/fogs?
- What is the effect of pollution on clouds, (ice)-fogs and boundary-layer structure?
- What is the role of the urban heat island effect and anthropogenic heat emissions on mixing and dispersion of pollutants?

2.4 What are the impacts, both locally and regionally, of high latitude urban pollution?

2.4.1 What are the estimated health impacts of Fairbanks air pollution?

Decades of epidemiological research links ambient air pollution with a range of deleterious health outcomes among varied populations in the U.S. and globally (e.g. Burnett et al., 2014; Cohen et al., 2017). Health effects result from both short-term and long-term exposure to ambient air pollutants. Based on epidemiology studies conducted in different environments and populations around the world, long-term exposure to $PM_{2.5}$ is associated with increased risk of ischemic heart disease, stroke, chronic obstructive pulmonary disease, lung cancer, and lower respiratory infection (Burnett et al., 2014), as well as diabetes mellitus (Bowe et al., 2018), asthma incidence (e.g. Zheng et al., 2015), and other outcomes. Long-term exposure to ozone is also associated with respiratory mortality and potentially cardiovascular mortality (Jerrett et al., 2009; Turner et al., 2016). Short-term exposure to both $PM_{2.5}$ and ozone is associated with increased rates or emergency room visits and hospital admissions. While air pollution epidemiological research has been limited in Arctic environments, the available research indicates some consistency with epidemiological studies conducted elsewhere, as they find significant associations between short-term $PM_{2.5}$ exposure and hospital admissions in Fairbanks (Kossover, 2010), as well as reduced cases of respiratory outcomes during a woodstove intervention in a rural mountain valley community (Ward et al., 2017). Additional research is needed to elucidate potentially synergistic effects of simultaneous exposure to air pollution and cold temperatures.

To our knowledge, no previous study has estimated the burden of disease from air pollution in Fairbanks, specifically. It is feasible to calculate such an estimate by combining relative risk estimates from epidemiological studies in other parts of the world with the locally-specific population characteristics, disease rates, and pollutant concentrations. Without long-term epidemiological cohort studies conducted in Fairbanks, there are two key assumptions in this type of calculation. These assumptions are common to the leading Global Burden of Disease assessments by the Institute for Health Metrics and Evaluation (Cohen et al., 2017) and the World Health Organization (Prüss-Ustün et al., 2016). First, one must assume that the relative risk estimates found in other parts of the world apply to the local population of Fairbanks, though there would be differences in population characteristics, such as health status, potential confounders that have not been adequately controlled in the epidemiological studies, and access to health care (e.g. distance from hospitals). Second, the $PM_{2.5}$ Integrated Exposure Response

curves that characterize health risks at different concentration levels assume that different components and mixtures of $PM_{2.5}$ have equal effects on health. Since the local characteristics of the population and air pollution sources and mixtures may differ in Fairbanks compared with the cohorts that were the subjects of past epidemiological studies, some consideration of the potential influence of these factors on estimated health impacts is warranted. However, information is available to generate policy-relevant estimates of the air pollution burden of disease in Fairbanks using state of the science methods that are consistent with the Global Burden of Disease studies, without waiting for resource-intensive and time-consuming epidemiology studies to be carried out locally.

2.4.2 How toxic is wintertime urban aerosol?

Nonattainment of the 24-hr $PM_{2.5}$ standard in Fairbanks is mostly caused by residential wood-smoke emissions during the winter heating season (see Section 1.4). Extensive research has shown that smoke from wildfires, prescribed burning and residential heating is a significant health risk (see reviews by: Zelikoff et al., 2002; Naeher et al., 2007; Torres-Duque et al., 2008). Many individual wood smoke constituents are known to produce both acute and chronic adverse effects (Zelikoff et al., 2002; Torres-Duque et al., 2008) and a wide range of specific toxicologic effects of wood smoke have been identified in both animal and human models (Naeher et al., 2007; Torres-Duque et al., 2008). Clinical observations of populations with continued exposures to smoke show chronic symptoms of cardiorespiratory diseases (Torres-Duque et al., 2008). The deleterious effects of wood smoke emissions can be most severe for susceptible populations, including children, asthmatics, elderly and individuals with preexisting cardiopulmonary disease (Zelikoff et al., 2002). Biomass burning smoke is recognized by the World Health Organization as a probable human lung carcinogen (Straif et al., 2006; IARC, 2010).

Emissions from biomass burning, including residential wood burning, are a complex mixture of both gases and particles that are modified over time due to changes in plume temperature and plume dilution. Interaction of both smoke vapors and particulates with sunlight, oxidants, particle surfaces and particle liquid water also alter the mass and chemical composition of smoke. Studies have shown that for incomplete combustion in general, toxicities of aerosols can increase with photochemical aging (Antiñolo et al., 2015; Verma et al., 2015).

The environment into which wood burning smoke is emitted in Fairbanks and other northern communities during winter is unique due to extreme low temperatures and diminished photochemically-driven reactions, resulting in reduced, and possibly different, oxidation processes compared to the typical ambient environments studied. Smoke to be studied during ALPACA may be closer to primary emissions relative to what is seen in other areas. ALPACA provides an opportunity to study smoke emitted into unique ambient conditions.

There are many approaches to characterizing the health hazards associated with smoke. Given the limited population of northern communities, a practical way to assess the hazards of Fairbanks smoke would be by linking it to the larger body of research on biomass burning health impacts generally performed in warmer climates. This could be done by contrasting the toxicity of Fairbanks winter aerosol to what is reported in other studies. Two broad approaches that would be readily accessible and allow contrasts from earlier studies are through smoke analysis by both animal models and various assays. Examples of two possible approaches are detailed below.

Kim et al. (2018) recently reported on the toxicity of laboratory-generated biomass burning smoke under controlled conditions that included smoldering and flaming. With no chemical aging, these experiments may reflect the types of conditions expected in Fairbanks. Based on exposure of mouse models to collected smoke condensates, PM from different fuels and combustion phases were found to have appreciable differences in lung toxic and mutagenic potency, with many fuels more toxic on a per mass basis than those reported for burning of other materials. The results point to significant toxicity of fresh emissions from flaming biomass combustion.

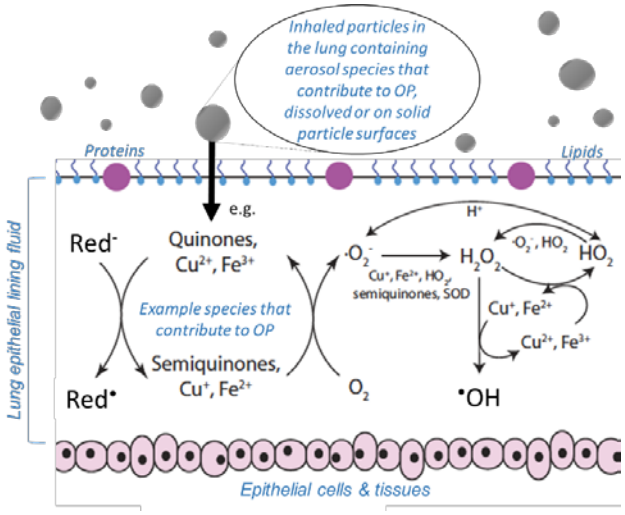

Figure 7. Schematic of loss of antioxidants (reductants) and formation of oxidants by components of inhaled particles in the lung epithelial lining fluid that can lead to oxidative stress. Translocation of soluble and insoluble components via the circulatory system can lead to similar chemical processes occurring at extrapulmonary sites. Red- represents a generic physiological reducing agent. Specific aerosol components, such as quinones and redox-active transition metals (e.g., Fe and Cu), catalytically transfer an electron from the reductant (Red-) to dissolved oxygen (O_2) leading to the formation of super oxide (O_2^-), which can be further converted by aerosol components to stronger oxidants, such as the hydroxyl radical (•OH). Such loss of antioxidant and generation of oxidants can lead to oxidative stress in living systems. This figure is adapted from Figure 1 of Lakey et al. (2016)

