UiT
THE ARCTIC UNIVERSITY OF NORWAY

Faculty of Science and Technology

Department of Engineering and Safety IVT

# On local and long-range transported air pollution in Svalbard

*From a single station to international measurement network*

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**Alena Dekhtyareva**

*A dissertation for the degree of Philosophiae Doctor – June 2019*

UNIVERSITY OF TROMSØ

# On local and long-range transported air pollution in Svalbard

*From a single station to international measurement network*

By
Alena Dekhtyareva

*Thesis submitted in fulfilment of the requirements for the degree of Philosophiae Doctor (PhD)*

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June 2019

*Dedicated to my beloved husband Vitaly and my kind and loving mother Irina.*

*"…I have got the Arctic illness,*
*And it means I have no choice,*
*As She took my heart and called me*
*By Her cool and windy voice.*

*So no matter where I travel,*
*On the threshold of any spring,*
*I still rave of the polar trails,*
*And I see the snowy dreams…"*

*(From the poem of*

*Robert Rozhdestvensky "Arctic Illness"/*

*Роберт Рождественский "Арктическая болезнь"*

*translated from the Russian by Alena Dekhtyareva)*

## Abstract

Climate change, health of the residents and ecosystems in the Arctic region are impacted by local and long-range transported air pollution. Local emissions in the Arctic are important, but overlooked issue. Despite there have been extensive modelling and measurement studies of long-range transport of short-lived climate forcers (SLCFs) to Svalbard, the effect of local emissions from diesel and coal power plants and ship traffic on the concentrations of these compounds in major settlements has not been investigated thoroughly.

The scope of this work is to study temporal and spatial evolution of air pollutant concentrations in Svalbard using the historical chemical and meteorological data collected in Ny-Ålesund and newly obtained observations from three sites: Ny-Ålesund, Longyearbyen and Barentsburg. Remote and local emission sources, concentrations of anthropogenic SLCFs and environmental factors that promote long-range transportation and accumulation of local air pollution in the Svalbard settlements have been investigated.

A strong seasonality in the concentrations of sulphur dioxide ($SO_2$), nitrogen oxides ($NO_x$), tropospheric ozone ($O_3$) and black carbon (BC) in Svalbard has been observed. Measurements in Ny-Ålesund revealed that in autumn, winter and spring the concentrations of $SO_2$, sulphate and particles of accumulation mode are dominated by the long-range transport of air pollution from remote and regional sources. In summer, the long-range transport of air pollution is limited, and local sources become more important. Indeed, ship traffic emissions in Longyearbyen and Ny-Ålesund promoted significant increase in $SO_2$ and $NO_x$ concentrations and slight decrease of the $O_3$ values. Measurements in Barentsburg revealed strong temporarily deterioration of local air quality because of adverse weather conditions promoting transport of polluted air from the local coal power plant to the town. The cases of enhanced accumulation of local ground-level pollution have been revealed in Longyearbyen as well. They have often coincided with long-range transport events when the advection of warm air from mid-latitudes to Svalbard promoted creation of strong temperature inversions and led to increased concentrations of BC detected by the ground-based instrument and in the vertical profiles below 1000 m.

Svalbard archipelago is an area with complex topography. This creates a pronounced spatial and vertical variation in the concentrations of SLCFs. Thus, the springtime $NO_x$ observations demonstrated that there is little correspondence between the data from the three stations. The concentrations of these compounds are controlled by local sources and mostly dependent on prevailing wind direction in each of the settlements. Comparison of the daily $SO_2$ and sulphate concentrations accumulated in filter samples collected at the low-altitude station in Ny-Ålesund and at the Zeppelin mountain observatory revealed a significant difference in the data obtained at different heights. The correspondence between the observations varies seasonally. It is the best in winter due to stronger winds, more efficient mixing and absence of additional local sources of pollution. In contrast, the correspondence between the two datasets is lowest in summer when insufficient ventilation of atmospheric boundary layer combined with increased emissions from local ship traffic promote accumulation of pollution in the settlement, while the station at the mountain top is often located above the cloud base level and is unaffected by the local emissions.

## Acknowledgements

First of all I would like to thank my beloved husband Vitaly for his patience and support. He has always been willing to stay with me till late evening at the University when needed, discuss my work on the way home, and help me with the practicalities at home. Besides, he has been a skilful and smart field assistant and helped me a lot with electronic equipment and data processing during my fieldwork in Longyearbyen in 2018.

I would like to thank my supervisors Associate Professor Kåre Edvardsen and Professor Rune Graversen for all the support which I received during the work on the articles included in current thesis and for giving me the opportunity to develop my own research ideas. I also appreciate your invaluable advice on scientific content and language corrections. In addition, Rune, I am very grateful for your patience, since the last year of the PhD work has been very busy for me and I have been responsible for the external research project while writing my last papers and the thesis at the same time.

I am very thankful to my supervisor Dr. Kim Holmén for fruitful discussions, critical reviews of my articles and logistical support during the fieldwork. Without his guidance in designing of the fields campaigns and help with the access to the data and research infrastructure in Svalbard none of the papers included in the current thesis would have seen the light.

Part of my PhD work has been done in Longyearbyen. There I have always felt myself welcomed at the University Centre in Svalbard (UNIS) where the UNIS staff and employees of the Norwegian Polar Institute provided me with all the practical support needed for my field studies. I am also very grateful for all the knowledge which I have obtained at the two PhD courses taken at UNIS. Although studies there have been challenging, the learning environment has been perfect and I still keep in touch with my fellow students.

I appreciate a lot the openness and engagement of the researchers from the Ny-Ålesund Atmosphere Flagship and Italian Aerosol Society who supported and encouraged my research initiatives. This allowed me to apply for an external grant from the Research Council of Norway and successfully manage my first extensive international scientific project.

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## Abbreviations

| **Chemical compounds and abbreviations** | |
|---|---|
| AOD | Aerosol optical depth |
| BC | Black Carbon |
| CO | Carbon monoxide |
| NO | Nitrogen monoxide |
| $NO_2$ | Nitrogen dioxide |
| $NO_x$ | Nitrogen oxides |
| OH | Hydroxyl radical |
| $O_3$ | Ozone |
| PAH | Polycyclic aromatic hydrocarbon |
| PAN | Peroxy acetyl nitrate |
| PM | Particulate matter |
| $SO_2$ | Sulphur dioxide |
| VOC | Volatile organic compound |
| $XSO_4^{2-}$ | Non-sea salt sulphate |

| **General abbreviations** | |
|---|---|
| AARI | Russian Arctic and Antarctic Institute |
| ABL | Atmospheric boundary layer |
| AWS | Automatic weather station |
| GT | Gross tonnage |
| LD | Limit of detection |
| RF | Radiative forcing |
| SLCF | Short-lived climate forcer |
| UNIS | University Centre in Svalbard |
| WRS-test | Wilcoxon rank sum test |

## List of appended papers

**Paper 1 Dekhtyareva A.**, Edvardsen K., Holmén K., Hermansen O., & Hansson H.-C., 2016. Influence of local and regional air pollution on atmospheric measurements in Ny-Ålesund. *International Journal of Sustainable Development and Planning*, 11 (4), 578–587. DOI: 10.2495/SDP-V11-N4-578-587

**Paper 2 Dekhtyareva A.**, Holmén K., Maturilli M., Hermansen O., & Graversen R., 2018. Effect of seasonal mesoscale and microscale meteorological conditions in Ny-Ålesund on results of monitoring of long-range transported pollution. *Polar Research*, 37 (1), 1508196. DOI: 10.1080/17518369.2018.1508196

**Paper 3 Dekhtyareva A.**, Hermanson M., Nikulina A., Hermansen O., Svendby T., Graversen R., & Holmén K., 2019. Springtime nitrogen oxides and tropospheric ozone in Svalbard: results from the measurement station network. *Manuscript ready for submission*

**Paper 4 Dekhtyareva A.**, Drotikova T., Nikulina A., Hermansen O., Chernov D.G., Mateos D., Herreras M., Petroselli C., Ferrero L., Gregorič A., 2019. Summer air pollution in Svalbard: emission sources, meteorology and air quality. *Manuscript ready for submission*

# Part I Thesis summary

## 1. Introduction and background

The Arctic region may be defined geographically by the Arctic Circle, climate, vegetation and marine boundaries (Figure 1). In the area north of the Arctic Circle, midnight sun and polar night last at least 24 hours continuously each year. The mean air temperature in July in the region set by the climatic boundaries is below 10 ℃. Vegetation boundaries are stated by the treeline, a transition zone between the boreal forest and tundra vegetation. The Arctic ecosystems are often interdependent and consist primarily of cold-adapted biota vulnerable to climate change that alters physical, biogeochemical, and ecological processes (Vincent *et al.*, 2011). On the marine boundaries of the Arctic, warmer and saltier waters from oceans to the south interact with surface waters from the Arctic Ocean, which have lower temperature and salinity. The area, limited by the red line in the Figure 1, is defined as the Arctic by the Arctic Monitoring and Assessment Programme from the perspective of monitoring and assessing the status of and threats to the environment and health of residents in the region (AMAP, 1998).

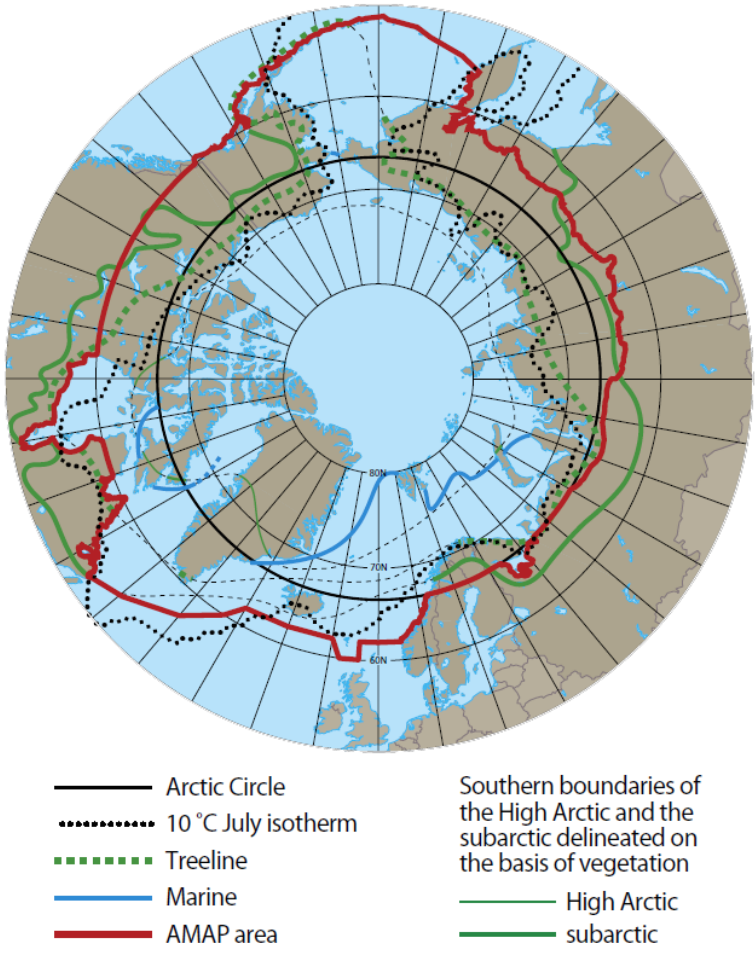


Figure 1 Definitions of the Arctic region (Fig. 1.1 in AMAP 2009)

The population density and urbanization rate varies significantly within the Arctic region (Nuttall, 2012). Lack of infrastructure poses limitations on the development of industrial activities in the Arctic, such as natural resource extraction, shipping and tourism. However, the climate change stimulates the melting of the sea ice, thus creating new opportunities for

improvement of physical connection within the Arctic (Christopher and Fast, 2008). Indeed, the area covered by the annual summer sea ice declines steadily, and ice-free summers are predicted to occur in the perspective of several decades. The oil and gas, shipping and fishing activities in the Arctic have increased to a notably large extent over the last years, and further development of these industrial sectors in the region is likely to happen (Dalsøren *et al.*, 2007).

As a consequence, emissions of such pollutants as nitrogen monoxide (NO), nitrogen dioxide ($NO_2$), nitrogen oxides ($NO+NO_2=NO_x$), sulphur dioxide ($SO_2$), carbon monoxide (CO), volatile organic compounds (VOCs) and BC (Black Carbon) are expected to rise (Dalsøren *et al.*, 2009; Peters *et al.*, 2011). Increased emissions from shipping activities lead to elevated concentrations of $NO_x$, $SO_2$, $O_3$ and BC and rise the number of fine particles with diameter less than 2.5 µm ($PM_{2.5}$). These substances also have negative effect on human health and are included in the standard air quality observations in urban areas. Long-term exposure to elevated concentrations of $PM_{2.5}$ can cause chronical cardiovascular diseases. Daily variations in BC concentrations are associated with short-term health changes. In addition to this, BC, as a component of PM, may carry toxic chemicals such as polycyclic aromatic hydrocarbons (PAHs) to the lungs and possibly introduce them to the systemic blood circulation (Janssen *et al.*, 2012).

Besides, $SO_2$ and $NO_x$ emissions may have negative impact on the ecosystems due to acidification of fresh-water, marine and terrestrial environments. The Arctic vegetation is especially sensitive to air pollution due to multiple factors of influence such as climate change and long exposure to sunlight in summertime (Eriksen *et al.*, 2012; Futsaether *et al.*, 2015). At the same time, nitrates produced from $NO_x$ can act as fertilizers to local ecosystems, especially if deposited in nutrients limited Arctic areas (AMAP, 2006).

Increasing anthropogenic activity in the region is one of the reasons to study the local emissions in order to investigate their influence on the near pristine Arctic environment.

In addition to local atmospheric emissions from the above mentioned activities, there is a seasonal long-range transport of air pollution to the region known as Arctic haze (Quinn *et al.*, 2007). $SO_2$ and non-sea salt sulphate ($XSO_4^{2-}$), along with BC, are the most studied compounds present in the Arctic haze. The air transport efficiency from mid-latitudes towards the North pole depends on the location of the Arctic front and varies seasonally (AMAP, 2006). This transport pattern is most pronounced in winter and spring when specific conditions affecting environmental fate of the atmospheric pollutants are present. For example, lack of sunlight during polar night restricts photochemical reactions, while low air temperatures slow down certain chemical reactions such as thermal decomposition of peroxy acetyl nitrate (PAN). At the same time, low atmospheric humidity decreases the hygroscopic growth of aerosol particles and hampers dry deposition since the efficiency of that process depends on the particle mass. The precipitation is rare, and wet deposition is scarce. These factors lead to prolonged lifetime of the particles in the air masses (Seinfeld and Pandis, 2006).

As the Arctic is warming faster than the rest of the world, especially in winter (Richter-Menge and Mathis, 2017), the conditions preventing removal of pollutants from the air masses during transportation are changing. Increased air temperature, humidity and

precipitation rate may intensify wet deposition of pollutants (Qi *et al.*, 2017). In addition to this, the concentrations of tropospheric ozone ($O_3$) have increased over last 100 years. Along with several competing climate-dependent factors such as amount of biogenic emissions, water vapour abundance and change of convection and lightning, $O_3$ may increase the atmospheric abundance of hydroxyl radical (OH), and consequently, the oxidative capacity of the atmosphere, causing reduction in the lifetime of air pollutants (Alexander and Mickley, 2015). Besides, the $SO_2$ emissions in European countries have declined over the last twenty-five years (Vestreng *et al.*, 2007), while emissions from Asian sources slightly increased (Lu *et al.*, 2010). Thus, it may have affected the concentrations of long-range transported pollutants measured in the Arctic.

The current work focuses on Svalbard, since the archipelago has unique characteristics, which allow us to study ongoing alteration in atmospheric composition and physical processes due to the change in anthropogenic activities and environmental response to the climate change.

Firstly, Svalbard is warming faster than the most of the Arctic territories (Isaksen *et al.*, 2016). Since the region is located on the marine Arctic boundary (Figure 1), several factors contribute to this accelerated warming rate observed there: change in the inflow and temperature of North-Atlantic water on the west coast of Spitzbergen island, sea ice decline, change in atmospheric circulation patterns and properties of air masses (Piechura and Walczowski, 2009; Maturilli, Herber and König-Langlo, 2013; Onarheim *et al.*, 2014; Isaksen *et al.*, 2016; Dahlke and Maturilli, 2017; Maturilli and Kayser, 2017).

Secondly, there are few regional and local sources of air pollution at Spitzbergen, the archipelagos biggest island, thus it is easier to estimate the change in amount of long-range transported and local air pollution, and study the effects of these factors on atmospheric physical and chemical processes.

Long-term observations of atmospheric compounds performed at the Zeppelin station in Ny-Ålesund, a research settlement in the north-western part of the island (Figure 2), allow us to investigate the change in efficiency of long-range transport of air pollutants from mid-latitudes to this region. Hirdman *et al.*, 2010 attributed the significant negative long-term trend in concentrations of elemental BC and sulphate aerosol observed at the Zeppelin station to the reduction in European emissions. However, change in environmental conditions may affect the lifetime of aerosols as well. For example, the aerosol scavenging efficiency varies for different cloud types: it is lowest for ice-phase clouds and increases for warmer mixed-phase clouds (Eckhardt *et al.*, 2015). The sea ice melting facilitates the vertical transfer of moisture which contributes to the liquid cloud phase and may result to the increase of mixed-phase clouds occurrence over the Arctic (Mioche *et al.*, 2015). Long-term radiosonde and ground-based observations in Ny-Ålesund revealed a strong increase in atmospheric temperature and humidity (Maturilli, Herber and König-Langlo, 2013; Maturilli and Kayser, 2017), and the long-term projections for precipitation and temperature in Svalbard indicate further increase (Førland *et al.*, 2011). Moreover, the precipitation over the Arctic is predicted to monotonically increase towards the end of the century (Kusunoki, Mizuta and Hosaka, 2015). Thus, because of changes in the properties of air masses arriving to Svalbard and reduction of European emissions, the concentrations of long-range

transported $SO_2$ and $XSO_4^{2-}$ have been decreasing and may further decrease in future. In contrast, local sources of emissions may play increasingly important role and deserve special attention.


Figure 2 Map of Svalbard

At the same time, not only long-range transport of air pollutants is affected by the climate change. Local meteorological processes are altered as well. For example, the frequency of decoupling of atmospheric boundary layer (ABL) from free troposphere has increased and the wind speed in the lowest 500m has reduced in all seasons in the period from 1993 to 2014 (Maturilli and Kayser, 2017). This may negatively affect the ventilation within ABL and lead to the accumulation of locally produced primary and secondary atmospheric aerosols of natural and anthropogenic origin. They play an important role in the cloud formation processes (Possner, Ekman and Lohmann, 2017; Jung *et al.*, 2018; Mahmood *et al.*, 2019) which, in turn, are altered by the observed change in air temperature and humidity (Maturilli and Kayser, 2017).