A major mechanism smoke PM is thought to cause adverse health effects is through oxidative stress, a biochemical imbalance in which the presence of oxidants (i.e., reactive oxygen species, ROS) in the body overwhelm its natural antioxidant defenses. A potentially potent mechanism for PM-induced oxidative stress is through the catalytic *in vivo* generation of oxidants in the lungs or other organs by certain particle components, such as humic-like substances (HULIS), a complex group of relatively high molecular weight organic compounds and a significant component of wood smoke (Graber and Rudich, 2006). Water-soluble transition metal ions (TMI), such as Fe(II) and Fe(III), are also a class of aerosol components that contributes to the so-called aerosol oxidative potential, OP (Lakey et al., 2016). They are also found in wood smoke emissions (Oakes et al., 2010), but to a much lower extent than HULIS. Note that water soluble Fe is also potentially important in SO_2 oxidation under dark conditions (Seinfeld and Pandis, 2006) (see Section 2.2.1). Measures of $PM_{2.5}$ oxidative potential (OP) with both cellular and acellular assays are of increasing interest as they can provide a more comprehensive and integrative health-relevant measure of ambient aerosol toxicity compared to traditional measurements, such as $PM_{2.5}$ mass concentration. Cellular assays widely utilized in aerosol toxicity work include the study of ROS generation by human, rat, or mouse lung epithelial cells (Cheung et al., 2010; Daher et al., 2012; Saffari et al., 2014; Decesari et al., 2017;

Shirmohammadi et al., 2017; Tuet et al., 2017). Acellular assays include electron spin resonance spectrometry, which measures the capacity of PM to release radical types of ROS such as environmentally persistent free radicals (i.e., semiquinone radicals), OH, and superoxides (Arangio et al., 2016). Assays composed of physiological antioxidants, such as ascorbic acid (AA) (Mudway et al., 2005) and glutathione (GSH) (Godri et al., 2011) or surrogates, such as the dithiothreitol (DTT) assay (Cho et al., 2005; Verma et al., 2014) involve measurements of the reductant loss when mixed with ambient particles as an indicator of reactions that lead to oxidative stress (see Figure 7, where AA, GSH or DTT represents Red-). In addition to these experimental techniques, there are efforts to develop a kinetic model of surface and bulk chemistry in the epithelial lining fluid, which explicitly treats mass transport and chemical reactions involving redox-active components (e.g., quinones, Fe, Cu), ROS, antioxidants and surfactants (Fig. 7; Lakey et al., 2016). It can estimate the production rates and concentrations of ROS in the lining fluid based on redox reactions, although it does not treat biological interactions and responses of the human immune system.

A range of studies has connected PM OP to adverse health impacts. Panel studies show that $PM_{2.5}$ OP is linked to markers of inflammation (Delfino et al., 2013) and microvascular dysfunction (Delfino et al., 2010; Zhang et al., 2016). Some population-based studies, which offer greater generalizability than panel studies, have found associations between various cardiopulmonary diseases and $PM_{2.5}$ OP (Yang et al., 2014; Bates et al., 2015; Weichenthal et al., 2016a, 2016b; Abrams et al., 2017). In Atlanta using the DTT assay, both receptor-based and chemical transport model source apportionment approaches indicate that elevated levels of OP across the metropolitan region were due to broadly-dispersed biomass burning and this PM OP was more strongly associated with cardiorespiratory adverse health effects than $PM_{2.5}$ mass (Bates et al., 2015; Abrams et al., 2017), demonstrating the both the utility of OP assays and importance of biomass burning on adverse health impacts. These considerations lead to the following questions.

- What are the levels of oxidative potential measured by various assays during winter pollution events and how do they compare to other regions?
- What is the $PM_{2.5}$ toxicity in terms of oxidative potential per mass during winter pollution events and how do they compare to other regions?
- What is the aerosol toxicity based on biological assays?
- What are the major chemical species driving the aerosol toxicity?
- Is the toxicity of northern climate winter pollution events unique relative to other locations?

2.4.3 What is the downwind fate of the pollution and what is the depositional footprint?

To assess the full scope of impacts of Fairbanks air pollution requires knowledge of the downwind fate and depositional footprint of the air pollution. It will also be necessary to understand the depositional footprint within the wider context of pollution transported from outside the region, including Arctic haze, and to elucidate the relative contributions of dry and wet deposition. Snowpack sampling is one means of assessing this, as snow samples provide a measurement of the integrated wet and dry fallout of pollutants from the atmosphere. Limited measurements of the electrical conductivity and the ionic composition of snow in the region surrounding Fairbanks indicated that the urban contamination is largely confined to the region within ~10 km of Fairbanks' inhabited boundaries (Shaw et al., 1993), consistent with findings

reported by Winchester et al. (1967) of halogens in snow. Huff et al. (2011) reported that mildly polluted air originating from Fairbanks is transported 20 km to the southwest by downslope drainage flow (Huff et al., 2011). The limited scope of spatial sampling in these studies combined with limited analysis leave numerous questions unanswered about pollution transport and deposition, including the role of aerosol age and particle size sorting (Winchester et al., 1967), depositional velocities (Huff et al., 2011), and secondary pollutant formation downwind of Fairbanks (Joyce et al., 2014).

Further research to address the depositional footprint of Fairbanks pollution should expand measured components and focus on greater spatial characterization of pollutants (prior studies were limited in their spatial scope), which can be achieved through ambient transect monitoring and more extensive snow sample collection together with measurements of vertical gradients of key species in the lowest levels of the boundary layer. These studies could address the questions:

- What are the removal processes for pollution outside of the source region?
- What chemical transformations happen downwind, and on what length scales?
- How does mixing affect downwind chemical transformations?
- What are the ecological impacts of pollution deposition in the region?

2.4.4 What are the health impacts of outdoor and indoor air pollution?

Outdoor concentrations of aerosol particles have been linked to many adverse health impacts (Dockery et al., 1993; Pope and Dockery, 2006; Bell et al., 2008), yet the vast majority of our time is spent indoors (Meng et al., 2005a; Turpin et al., 2007). Consequently, most of our exposure to aerosol is in the indoor environment, but in many cases that is as a result of outdoor air pollutants getting indoors (Meng et al., 2005b; Johnson et al., 2017) especially in indoor spaces without large aerosol sources. Since high aerosol concentration periods in the wintertime in Fairbanks, AK are linked to extreme cold, people are almost certainly indoors the vast majority of the time. Figure 8 shows that high pollution episodes in wintertime Fairbanks are associated with low outdoor temperatures and an increase in the $PM_{2.5}/\Delta CO$ ratio indicating lower combustion efficiency typical of woodstoves (Heringa et al., 2011, 2012). Since this combustion process occurs indoors and many woodstove seals leak, indoor concentrations may be higher than outdoor concentrations for homes which utilize wood fuel for heating (McNamara et al., 2017; Ward et al., 2017). From a health and exposure perspective, understanding the differences in exposure for homes which utilize wood heat compared to those home that do not is an important science question. Chemically resolved indoor and outdoor measurements of aerosols and trace gases would help to answer those questions. Beyond the health and exposure issues, there are interesting scientific questions that can be addressed as well. Given the strong temperature differences between indoors and outdoors, there will be a difference in the chemical composition of wood smoke particulate matter if concentrations are similar. Temperature gradients between indoors and out can strongly drive partitioning of semi-volatile species in the aerosol (Johnson et al., 2017). For organic aerosol the volatility basis set framework (Donahue et al., 2006) can provide information on what to expect from relative temperature difference for indoor wood smoke at 20°C to outdoor temperatures approaching -40°C. Under typical conditions, this temperature change from outdoors to indoors would be equivalent to an increase in volatility of several decades in $\log_{10}$ of saturation concentration. Aerosol composition measurements made indoors and outdoors would help to elucidate the wood smoke volatility profiles at these extremely low temperatures and provide useful information for regulatory and

general atmospheric chemistry models. Typically, wood smoke and biomass burning emission measurements have been made at relatively high ambient temperatures with further heating (e.g. Huffman et al., 2009). Probing the lower ambient temperatures seen in high latitude areas adds important observational data for these emissions in sensitive environments.

Exploring the volatility profiles at a range of temperatures would also help resolve the disconnect between typical $PM_{2.5}$ measurements used in health studies and what outdoor exposure actually is in cold environments. Filter-based, Federal Reference Method $PM_{2.5}$ measurements are made by collecting a filter in ambient conditions and then transferring it to a laboratory where the filter is weighed after being re-conditioned to 22°C and 35% relative humidity. For most health effects studies of $PM_{2.5}$, the conditions of the $PM_{2.5}$ measurement are not much different than the conditions of outdoor human exposure. However, that is not the case for much colder Arctic locations during winter. A better understanding of the volatility profiles will help us understand the difference between human exposure in cold Arctic weather compared to conditions represented in most epidemiological studies. In addition, the changes occuring to the $PM_{2.5}$ as it enters the human airways are more dramatic when the outdoor weather is very cold and dry. A better volatility profile will aid understanding of how the particles and gases affecting the human system are different than what is measured at ambient conditions or in a gravimetric laboratory measurement. These considerations lead to these questions:

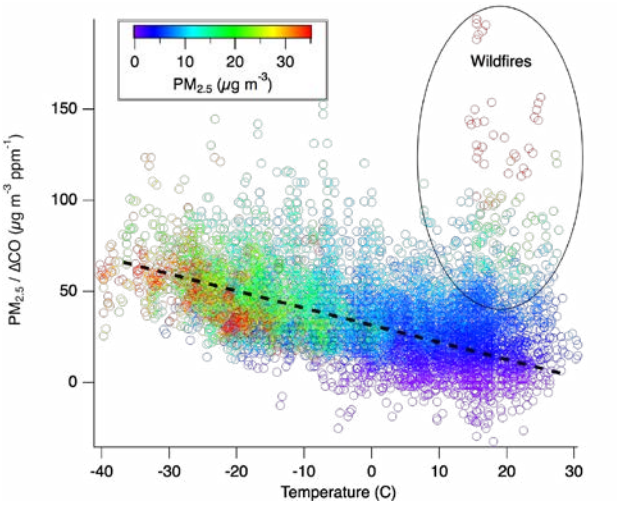

Figure 8. Hourly $PM_{2.5}$ to ΔCO ratios for Fairbanks AK during 2017. The values are colored by the ambient $PM_{2.5}$ concentration up to 35 μg m^{-3}.

- What are the pollution levels inside homes using woodstoves in Arctic conditions?
- What is the volatility profile for the organic aerosol both outside and inside houses using a woodstove?
- Based on the volatility, how are the emissions from woodstoves different in Arctic climates than in mid-latitude climates and how do the indoor and outdoor emissions compare?
- How does outdoor exposure to $PM_{2.5}$ differ in Arctic climates relative to climates where most epidemiological studies have been conducted?
- How does the linkage between exposure and Federal Reference Method $PM_{2.5}$ measurements differ in Arctic climates relative to climates where most epidemiological studies have been conducted?

2.4.5 What are the potential climate effects associated with wintertime air pollution?

Wintertime air pollution in the Arctic may be responsible of potential local climate effects through aerosol-cloud interactions or heat island effects.

The remote Arctic experiences higher levels of particulate pollution between winter and late spring (Quinn et al., 2007). They are often the result of intrusions of aerosols termed "Arctic haze", generally concentrated in the lowest few kilometers of the Arctic atmosphere (Brock et al., 1989). The deposition of black carbon from anthropogenic emissions, originating either locally in the Arctic or transported from mid-latitudes has an important, but as yet poorly quantified effect on Arctic climate via albedo feedbacks (AMAP, 2015). Arctic pollution aerosols may also modify the microphysical properties of clouds, especially increasing droplet number concentration and decreasing cloud droplet effective radius through changes in cloud condensation nuclei (CCN) (Shaw, 1986; Curry et al., 1996). While most global and lower latitude studies have focused on shortwave indirect effects, efforts dedicated to the polar regions have suggested the particular importance of indirect aerosol effects on the longwave radiative fluxes in the Arctic.

A number of studies (e.g. Garrett et al., 2002; Garrett and Zhao, 2006; Lubin and Vogelmann, 2006; Menon et al., 2008) have examined a possible impact of Arctic aerosols on regional climate in winter through changes in the longwave thermal emissivity of low-level clouds. Whereas low- and middle-latitude clouds are thicker and less clean, Arctic clouds often have relatively low liquid water paths and hence tend to emit as graybodies. As a result, Arctic clouds are expected to be particularly susceptible to aerosol-induced changes in emissivity (Zhao et al., 2012). A recent work of Zhao and Garrett (2015) using direct measurements of the surface radiative flux balance from Barrow in Alaska found an average 5.8 W/m^2 increase in monthly mean surface cloud longwave radiative forcing associated with elevated haze levels, with a maximum increase of 12.9 W/m^2 in February.

Roughly 50%–70% of the net effect of aerosols on surface cloud radiative forcing is caused by changes in cloud particle effective radius for liquid clouds. Interestingly, the remainder suggests some amplification of the aerosol first indirect effect caused by unidentified atmospheric feedbacks that enhance cloud water path (Tietze et al., 2011). This can be the consequence of reduced precipitation or feedbacks between cloud radiative forcing and atmospheric dynamics (Garrett et al., 2009). Arctic pollution aerosol may also impact ice nucleation by modifying the characteristics of INPs. However, aerosol-ice microphysical interactions, and their potential impact on the regional cloud cover, thermodynamics, and mesoscale dynamics in the Arctic, remain highly uncertain.

Air pollution in high latitudes is closely connected with meteorological conditions (Section 2.3.1) (Mayfield et al., 2013; Fochesatto et al., 2015) due to particular properties of Arctic boundary layer (deep inversions, lack of solar radiation during winter time etc.). That is why in Arctic cities urban heat island (UHI) and urban wind speed are natural predictors of air quality regime. Urban heat island properties in high latitudes can be derived from satellite imagery (Konstantinov et al., 2015) but the most direct way is to use in-situ temperature measurements.

During Urban Heat Island Arctic Research Campaign (UHIARC) (Konstantinov et al., 2018) in Eastern Arctic strong warm temperature anomalies were revealed in cities of European part of Russia (Apatity, Vorkuta) and Western Siberia (Salekhard, Nadym, Novy Urengoy). Such persistent temperature anomalies are frequently referred to as urban heat islands (UHIs). The

mean wintertime magnitude of these temperature anomalies (the UHI intensity) was found to be between 0.8˚C and 1.4˚C. Extreme UHI intensities up to 7˚C were observed during cold anticyclonic weather conditions. These UHIs may reduce the strength of surface-based inversion layers, increasing vertical dispersion of pollution and potentially decreasing surface pollutant concentrations. Low population areas, which would be expected to have weaker UHI effects, may thus have stronger surface-based inversions causing worse dispersion and higher pollution. Positive social impacts of the UHI could be expected for the Arctic cities due to the possible mitigation of harsh frost conditions and decreasing wintertime fuel consumption (Davies et al., 2008; Kolokotroni et al., 2009). In summary, these considerations lead to the following questions:

- How might the outflow from Arctic cities and industrial facilities affect regional haze and thus cause aerosol-induced climate effects?
- How might urban heat islands moderate surface-based inversions and thus affect dispersion of pollution?