Current study focuses on short-lived climate forcers (SLCFs) which concentrations are increasing because of fossil fuel combustion: $SO_2$, $NO_x$, $O_3$ and BC. The change in energy flux at the tropopause or at the top of the atmosphere caused by a specific climate driver is called the radiative forcing (RF) (IPCC, 2013). When the increased concentration of the forcer increases the difference between the energy absorbed by the Earth and radiated back to space, the RF is positive and leads to atmospheric warming. In contrast, the driver has

negative RF, when its increased concentration leads to cooling of the atmosphere. Thus, $SO_2$ and $NO_x$ have negative RF due to formation of light scattering aerosols containing sulphates and nitrates (Figure 3).

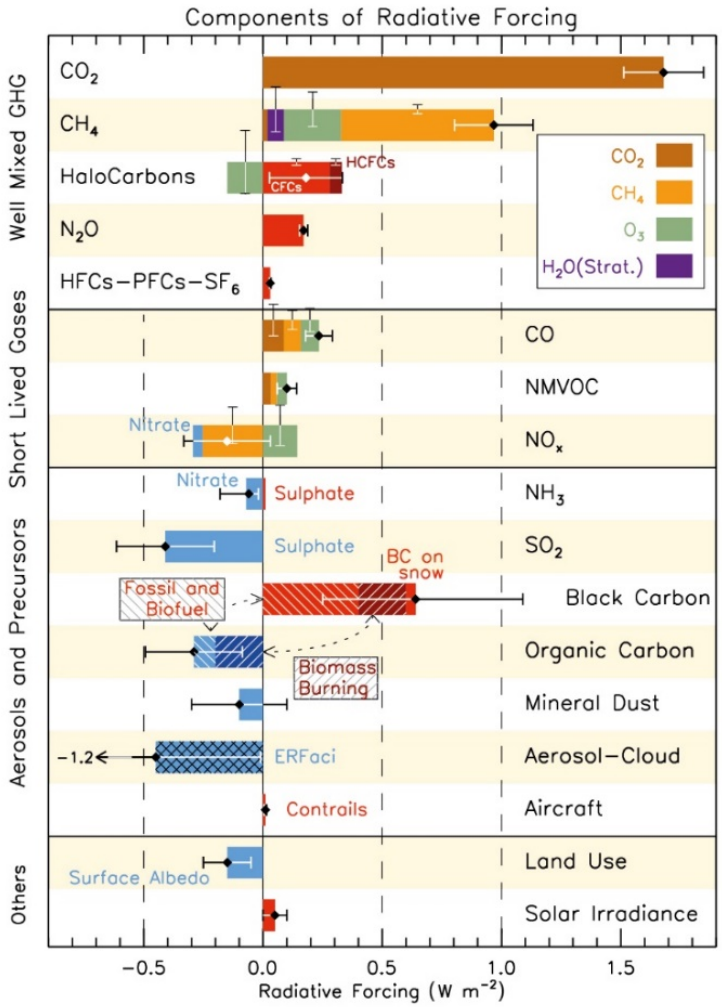


Figure 3 Radiative forcing bar chart for the period 1750–2011 based on emitted compounds (gases, aerosols or aerosol precursors) or other changes (Fig. 8.17 in IPCC, 2013)

Although the total aerosol-cloud interaction has negative radiative forcing of climate (IPCC, 2013), thin Arctic clouds in winter and early spring have positive radiative forcing due to increased downward long-wave radiation, and this effect is enhanced when the anthropogenic aerosols are present (Garrett and Zhao, 2006). In turn, the light absorbing aerosols have significant positive radiative forcing through aerosol-radiation interactions and when deposited on snow and ice because of reduction of surface albedo (IPCC, 2013). However, $NO_x$ also have a positive RF due to formation of tropospheric $O_3$, a potent greenhouse gas, in presence of CO and VOCs. BC is a component of light-absorbing aerosols, and thus has strongly positive RF. In the real atmosphere the RF of aerosols depends on the ambient relative humidity, which varies strongly horizontally and vertically, aerosol size distribution and refractive index that depends on aerosol composition (Myhre *et al.*, 2004).

A recent modelling study of Sand et al. 2015 stated that the major contribution to the Arctic warming comes due to Asian emissions of the SLCFs, which increase the heating rate in the source region, and therefore affect the equator-pole temperature gradient. However, the regional sensitivity to local emissions within the Arctic is very high due to enhanced warming impact from BC deposited on snow and ice covered surfaces. Indeed, the simulations with additional ship emissions in the Arctic showed significant local increase in RF due to BC deposition over the central Arctic Ocean, but the net cooling effect from the aerosols and their precursors is expected (Gilgen *et al.*, 2018). Similarly, Ødemark *et al.*, 2012 have estimated positive RF from increased BC and $O_3$ concentrations, but the total negative RF due to formation of sulphate and nitrate containing aerosols from emissions of $SO_2$ and $NO_x$ because of shipping activity.

## 1.1 Research motivation and problem statement

There are two factors that make it challenging to assess the current environmental impact of local Arctic emissions and make prognoses for the future. First one is the uncertainty in emission inventories, since, in addition to existing stationary emission sources, there is an ongoing increase of local emissions from shipping. Second factor is the uncertainty in environmental fate of air pollutants due to the lack of meteorological and air pollution observations in the region. Previous studies state that air pollution from local emission sources is an important, but an underestimated issue, and that pollution levels within the region may exceed air quality standards, pose a negative impact on the health of residents and environment (Schmale et al., 2018).

Similarly, the long-range transport of $NO_x$, $SO_2$, $O_3$ and BC to Svalbard has been studied extensively, while little attention has been given to the local sources of these compounds and meteorological conditions promoting in-situ pollution accumulation. For example, last and the only study about influence of shipping emissions on air quality in Svalbard has been based on ten years old data from Ny-Ålesund, while no extensive air quality studies have been performed previously in other Svalbard communities.

Ny-Ålesund is located more than 100km away from the biggest Svalbard settlements. The remoteness and measures, which are applied to reduce anthropogenic impact on the research activity, offer unique opportunities for monitoring of background air composition (The Research Council of Norway, 2019).

However, in contrast to the near pristine Ny-Ålesund environment, there are also places in Svalbard where the anthropogenic activity may significantly affect local air quality. The two mining towns, Longyearbyen and Barentsburg, are located to the south-east from the research settlement. Although the installation of exhaust treatment system on the coal power plant in Longyearbyen led to dramatic reduction of emissions there (Miljødirektoratet, 2019), the Barentsburg coal power plant is still the biggest point source of $SO_2$ in Norway (Miljødirektoratet, 2018). However, no air quality measurements have been available to assess the magnitude of pollutant concentrations accumulating under different meteorological conditions in these towns.

The current work allows us to combine the air quality studies with monitoring of SLCFs in Svalbard and assess the current concentrations of anthropogenic SLCFs and environmental factors that promote long-range transportation and accumulation of local air pollution in the Svalbard settlements. This study attempts to create a network from existing and temporarily pilot stations and assess the measurement results obtained using the conventional and portable low-cost sensors in Svalbard.

## 1.2 Research questions

Based on the proposed problem statement, five research questions have been produced:

1. What causes variation in the pollutant concentrations on a different temporal scale (seasonally, daily and diurnally)?
2. How do the pollutant concentrations vary spatially between the three main Svalbard settlements?
3. What is the current influence of ship traffic on air quality in Svalbard settlements?
4. What meteorological phenomena affect the ground level concentration of measured compounds and their vertical distribution in the ABL?
5. What are the advantages and disadvantages of usage of different measurement techniques for air pollution monitoring in the Arctic?

## 1.3 Research objectives

The following objectives have been performed to answer to the research questions stated above:

- Investigate the long-range transport of air pollution to Svalbard and explore the existing techniques to study it.
- Analyse data series to identify factors affecting long-term observations of long-range transported pollution.
- Perform ground-based $SO_2$, $NO_x$, $O_3$ and BC measurements in Longyearbyen and analyse the acquired data along with the measurement results from Ny-Ålesund and Barentsburg.
- Identify major emission sources in all three settlements and their influence on local air quality.
- Test portable sensors to measure air quality and meteorological parameters and assess performance of these sensors.
- Perform vertical meteorological and air quality measurements in Longyearbyen and identify what affects the vertical distribution of air pollutants.

## 2. Methodology

The current study focuses on measurements of four SLCFs in Svalbard: $SO_2$, $NO_x$, $O_3$ and BC. Paper I and II discusses $SO_2$ and $NO_x$ sources, chemical transformations and factors affecting their ambient concentration in Ny-Ålesund. Paper III considers springtime $NO_x$ and $O_3$ concentrations measured in Longyearbyen, Ny-Ålesund and Barentsburg in 2017 and atmospheric chemistry of these compounds. Paper IV presents extensive summer measurements of all four compounds performed in the three major Svalbard settlements in 2018 and determines contribution of various local and long-range sources to air quality in Spitzbergen.

### 2.1 Chemistry of $SO_2$, $NO_x$, $O_3$ and BC

$SO_2$, $NO_x$ and BC are emitted directly in the process of fossil fuel combustion, while $O_3$ may be produced in the presence of $NO_x$, VOCs and CO. The atmospheric cycles of nitrogen compounds, $O_3$ and sulphur compounds are illustrated in Figure 4, Figure 5 and Figure 6, respectively. This work focuses on the small part of the reactions from combustion to NO and $NO_2$ formation and reactions which may lead to increasing and decreasing of $O_3$ concentrations in the troposphere (Figure 4).

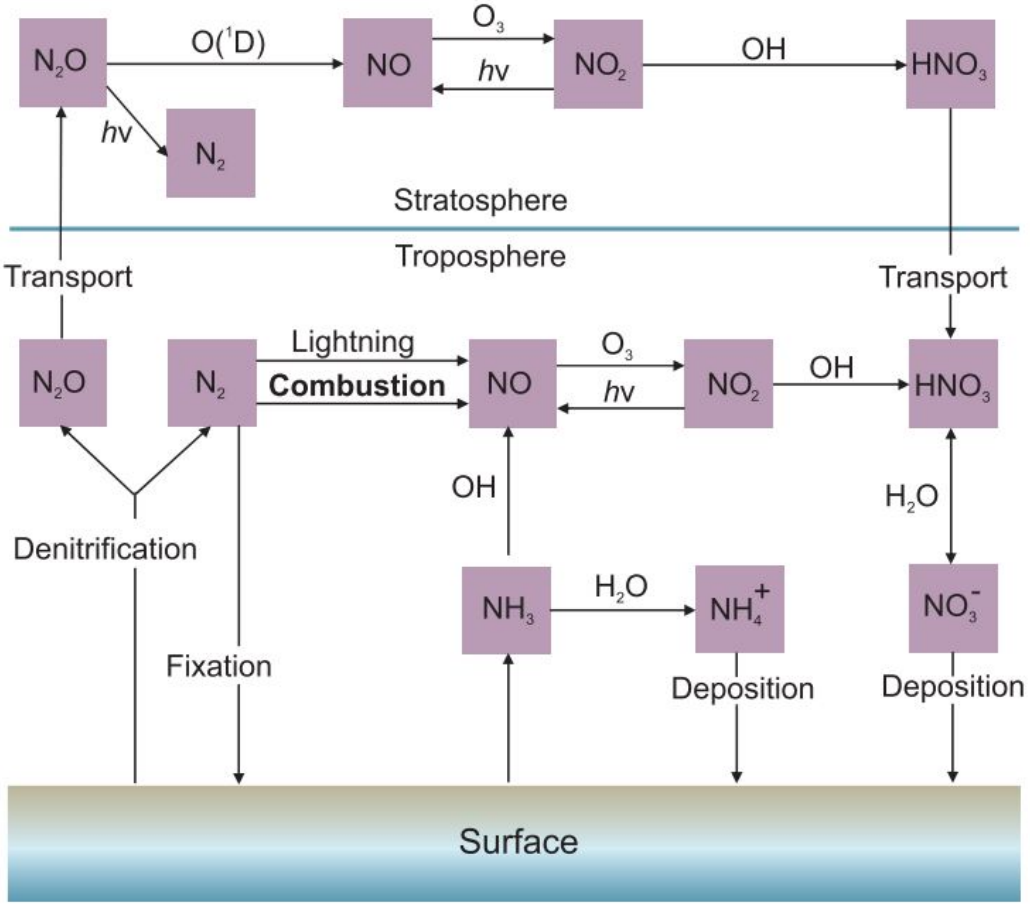


Figure 4 Atmospheric nitrogen cycle (Fig. 3.2 in AMAP, 2006)

The formation of tropospheric $O_3$ is a non-linear process depending on the ratio between $NO_x$ and VOCs (hydrocarbons (RH) in Figure 5 (Fan and Jacob, 1992; Jacob, 2000; Monks, 2005)). The reactions between $O_3$ precursors ($NO_x$, CO and RH), which may lead to $O_3$ production in the presence of sunlight, are depicted by the two cycles on the right side of the Figure 5. In the VOCs-limited regime, $O_3$ concentration may decrease due to titration with excess of NO. The production of $O_3$ in the $NO_x$-limited regime is independent on VOCs amounts and increases with rising of $NO_x$ concentration. Despite the fact that the concentrations of non-methane hydrocarbons increases with latitude due to long-range transport of pollution (Helmig *et al.*, 2016), the average values in the pristine Arctic environment are lower than in industrial areas, and therefore VOCs-limited regime is expected close to big sources of $NO_x$ such as ships and fossil-fuelled power plants. Further downwind from the source, $NO_x$ are removed from the plume faster and $NO_x$/VOCs ratio sufficient for $O_3$ production may be obtained. Similarly, the $O_3$ production is more efficient in the Arctic, downwind from boreal fires, than in the vicinity of the biomass burning areas (Monks *et al.*, 2015). In the left side of the Figure 5, the heterogeneous photochemical reactions with bromine species on snow or sea-ice surfaces, which may result to springtime tropospheric $O_3$ depletion in the Arctic, are illustrated (Fan and Jacob, 1992; Monks, 2005).

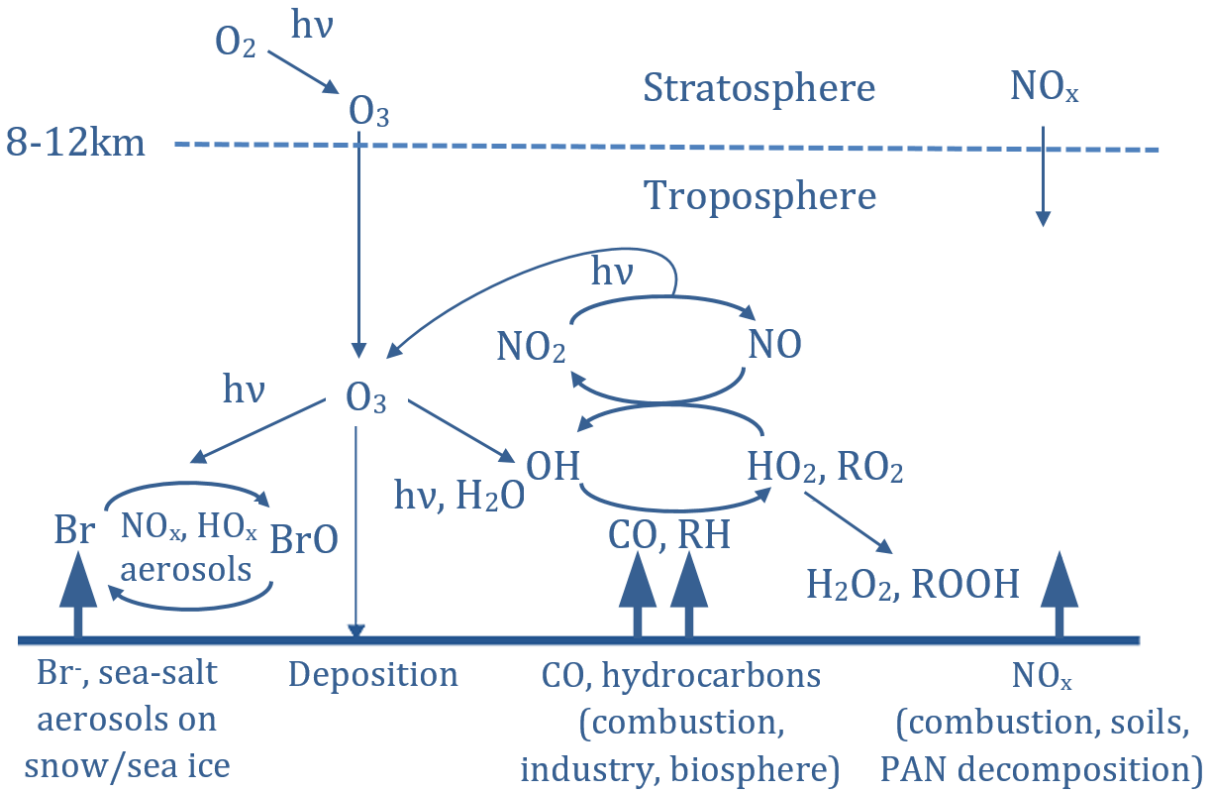


Figure 5 Tropospheric $O_3$ chemistry where $HO_x$ and $RO_2$ are peroxy radicals, R is alkyl radical, $H_2O_2$ and ROOH are hydrogen and organic hydroperoxides, respectively.

The part of the atmospheric sulphur cycle shown in Figure 6, which describes emissions of $SO_2$ and its precursors and formation of sulphate aerosols, in the troposphere is studied in the current work.