3 ALPACA study design

3.1 Targeted study questions for ALPACA:

The discussion above motivates that there are many open questions regarding wintertime air pollution processes under cold and dark conditions. The key observational gaps that have been identified above need to be closed to make progress on answering these questions. Therefore, we propose a field investigation in Fairbanks, Alaska in January/February 2021. This field study needs to bring together multiple investigators and overlaps the mandates of multiple funding agencies, so coordination of the project is necessary. We propose a core set of observations that are designed to answer open physical and chemical questions regarding this problem. The identified key questions are:

Question 1: What is the role of primary versus secondary emission sources impacting ground-level air quality?

Question 2: What chemical processes are occurring that transform gaseous precursor sulfur, nitrogen, and organic species into secondary aerosol?

Question 3: What role does low temperature play in these pollution problems, including semi-volatile partitioning, ice formation, and the confounding factor of emission source changes with temperature?

Question 4: What affects the boundary layer structure? How does that structure affect dispersion and chemistry?

Question 5: Can regional models simulate emissions, dispersion, and chemistry in the cold and dark?

This study will improve our understanding of air pollution in cold and dark places and has many impacts on societally relevant questions. Therefore, the core study is also coupled to impacts and forms the basis for further questions:

Question 6: How can this information be used to improve public understanding of this problem, answer questions from stakeholders, and improve air quality?

Question 7: What are the health and toxicological effects of these particles?

Question 8: What are the downwind impacts on ecosystems and regional climate?

These questions and field activities are summarized in Figure 9.

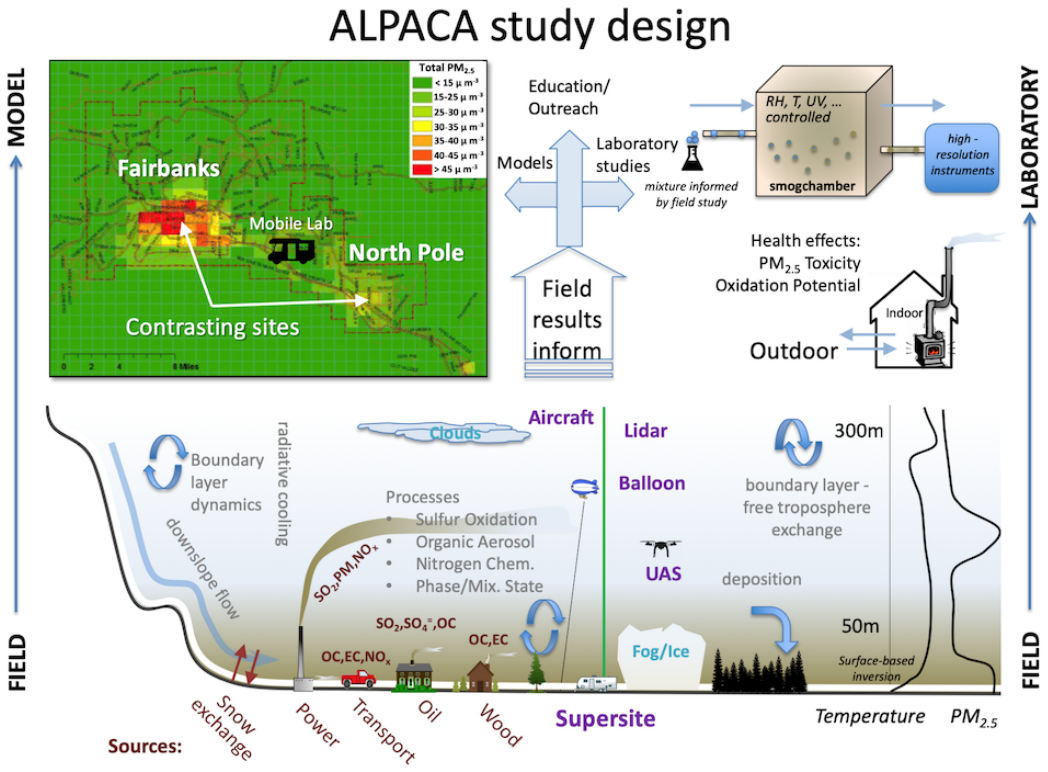

Figure 9. Schematic of the ALPACA field campaign. Modeled $PM_{2.5}$ concentrations on the top left are from Alaska DEC (2014b).

3.2 Vertical profile observations:

In order to comprehensively address the questions introduced in the above sections, it is clear that vertical profiling of the lower atmosphere is critical. Over recent years a variety of unmanned aerial systems (UAS), including both fixed- and rotary-wing aircraft as well as tethered balloons have been used to provide such information (e.g. de Boer et al., 2018) in the Arctic and beyond. Thanks to rapid development of both aircraft systems and sensors, UAS are providing revolutionary perspectives of the vertical and horizontal variability of key quantities, while simultaneously offering flexible deployment options allowing the user to sample previously inaccessible portions of the atmosphere. An example of this that is very relevant to the ALPACA effort is the ability of UAS to sample the lowest portions of the atmosphere, (e.g. surface to 100 m altitude), in great detail. This low altitude sampling is difficult to do using larger research aircraft due to safety concerns associated with extended low-altitude flight. Additionally, UAS offer a less-intrusive measurement platform than traditional manned research

aircraft, whose repeated presence at low altitudes would likely constitute a disturbance to residents of an urban study area. Piloted aircraft could potentially be used to measure higher altitudes and limited profiling at airports by using missed approaches.

In order to support the ALPACA effort, UAS and tethered balloons would be instrumented to measure aerosol chemical and physical properties, gas concentrations, winds and turbulence, and thermodynamic quantities (See Table 2). Such a configuration would allow the team to sample the structure and evolution of these quantities throughout the depth of the inversion layer. Routine profiling of the atmosphere between the surface and 1 km altitude would document the evolving state of this atmosphere and could act as a highly-portable tower system to provide information to document the role of the surface and winds in mixing the lower atmosphere, as well as the associated impacts on pollution concentrations, the evolving particle size distribution, and ongoing chemical reactions. Such profiling could occur nearly continuously during inversion events, and on a 1-2 time per day timescale at other times in order to both provide a background state and to capture the onset and decay of such events. Similarly, the ability to sample across horizontal scales can offer insight into key gradients governing pollution accumulation and transport, existing in both atmospheric and surface properties.

Table 2. Proposed UAS-based and tethered-balloon-based measurements

Measurement	Technique	Questions
Thermodynamic soundings: Temperature, humidity, pressure, wind	UAS-borne meteorological sensors	4,5
Remote sensed surface temperature and IR fluxes	IR radiometry	4
$PM_{2.5}$ and chemical measurements	Optical particle counters, In-situ sensors	1,4,5
Turbulent fluxes of sensible and latent heat and momentum	Acoustic anemometry, fast temperature sensors	4
Reactive gases, sulfur dioxide (SO_2), nitrogen dioxide (NO_2), ozone, etc.	Electrochemical sensors, optical absorption spectroscopy	1,2,4,5
Cloud particles, INPs	Particle counters, samplers	3,4

We envision that UAS measurements could cover a variety of scales. For one, these systems could be paired with supersites and mobile labs to enhance information content on vertical structure over these surface sites. Additionally, they could be operated from hilltops surrounding the downtown Fairbanks area in order to provide a background state to the polluted atmospheric core. Finally, they could be operated downstream of the downtown corridor in order to map the outflow of Fairbanks pollution into the larger environment.

Tethered or remote controlled balloons (e.g. Spirig et al., 2004; Neff et al., 2008; Ferrero et al., 2016; Creamean et al., 2018) can provide vertical profiling with longer duration measurements than typical UAS and could be deployed at a supersite or other locations of interest. Chemical sensors for UAS and/or tethered balloons are being actively developed at this time and can provide vertical profiles of aerosol particles and reactive species such as ozone, NO_2, and SO_2. These measurements will be used to determine the relationship between thermodynamic stability and pollutant dispersion. Particularly in the case of SO_2 (Morris et al., 2010), different sources (e.g. powerplant emissions from high stacks versus home heating emissions from low stacks) and their mixing or lack of mixing can be probed by these vertical profiles. The observations will also be critical for testing models.

3.3 Supersite observations:

Because many open questions involve chemical and physical interactions between gases and particles, co-located observations of many species are needed to observe and understand these interactions. The observations will also be used to validate and improve chemical modeling efforts. Table 3 shows a listing of species to be measured. Measurements would be taken at high time resolution (hourly or sub-hourly), although some filter sampling may require longer durations. To connect this effort to past studies in Fairbanks, this supersite should be located near the NCORE sampling site. Past work has shown that the State Office Building site is sufficiently close to NCORE to be considered equivalent with respect to fine particulate matter ($PM_{2.5}$). It is useful to have a second supersite to compare different source mixtures, and the North Pole location at Hurst Road is an excellent choice.

Specifically related to the questions of emission sources and their impacts on ground-level air quality, the gas and particle measurements will give us greater insight. The basic chemical and aerosol monitoring at NCORE would be enhanced by a larger number of gases measured including reactive species such as nocturnal nitrogen oxides (e.g. NO_3, N_2O_5), radical precursors (HONO, HCHO) and aerosol mass spectrometry (which has not been carried out in Fairbanks before). The AMS and single-particle mass spectrometers will give high-time-resolution observations that greatly improve upon the 24-hour filter samples that have been used in the past and will give greater chemical detail, with single-particle measurements also resolving internal/external mixing effects. Vertical profiling by long-path DOAS, balloons, LIDAR, and/or UAS will give data that can validate point source plume dispersion models and thus will address the question of which emissions impact ground-level air quality.

Prior work in Fairbanks has seldom had sufficient constraint on radical budgets and reactive species to address pollutant transformations under cold and dark conditions. While there have been some measurements of nocturnal nitrogen oxides at the ground level (Ayers and Simpson, 2006; Apodaca et al., 2008; Huff et al., 2011), those results along with 1-D modeling (Joyce et al., 2014) indicate that the near-surface vertical profile and mixing effects are important, thus motivate the need for vertical profiling, potentially using long-path DOAS, LIDAR, balloons, and near-surface towers. Sulfur chemistry has not been explored in Fairbanks, Alaska, despite the fact that sulfate is a significant part of $PM_{2.5}$ and that chemical models cannot reproduce these observations (Mölders and Leelasakultum, 2012). Sulfate, along with ammonia, affects aerosol pH, which can then affect partitioning of nitric acid/nitrate. Therefore, measurements of ammonia and ammonium, nitrate and gas-phase nitric acid, none of which have been done in Fairbanks, are necessary. These complex interactions between inorganic acids and bases need to be considered in designing pollution mitigation strategies.

During a 6-week field campaign in wintertime, there is very likely to be a large range of temperatures likely approaching -40°C and going up to near freezing. This range of conditions provides a natural laboratory for detailed study of the semi-volatile partitioning of organics through a combination of gas phase / particle phase measurements (e.g. TAG+TOF-MS, FIAGERO+TOF-MS, CHARON-PTR-TOF-MS, and combinations of AMS / single-particle MS and PTR-MS). These data will address the role of low temperature in these pollution problems. The proposed combination of aerosol composition measurements will provide a comprehensive understanding of the full aerosol population and chemical species, allowing detailed process-level questions to be probed. Combined CCN, INP, and polarization LIDAR work will improve our understanding of cloud particle phase and thus allow exploration of the role of phase on

pollution transformations. Gas-phase and snow chemical composition measurements will probe the role of air-snow interactions in the unique urban snow setting.

Table 3. Species to be measured at the downtown Supersite

Measurement	**Technique**	**Questions**
Basic measurements: Temp., humidity, pressure, wind	In-situ sensors (existing at NCORE)	4,5
Cloud base height	Celiometer	3,4
Inversion strength	Tower / tethered balloon	4,5
Liquid water/ice content, fog occurrence	PVM, FSSP, or other	3
Radiation (shortwave and longwave)	Radiometers	3,4
Photolysis rates	2-pi radiometer	2,5
O_3, CO, NO, NO_2, NO_y, SO_2	EPA methods (existing at NCORE)	2,5
High specificity NO_2	Blue converter or CRDS	1,2,5
NO_3, N_2O_5	CRDS	2,5
NH_3, HNO_3 (gas phase), HONO	CIMS, mist chamber/IC	1,2,5
Speciated VOCs	PTR-MS; CIMS	1,2,5
Hourly PM2.5 mass concentration, regulatory monitor	BAM, FRM (existing at NCORE)	1,5,6
Aerosol composition (24-hour) using XRF, IC, TOT/TOR EC/OC methods	URG-3000, SASS (at NCORE)	1,2,3,5
Hourly metal and sulfur composition by XRF	Horiba PX-375 or similar	1,2,5,6
Non-refractory PM_1 aerosol composition	Various types of aerosol mass spectrometers	1,2,5,6
$PM_{2.5}$ / PM_{10} composition	offline aerosol mass spectrometry	2,3
Particle number size distribution measurements	SMPS, OPC, APS	1,3,5
Aerosol particle mixing state; refractory aerosol composition	Single-particle mass spectrometer	1,2,3,5
Size/time-resolved composition (~hourly)	Cascade impactor	1,2,5
Gas-particle partitioning & OA molecular composition	TAG+TOF-MS; FIAGERO+TOF-MS; CHARON-PTR-TOF-MS	1,2,3,5,6
Black carbon concentration	SP2	1
Isotopic measurements (sulfate, nitrate, ammonium, carbon)	Filter samples/post-analysis	1,2
Water-soluble particle composition	PILS	1,2,5
$PM_{2.5}$ Oxidative Potential (OP)	Sampler / post analysis and/or online system	7
PM2.5 toxicity	Sampler / post analysis and/or online system	7
Ice nucleating particles (INPs)	Chamber and/or filters for post-analysis	3,4
Cloud condensation nuclei (CCN)	CCNc	3,4
Vertical profiling NO_2, HONO, HCHO, O_3, NO_3	Long-path DOAS	1,2,5
Aerosol particle vertical profiling	Micro-LIDAR (e.g. LATMOS system at 532 and 808nm)	1,4,5
Turbulence (on modeling gridscale ~1km)	Laser scintillometers	4
Vertical temperature structure	SODAR (e.g. Remtech)	4
Near-surface monitoring of gas/particles	Walk-up tower	1,4
Snowpack composition	Ion chromatography	1,2,5

Detailed measurements of atmospheric stability and mixing would also be carried out at the supersite by towers, scintillometers, and vertical profiling on UAS, tethered balloons, and LIDAR/radars. These would be coupled with surface energy balance measurements and cloud

radiative properties to determine their effects on pollutant mixing and transformations. These aspects are critical because they are necessary to determine airflow from sources, and to resolve thermodynamic and turbulent boundary layer structures that affect pollutant dispersion and mixing. In the preparation for the ALPACA experiment, it is necessary to determine turbulence, winds, and flow structures (e.g. downslope net flow) around the selected sites. Surface radiation balance plays a strong role in formation of shallow surface-based inversions and needs study in the region of potential supersites.