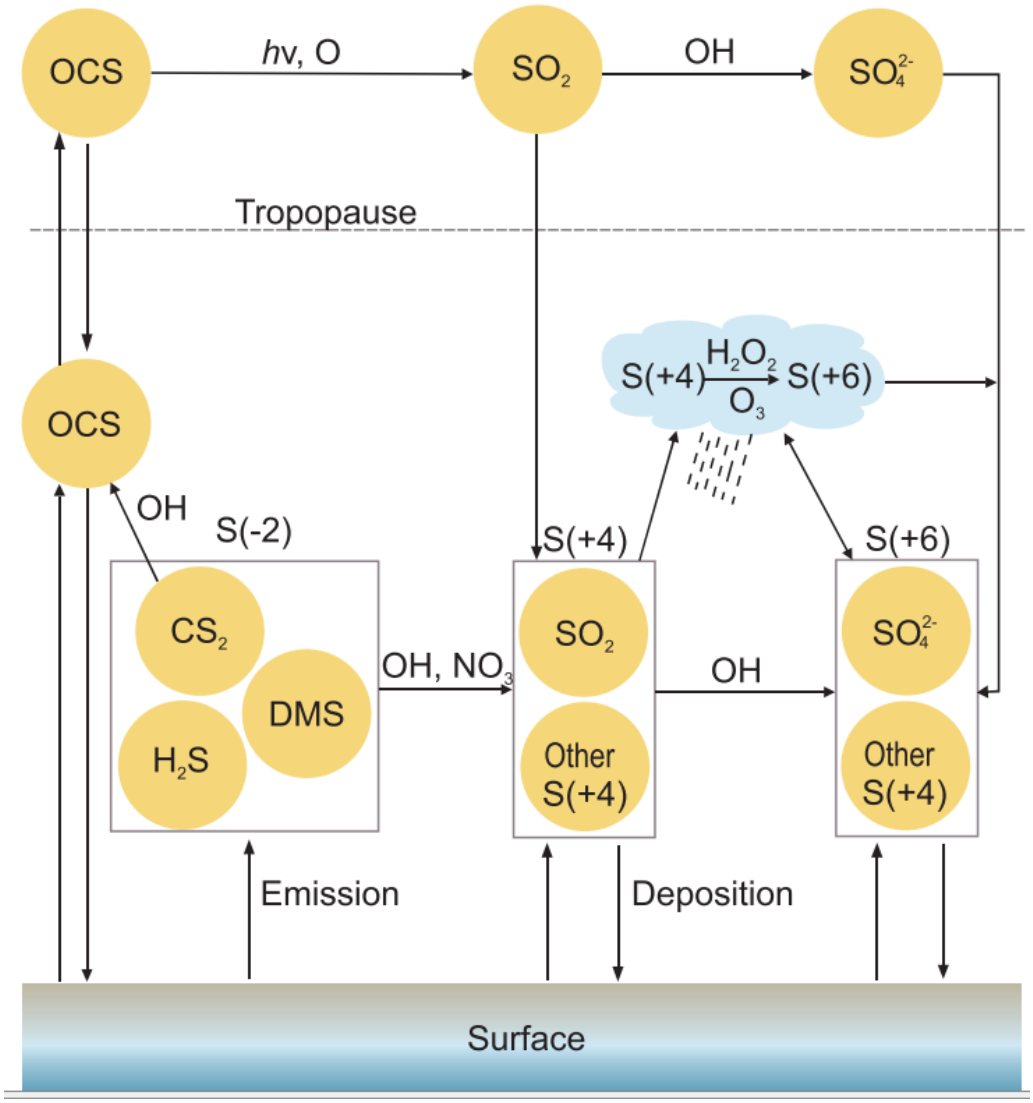


Figure 6 Atmospheric sulphur cycle (Fig. 3.1 in AMAP, 2006)

Another compound, which has been studied in the present work, is BC. It is a main component in soot. The soot is formed in a process of incomplete combustion, and the formation efficiency depends on the ratio of carbon to oxygen (C/O) in the mixture of hydrocarbons and air. For example, if C/O= m/2a, following combustion stoichiometry is obtained (Seinfeld and Pandis, 2006):

$$C_m H_n + aO_2 \rightarrow 2a\ CO + 0.5\ n\ H_2 + (m - 2a)C_s \quad (1)$$

where Cs is the soot formed.

## 2.2 Meteorological processes affecting air pollution transport and in-situ pollution dispersion

As Arctic front extends southerly during winter and spring (Figure 7), the long-range transport of pollutants intensifies (Stohl, 2006).

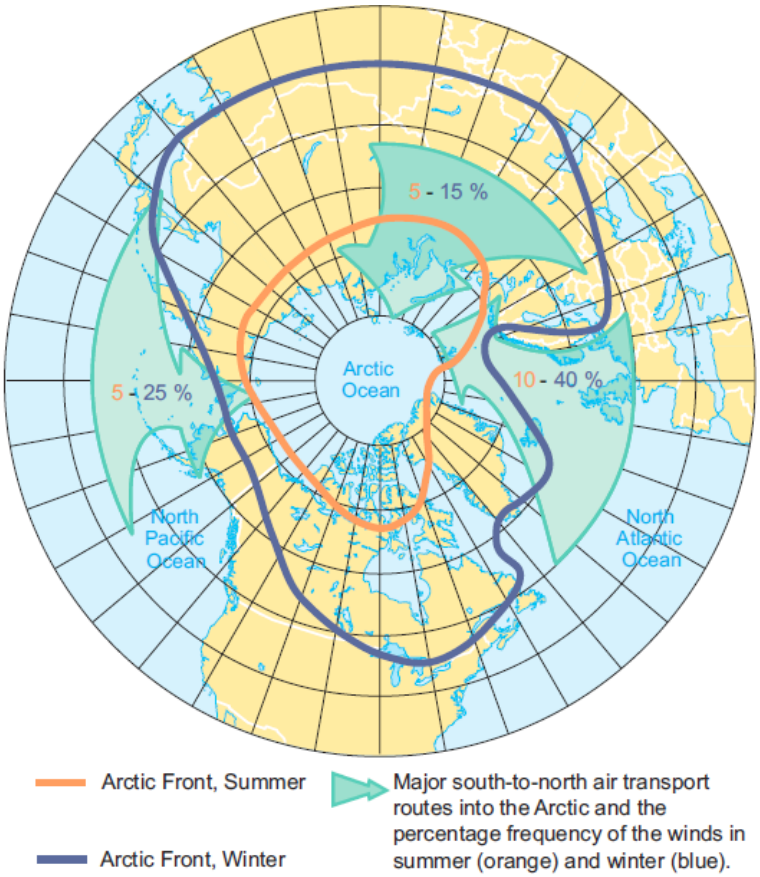


Figure 7 Mean winter and summer position of the arctic front defining the percentage frequency of major south-to-north transport routes into the Arctic in summer (July) and winter (January) (Fig. 4.1 in AMAP, 2006)

In these seasons, the Arctic haze, an anthropogenic aerosol consisting primarily of sulphate containing particles in accumulation mode, has been repeatedly observed in Svalbard and other Arctic regions (Heintzenberg, Hansson and Lannefors, 1981; AMAP, 2006; Quinn *et al.*, 2007). Low air temperature and humidity and lack of sunlight during the polar night extend the lifetime of SLCFs due to reduction of wet scavenging and limited photochemical oxidation (Seinfeld and Pandis, 2006).

At the same time, there are local year-round air pollution sources in Svalbard, which may increase the concentrations of SLCFs in the ABL such as coal power plants in Barentsburg and Longyearbyen and diesel generator in Ny-Ålesund, and seasonally important sources, namely, the ship traffic and biogenic emissions of $SO_2$ precursors. Calm winds and temperature inversions reduce the efficiency of air pollution dispersion (Arya, 1999). Although the median wind speed is lowest in summer and increases in winter in Svalbard (Maturilli, Herber and König-Langlo, 2013), the frequency of occurrence of temperature inversions is higher in winter as well, because the air is more often stably stratified due to radiative cooling of snow and ice-covered surfaces (Vihma *et al.*, 2011). However, it has been shown that the reduction of the sea ice extent around Svalbard increases the sea-atmosphere energy transfer and decreases the efficiency of inversion formation under the

same high-pressure situations with calm winds, which favour this process over the sea ice (Isaksen *et al.*, 2016).

Using the methodology proposed by Vihma *et al.*, 2011, the temperature inversions have been identified in the radiosonde and tethered balloon profiles as layers thicker than 10 m where the air temperature increases with height on more than 0.3 °C. Additional method to determine the stability in the ABL, suitable for the sites where the airborne measurements have not been performed, but the meteorological observations at two different heights have been available, is to calculate the Richardson number as the ratio between the buoyancy and wind shear terms.

The gradient Richardson number (Arya, 1999) has been calculated for the case study in Barentsburg described in the Paper IV:

$$Ri_m = \frac{g}{T_0} \frac{\overline{\Delta\theta} z_m}{\overline{\Delta u}^2} \ln\left(\frac{z_2}{z_1}\right) \quad (2)$$

where $T_0$ is the mean temperature for two heights ($z_1$=70 m and $z_2$=255 m), $z_m = (z_1 \cdot z_2)^{1/2}$ is the geometric mean height, $\overline{\Delta\theta}$ - potential temperature difference between $z_1$ and $z_2$, $\overline{\Delta u}$ is wind speed difference between $z_1$ and $z_2$.

The potential temperature, in turn, has been calculated as (Arya, 1999)

$$\theta = T\left(\frac{1000}{p}\right)^k \quad (3)$$

where T is the measured temperature in K, p is atmospheric pressure in millibars, k≈0.286 is the ratio between the specific gas constant R=287.04 J $K^{-1}$ $kg^{-1}$ and the specific heat capacity for dry air at constant pressure $C_p$≈1005 J $K^{-1}$ $kg^{-1}$.

## 2.3 Observations

### 2.3.1 Measurement sites

Three different measurement sites have been chosen in this work: Ny-Ålesund, Barentsburg and Longyearbyen (Figure 2). There is an established research infrastructure within the field of the monitoring of atmospheric composition in the first two settlements. High-quality long-term measurements of background air composition, including $O_3$ and BC concentration, are performed by the Norwegian Institute of Air Research at the Zeppelin station located at the mountaintop (474 m a.s.l.) two kilometres to the south-east from Ny-Ålesund, while $SO_2$ and $NO_x$ monitors have been installed in the middle of the village to study the local air quality since 2008. In Barentsburg, $SO_2$, $O_3$, $NO_x$ and meteorological measurements are performed continuously by the Russian Arctic and Antarctic Institute (AARI) since 2017. Aerosol observations such as BC and aerosol optical depth data are collected by the AARI specialists for the V.E. Zuev Institute of Atmospheric Optics of Siberian Branch of the Russian Academy of Science. Although there are several automatic meteorological stations around Longyearbyen operated by the University Centre in Svalbard (UNIS), there is no continuous measurements of atmospheric composition in the town. Short-term observations of $NO_x$ and

$NO_x$, $SO_2$, $O_3$ and BC were performed by Alena Dekhtyareva in Longyearbyen in spring 2017 and summer 2018, respectively.

### 2.3.2 Stationary and portable measurement equipment

Hourly data from stationary chemiluminescence $NO_x$, UV fluorescence $SO_2$ and UV photometric $O_3$ analysers and aethalometers have been studied in the current work. The data from different instrument models with the same measurement principle have been available from Ny-Ålesund, Longyearbyen and Barentsburg. This adds some uncertainty to the comparison of measurement results from the three settlements in addition to the different calibration procedure employed there. Beside the hourly data, daily $SO_2$ and $XSO_4^{2-}$ concentrations accumulated in filter samples collected in Ny-Ålesund have been analysed (NILU, 1996). The data from the condensation particle counters and sun photometers have been studied in addition to the main measurements stated above.

Several portable environmental sensors have been used during the fieldwork in Longyearbyen in spring 2017 and summer 2018.

A broad variety of low-cost sensors is available in the market, however, the performance of the sensors varies significantly (Jiao et al., 2016; Castell et al., 2017). During the fieldwork in Svalbard, three types of sensors for gaseous and particle measurements (Cairpol $NO_2$ electrochemical sensor, MiniDISC particle counter and microaethalometer AE51 for BC measurements) and the portable weather trackers Kestrel 5500 have been used.

The low-cost mobile gas sensors is a new technological solution for environmental monitoring (Jiao et al., 2016). Advantages of these devices is that they are portable and relatively inexpensive, while disadvantages are decrease in sensitivity with time and measurement interference with other gases. Therefore, the sensors cannot be used alone to measure ambient air concentration without a reference monitor. Thus, combined usage of stationary reference device and mobile sensors may cover broader spectrum of detectable concentrations and may be used for the observations close to the pollution source.

Cairpol is a portable $NO_2$ sensor for air pollution studies. The sensor may give reliable $NO_2$ measurement results when higher concentrations than those that are typical for ambient air on rural site are sampled. Therefore, this sensor may be suitable for evaluation of emissions from snowmobiles in the immediate vicinity of the source of pollution.

Main disadvantage of portable gas and particle sensors is their high limit of detection (LD). According to the instruments manufacturers, LD of Cairpol $NO_2$ sensors is 20ppb, LD of AE51=50-100 $ng \cdot m^{-3}$ (5 minute resolution), while measurement range of the MiniDISC particle counter is $10^{3}$-$10^{6}$ particles per $cm^3$ (Fierz *et al.*, 2011). Although high reproducibility have been obtained for microaethalometers in previous studies (Cai *et al.*, 2014), the performance depends on the concentrations (Ferrero *et al.*, 2016), and measurements obtained in the environment with lowest concentrations have the highest noise ratio, and further data post-processing may be needed (Hagler *et al.*, 2011).

The portable meteorological sensors are more suitable for operations in the Arctic conditions. For example, according to the manufacturers' specifications, the range and accuracy of wind speed and air temperature measurements by Kestrel 5500 Weather Meter and Onset stationary sensors are shown in Table 1.

Table 1 Accuracy and measurement range for the stationary Onset sensors installed at the UNIS roof and Kestrel 5500 sensors

| Parameter | Accuracy | Measurement range |
|---|---|---|
| Air temperature, Kestrel 5500 | ±0.5°C | -29.0°C to 70.0 °C |
| Air temperature, Onset S-THB-M002 | ±0.21°C (0°C to 50°C) | -40°C to 75°C |
| Wind speed, Kestrel 5500 | ±0.1 $m \cdot s^{-1}$ | 0.6 $m \cdot s^{-1}$-40 $m \cdot s^{-1}$ |
| Wind speed, Onset S-WCA-M003 | ±0.5 $m \cdot s^{-1}$ (u<17 m / s) | 0 to 44 $m \cdot s^{-1}$ |

The comparison of the Kestrel measurements with the data from the Onset sensors is presented further in the current thesis (part 3.4).

## 2.4 Reanalysis and trajectory model data

ERA-Interim and ERA5 data have been used to assess the synoptic-scale meteorological conditions over Svalbard for the periods of interest. ERA-Interim has a six hours temporal and coarse spatial resolution. In contrast, in ERA5, a new version of the global reanalysis dataset with hourly output frequency, the horizontal and vertical resolutions have increased from 79 km to 31 km and from 60 to 137 levels, respectively. (Dee et al., 2011; Hersbach and Dee, 2016).

Despite the main focus of the current work is local pollution in Svalbard, the backward trajectory modelling has been used to study long-range transport of SLCFs to the measurement sites. FLEXTRA and HYSPLIT are 3-dimentional trajectory models driven with the meteorological data with a spatial resolution of 1.25 degree from the European Centre for Medium-Range Weather Forecasts and 2.5 degrees from global NCEP/NCAR Reanalysis, respectively (Stohl, 1998; Stein et al., 2015). The temporal resolution of the input meteorological data in both models is six hours.

## 2.5 Statistical approach

### 2.5.1 Kolmogorov-Smirnov test for normality

The Kolmogorov-Smirnov test for normality has been used to check if the data in x population are normally distributed (Lilliefors, 1967). The test result is the maximum absolute difference between the empirical cumulative distribution function $S_N(X)$ calculated from *x* and the cumulative distribution function $F^*(X)$ for a standard normal distribution:

$$D = \max_x |F^*(X) - S_N(X)| \quad (4)$$

The D is calculated and the p-value, the probability of observing a test result as extreme as the observed value under the hypothesis that the data in vector x comes from a standard normal distribution, is obtained. If the p-value is less than 0.05, the hypothesis is rejected.

The function *kstest* in the MATLAB software has been applied to perform the calculations (MathWorks, 2019b).

### 2.5.2 Wilcoxon rank sum test

To compare the two samples from the observational dataset grouped according to some principle, for example, presence or absence of some environmental factor, the two-sided hypotheses that the two populations are equal may be tested using t-test or Wilcoxon rank sum test (WRS-test). The WRS-test has been used in Paper II, III and IV instead of t-test because the former performs better for the discrete samples and the data, which are not normally distributed (Krzywinski and Altman, 2014).

The ranks in the two independent samples of sizes $n_X$ and $n_Y$, which have been taken from populations X and Y and ordered in the combined sample with size $N = n_X + n_Y$ from smallest to largest, may be used to define, which of the populations has the highest median value. If the sum of ranks in the sample from X population are higher than from the second sample, then the median of the X population is generally higher than the median of the Y population (Gibbons and Chakraborti, 2003).

The WRS-test is equivalent to the Mann-Whitney U-test. We find the Mann-Whitney U-test statistic from the sum of the ranks for the observations, which came from the sample X:

$$U_X = R_X - \frac{n_X(n_X+1)}{2} \tag{5}$$

where $R_X$ is the sum of the ranks in sample X.

Similarly, we find the U-value for the sample Y:

$$U_Y = R_Y - \frac{n_Y(n_Y+1)}{2} \tag{6}$$

Since there is a connection between the ranks of the two samples such that $R_X+R_Y=N\,(N+1)/2$ and $U_X + U_Y = n_X n_Y$, the MATLAB function calculates only the rank sum of the first sample.

The WRS-test is related to the U-test as:

$$W = U + \frac{n_X(n_X+1)}{2} \tag{7}$$

The smallest value of U is used to define the significance of the result using the significance tables for small samples or z-statistic in case of large samples.

The full description of the MATLAB function *ranksum* is given in the MathWorks web-page (MathWorks, 2019d).

### 2.5.3 Pearson and Spearman correlation

The MATLAB function *corr* (MathWorks, 2019a) has been used to calculate the Pearson correlation coefficient to check if there is any statistical significant linear relationship between the same parameters measured at different stations such as the correlation between $NO_x$ in Longyearbyen and Ny-Ålesund or by different equipment, for example, correlation between BC values obtained by the AE33 and AE51 aethalometers.

The Pearson correlation coefficient for the two variables x and y is following:

$$r_{yx} = \frac{\sum_{i=1}^{n}(x_i-\bar{x})(y_i-\bar{y})}{\sqrt{\sum_{i=1}^{n}(x_i-\bar{x})^2 \sum_{i=1}^{n}(y_i-\bar{y})^2}} \quad (8)$$

In contrast, the Spearman correlation is used to test for monotonic relationship between the two variables (Chalmer, 1986). The partial Spearman (rank) correlation coefficients have been calculated to test if the concentrations of atmospheric compounds are related to the meteorological parameters in Paper III.