The data measured at the supersite will greatly improve our observations of chemical species and therefore help test and improve those models. The shallow vertical structure of the pollution in Fairbanks represents a challenge to regional modeling, and an important test case for addressing how these models can simulate dispersion and chemistry. The measurements of gaseous and aerosol composition, including volatility of organics will greatly improve our understanding of emissions, which can then be integrated into improved modeling.

The downtown supersite will also be a highly visible aspect of the work that can be used to increase public awareness of the problem and what the public can do to improve air quality. We would develop outreach materials that use the advanced measurements at the site to improve public understanding. Samples taken at the site, which can be put into a much richer context through the complementary measurements, would be used in subsequent toxicological studies to address health impacts of Fairbanks $PM_{2.5}$.

3.4 Horizontal / mobile laboratory observations

The strengths of deployment of sampling equipment at a fixed supersite such as that described above include the ability to co-locate multiple pieces of instrumentation for quantification of complementary species and the ability to have extended temporal coverage to improve data statistics. However, during any air quality field campaign, there also are benefits to application of flexible mobile sampling platforms ranging from a mobile laboratory equipped with state-of-the-art instrumentation to simpler systems making use of portable devices equipped with small sensors or filter sampling. The primary benefits are related to the ability to cover a larger geographic area with respect to data collection, to perform, Lagrangian studies downwind of specific emission sources, to investigate plume dispersion and chemistry, to identify pollution ‘hot spots’ not identified by stationary monitoring networks (e.g., Apte et al., 2017), and to adapt sampling location(s) based on variability in meteorology. Previous flexible mobile air quality sampling campaigns have focused predominantly on urban areas in mid-latitude populated areas such as Zurich, Pittsburgh, and Oakland (e.g., Mohr et al., 2011; Li et al., 2016; Apte et al., 2017), although some work has been done at high latitudes (Pirjola et al., 2017). Example instrumentation deployable on a mobile platform is summarized in Table 4.

Depending on the mobile platform deployed and the analytical equipment integrated, various scientific questions can be addressed. Here, we focus on those relevant to wintertime air quality in Fairbanks. It should be stressed, however, that due to the flexibility of these mobile platforms, analogous scientific questions may be posed for any domain in which these platforms are utilized, including other high-latitude urban areas.

Deployment of a mobile laboratory in the near downwind area of a known source would allow for characterization of how air pollutant concentrations are impacted by that source. In the case of high-latitude winter, it would also allow for comparison of collected data to that collected in

more temperate regions. For example, in areas impacted by wood smoke, aerosol mass spectrometer data that provide spectra of biomass burning aerosol (BBOA) can be collected. Comparison of the chemical nature of BBOA in each location would allow determination of whether or not the cold temperatures in a location such as Fairbanks cause condensation of more volatile species than would be in the particle phase in more temperate regions.

Table 4. Example instrumentation deployable on a mobile air quality platform.

Species	Example Instrumentation	Questions
Size-resolved composition of non-refractory particulate matter (PM)	High-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS); aethalometer (BC)	1,3
Ozone (O_3)	UV photometer O_3 analyzer	2
Carbon monoxide (CO)	CO analyzer (Cavity-based absorption)	1
Carbon dioxide (CO_2)	IR absorption CO_2 analyzer	1
Sulfur dioxide (SO_2)	Pulsed fluorescence SO_2 analyzer	1,2
Nitrogen oxides (NO, NO_2, NO_x), NO_y	Chemiluminescence NO-NO_2-NO_x analyzers	1,2
Volatile organic compounds	Proton transfer-reaction (PTR) mass spectrometer	1,2
Temperature, relative humidity, pressure, wind speed/direction, light, boundary layer height	Meteorological station; radiometer; ceilometer	1,4

Similarly, atmospheric chemistry within a plume can be investigated through a Lagrangian application of a mobile platform – in this case at ground level. For example, near an expected source of sulfur the sulfur dioxide (SO_2) to total sulfur ratio can be measured at high temporal resolution downwind using an SO_2 monitor and an aerosol mass spectrometer. With the wind speed and distance downwind of the source, the time since emission of the sulfur can be estimated, and the time scale for the conversion of the SO_2 to sulfate can be found. The dependence of this time scale on other parameters measured by the instrumentation aboard the mobile laboratory can also be investigated, providing insight into the processes occurring.

Vertical pollutant distribution can be investigated by a mobile facility, as well. For example, in an area such as Fairbanks where mixing height is expected to be very shallow, the mobile facility can be driven repeatedly up and down a hill (such as that between two supersites) to consider the vertical gradient in pollution – which will give an indication of the mixing level height, as well as confirm the extent of horizontal dispersion below and above the mixing height. Correspondingly, horizontal dispersion can be investigated downwind of a known source by repeatedly driving cross-wind patterns to identify horizontal plume dimensions. These measurements will provide corroborating data for simulations of physical transport in the troposphere. Measurements in snow samples will also provide information about the deposition of pollutants to the snowpack and could be carried out prior during and after the campaign to provide a wider context.

Despite the advantages associated with mobile sampling, there are several challenges related to this approach that must be overcome with any deployment. Regardless of location, adequate power to operate the equipment must be available and should not, if possible, be generated in a manner that could lead to self-contamination. In addition, data must be adjusted spatially to account for differences in vehicle location when the sample(s) entered the inlet(s) and that when instrumentation detects the sample (a so-called lag time). Specific challenges associated with deployment in wintertime in high-latitude areas (beyond delivery of the laboratory to the

location) include those related to temperature (e.g., differences between temperature inside the mobile laboratory and the ambient atmosphere; availability of warm storage areas for the mobile laboratory when not in operation to avoid multiple start-up/shut-down of certain instrumentation).

3.5 Coordinated laboratory studies

The discussions above have shown that there are significant knowledge gaps in fundamental chemistry in the following areas:

- Sulfur oxidation mechanisms
- Semi-volatile partitioning
- Particle phase reactions that may convert semi-volatile species to less-volatile species
- pH and phase effects on chemical reactions
- Heterogeneous nitrogen chemistry (e.g. N_2O_5, $ClNO_2$, etc.)

These questions will partly be addressed by the field measurements. However, the interpretation of ambient measurements is often challenging due to the complicated interplay between chemical reactions and particle phase properties that are continually changing in the atmosphere. To single out potentially important reaction pathways and physical properties, we suggest undertaking a series of structured laboratory chamber experiments. Ideally, the chamber experiments will come after the ambient observations so that as new mechanistic questions arise, they can be studied in detail. We aim to understand the order of importance of specific reactions and will also aim to derive reaction coefficients to inform the set-up of the chemical mechanisms in the models.

In order to simulate chemical reactions under wintertime Arctic conditions, it is essential to make observations of the gas and particulate phase in the field including the chemical composition, abundance, relative humidity, radiation and temperature. These measurements will inform the design of the chamber studies, which afford greater control over temperature (covering at least –20°C to +10°C), relative humidity (10 % to 100 %), and UV radiation.

Sulfur oxidation mechanisms: Following the discussion from Section 2.2.1, the main experiments to be conducted need to address the question of what oxidizers are responsible for the formation of secondary particulate sulfate. This would include quantification of secondary sulfate yield with the above mentioned varied physical parameters to test (i) photochemical pathways with a range of OH and O_3 concentrations, (ii) aqueous phase reactions with a dedicated set of experiments focused on the role of RH and particle phase, and (iii) the introduction of HCHO (Moch et al., 2018) and transition metals such as iron (Warneck, 1999), manganese (Berglund and Elding, 1995) and copper (Conklin and Hoffmann, 1988a, 1988b) to explore non-photochemical pathways. Oxidation of SO_2 by NO_2 is highly pH dependent, so it is not clear if this process happens in Fairbanks or not (Guo et al., 2017b). Aerosol mixing state is a key component of these particle-phase chemical mechanisms.