For example, the Spearman partial correlation for the two variables x and y controlling for the variable z is calculated as:

$$\rho_{yx.z} = \frac{\rho_{yx}-\rho_{yz}\cdot\rho_{xz}}{\sqrt{1-\rho_{yz}{}^2}\cdot\sqrt{1-\rho_{yx}{}^2}} \quad (9)$$

where $\rho_{yx} = 1 - \frac{6\sum d_{yx}{}^2}{n(n^2-1)}$, $\rho_{yz} = 1 - \frac{6\sum d_{yz}{}^2}{n(n^2-1)}$ and $\rho_{xz} = 1 - \frac{6\sum d_{xz}{}^2}{n(n^2-1)}$ are the Spearman correlation coefficients for variables x and y, y and z and x and z, respectively, calculated for $d_{yx}$, $d_{yz}$, $d_{xz}$, *the* difference between the ranks of the two variables x and y, y and z and x and z, accordingly, and *n* is the length of each variable.

The partial correlation has been calculated using the MATLAB function *partialcorr* (MathWorks, 2019c).

### 2.5.4 Monte Carlo method

Monte Carlo method has been used to test the significance of the relationships between the two variables when the correlation coefficient between them has been low (r<0.2) (Graversen, 2006). A new artificial variable with the same power spectra as one of the two variables of interests, but with the shifted phase has been created, and the correlation coefficient has been calculated. The procedure has been repeated 5000 times and the percentage of the correlation coefficients, which are higher than or equal to the original one calculated for the two variables of interest, has been found. The percentage indicates significance of the correlation.

# 3. Discussion of the results

## 3.1 Paper I

**Dekhtyareva A.**, Edvardsen K., Holmén K., Hermansen O., & Hansson H.-C., 2016. Influence of local and regional air pollution on atmospheric measurements in Ny-Ålesund. *International Journal of Sustainable Development and Planning*, 11 (4), 578–587. DOI: 10.2495/SDP-V11-N4-578-587

The main author has been responsible for the work on the article. The contribution of each of the authors listed above is stated in the *Table 1*Table 2. In the Table 2,Table 3,Table 4 and Table 5 the conception is an idea for the research; design is the study planning; supervision is taking the responsibility for the work on the article; funding and materials include personnel, logistical and technical support needed for the study; critical review is the reviewing of the article for its intellectual content.

Table 2 Authors contributions in Paper I

| The authors contributions | D.A. | E.K. | H.K. | H.O. | H. H.-C. |
|---|---|---|---|---|---|
| Conception | + | | + | | |
| Design | + | | + | | |
| Literature review | + | | | | |
| Supervision | + | | | | |
| Funding / materials | | | | + | + |
| Data collection | | | | + | + |
| Data processing | + | | | + | + |
| Analysis and results interpretation | + | + | + | | |
| Writing | + | + | + | | |
| Critical review | | + | + | | |

The paper discussed lifetimes of $NO_x$, $SO_2$ and aerosol particles of different size and seasonal variation of their concentration in Ny-Ålesund from 2008 to 2010. The importance of the Zeppelin Observatory as an international research station for monitoring of background air composition is also stated in the paper. Diesel power plant and ships in Ny-Ålesund and coal power plants in Longyearbyen and Barentsburg are defined as local and regional emission sources, respectively, which may affect the concentration of compounds measured at the Zeppelin station. FLEXTRA air mass trajectory have been used to identify cases when the air masses arriving at the Zeppelin station may have been impacted by the long-range, regional and local emission sources.

It has been observed that the summer wind conditions measured in Ny-Ålesund and at the Zeppelin station as well as at the Svalbard airport in Longyearbyen differ significantly from other seasons: the mean wind speed is lower and onshore wind is observed more often. The westerly wind prevails in summer in Longyearbyen, thus the influence of towns pollution on the measurements at the Zeppelin station is unlikely. In contrast, south-easterly and south-westerly wind may bring regional pollution to Ny-Ålesund. The lack of meteorological observations in Barentsburg restricted analysis of seasonal wind patterns in this settlement.

The seasonality in $SO_2$, $NO_x$, $XSO_4^{2-}$ concentrations and particle size distribution have been explained by the influence of different emission sources and change in environmental conditions in Svalbard. Higher concentrations of smaller particles have been observed in summer at the Zeppelin station, while accumulation mode particles has prevailed in spring. Local $NO_x$ sources have been important in summer and winter, while long-range transported pollution has dominated in autumn and spring. $SO_2$ concentrations have been the highest in winter and spring due to long-range transport of pollution from regional and remote sources.

To clarify the influence of regional pollution sources on air quality in Svalbard and measurements at the Zeppelin station, local air quality monitoring campaigns and sampling of the plume from the coal power plants in Barentsburg and Longyearbyen have been recommended. It has been stated that the results of these measurements may be further used for the plume modelling to study the environmental fate of air pollutants emitted from the largest sources in Svalbard.

In addition to this, the uncertainty in future emission scenarios from ships and power plants in Svalbard and the need for follow-up measurements in all three settlements have been stated in the paper.

## 3.2 Paper II

**Dekhtyareva A.**, Holmén K., Maturilli M., Hermansen O., & Graversen R., 2018. Effect of seasonal mesoscale and microscale meteorological conditions in Ny-Ålesund on results of monitoring of long-range transported pollution. *Polar Research*, 37 (1), 1508196. DOI: 10.1080/17518369.2018.1508196

The work on the article has been managed by the main author. Table 3 indicates the contribution of each of the authors listed above*Table 1*.

Table 3 Authors contributions in Paper II

| The authors contributions | D.A. | H.K. | M.M. | H.O. | G.R. |
|---|---|---|---|---|---|
| Conception | + | + | | | |
| Design | + | + | | | |
| Literature review | + | + | | | |
| Supervision | + | | | | + |
| Funding / materials | | + | + | + | |
| Data collection | | | + | + | |
| Data processing | + | | + | + | |
| Analysis and results interpretation | + | | | | |
| Writing | + | + | | | + |
| Critical review | | + | + | | + |

The seasonality in concentrations of particles and gases in Ny-Ålesund has been described in Paper I. However, the vertical distribution of measurement compounds has not been discussed. The settlement is located in the area with complex topography, and local meteorological processes differ at the various measurement altitudes. The vertical

distribution of local and long-range transported aerosols is dependent on the ABL dynamics and is controlled by the mesoscale and microscale meteorological phenomena.

Paper II investigates correspondence between the daily $SO_2$ and $XSO_4^{2-}$ concentrations detected in the filter samples in Ny-Ålesund (8 m a.s.l.) and at the Zeppelin station (474 m a.s.l.) and analyses the seasonality in the influence of the different environmental factors on the concentrations measured on the two sites. The microscale and mesoscale meteorological conditions have been studied using observations at the two stations and ERA-Interim reanalysis dataset, respectively. In addition to this, the daily radiosonde soundings have been used to investigate the atmospheric stability and wind conditions in the first 500 m of the ABL.

The correlation between the daily $SO_2$ and $XSO_4^{2-}$ data sets from the Ny-Ålesund and the Zeppelin stations has been calculated for different seasons. There is no significant correlation between the $SO_2$ data sets from the two stations in summer, while it is very strong in winter. The values of Pearson correlation coefficient in autumn and spring are intermediate to moderate. The correlation between the $XSO_4^{2-}$ data sets is significant for all seasons, but it is the lowest for the summer data.

The seasonal influence of four major factors on the observations on both sites have been investigated. Three of them may introduce disturbance in the correlation between the data at the two stations, increasing ground-level concentration of pollutants in Ny-Ålesund, while having no effect on the measurements at the Zeppelin station: temperature and humidity inversions, directional wind shear and local summertime emissions from ship traffic. In contrast, the wind speed shear is the factor that may reduce the difference between the two datasets due to enhanced mixing and more effective dispersion of local pollutants. The significance of impact of different factors has been verified by applying of the WRS-test on the two groups of measurements from each of the stations for the days when the specific factor of influence has been present and absent, respectively.

The diagram of the statistically significant factors of influence based on the results of the WRS-test is shown in Figure 8. One can see that all the factors except the directional wind shear are affecting the concentration in Ny-Ålesund in different seasons. Lowest correlation between the datasets at the two stations in summer may be explained by the fact that the Ny-Ålesund data is influenced by several different factors: emissions from ship traffic and insufficient dispersion of local pollution when there is no vertical wind speed shear and strong humidity inversion is present. However, they do not have any significant effect on the median concentrations at the Zeppelin station. In contrast, the significant influence on the concentrations both at the Zeppelin station and in Ny-Ålesund has been observed only for the temperature inversions in spring. The temperature inversions in spring have been formed due to radiative cooling when cold air masses have been transported to Svalbard from east-north-east, and higher concentrations of $SO_2$ and $XSO_4^{2-}$ have been observed in these days.

The correlation between the datasets at the two stations varies due to the influence of different micrometeorological phenomena and local pollution. Modelling of these environmental factors is still challenging, and it needs to be considered when one compares

modelling results with measurements taken at different heights in the area with complex topography.

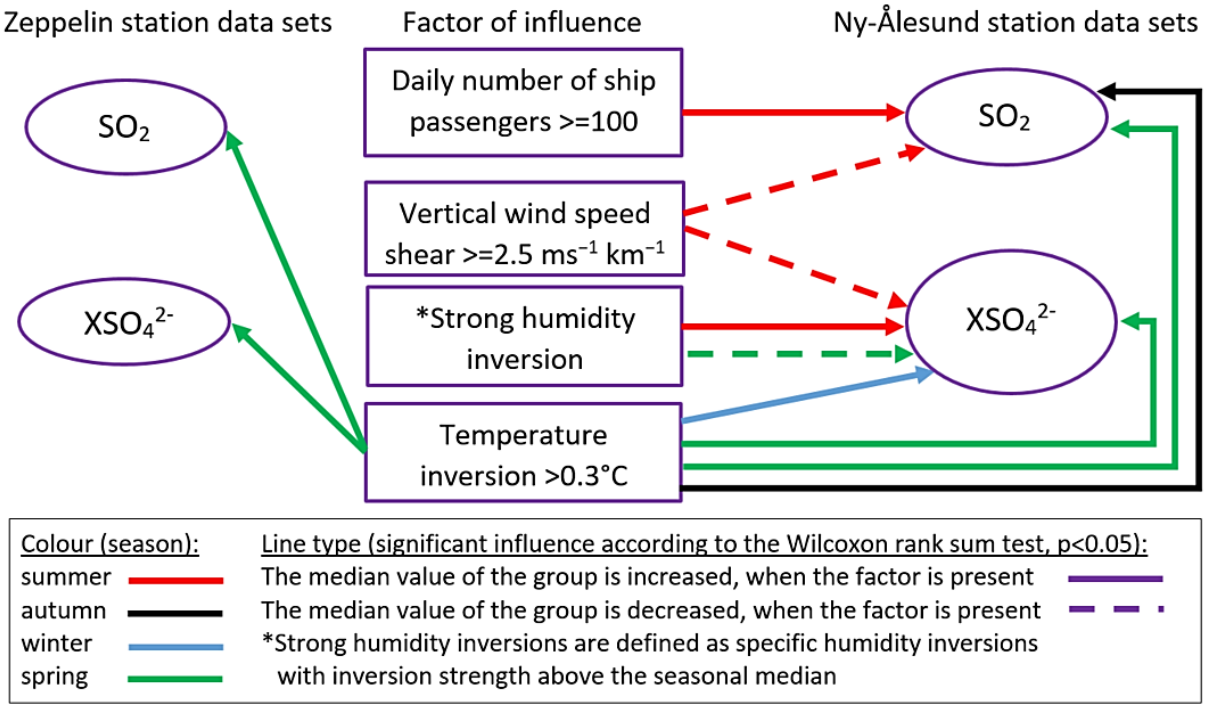


Figure 8 Diagram of the statistically significant factors of influence based on the results of the WRS-test ($p < 0.05$)

## 3.3 Paper III

**Dekhtyareva A.**, Hermanson M., Nikulina A., Hermansen O., Svendby T., Graversen R., & Holmén K., 2019. Springtime nitrogen oxides and tropospheric ozone in Svalbard: results from the measurement station network. *Manuscript ready*

The work on Paper III has been managed by the main author. The contribution of each of the authors listed above is stated in the Table 4*Table 1*.

The importance of long-range transported $NO_x$ for springtime $O_3$ chemistry in the Arctic has been stated in several papers (Beine, Jaffe, Herring, *et al.*, 1997; Beine, Jaffe, Stordal, *et al.*, 1997; Custard *et al.*, 2015). However, only few studies investigate the relationship between $NO_x$ and $O_3$ near the pollution sources within the Arctic (Beine *et al.*, 1996; Custard *et al.*, 2015). Furthermore, the emissions from snowmobile traffic in Svalbard have not been studied until present time. Paper I underlined the necessity of local measurements in Longyearbyen and Barentsburg. In Paper II, the role of complex topography and local micrometeorological processes in creating the difference in concentrations of measured compounds at the two stations located at the distance of two kilometres from each other and at different altitudes have been discussed.

Table 4 Authors contributions in Paper III

| The authors contributions | D.A. | H.M. | N.A. | H.O. | S.T. | G.R. | H.K. |
|---|---|---|---|---|---|---|---|
| Conception | + | | | | | | + |
| Design | + | | | | | | + |
| Literature review | + | | | | | | |
| Supervision | + | | | | | | |
| Funding / materials | + | + | + | + | + | | + |
| Data collection | + | + | + | + | + | | |
| Data processing | + | | + | + | + | | |
| Analysis and results interpretation | + | | + | | + | + | |
| Writing | + | + | | | | + | + |
| Critical review | | + | | | | + | + |

The measurements in Longyearbyen were financed via the Arctic field grant for which Alena Dekhtyareva had applied in October 2016. The project "Monitoring of nitrogen oxides from stationary and mobile sources at Svalbard" had been funded in January 2017. The proposal incorporated testing of portable $NO_2$ sensors to monitor the emissions from snowmobiles and comparison of the results with the standard stationary $NO_x$ monitor.

The mobile $NO_2$ Cairpol sensor and Kestrel 5500 Pocket Weather Tracker were used during the fieldwork trip on snowmobiles to Mohnbukta organized by the UNIS course AT-831 "Arctic Environmental Pollution: Atmospheric Distribution and Processes" on the 05$^{th}$ of May 2017 (Figure 9). Kestrel weather station has been temporarily installed in Sassendalen (001), Mohnbukta (002) and Koningsbergbreen (003).

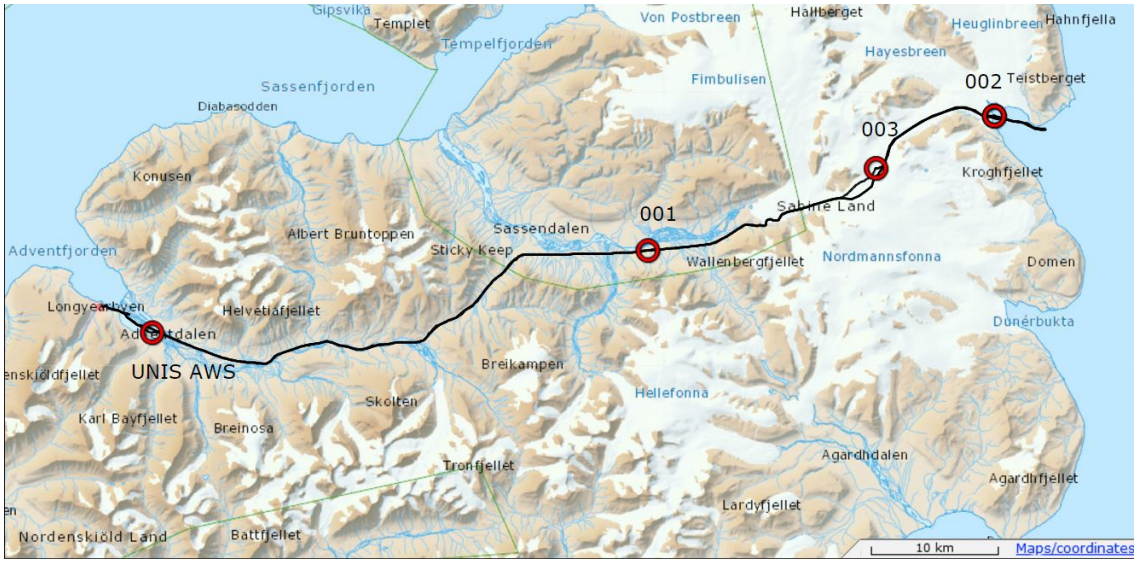


Figure 9 Snowmobile route produced using GPS log. The locations of Kestrel stations (001-003) and UNIS automatic weather station (AWS) are shown by the red circles

The Kestrel tracker has been installed on a tripod for short-term stationary measurements during the stops (Figure 10a). The Cairpol $NO_2$ sensor has been attached to arm of the author during the snowmobile trip (Figure 10b).

The surface wind speed and direction depended strongly on local topographical features, and channelling along the glaciers and valleys has been observed. Most of elevated $NO_2$ values have been detected when the snowmobiles stopped at the measurement stations (up to 24 ppb), while concentrations of the measured compound have been low during the ride (0 ppb).

(a) (b)


Figure 10 a) Kestrel station installed in Mohnbukta; b) project manager Alena Dekhtyareva with Cairpol $NO_2$ sensor attached to the arm to measure $NO_2$ concentration during the field trip

Negligible concentrations of $NO_2$ have been observed during the ride most probably because NO is formed first in the process of fuel combustion, and then it is further converted to $NO_2$ (Seinfeld and Pandis, 2006). The conversion rate depends on the concentration of NO, therefore, when several snowmobiles stop simultaneously, NO accumulates and we can see the production of $NO_2$. The snowmobile with the sensor has been the second from the end in the motorcade of 13 snowmobiles, therefore the air polluted by the emissions from the 11 snowmobiles, which arrived first, has been sampled.

The measurement results obtained from the portable sensors have not been included in the Paper III, since the *in-situ* calibration with the stationary $NO_x$ monitor showed very low correlation between the two instruments ($r=0.22$, $p=0.003$). The Cairpol $NO_2$ sensor

underestimated the concentrations measured at UNIS during the case study when the ships have been near the port of Longyearbyen. The 1-minute $NO_2$ concentrations detected by the stationary $NO_x$ monitor have been up to 26.7 ppb, while Cairpol sensor showed 0 ppb during the whole time of calibration with only three 1-minute values of 1 ppb.

While the portable sensor data are not valid for publication, the measurement results obtained from the stationary $NO_x$ monitor are included in Paper III where they are combined with the data from Ny-Ålesund and Barentsburg.

The main aim of Paper III is to analyse $NO_x$ and $O_3$ observations from the three Svalbard settlements, Ny-Ålesund, Longyearbyen and Barentsburg, in order to study the spatial variability in the concentrations of measured compounds and the effect of emissions from various local sources on the measurement results. Synoptic and micrometeorological conditions affecting the values of observed compounds have been studied using the ERA5 reanalysis dataset, ground-based observations and radiosonde soundings from Ny-Ålesund.

The $NO_x$ concentrations in the three settlements are mostly influenced by the local atmospheric circulation controlling the frequency of transport of polluted air masses from the local sources to the measurement stations. However, the synoptic-scale situations, which promote light winds conditions and formation of temperature inversions, decrease the efficiency of dispersion of local pollutants in the ABL and increase the concentration of $NO_x$ at all stations. In addition to influence from the snowmobile and power plant emissions in Longyearbyen, elevated concentrations of $NO_x$ have been detected when ships have been near the harbour and further investigation of the effect of ship emissions on the air quality in town has been recommended.

In contrast to $NO_x$, the local emission sources in Barentsburg and Ny-Ålesund affect the $O_3$ values insignificantly, and the concentrations are controlled by the prevailing synoptic-scale situation and long-range transport of air masses. Several cases of transport of $O_3$ depleted and enriched air masses have been described and studied with the help of HYSPLIT air mass trajectories.

The main weakness of the study has been the absence of measurements of halogenic species and VOCs playing important role in both $O_3$ and $NO_x$ chemistry in the Arctic. For example, Custard *et al.*, 2015 performed comparison of measurements and modelling of $O_3$ depletion events in low and high $NO_x$ environments and revealed decreased $O_3$ net loss rate in high $NO_x$ environments due to reactions with BrO. However, no such effect has been noticed on the $O_3$ concentrations measured during the $O_3$ depletion events in Ny-Ålesund and Barentsburg studied in Paper III, where only titration of $O_3$ with NO has been pronounced. The modelling of $O_3$/$NO_x$ species could have been done in Paper III as well.

The paper uses outdated emission data from Vestreng, Kallenborn and Økstad, 2009. The only newer data available is yearly reported emissions from the coal power plants in Longyearbyen and Barentsburg published at the https://www.norskeutslipp.no/. However, there is no newer data about local emissions from ships, snowmobiles and generators in all three settlements. If these data would have been available, the local flux of $NO_x$ in Svalbard could have been compared to the horizontal flux of long-range transported $NO_x$ coming to

Svalbard. Then the performed $NO_x$ measurements could have been used to correct the emissions estimates.

In addition to this, the precise estimations of the variability in local emissions in Longyearbyen and Barentsburg are needed. For example, it is unclear if the diesel generator has been working 16.05.2018 during the case study in Longyearbyen, but there is no publicly available information on it.

There have been particular observational challenges in this study, which add some uncertainty to the interpretations of the measurement results: $NO_x$ and $O_3$ monitors have been located at different places in Ny-Ålesund, $NO_x$ monitor in Barentsburg has not been calibrated in the same manner as in Ny-Ålesund and Longyearbyen, and there have been no $O_3$ measurements in Longyearbyen. These issues have been taken into consideration in the process of preparing of the fieldwork to collect summer data in 2018 utilized in Paper IV.

## 3.4 Paper IV

**Dekhtyareva A.**, Drotikova T., Nikulina A., Hermansen O., Chernov D.G., Mateos D., Herreras M., Petroselli C., Ferrero L., Gregorič A., 2019. Summer air pollution in Svalbard: emission sources, meteorology and air quality. *Manuscript ready*

The work on Paper IV has been supervised by the main author. The contribution of each of the authors listed above is stated in the *Table 1*Table 5.

Table 5 Authors contributions in Paper IV

| The authors contributions | D.A. | D.T. | N.A. | H.O. | C.D.G. | M.D. | H.M. | P.C. | F.L. | G.A. |
|---|---|---|---|---|---|---|---|---|---|---|
| Conception | + | | | | | | | | | |
| Design | + | | | | | | | | | |
| Literature review | + | | | | | | | | | |
| Supervision | + | | | | | | | | | |
| Funding / materials | + | + | + | + | + | + | + | + | + | + |
| Data collection | + | + | + | + | + | + | | + | | + |
| Data processing | + | + | + | + | + | + | + | + | | + |
| Analysis and results interpretation | + | + | + | | | + | + | + | + | + |
| Writing | + | + | | | | + | | | | |
| Critical review | | + | | | | + | | + | + | + |

The importance of summertime $NO_x$ observations in Longyearbyen to quantify the influence of ships emissions on the local air quality has been defined in Paper III. Thus, to perform the measurement campaign there and compare the data with measurements in Ny-Ålesund and Barentsburg, application for the project “Strengthening cooperation on air pollution research in Svalbard” had been sent by the author of the current work to the Research

Council of Norway in November 2017. The pilot study in Longyearbyen was incorporated in the project proposal for the Svalbard Strategic Grant and was funded in January 2018.

As a result, a ground-based measurements have been performed at UNIS and tethered balloon meteorological and BC soundings in Adventdalen valley in the period from June to August 2018. The BC, $SO_2$, $NO_x$, $O_3$ and atmospheric optical depth (AOD) observations from Longyearbyen have been compared with the data from Barentsburg and Ny-Ålesund. The data from airborne measurements from Adventdalen have been compared with the radiosonde soundings from Ny-Ålesund.

Significant increase of $SO_2$ and $NO_x$ concentrations and decrease of tropospheric $O_3$ values have been observed due to ship traffic emissions in Longyearbyen and Ny-Ålesund. In Barentsburg, the coal power plant has the most significant impact on the air quality, and at times, the pollution level exceeds Norwegian and Russian air quality standards. Long-range transport events have been identified using CO and $O_3$ data from the Zeppelin station and AOD values from Ny-Ålesund. It has been observed that warm air advection from mid-latitudes to Svalbard brings air enriched in $O_3$ and CO in summer, but it also creates strong temperature inversions, beneath which air pollution from the local sources may be trapped and higher concentrations of BC may accumulate.

In addition to the hourly $SO_2$, $NO_x$, BC and $O_3$ observations in Longyearbyen, daily filter samples for PAH analysis have been collected there. The selection of the samples for analysis based on the daily BC concentration has been determined by the fact that PAHs are both precursors for soot formation in the fossil fuel burning process and may be further absorbed by the combustion particles. Firstly, the fuel is pyrolyzed and/or oxidized into hydrocarbon molecules with lower number of carbon atoms such as acetylene and PAHs. Secondly, these gas molecules are polymerized to produce larger PAHs molecules, and when the concentration of these reaches its peak values, soot nuclei may form as a result of reactive collisions between these molecules. Thirdly, the growth of soot nuclei continues until they exceed 10 nm in diameter and start to coagulate and form chain-like structures. When the combustion products cool down, the PAHs are effectively absorbed by the soot particles and may accumulate in high quantities (Seinfeld and Pandis, 2006).

The dominating PAH compound measured in the filters in Longyearbyen in summer has been naphthalene. This result may be partly influenced by the selection of samples collected in the days with highest BC concentrations. Fuels with high naphthalene content have higher sooting tendency (Seinfeld and Pandis, 2006), and therefore there is a natural prerequisite to occurrence of higher naphthalene content in the samples when the concentration of BC, a main component of soot, has been higher. At the same time, naphthalene accumulating on the soot particles may be both from surviving combustion and formed pyrosynthetically (Rhead and Pemberton, 1996).

The PAHs and BC measurements from Ny-Ålesund have not been available at the time of working on Paper IV, therefore concentrations of these compounds have not be compared with the data from Longyearbyen.

Airborne observations performed during the fieldwork in 2018 allowed identifying how different weather regimes affected the stratification of the ABL in Adventdalen and Ny-

Ålesund and BC concentration in the profiles. However, more detailed analysis of the BC profiles and classification on different profile types could have been done as in the work of Ferrero *et al.*, 2016.

Several portable sensors have been used during the field campaign in Longyearbyen: MiniDISC particle counter, Kestrel 5500 weather tracker and AE51 microaethalometer.

The performance of the Kestrel 5500 weather tracker has been assessed *in-situ* using the data from the Onset AWS installed at the UNIS roof. 1-minute Kestrel data have been averaged to the 12-minute sampling interval of the UNIS AWS. The two meteorological parameters, which have been used the most in Paper IV, are wind speed and temperature. The comparison of the data from two portable sensors (serial numbers: 17 and 15) with the data from the AWS is shown in Figure 11a) and Figure 11b), respectively.

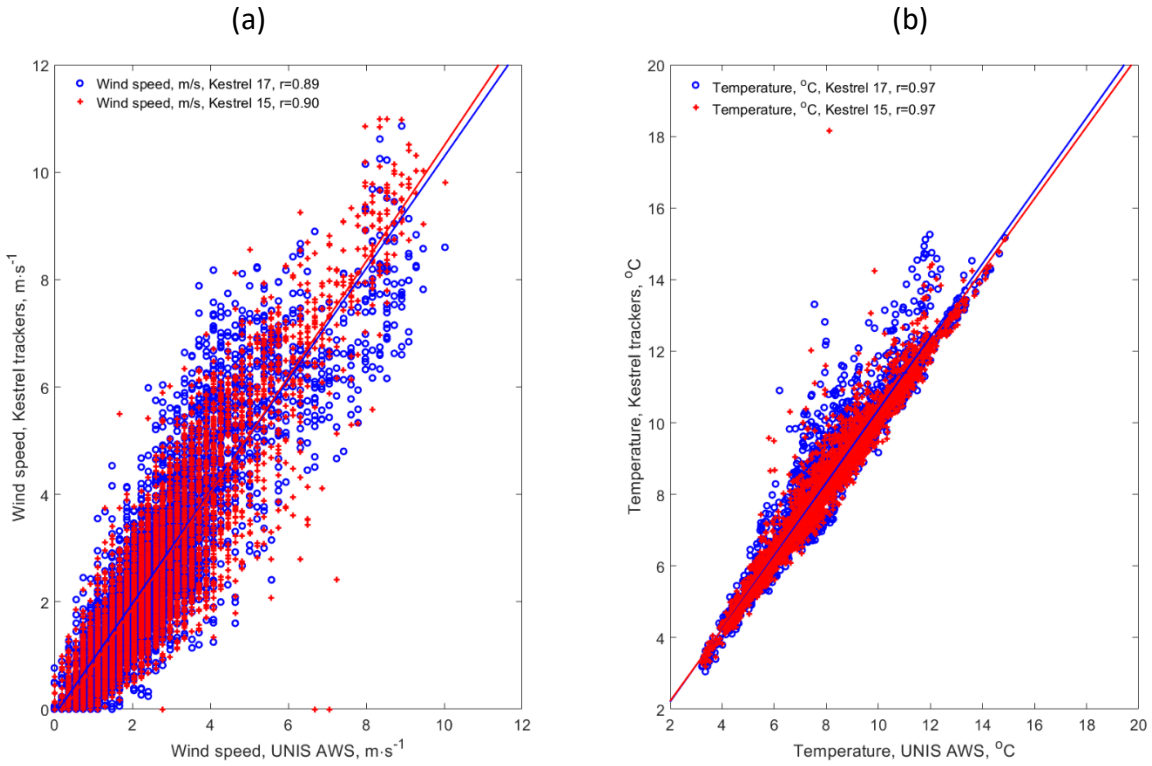


Figure 11 Comparison of Kestrel and AWS data and correlation coefficients for the: a) wind speed; b) air temperature

One can see that the correlation between the data from the Kestrel trackers and AWS is very strong. While the measurement accuracy varies from -3.1 $m \cdot s^{-1}$ to 4.2 $m \cdot s^{-1}$ and from -7 $m \cdot s^{-1}$ to 3.8 $m \cdot s^{-1}$ for Kestrel 17 and Kestrel 15 trackers, respectively, the median difference in wind speed between the data from the portable sensors and AWS has been -0.12 $m \cdot s^{-1}$ and on -0.16 $m \cdot s^{-1}$ for Kestrel 17 and Kestrel 15 trackers, respectively. This result is close to the instruments accuracy of 0.1 $m \cdot s^{-1}$. However, it worth to note that the median underestimation of the wind speed by both portable sensors is ~0.2 $m \cdot s^{-1}$ under low wind speed conditions (<2 $m \cdot s^{-1}$), and sensors performance improves when the wind speed is above 2 $m \cdot s^{-1}$. The precision of the measurements in terms of standard deviation is 1.9 $m \cdot s^{-1}$ for the Kestrel sensors and 1.6 $m \cdot s^{-1}$ for the AWS. The measured accuracy of the

temperature sensors varied in the range of -1.0 ℃- 5.8 ℃ and -0.79 ℃- 16.2 ℃ for Kestrel 17 and Kestrel 15, respectively, with median values of 0.18 ℃ for both Kestrel sensors, which is within the accuracy range stated in the Table 1. The observed warm bias may be reduced by the shielding of the portable sensors from the sun, however, then it may be challenging to keep the wind measurements unobstructed in this case. The precision of temperature measurements is ±2.0 ℃, ±1.9 ℃ and ±1.9 ℃ for Kestrel 17, Kestrel 15 and AWS, respectively.

Unfortunately, no stationary particle counter has been installed in Longyearbyen to compare its data with MiniDISC measurements, since the MiniDISC sensor has been not planned to be used in the campaign. The sensor has been offered for the tethered balloon measurements in addition to AE51 by the project partners during the fieldwork.

The performance of the microaethalometer AE51 is discussed in the Discussion part of Paper IV.

Different types of the ship traffic data have been available for Paper I, II and IV. In the first two papers, the port calls in Ny-Ålesund have been registered manually by the harbour authorities in Ny-Ålesund and the number of passengers has been indicated. In the last paper, the ship traffic log has been based on the automatic identification system data reported to the marinetraffic.com where the gross tonnage (GT) for most of the ships may be found as well. However, the number of passengers has not been available in 2018. The difference between the approaches to the ship traffic analysis makes it challenging to compare the absolute influence of the ship traffic on the concentrations of $SO_2$ and $NO_x$ in 2009, 2010 and 2018. In addition to this, in Paper I only passenger ships are taken into account, while in Paper IV all ships with the GT>100 are considered. Another difference is that only ships registered in the Ny-Ålesund port log obtained from marinetraffic.com have been considered in 2018, while ships anchored in the fjord have not been taken into account.

Furthermore, in Paper I only values of $SO_2$ and $NO_x$ above LD=0.4 ppb are considered. If we use all the summer data from 2008, 2009 and 2010 including those below LD as it is done in Paper IV, we will see much stronger influence of ships emissions: increase of mean $SO_2$ values on 59% and increase of mean $NO_x$ values on 28%, and the result will be comparable with the data presented in Paper IV.

The comparison of summer concentrations (July and August) in 2009, 2010 studied in Paper I and 2018 using the WRS-test reveals significant reduction in concentrations of $SO_2$ and $NO_x$ in Ny-Ålesund in 2018. However, the median wind speed has been slightly higher in 2018 (2.1 $m \cdot s^{-1}$) than in 2009 (1.7 $m \cdot s^{-1}$) and 2010 (1.9 $m \cdot s^{-1}$), thus the dispersion of local pollution might have been more efficient in 2018.

The highest median $NO_x$ concentrations were observed in 2010, when north-westerly wind has been detected more often than in other two years (Figure 12a), while the highest mean concentrations were observed in summer 2018. This may indicate, that the emissions from sources located to the north from the station increased although they do not affect the median summer concentrations strongly due to prevailing south-easterly wind (Figure 12a).

Indeed, the mean concentration of $NO_x$ coming from northerly direction increased from 32.0 $\mu g \cdot m^{-3}$ in 2009 to 54.4 $\mu g \cdot m^{-3}$ in 2018 (Figure 12b).

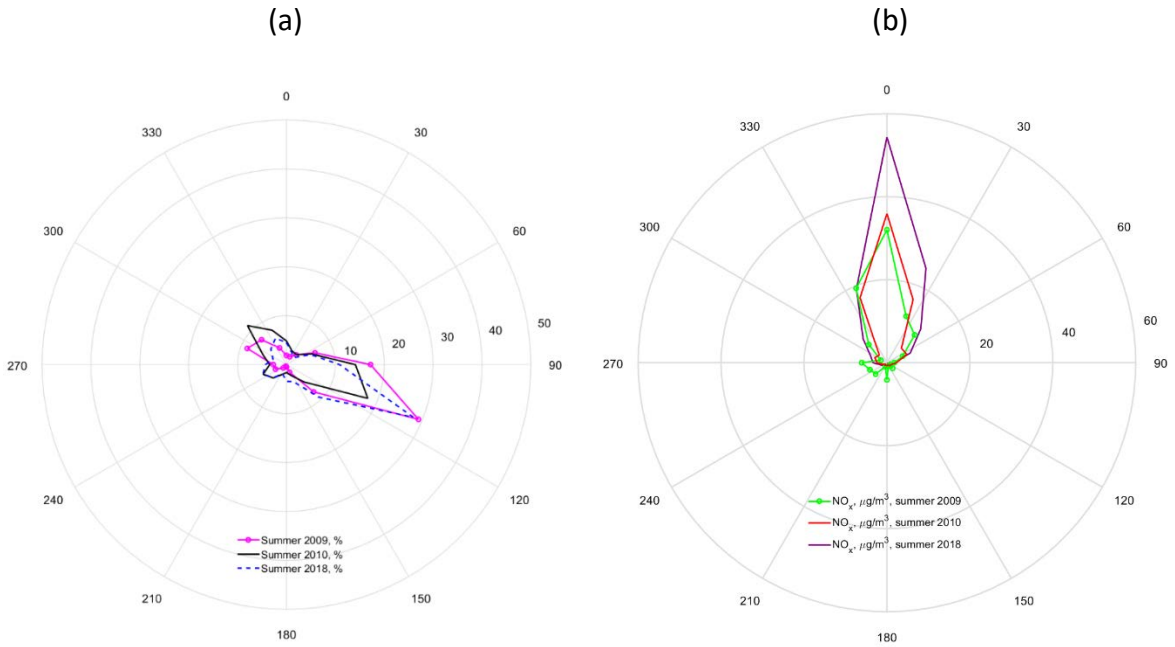


Figure 12 a) Summer wind roses for 2009, 2010 and 2018; b) $NO_x$ concentration averaged over wind directions for 2009, 2010 and 2018

Although, in Paper IV we assume that the emissions from the diesel generator in Ny-Ålesund have not changed significantly since 2013, one can see that the concentrations of $NO_x$ observed when the wind was coming from the north were higher in summer 2018 than in summer 2010 and 2009 even in absence of ships (Figure 13a). This means that updated information about emissions from the power plant is needed, and the reduction of emissions may be recommended to decrease the disturbance of atmospheric measurements in the settlement as it has been stated in previous reports (Shears *et al.*, 1998; Sander, Holst and Shears, 2006; Sander, 2014). The mean concentrations of $SO_2$ for the hours when the ships were present in Ny-Ålesund reduced from 0.38 $\mu g \cdot m^{-3}$ and 0.28 $\mu g \cdot m^{-3}$ in 2009 and 2010, respectively, to 0.16 $\mu g \cdot m^{-3}$ in 2018 probably due to the restrictions on use of heavy fuel oil in Ny-Ålesund since 2015. However, the change in distribution of average $SO_2$ concentrations over wind directions may be noticed in 2018 indicating a possible new source of $SO_2$ located to the south-west from the measurement station (Figure 13b). This needs to be investigated further.