Semi-volatile partitioning, particle pH and particle phase reactions: The partitioning of organic semi-volatile species is subject to a complex interplay of ambient physical conditions (temperature, RH), particle pH, phase, and mixing state of particles. The last three aspects modulate the type of uptake reactions and limitations and have been partly discussed in Sections 2.2.3 and 2.2.4. It is important to realize that not all combinations of variables can be tested, hence the input from the field observations are essential to prioritize chamber experiments. One basic set of experiments will be the quantification of semi-volatile species uptake under the range

of temperature and RH conditions listed above. The concentration of each individually chosen species will then be monitored in the gas and particulate phase. Particularly varying RH as a surrogate for the particle phase (solid to liquid) will inform about potential reaction mechanisms. For example, Li et al. (2015) investigated ammonium uptake in SOA particles in laboratory studies and found that at low RH (solid particles) uptake is limited by diffusion while at higher RH other limitations play a role. These measurements will be complemented by both isothermal evaporation and thermal-desorption methods to probe the actual volatility of the particles, which will be used as a feedback to determine how particle phase state impacts the volatility of the SVOCs present over Fairbanks.

However, partitioning does not only depend on physical parameters but also on the chemical properties of the aerosol particles. Reactions in the particle phase can increase or decrease the partitioning of a species into the particle phase. For example, Gong et al. (2018) find that the uptake of levoglucosan to organic particles and ammonium sulfate is different over a range of RH conditions. For aqueous sulfate particles the uptake decreases with increasing RH, which was attributed to salting mechanisms. For organic particles, the uptake increased between 10 and 40 % RH and then stagnated. Here, kinetic and thermodynamic limitations were implied for below 40 % and greater 40 %, respectively. Mixing state of the particles, as discussed in Section 2.2.5 may also affect these processes and needs further laboratory investigation.

For acidic particles, SOA can be formed either through condensation onto the particle or through acid-catalyzed reactive uptake forming, presumably, low-volatility organo-sulfates (Pye et al., 2013; Lopez-Hilfiker et al., 2016). Acidic particles have also been shown to enhance multi-phase oligomer formation in α-pinene SOA (Jang et al., 2002), which could alter the phase-state and resulting volatility. Studies have shown that thicker coatings of SOA limit the acid-catalyzed reactive uptake pathway (Riva et al., 2016; Zhang et al., 2018). Based on these previous laboratory studies and with the input from the ALPACA field observations, a series of experiments can be designed to understand the physico-chemical properties, and potentially health impacts of the aerosol over Fairbanks in the wintertime and their implications.

Heterogeneous nitrogen chemistry: As discussed in Section 2.2.2, cold temperatures, high NO_x inputs, and low photolysis rates combine to provide favorable conditions for heterogeneous nitrogen chemistry via N_2O_5 and/or HONO. However, removal of ozone by pollution-sourced NO can lead to reduced or near-zero ozone mixing ratios at the surface, making this chemistry highly dependent upon vertical mixing (Joyce et al., 2014). When N_2O_5 is present, it undergoes heterogeneous reaction with water (e.g. Brown et al., 2006b) to form nitric acid and remove NO_x, and with aerosol chloride to form nitryl chloride ($ClNO_2$) (e.g. Osthoff et al., 2008; Thornton et al., 2010). The role of potential aerosol coatings on modifying this surface reactivity has been shown (e.g. Thornton and Abbatt, 2005; McNeill et al., 2006), but needs more study, particularly under the cold temperature conditions of Fairbanks, Alaska. The role of particle phase and/or viscosity on these heterogenous reactions clearly needs further investigation in the laboratory. Heterogenous formation of HONO (Stutz et al., 2002; VandenBoer et al., 2013; Xing et al., 2018) could provide a photolabile radical precursor that can produce radicals even at very low insolation of Fairbanks wintertime. However, the mechanism of HONO formation needs further laboratory investigation, particularly under cold temperatures relevant of Fairbanks.

3.6 Modeling:

A three-component modeling approach is proposed that addresses the need to build predictive capabilities for both meteorological and chemical aspects of the atmosphere. The components include:

3.6.1 Emission inventory improvement and testing:

As a part of the State Implementation Plan (SIP) modeling efforts, a detailed emission inventory has been developed (Alaska DEC, 2014a). This inventory includes temporal emissions profiles for each source and distributes emissions into altitude layers based upon stack heights and plume rise estimates. The inventory also includes some Fairbanks-specific speciation profiles, although they were measured at warmer temperatures than experienced locally, as described in Section 2.1.1. Even with this inventory, there are still challenges and an accurate inventory is particularly necessary because past measurements indicate pollution is not strongly aged in Fairbanks. For example, sulfur is not very highly oxidized in Fairbanks (sulfur oxidation ratio = ~5%, Nattinger, 2016) and current data indicates a low values of the organic matter to organic carbon ratios (OM/OC~1.4), indicating more primary-like organic speciation. This inventory should also take into account variations in woodstove emissions throughout the burn cycle, variations in fuels (e.g. fuel moisture), and variability caused by operators and behaviors, potentially including catalytic converters and electrostatic precipitators. These aspects could affect the "source profile" of woodstove emissions. The inventory will be tested by mobile measurements that can target specific areas that are dominated by certain parts of the area's source mixture. Mobile and/or vertical measurements (e.g. UAS or balloons) can be used to probe emissions from point sources and determine how well the inventory represents these sources. It is also important to use models to determine when and where these elevated sources affect ground-level air quality. Longer-term measurements at the supersites will assist in determining appropriate temporal distribution functions for the emissions inventory, which is necessary for their inclusion in subsequent modeling efforts.

3.6.2 Reduced dimensionality models:

Vertically resolved (1-dimensional) and box (0-dimensional) models, including source apportionment models (e.g. Kotchenruther, 2016), photochemical models (e.g. Joyce et al., 2014), thermodynamic models (e.g. ISORROPIA Fountoukis and Nenes, 2007) and other box and 1-D models with a range of chemical complexity (e.g. Master Chemical Mechanism (MCM), CiTTyCAT (Ridley et al., 2017; Galeazzo et al., 2018), can be used to provide insight into factors controlling the chemical environment in Fairbanks. Such models can be used to test and compare detailed aerosol or chemical mechanisms. The possibility of including more detailed chemical processes in these dynamically simple models makes it possible to include detalied isotope mechanisms (e.g. Galeazzo et al., 2018) or to investigate sensitivities to mechanistic or kinetic uncertainties (e.g. Ridley et al., 2017). Simple 1-D models can be used to examine vertical stratification and mixing within the boundary layer and to the free troposphere.