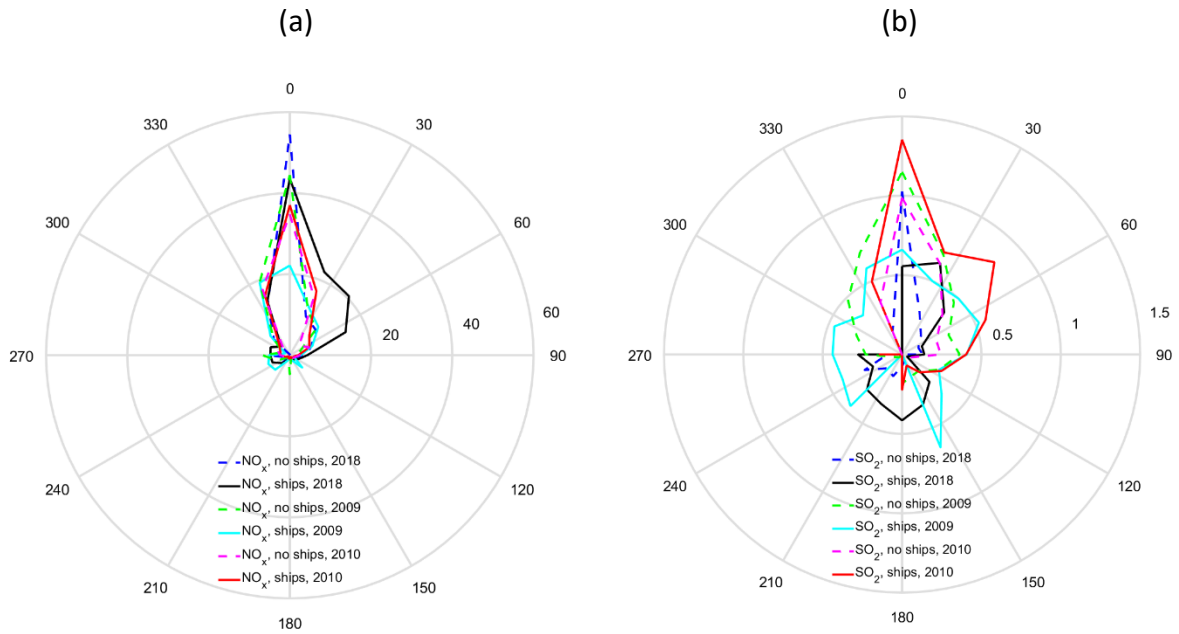


Figure 13 $NO_x$ (a) and $SO_2$ (b) concentrations ($\mu g \cdot m^{-3}$) averaged over wind directions in presence and absence of ships in July and August 2009, 2010 and 2018

## 3.5 Summary of the appended papers

Table 6 describes how the appended papers address the research questions stated in the current study with weakest relationship denoted by "+"sign and strongest by the "+++" sign, respectively. Although the thesis focuses on four different SLCFs measured in the three Svalbard settlements, only Paper IV intends to cover the whole range of compounds on all measurement sites. This is because the studies in Longyearbyen have been done with an external financial support that has been received in the third year of the current PhD project.

Table 6 Appended papers addressing the research questions

| Paper | Measurement sites | Compounds | Research questions | | | | |
|---|---|---|---|---|---|---|---|
| | | | 1 | 2 | 3 | 4 | 5 |
| I | Ny-Ålesund | $SO_2$, $SO_4^{2-}$, $NO_x$ | +++ | | ++ | + | + |
| II | Ny-Ålesund | $SO_2$, $SO_4^{2-}$ | +++ | | + | +++ | ++ |
| III | Ny-Ålesund, Longyearbyen, Barentsburg | $NO_x$, $O_3$ | ++ | +++ | + | ++ | ++ |
| IV | Ny-Ålesund, Longyearbyen, Barentsburg | $SO_2$, $NO_x$, $O_3$, BC | ++ | +++ | +++ | +++ | +++ |

Paper I studies the seasonal evolution of $SO_2$, $NO_x$ and particle concentrations and indicates different remote and local sources, which may affect the concentrations, measured in the settlement. Paper II compares the filter data obtained at two different elevations and studies the processes which may cause discrepancies between the two datasets in different seasons. The long-term measurements have been analysed in both papers, and thus, they can answer to the research question 1 about the seasonal and daily variation in the pollutant concentrations.

However, Paper I and II are based on the data from one measurement site, Ny-Ålesund; thus, they cannot answer to the research question 2 and describe how the pollutant concentrations vary spatially between the three main Svalbard settlements. Paper III and IV are based on short-term measurement campaigns, which capture particular atmospheric composition and meteorological events present during the measurement periods in spring 2017 and summer 2018. The advantage of the field observations has been that higher spatial data coverage has been obtained with three stations around Svalbard. Thus, the research question 2 could be answered only in Paper III and IV. On the other side, the total duration of the measurements has been short, around two months for the data presented in each of the papers. Therefore, the last two papers are to a lesser extent related to the research question 1 since only daily and diurnal variation in the pollutant concentrations have been studied due to the short fieldwork period.

Research question 3 about the current influence of ship traffic on air quality in Svalbard settlements is touched in all papers, however, only Paper IV focuses on the impact on all SLCFs concentrations in both Ny-Ålesund and Longyearbyen.

All papers study the effect of different meteorological phenomena on the ground-level concentration of measured compounds, but only Paper II and IV additionally analyse the impact of microscale and synoptic scale meteorology on the vertical distribution of air pollutants in the ABL and addressed the research question 4 to a higher extent.

Data obtained using different measurement techniques is analysed in the papers, and all of them to some extent describe the advantages and disadvantages of different methods, however, broader variety of methods has been assessed in Paper IV, thus the relationship of this paper to the research question 5 is stronger.

# 4. Research contributions and suggestions for future work

## 4.1 Research contributions

Svalbard is a unique laboratory for environmental studies. Despite its remote location, the consequences of the changes in mid-latitude emissions and global climate are dramatic there. In addition to this, its near-pristine environment is affected by increasing amount of anthropogenic activities in the Arctic region. Studies of changes in both local and long-range transported air pollution performed in Svalbard are of high importance since it is a harbinger of the changing Arctic.

The current work focuses on measurements in Svalbard in all seasons, and hereby provides the opportunity to study the seasonality of air pollution varying due to long-range transport patterns, atmospheric chemistry and intensity of local emissions in the major settlements. The main emission sources in Svalbard are local power plants, however, substantial additional deterioration of the air quality by ship emissions has been revealed in Ny-Ålesund and Longyearbyen. The data from chemical observations performed in the current study under different meteorological conditions can be used to understand the environmental fate of air pollutants better and can be applied to model their future concentrations due to alterations in long-range transport patterns, local emissions, synoptic-scale conditions and micrometeorology.

As it is illustrated in the Table 6, the four appended papers address the five research questions stated in the chapter 1 of the current thesis in a specific way. The main findings from the papers giving summarized answers to the research questions are presented in the current chapter. The following subchapters are dedicated to each of the research questions.

### 4.1.1 Causes of the pollutant concentrations variation on a different temporal scale

The seasonality of sulphur species and $NO_x$ concentrations in Ny-Ålesund is investigated in Paper I and Paper II. The findings confirm the results from previous studies (e.g. Heintzenberg and Leck, 1994) showing that the highest concentrations of $SO_2$ and $XSO_4^{2-}$ are observed in winter and spring when the long-range transport of air pollution to Svalbard prevails, however, in Paper I, specific attention is given to possible pollution transport from regional sources in Longyearbyen in these seasons. This is due to the prevailing SE-wind direction observed there in this time of year. In addition to this, high $NO_x$ concentrations were observed in Ny-Ålesund in winter even in absence of long-range transported pollution indicating importance of local emissions.

The two papers also show that there is a seasonal variation in local wind speed and direction. In summer, in contrast to other seasons, the onshore wind direction in Ny-Ålesund and Longyearbyen is more frequently observed. This creates conditions favourable for transport of the air masses influenced by the marine traffic and biogenic emissions to the measurement stations. At the same time, low wind speed conditions reduce the efficiency of mechanical mixing of the ABL and promote accumulation of local pollution in the settlements. Indeed, the comparison of NO and $NO_2$ observations from Ny-Ålesund studied in Paper III and Paper IV revealed that the summer median levels of these compounds are

more than two times higher than the spring ones. However, as it is shown in Paper II, mixing height obtained from the radiosonde soundings in Ny-Ålesund has often been lower than the altitude of Zeppelin observatory. This indicates that the chemical measurements performed there are less influenced by the local summer emissions than the sea-level station in the settlement.

The daily variations in $SO_2$ and $XSO_4^{2-}$ concentrations at both stations depend on the source region of the air masses arriving to Svalbard. The most polluted air masses have been brought from the east and south-east in winter and spring. According to the spring and summer tethered balloon measurements performed by Ferrero *et al.*, 2016, the aerosols are often inhomogeneously distributed in the ABL in Ny-Ålesund. The soundings represent the aerosol profile structure at the time of observations; however, diurnal evolution of the ABL structure is unknown. In contrast, the high-volume air sampling comprises continuous accumulation of particles and gases in the filter. The comparison of the filter data in Ny-Ålesund performed in Paper II indicates that the layering effect is persistent on a daily scale, and there is a difference in the concentration of daily samples collected at different elevations in the settlement. For example, in days with strong humidity inversion in spring when the moist marine air has arrived to Ny-Ålesund, the concentrations of non-sea salt sulphate have been lower at the sea-level station than at the Zeppelin mountain observatory. In summer, there is also day-to-day variation in the intensity of local shipping emissions in Longyearbyen and Ny-Ålesund and in the efficiency of pollution dispersion. In the days with strong temperature inversions such that occurred in summer 2018 due to transport of extremely warm air masses from Scandinavia to Svalbard, the highest concentrations of BC and $NO_2$ have been observed in Longyearbyen.

The diurnal variation of concentrations in Ny-Ålesund, Longyearbyen and Barentsburg is studied in Paper III and Paper IV. The concentrations of $NO_x$ and $SO_2$ have increased in the daytime and decreased in the nighttime following the diurnal variations of the anthropogenic activities in the three Svalbard settlements, while the concentrations of $O_3$ have shown significant daytime $O_3$ titration with $NO_x$ only in Longyearbyen indicating that the emission quantities of nitrogen oxides are higher there.

**In summary**: There is a strong seasonality in long-range transport of air pollution to Svalbard with peak concentrations of sulphur compounds observed in winter and spring. However, it has also been shown that the local emissions have a pronounced seasonal and diurnal pattern as well. In summer, the ship traffic emissions have significant influence on the air quality in Longyearbyen and Ny-Ålesund. Moreover, median wind speed is lowest in summer, which reduces the ABL mixing, promotes accumulation of locally emitted pollution, and creates discrepancies in the filter measurements obtained at two heights in Ny-Ålesund, at the sea-level station and at the Zeppelin mountain observatory.

### 4.1.2 Spatial variation of the pollutant concentrations between the three main Svalbard settlements

The springtime and summertime spatial variation of the pollutant concentrations between Ny-Ålesund, Longyearbyen and Barentsburg is studied in Paper III and Paper IV, respectively. The concentrations of SLCFs in Svalbard settlements depend on the long-range transport of

air pollution, intensity of emissions from local stationary and mobile sources and local meteorological conditions. The main challenge for the comparison of the data from the three settlements is the usage of different instrumentation and calibration routine. Similar equipment has been used in Longyearbyen and Ny-Ålesund. The instruments have been calibrated at the same laboratory at the Norwegian Institute for Air Research before transportation to Svalbard, and the same sampling and zero and span calibration procedure has been followed during the observations at the both sites. In contrast, different monitors are deployed in Barentsburg and the in-situ calibration routines and calibration frequency are different. The transportation of calibration $SO_2$ and NO gases by boat from Longyearbyen to Barentsburg presents a logistical difficulty. Thus, in this settlement, the in-situ span calibration with the standard gases used in Longyearbyen has been done by the UiT personnel only once in the summer field campaign 2018. During that calibration procedure an overestimation of the $SO_2$ and NO concentrations by 22% and 30%, respectively, has been revealed. Normally, if the calibration period is one week as it has been in Ny-Ålesund and Longyearbyen, the data may be scaled linearly according to the values obtained in the span and zero check. This has not been done with the Barentsburg data as only one reference point was obtained. This surely affects the measurement results, thus the direct comparison of the concentration magnitude at the three sites is challenging. However, one can compare the $NO_x$, $SO_2$ and $O_3$ values from Ny-Ålesund and Longyearbyen presented in Paper IV. In Ny-Ålesund, median summertime NO and $NO_2$ values have been approximately 6 and 9 times lower, accordingly, than in Longyearbyen. In this settlement, the pronounced titration of $O_3$ with $NO_x$ resulted in reduction of median $O_3$ concentrations by 12% from the median background value observed at the Zeppelin station. Median and mean $SO_2$ values in Longyearbyen have been much higher than in Ny-Ålesund, but the air quality standards have not been exceeded in any of the two settlements. In contrast, the highest median $SO_2$ values were detected in Barentsburg and extremely high concentrations of this compound were observed there on the 10$^{th}$ of July 2018. If we take into account possible overestimation of the concentration by the monitor, the recalculated average $SO_2$ value for that day would be 119 $\mu g \cdot m^{-3}$, which is 88 times higher than the maximum daily value detected in Ny-Ålesund. Thus, further follow-up air quality studies with standardized calibration routine are urgently needed in Barentsburg.

**In summary:** There is a broad span of SLCFs concentrations in the three major settlements in Svalbard. As expected from the emission inventories presented in Paper IV, the lowest median concentrations of $SO_2$ and NO have been detected in Ny-Ålesund, intermediate in Longyearbyen and highest in Barentsburg. However, the median BC values in Barentsburg are lower because the car traffic is less intensive and the wind flow bringing pollution from the local coal power plant is less frequent there than in Longyearbyen. The highest mean values of $SO_2$, NO and BC were observed in Barentsburg. However, additional long-term studies with well-established measurement and calibration routines are needed to assess the frequency of occurrence of extremely high SLFCs concentrations there.

### 4.1.3 Influence of ship traffic on air quality in Svalbard settlements

The analysis of the port call data performed in Paper IV revealed that marine traffic emissions significantly increase concentrations of $SO_2$ and $NO_x$ in Ny-Ålesund and Longyearbyen. The concentrations of PAH and BC have increased due to ship emissions in

Longyearbyen as well. Moreover, the magnitude of total PAH concentrations has correlated positively with the ship size. A slight $O_3$ titration with $NO_x$ in Longyearbyen and Ny-Ålesund has been observed in the period of two hours before arrival to two hours after departure of the ships with total gross tonnage exceeding 100. In Paper I and Paper II, significant influence of ships emissions on the concentrations of $SO_2$ and $XSO_4^{2-}$ in filter samples collected in Ny-Ålesund and $XSO_4^{2-}$ and particles with diameter from 50 to 100nm observed at the Zeppelin station has been revealed.

**In summary**: The impact of ship emissions on air quality in Svalbard settlements is pronounced in summer. This is revealed for all range of SLCFs studied in the current work as well as for the concentrations of particles and PAH. Since the efficiency of the ABL ventilation is lowest in summer, the influence on air quality is proportional to the ship size and total number of ships. Thus, the ship traffic restrictions limiting the total number of ships of a specific size visiting port simultaneously are efficient measures, which could be applied in future to reduce the SLCFs concentrations in this season.

### 4.1.4 Meteorological phenomena affecting the ground level concentration of measured compounds and their vertical distribution in the ABL

As it is stated in Paper III and Paper IV, the ground-level concentrations of $SO_2$, $NO_x$ and BC depend strongly on prevailing wind direction at each of the measurement locations. Due to topographic channelling of the wind along the fjords and valleys, elevated pollution levels may be observed in one of the Svalbard settlements and absent in others under the same synoptic weather conditions. At the same time, low wind speed and temperature inversions are the conditions promoting accumulation of air pollutants in the ABL at all stations.

In Paper III, it is observed that the concentrations of $NO_x$ have been higher in calm and cold days due to radiative temperature inversions and supressed ABL mixing in spring. In contrast, higher concentrations of $O_3$ have been observed in Barentsburg and at the Zeppelin station when warmer air masses have been transported from mid-latitudes to Svalbard, meanwhile colder air arriving from the north has been depleted in $O_3$.

In Paper II, strong seasonality in correspondence between the $SO_2$ and $XSO_4^{2-}$ data obtained at two heights in Ny-Ålesund is revealed due to influence of local micrometeorological conditions. The lowest correlation between the two datasets is observed in summer due to insufficient mixing of the ABL and additional local ground-level emissions from the ship traffic.

The tethered balloon measurement results presented in Paper IV, revealed cases of insufficient ABL ventilation and pollution accumulation in Longyearbyen in days when the temperature inversions occurred due to advection of warmer air masses from mid-latitudes to the archipelago. During these events, the background concentration of $O_3$ has increased as well due to long-range transport of air pollution. The frequency of occurrence of such events in summer needs to be investigated and taken into account along with the development of shipping emissions in the Svalbard zone.

**In summary:** The long-range transport events, topographic wind channelling, temperature inversions due to radiative surface cooling in winter and spring, and advection of warmer air masses from mid-latitudes to Svalbard in summer are the factors affecting the ground level concentrations of SLCFs and their distribution within the ABL.

### 4.1.5 Advantages and disadvantages of usage of different measurement techniques for air pollution monitoring in the Arctic

Additional practical achievements of this study are testing of portable and conventional sensors for meteorological and chemical observations in the Arctic environment and attempt to create a measurement network in Svalbard by combination of the data from existing research stations, Ny-Ålesund and Barentsburg, and establishing of new temporal station in Longyearbyen for spring and summer measurements in 2017 and 2018, respectively.

Conventional measurement techniques such as high volume filter samplers, gas monitors and aethalometers have been widely used for the air quality studies and monitoring of long-range transported pollution. Thus, the stations where these instruments are deployed may be easier included in an observational network, the information about the instruments' performance under different environmental conditions is available, and there is plenty of reliable reference measurements to compare with.

However, the usage of conventional ground-based and airborne instruments has a number of disadvantages. A common issue for all ground-based gas monitors and aethalometers are restrictions regarding the location of the measurement station. There should be a continuous and reliable power supply and the instruments must be installed in a heated room with inlets placed outside of the window or secured on the roof. Thus, these measurement techniques allow observations of the SLCFs concentration only in one location and at one vertical level. Sun photometer data contain information about aerosol content in the air column above the station, but the frequency of valid observations is low due to cloudy conditions often observed in summer in the Arctic. Tethered balloon observations allow receiving valuable data about vertical variation in particle concentration and meteorological parameters, but the measurements are time-consuming, and, due to weather and aircraft traffic restrictions, there is rarely a possibility to perform more than one up and down sounding per day. The radiosonde and ozone sonde measurements are less time-consuming, but much more costly, thus the frequency of observations is normally daily and weekly, respectively.

High LD is a common issue for both conventional measurement techniques such as stationary $SO_2$ and $NO_x$ monitors and filter sampling described in Paper I and Paper II, respectively, and portable sensors, $NO_2$ electrochemical sensor and microaethalometer used in work on Paper III and Paper IV, accordingly. This problem may be solved using zero- and span-calibration for the stationary monitors and setting longer sampling time for filters and aethalometers. However, further development of reliable portable sensors with high temporal resolution of measurements is needed for applications in airborne observations and creating a network of stations equipped with the low-cost instruments. In addition to this, the *in-situ* assessment of the performance of these new monitors is needed in the Arctic.

**In summary:** There are advantages and disadvantages of various measurement techniques used for air pollution measurements in the Arctic. Thus, to combine different methods and study concentrations of different SLCFs simultaneously at different locations and altitudes, an international cooperation and joint field campaigns, such as the pilot study performed in summer 2018 in Svalbard, are needed. This allows setting the measurement results in a broader perspective and permits investigation of the state of environment on a regional scale. However, such extensive studies with conventional monitors are costly and need external financial support. Therefore, further development of reliable portable low-cost sensors would allow for increasing the number of observations at different locations in the region. In this case, the measurements could be included in various educational and scientific activities routinely performed at UNIS and research stations around Svalbard.

## 4.2 Suggestions for future work

The meteorological data from daily tethered balloon soundings performed in Longyearbyen in 2018 may be combined with continuous LIDAR observations performed in Ny-Ålesund to study the effect of temporal evolution of the ABL height on the concentration of air pollutants in Longyearbyen and Ny-Ålesund.

There is a great interest in the scientific community in combining the studies of the atmosphere by means of airborne instruments and high-resolution models as they give insight into lower atmosphere layers that are not monitored by the ground-based stations (Kral *et al.*, 2018). The radiosonde measurements are routinely used to supply vertical data for the numerical weather prediction models; however, the temporal and spatial resolution of these measurements is poor. Recently several campaigns have been performed in the polar regions using tethered balloon and Unmanned Aerial Vehicles (UAVs) for meteorological and aerosol measurements (Argentini *et al.*, 2003; Vihma *et al.*, 2011; Jonassen *et al.*, 2015; Moroni *et al.*, 2015; Kral *et al.*, 2018; Leibniz Institute for Tropospheric Research (TROPOS), 2018). The aerosol and turbulence data from the airborne instruments are unique, since they allow to observe local phenomena at high temporal resolution and assess the capability of the large-scale model to reproduce processes important for the environmental fate of air pollutants (Bärfuss *et al.*, 2018; Kral *et al.*, 2018; Leibniz Institute for Tropospheric Research (TROPOS), 2018). However, although some tests on the performance of the portable environmental sensors in Svalbard have been done in the current work, further investigations are needed with more frequent inter-calibration *in-situ* measurements at different concentrations and ambient conditions in order to use the low-cost sensors for airborne measurements described above.

In addition to this, unified approach to the analysis of the ship traffic data is needed to assess the evolution of impact from ship emissions on the concentrations of measured compounds in all three settlements. Besides, the emissions from ship traffic around Svalbard may cause diffuse signal in measurements at the stations due to more diluted concentrations of air pollutants, which may mimic long-range transport events in summer. There have been a number of ship-based campaigns performed at the research vessel Oceania (Ferrero *et al.*, 2019). The data from these campaigns may be compared with the long-term homogeneous ship traffic dataset for the Svalbard zone that may be obtained for research purposes from the Norwegian Coast Guard. Analysis of this dataset together with

the data from research cruises would allow estimating the contribution of marine traffic to the background concentrations of air pollutants in the Arctic.

Long-term measurements in the Arctic emission hot spots such as Barentsburg and plume modelling for different atmospheric conditions are needed to put these observations into a broader perspective. The local emissions of air pollutants in the Arctic are expected to rise with increasing industrial activity, however, the pollutants' lifetime may be decreasing at the same time due to ongoing changes in environmental conditions such as increased air temperature and humidity. High-resolution plume and deposition modelling may be performed to define where the local pollutants from ships and power plants are transported and deposited.

There is an established system for long term monitoring of air quality in Ny-Ålesund, however, no modelling of air quality has been applied (Sander, 2014). According to the strategy proposed by the Research Council of Norway for further development of Ny-Ålesund (The Research Council of Norway, 2019), there is an intent to make it "a world leading observation and research platform for natural sciences". Thus, both the Norwegian and international research projects would benefit from open access high-resolution modelling system for air quality with coupled chemistry and meteorology that would allow to correct the atmospheric measurements performed in the settlement and facilitate scientific activities.

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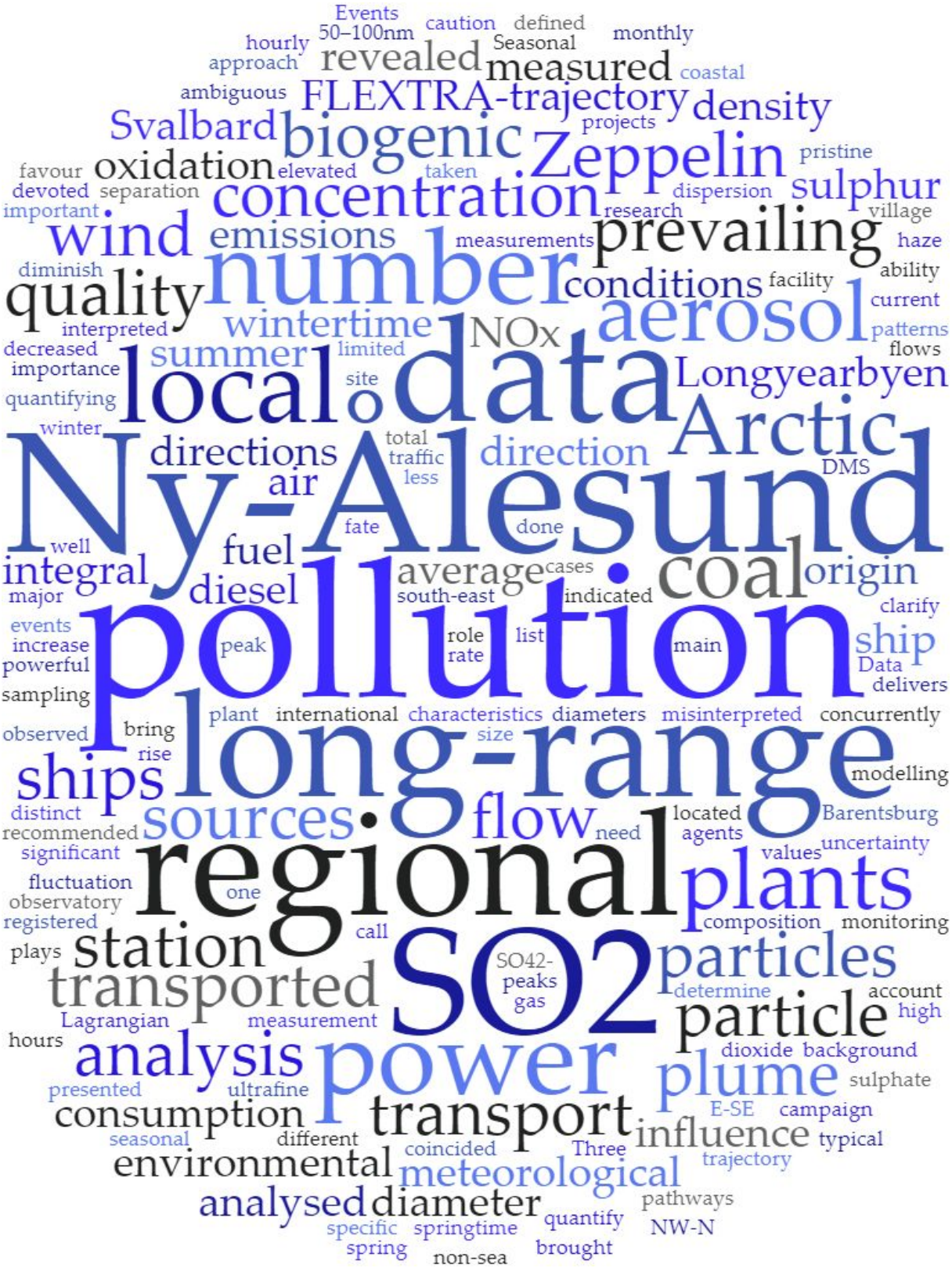
# Part II Appended papers

**Paper I**


Events
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Zeppelin
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dispersion
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current
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wintertime
NOx
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summer
limited
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events
pollution
role
list
ship
increase
peak
rate
main
Data
powerful
delivers
sampling
plant
international
characteristics
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SO42-
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gas
determine
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particle
Lagrangian
measurement
high
hours
analysis
power
dioxide
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sulphate
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# INFLUENCE OF LOCAL AND REGIONAL AIR POLLUTION ON ATMOSPHERIC MEASUREMENTS IN NY-ÅLESUND

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ABSTRACT

The Zeppelin observatory is a research station near the village Ny-Ålesund in Svalbard. The facility delivers data to international projects devoted to high data quality monitoring of the background air pollution in the Arctic. An approach for quantifying the influence of local and regional pollution on measurements that may be misinterpreted as long-range transported one, is presented here.

The hourly gas and aerosol data measured in Ny-Ålesund and at the Zeppelin station, respectively, have been analysed along with the meteorological data from Ny-Ålesund, Zeppelin station and Longyearbyen (south-east of Ny-Ålesund).

Seasonal fluctuation of the average measured values of $SO_2$ and $NO_x$ has been observed. Three main wind directions coincided with the peak concentration of $SO_2$ and $NO_x$. The NW-N flow may bring local pollution from ship traffic and diesel power plant as well as biogenic $SO_2$ from the oxidation of DMS. The monthly average number of particles with diameter characteristic for ship plume (50–100 nm), was elevated for the hours when ships have been registered in the local call list. The number concentration of particles with diameter 200 nm, typical for Arctic haze events, and concentration of non-sea salt sulphate rise during springtime. The FLEXTRA-trajectory analysis indicated that most pollution brought by E-SE and SW flows may be of long-range and/or regional origin. Events with these flow directions need to be interpreted with caution.

*Keywords: aerosol, Arctic, local pollution, long-range transport, trajectory.*

## 1 INTRODUCTION

Nitrogen oxides $NO_x$, ($NO+NO_2$), and sulphur dioxide $SO_2$ are emitted in large amounts from combustion of various fossil fuels worldwide. Monks [1] among others stated that $NO_x$, in the presence of volatile organic compounds (VOC) and/or carbon monoxide CO, are responsible for the production of tropospheric ozone, $O_3$, through the photochemical reactions in urban smog. In turn, as was reported by the Intergovernmental Panel on Climate Change (IPCC) [2], tropospheric $O_3$ is an important greenhouse gas, and being a strong oxidant, it effects the concentration of other greenhouse gases. Moreover, both $NO_x$ and $SO_2$ are acidifying agents, and, according to the Arctic Monitoring and Assessment Programme (AMAP) reports [3, 4], their deposition may have a strong negative effect on many terrestrial ecosystems, specifically vulnerable in the Arctic under rapidly changing climatic conditions, while the impact of emissions of acidifying agents from increasing shipping traffic in the Arctic on marine coastal ecosystems needs to be investigated.

Jacob [5] stated that the lifetime of $NO_x$ is on the order of one day in the lower troposphere at mid-latitudes, and Lelieveld *et al*. [6] reported the lifetime of $SO_2$ and non-sea salt sulphate

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to be approximately 2 days and 5 days, respectively. Therefore, according to the "classic" air pollution literature, the sources of $NO_x$ and $SO_2$ are mostly regional or local. Furthermore, in mid-latitudes, the local air pollution is mainly characterized by higher concentrations, while long-range transported pollution is considered to be more dispersed. However, physical conditions, such as low air temperatures and low humidity, limited turbulent mixing and the absence of sunlight during the polar night, as well as atmospheric dynamics, namely the position of the Arctic front, increase lifetime of pollutants during winter- and springtime Stohl [7]. Due to this, the lifetimes of $NO_x$ and $SO_2$ were estimated by Beine *et al*. [8] and Lee *et al*. [9] as 10 days and 4 days north of the Arctic front, respectively.

During winter the Arctic front barrier, formed by the surfaces of constant potential temperature increasing with height, extends further south (up to 50°N). Consequently, according to Quinn *et al*. [10], in the winter and spring the Arctic haze, polluted air masses transported mainly from Europe and Asia, may be observed in polar regions. On the other hand, during summer the Arctic front is located further north and air may remain continuously north of 80°N in the lower troposphere up to 14 days. Results from several studies, [7,11,12], show that this prevents the transport of pollutants from Eurasia during this season, and local aerosol sources on Svalbard are considered to be more important during this time of the year.

Svalbard's archipelago is nearly a pristine Arctic environment with only a few local and regional anthropogenic pollution sources. Therefore, much of the atmospheric research activities there are devoted to the monitoring of long-range transported pollution in the Arctic.

The Zeppelin Observatory is the Norwegian atmospheric monitoring station situated on a mountain ridge 2 km away from a small research settlement Ny-Ålesund. The station is of high importance for the Global Atmosphere Watch, The European Monitoring and Evaluation Programme (EMEP), AMAP and many other research projects due to unique opportunities for monitoring of background air composition, meteorological and climatological studies.

The main purpose of this article is to discuss ambiguities related to the process of identification of possible sources of air pollution on Svalbard and present an approach for quantifying the influence of local and regional pollution on measurements in Ny-Ålesund.

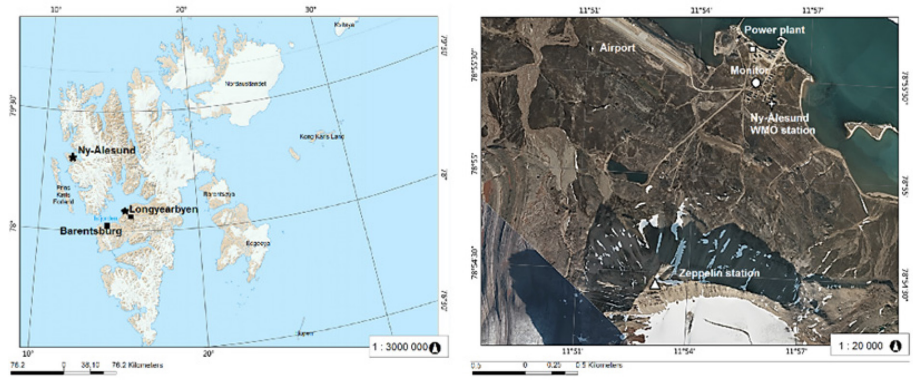
## 2 AIR POLLUTION SOURCES ON SVALBARD

Coal-fired power plants are operated in the two settlements of Longyearbyen and Barentsburg located to the south-east of Ny-Ålesund. According to Vestreng *et al*. [13], these are the largest year-round anthropogenic point sources of $SO_2$ in Svalbard. Both have seasonally variable emission rates, Fig. 1a.

According to the environmental impact assessment of Ny-Ålesund as an international scientific research and monitoring station (Shears *et al*. [14]), the power plant fuelled by low sulphur diesel, is the largest local year-round point source of $NO_x$ in Ny-Ålesund. It too has seasonally variable emission rates. The monitoring station is installed south of the power plant, Fig. 1b.

In addition to this, combustion engines on tourist ships produce fumes that contain $NO_x$, $SO_2$ and particulate matter. Shears *et al*. [14] and Eckhardt *et al*. [15] noted that these summertime local sources of emissions have significant impact on atmospheric measurements in Ny-Ålesund. The impact rate depends on emission rates (ship's size and number of ships present in the fjord simultaneously) and atmospheric conditions.

The overview of the emissions from the main sources is given in Table 1. The emission rates for the first three sources and for the last two sources in the table are calculated from hourly and annual values defined in Shears *et al*. [14] and Miljødirektoratet [16], respectively.




(a) (b)

Figure 1: Study area: (a) map of Svalbard showing Barentsburg and Longyearbyen and meteorological stations as squares and stars, respectively, and (b) local map of Ny-Ålesund showing power plant, monitor, Ny-Ålesund WMO station and Zeppelin station as square, circle, star and triangle, respectively.

Table 1: Main sources of emissions in Ny-Ålesund and on Svalbard.

| | Source | $SO_2$,·$10^{-5}$ $kgs^{-1}$ | $NO_x$,·$10^{-5}$ $kgs^{-1}$ |
|---|---|---|---|
| 1 | Diesel generators and central heating in Ny-Ålesund | 32 | 117 |
| 2 | Small ships in Ny-Ålesund | 14 | 83 |
| 3 | Cruise ships in Ny-Ålesund | 1444 | 3889 |
| 4 | Barentsburg power plant (average 2010–2013) | 7191 | 447 |
| 5 | Longyearbyen power plant (average 2009–2014) | 1368 | 490 |

## 3 MATERIALS AND METHODS

The ground-based $SO_2$ and $NO_x$ observations have been sorted according to the prevailing wind direction and compared with ship traffic statistics. When concentrations of $SO_2$ and $NO_x$ were above a lower detectable limit (LDL), the FLEXTRA-trajectory model output was examined for distinguishing between local and long-range transport of pollutants. By analogy with Stohl [7], if air resided exclusively north of 70°N during the previous 7 days for trajectory arriving at altitude 500 m to the Zeppelin station, it was considered to be no long-range transport of pollutants (NLRT case). If the trajectory data were missing, or there were values of latitude <70°N, it was assigned as a possible long-range transport (LRT) case. According to Brock *et al*. [17], the sub-0.1-μm particles often prevail in the particle number population of aged coal power plant plumes several hours after the emission. In addition, experimental studies of Petzold *et al*. [18] show that the combustion particles have modal diameters centred at 50 nm and 100 nm for raw emissions and for a plume age of 1 hour, respectively. Therefore, the particle mode with d = 50–100 nm has been chosen to check the

possible year-round influence of coal power plant and summertime ship emissions on Zeppelin measurements.