3.6.3 Three dimensional models:

Regional models, run at fine spatial resolution of ~1 km, ideally with multiple vertical layers below 20m, would provide a test-bed to examine if current scientific understanding can explain

observations collected during ALPACA. An essential step will be to evaluate the ability of modeling systems to capture SBL dynamics and pollutant interactions. Meteorological models, such as WRF, will be evaluated against available data, in particular providing information about dynamical structure and physical processes in the boundary layer and exchange with the free troposphere in order to test schemes for boundary layer dynamics including turbulent mixing, the radiative budget and cloud/fog formation. An important aspect will be the ability of meteorological models to capture to the build-up and breakdown of stable conditions, which are important for pollutant episodes. To aid in the understanding of stable boundary layer dynamics, large eddy simulation (LES) models may also contribute. Larger-scale global modeling can also be used to assess the large-scale synoptic forcing of stable conditions, and factors affecting its predictability. Knowledge of large-scale meteorological drivers as well as emission inputs such as those from the Alaska DEC and improvements described in Section 3.6.1 will be used to build on prior studies examining pollution in the Fairbanks region (Mölders and Kramm, 2010; Tran and Mölders, 2011, 2012; Leelasakultum et al., 2012; Mölders and Leelasakultum, 2012; Alaska DEC, 2014a). A variety of 3-D photochemical and aerosol models will be evaluated using the available data for the period of the intensive field observations. These include models such as WRF-Chem (e.g. Marelle et al., 2017) and CMAQ (https://www.epa.gov/cmaq), which include detailed treatments of aerosol formation mechanisms. These models can be used to test hypotheses about secondary aerosol formation and could, for example, make use of isotopes, to examine the role of certain formation mechanisms (e.g. sulfate aerosols). Models will also be used to answer questions in terms of the impacts of poor air quality on human health and deposition of pollutants to the environment by connecting atmospheric processing to deposition and providing temporal and spatial coverage where observations are not available. Such models will also allow investigation of the interactions between pollutant radiative effects and boundary layer stability and dynamics, as well as the role of aerosols in controlling cloud processes and indirect radiative effects. Coarser resolution models covering a greater spatial extent (regional to Arctic-wide) can be used to understand the regional Arctic composition, climate, and ecosystem impacts downwind of Fairbanks and within the greater Arctic area.

To support these modeling efforts, supersite measurements that include aerosol and gas speciation, size distributions, and gas and aerosol concentrations in combination with several satellite sites having more limited chemical and meteorological information and detailed laboratory studies are recommended. Micro/boundary layer meteorology measurements from towers, remote sensors and aerial systems (e.g. UAS) are needed to understand the fundamental processes related to development and maintenance of the stable lower atmosphere and to support analysis of chemical observations. Specific measurements of interest include turbulent energy and momentum fluxes (e.g. scintillometer measurements, UAS), the influence of orography-driven waves (measure perturbations in pressure, wind speed, temperature, and tracers (e.g. CO_2, CO) on horizontal spatial scales relevant for model grids), and the vertical structure of key quantities in and above the stable boundary layer (LIDAR, SODAR, radar).

3.7 Impacts of pollution

The high pollutant concentrations in Fairbanks in the winter has the potential to impact human and ecosystem health significantly. Therefore, we propose a series of studies to address these impacts.

3.7.1 Health

This study aims to address two major health related questions as follows:

How does the toxicity of the special mixture of aerosol in the Fairbanks area compare with other regions?

Reactive Oxygen Species (ROS) measurements, cellular assays, measurements of specific toxic species will provide insight into whether the distinct mixture of species in the particulate matter in Fairbanks is more or less toxic than the mixtures found in regions where extensive health studies have already been completed. A prior study of the association between air quality and hospital visits from 2003-2008 indicated potential association between $PM_{2.5}$ and cerebrovascular disease (Kossover, 2010) but lacked sufficient sample size to allow definite conclusions. We now have a greater volume of correlated $PM_{2.5}$ and health outcome data and should revisit the prior epidemiological work.

How does potential indoor and outdoor exposure differ for homes in the Fairbanks area?

Due to the extreme cold, many people spend the majority of their time indoors during winters in the Fairbanks area. Therefore, indoor air exposure may dominate human exposure and it is important to evaluate the difference in the aerosol composition and potential toxicity in both indoor and outdoor air (See Section 2.4.4). Additional health studies may also include evaluation of individual case studies and evaluating biomarkers from dogs in collaboration with veterinary medicine (One Health). A key uncertainty for health impacts in Arctic settlements is low understanding of underlying health issues affecting the local populations and how these affect susceptibilities to air-quality-related health risk.

3.7.2 Ecosystems

Section 2.4.3 described open questions regarding the downwind fate of pollution and motivates the following questions and studies:

How much pollution deposits in the immediate Fairbanks area and by what mechanisms?

Passive deposition monitoring and surface snow sampling at locations were ambient atmospheric concentrations are measured will provide a first estimate of the mechanisms responsible for deposition in the immediate Fairbanks area. These studies can provide estimates of the split between wet and dry deposition.

What is the extent of the impact from Fairbanks to the surrounding areas?

Deposition monitoring and surface snow sampling along transects in the upwind and downwind directions from Fairbanks will provide measurements of the extent of the impact to the surrounding region from transport and deposition. These measurements will also be coordinated with existing snowpack sampling at long-term ecological research (LTER) stations near Fairbanks (e.g. Bonanza Creek Experimental Forest and Caribou-Poker Creeks Research Watershed LTER sites). In addition, modeling will also provide estimates of the horizontal and temporal contributions to deposited pollutant mass.

3.7.3 Community Engagement

During this study, we intend to engage the public in helping to refine some of the research questions to improve the likelihood of successful communication of the results and adoption of new practices and/or policies as a result of the findings. Involvement from the community and stakeholders will also ensure that the research design adequately addresses the health impacts based on real world behaviors. Potential populations to engage include high school students and parent groups.

To successfully improve air quality in Fairbanks several types of knowledge need to be used **FIX** (Schmale et al., 2014). Practical knowledge is important for substantive and instrumental reasons to judge, for example, local behaviors (e.g. wood moisture) that lead to emissions, as well as compliance with burn bans. Scientific knowledge is necessary to refine estimates of the balance of emission sources that impact ground-level pollution and to improve predictions of air quality. Additionally, next to political motivations, normative considerations play a role when it comes to acceptability of air pollution mitigation measures and income disparity issues.

Solving such a "real-world" problem is ideally tackled with an approach that involves citizens, practitioners, decision-makers and scientists to co-design the solution options based on the different knowledge systems required. Although definitions vary in the inclusion of specific methods and components, such approaches are generally known as transdisciplinary research (e.g. Dilling and Lemos, 2011; Lang et al., 2012; Mauser et al., 2013; Polk, 2015). To this end, a Fairbanks stakeholder group tasked to develop community-based solutions to these air quality problems was established in June 2018 and completed work in November 2018. Members of the ALPACA organizing committee were involved in the stakeholder group, facilitating connection between the research community and concerned citizens. Information on this group is available online: http://fnsb.us/transportation/Pages/stakeholders.aspx. Although that process is completed, we see a need to maintain these discussions throughout the time of the ALPACA field campaign. This continued dialogue will inform the scientific study design. In the second stage, the results from the ALPACA field study as well as discussions with citizens, decision-makers and regulators will be used to suggest further solution options for better air quality, and these ideas then need to be enacted. The outreach aspect of this study will also improve public understanding of the problem and hopefully improve compliance with these air quality improvement mitigation strategies.

4 Conclusions

This whitepaper has compiled open questions regarding air pollution and its atmospheric chemistry under cold and dark conditions. We found a large number of outstanding questions (Section 2) and that current data is lacking to answer these questions. Therefore, we propose a field experimental program under the PACES umbrella organization addressing high latitude wintertime air pollution chemistry. The first part of this field program is the Alaskan Layered Pollution And Chemical Analysis (ALPACA) study to be carried out in Fairbanks, Alaska, USA. This campaign selected a set of most compelling questions (Section 3.1) and measurements, modeling, and laboratory studies to be carried out. In addition to closing knowledge gaps, this campaign addresses public concerns, provides needed information for designing air quality improvement plans, and can assist in understanding health effects of wintertime air pollution. The ALPACA project also acts as a reference point for pollution under cold and dark conditions

that can be compared to other mid-latitude wintertime pollution studies (e.g. Salt Lake City, etc.). It is clear that the scope of this field study overlaps the mandates of multiple funding agencies, which was the reason that we developed this whitepaper. The next step is for groups of investigators to make proposals to appropriate funding agencies.

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