### 3.1 Measurements description

The $SO_2$ and $NO_x$ data were collected by Norwegian Institute for Air Research (NILU) during the project Local Air Quality Monitoring 2008–2010 in Ny-Ålesund [19] from 14.07.2008 to 24.08.2010. Both analysers used in the project, had LDL = 0.4 ppb. Most of the data have been below this value, which is too high for the near pristine environment of Ny-Ålesund. Therefore, the data equal to or higher than LDL have been considered as peaks in this work. The aerosol measurements presented here have been performed by Stockholm University at the Zeppelin station. The daily filter samples data collected by NILU at the Zeppelin station are part of the "Cooperative programme for monitoring and evaluation of long-range transmission of air pollutants in Europe" (EMEP). The non-sea salt sulphate has been defined according to the non-sea salt sulphate correction algorithm presented in the WMO report [20]. An overview of the chemical and aerosol data is shown in Table 2.

Combined analysis of ground-based hourly meteorological data from the monitor with gas analysers and Ny-Ålesund WMO station (78.9230 N, 11.9333 E), operated by NILU and Norwegian Meteorological Institute, respectively, provides coverage for the whole period of air quality measurements. In addition, data from the Zeppelin station and from the Svalbard lufthavn (Svalbard airport) (28 m a.s.l., 78.2453 N, 15.5015 E) have been used, Fig. 1a and b.

### 3.2 Model data description

The FLEXTRA 3D backward trajectories (www.nilu.no/trajectories) used in this study are provided by NILU. They run 7 days backward in time and are based on ECMWF (European Centre for Medium Range Weather Forecasts) meteorological data with spatial resolution of 1.25° and temporal resolution of 6 hours. The nearest trajectory to the time of the peak has been chosen, and when the peak value falls in between two trajectories both have been assessed. If at least one of them passed south of 70°N, then it was considered to be an LRT-case. The modelled data are available for the whole period of interest. Four percent of data is missing (most of the dates in December 2008 and several other discrete days in 2008). As reported by Stohl [26], the position errors in FLEXTRA are in the order of 20% of the travelled distance

Table 2: Data analysed in the paper.

| Equipment and reference | Measured component and units | Location |
|---|---|---|
| Chemiluminescence $NO_x$ analyser (model 200E) [21] | NO, $NO_2$, $NO_x$, $\mu g/m^3$ | Monitor, 78.9247N, 11.9262E |
| UV Fluorescence $SO_2$ analyser (model 100E) [22] | $SO_2$, $\mu g/m^3$ | |
| Condensation particle counters (TSI CPC 3025 and TSI CPC 3010) [23, 24] | Integral aerosol number density and size distribution, $cm^{-3}$ | Zeppelin station, 78.9073N, 11.8859 E |
| Aerosol filter [25] | $SO_4^{2-}$(p), $\mu g\ S/m^3$ | |

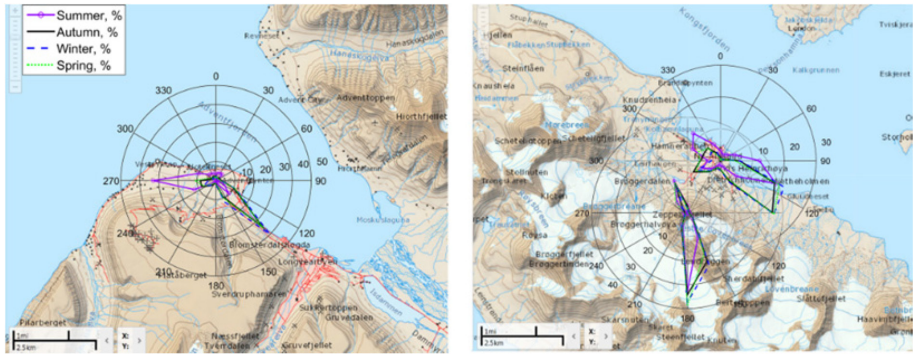



Figure 2: Seasonal wind roses: (a) Svalbard lufthavn station north-west of Longyearbyen and (b) Ny-Ålesund WMO (upper wind rose) and Zeppelin (lower wind rose) stations.

and the performance varies with the meteorological conditions, but the approximate pathway of the air mass may be estimated.

## 4 RESULTS AND DISCUSSION

The seasonal wind roses for the period of measurements are plotted (Fig. 2) for 16 wind directions (22.5° for each sector).

One can see that the prevailing wind direction differs significantly for summer at all stations comparing to other seasons. Most likely, the reason for this is that increasing temperature differences between the land and water facilitate the formation of on shore circulation. The lowest mean seasonal wind speed is observed in summer also. The seasonal wind speed rises gradually and is highest in winter likely due to enhancing influence of mesoscale cyclonic activity, as discussed in Maturilli *et al*. [27]. Additionally, the topographical wind channelling plays important role year-round at all three stations.

The change of wind direction and speed may influence the dispersion of pollutants. The plume from the power plant located to the south-east of the Svalbard lufthavn station in the Longyearbyen town may be trapped in the valleys nearby, Longyeardalen and Adventdalen, due to the prevailing westerly wind direction in summer (Fig. 2a). Therefore, this pollution source is unlikely to influence the measurement results in Ny-Ålesund during this season.

The Zeppelin station is located at the height of 474 m above sea level. The most frequently observed wind direction at the Zeppelin observatory is south and south-south-east due to shadowing effect of nearby mountains, Fig. 2b. From the wind roses for Ny-Ålesund and Zeppelin stations, one can see that the wind direction varies significantly, even within distance of 2 km, due to complex topography.

Analogously, although Longyearbyen and Barentsburg are located in the inner part of the same fjord, Isfjorden, the data from Svalbard lufthavn station most probably can't be utilized for assessment of spreading of pollution from Barentsburg power plant, and separate dataset for Barentsburg is needed (see Fig. 1a).

However, the on shore circulation may bring the pollution from the ships cruising in the Kongsfjorden or attached to pier in Ny-Ålesund in summer, Fig. 2b.

Indeed, despite only 31% and 8% of all $NO_x$ and $SO_2$ hourly data being defined as peak, seasonal fluctuation of the average measured values of $SO_2$ and $NO_x$ has been observed with increasing concentration of gases every summer and winter, Fig. 3. Mean $SO_2$ and $NO_x$ concentrations and number of particles with d = 50–100 nm values were higher on 21%, 16% and 55%, respectively, for time interval of 2 hours before arrival to 2 hours after departure registered in the cruise call list comparing to hours without ships. Elevated values of $SO_2$ in presence of ships mainly coincide with NNW-N wind, which is natural, because the harbour is located north of the monitor (Fig. 1b). However, one peculiar feature of local pollution dispersion pattern has been revealed. Although there are glaciers 3 km south-west from monitor and, according to meteorological studies [27], wind from this direction is of katabatic origin, SW-WSW wind brings at times $SO_2$, $NO_x$ and particles emitted by big anchored ships according to the cruise calls list. These cases are rare but they affect the mean summer concentration of $SO_2$ for this wind direction significantly. Mean $NO_x$ concentrations were approximately five times higher year-round when NW-NNW wind had been observed due to emissions from diesel power plant north of the monitor in Ny-Ålesund.

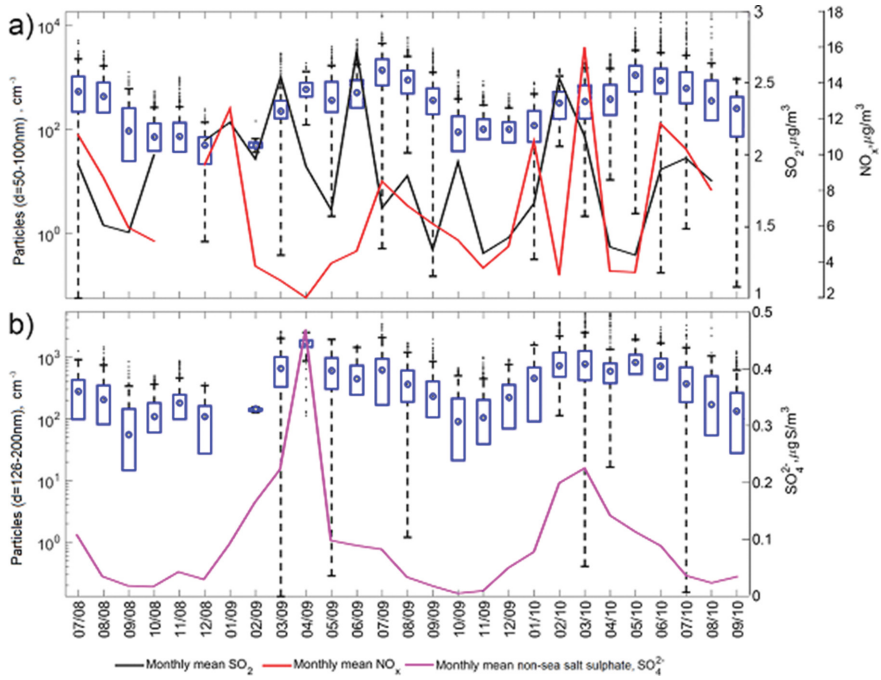



Figure 3: Monthly statistics of integral aerosol number density (box and whiskers plots) and gas analysers and filter samples data (lines): a) integral aerosol number density for particles with d = 50–100 nm, cm-3, and monthly mean $SO_2$ and $NO_x$ from gas analysers, μg/m$^3$; b) integral aerosol number density for particles with d = 126–200 nm, cm-3, and monthly mean non-sea salt sulphate from filter samples, μg S/m$^3$. The logarithmic scale has been used for aerosol data (left y-axis on both figures). The central mark is the median, the edges of the box are the 25th and 75th percentiles, the whiskers extend to the most extreme data points not considered outliers, and statistical outliers are plotted individually on each box.

The integral aerosol number density of particles with size 10–40 nm increases every summer significantly, which indicates boundary layer nucleation events. High mean number of particles were at times observed for northerly wind direction and coincided with $SO_2$ peak even in absence of ships. This supports previous studies of fine particle composition performed by Heintzenberg and Leck [28] that revealed the importance of marine biogenic sources of sulphur compounds on Svalbard.

The concentrations of particles with size 50–100 nm decrease in wintertime and start increasing again in spring with the peak in summer, Fig. 3a. One possible reason for this is that there is no influence of coal power plant emissions on measurements in Ny-Ålesund. However, one may notice that the growth in particle number of this mode starts in February, when the polar night season on Svalbard is over, although there is still no direct sunlight. According to Brock *et al*. [17], the particulate sulphate formation in coal power plant plumes takes place mostly through oxidation of $SO_2$ by OH. The oxidation may occur in aqueous phase as well. However, both processes are restricted due to Arctic environmental conditions in winter, thus the number of particles of this mode is not an applicable parameter for determination of regional pollution on Svalbard in winter. The number of particles with diameter 126–200 nm rises during springtime, and similar pattern is observed for non-sea salt sulphate from filter samples collected at the Zeppelin station, Fig. 3b. This is in good agreement with Seinfeld and Pandis [29] who stated that the Arctic haze phenomena is the long-range transported polar aerosol consisting to a large extent of non-sea salt sulphate of anthropogenic origin.

The trajectory analysis revealed that during summer there is no difference in $NO_x$ mean value whether it was a LRT or a NLRT case, and the concentration depends solely on the wind direction. Concentration decreases during autumn and spring, and mean $NO_x$ value was higher for LRT cases. During winter, values of $NO_x$ for NLRT cases are higher than for LRT ones, probably due to increasing of emissions from the diesel power plant because of enhanced fuel consumption and limited dispersion because of prevailing stable stratification of atmospheric boundary layer. $SO_2$ values for LRT cases were higher for winter and spring and lower than NLRT cases for autumn and summer. In general, the trajectory analysis indicated that most of pollution brought by E-SE and SW flows, 66% and 60%, respectively, may be of long-range and/or regional origin (possibly from two coal power plants located to SE from Ny-Ålesund). There were some cases in wintertime when trajectory analysis did not show possible long-range transport of pollution, however, elevated concentrations of $SO_2$ have been measured and SE wind direction has been detected both in Ny-Ålesund and at the Svalbard lufthavn station several hours earlier. Therefore, either there was influence of regional pollution from coal power plants, or modelled trajectory was erroneous. In order to check this, simple backward trajectory FLEXTRA-model results may be replaced in future work by Lagrangian particle dispersion model results, for example, FLEXPART, as it has been suggested by Stohl *et al*. [30] for more accurate interpretation of measured data. Additionally, case study modelling using a plume dispersion model may be done for the dates of interests for power plants in Longyearbyen and Barentsburg.

The changes in the amount of emissions from ships and power plants are expected. A three-stage treatment system of emissions to reduce $NO_x$, particles and $SO_2$ is planned to be installed in the Longyearbyen coal power. Barentsburg power plant has been stated as the biggest $SO_2$ source in Norway, it is currently operated with a permission for emissions of up to 2400 tons $SO_2$/year as reported by Norwegian Environmental Agency (Miljødirektoratet) [31]. As there is a high rate of uncertainty in the future emission scenarios for the power plants, field campaigns should be performed.

It is restricted to use heavy fuel in Ny-Ålesund since January 1, 2015, and only marine gas oil (MGO) with maximum sulphur content 1.5% is permitted. The emission restrictions will give a steep decline in the number of port calls by cruise ships. However, the $NO_x$ and particle pollution may still be present in summer even if big ships will use MGO instead of heavy fuel. The present study should be continued to quantify the magnitude of improvement in air quality resulting from the emission restriction.

## 5 CONCLUSIONS

The air quality and meteorological data from Ny-Ålesund have been analysed concurrently. The distinct characteristics for the near pristine coastal Arctic site have been defined, such as seasonal patterns in prevailing wind direction and aerosol integral number density specific for different particle diameters and importance of long-range transported pollution.

FLEXTRA-trajectory analysis revealed that most of the total number of $SO_2$ peaks from SE and SW flow may be of long-range and/or regional origin with prevailing LRT cases during winter and spring.

During wintertime, there are no ships or biogenic sources in Ny-Ålesund and due to environmental conditions the oxidation of $SO_2$ to $SO_4^{-2}$ is limited. Because of this, the long-range transport of $SO_2$ plays major role. However, the same conditions favour regional transport of sulphur dioxide from sources located SE from Ny-Ålesund due to prevailing wind direction. In addition, the fuel consumption at the coal power plants may increase during wintertime. The separation of LRT and regional emissions is therefore ambiguous. Data from Hornsund or Hopen could be very powerful agents to diminish this uncertainty in our ability to quantify the regional influence on the quality of the Ny-Ålesund data.

During summer, long-range transported pollution is less important. Despite decreased fuel consumption at the diesel and coal power plants, local pollution from ships in Ny-Ålesund has been significant during summer. However, analysis of integral aerosol number density of particles with size 10–40 nm and $SO_2$ data revealed that biogenic sulphur and ultrafine particle sources have to be taken into account also.

The Lagrangian particle dispersion and plume modelling may be done to clarify the pathways and environmental fate of regional pollution from coal power plants on Svalbard. The measurement campaign for sampling of the plume from the coal power plants in Barentsburg and Longyearbyen may be recommended to determine current rate and composition of emissions.

## ACKNOWLEDGEMENTS

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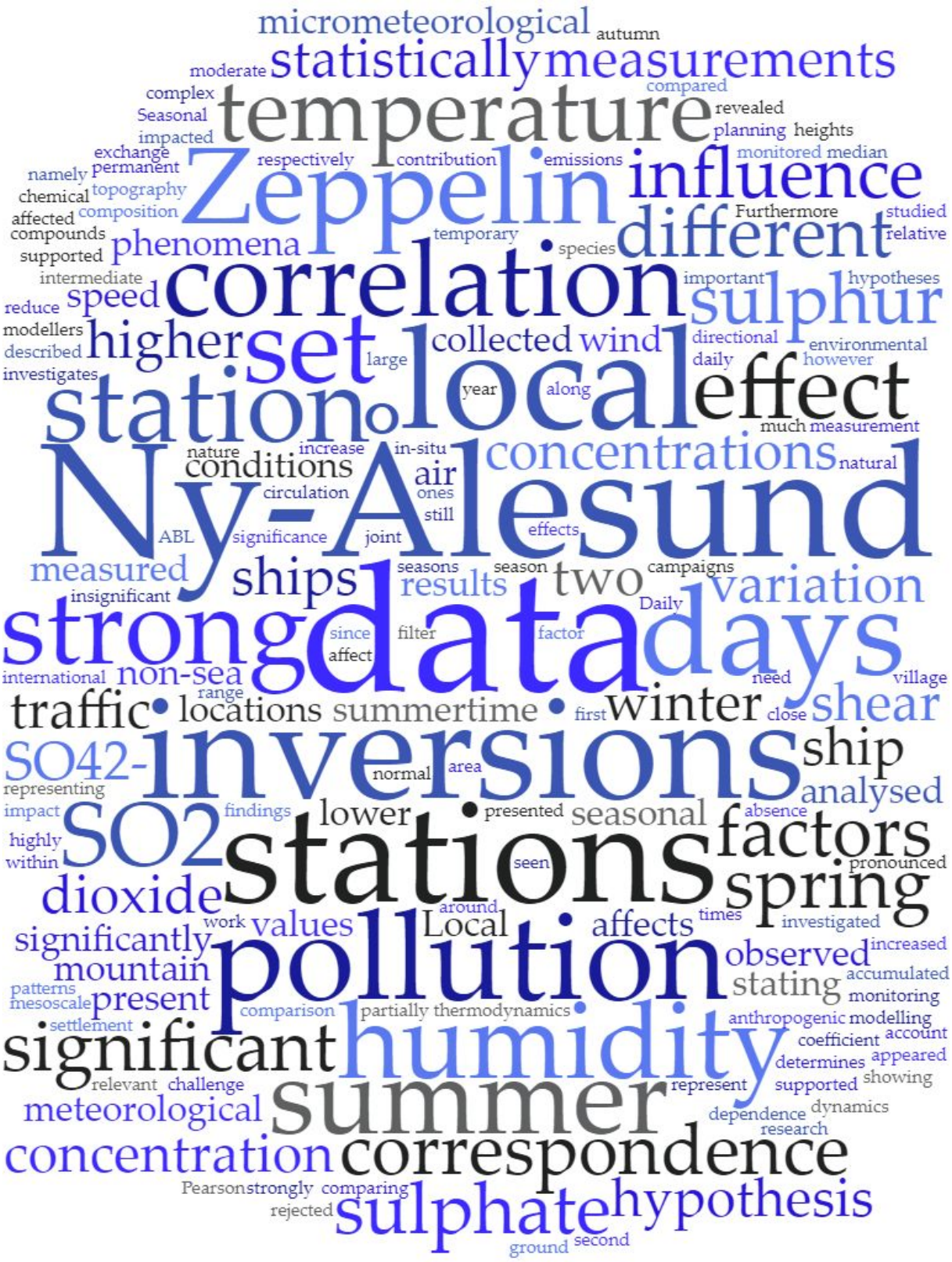
## Paper II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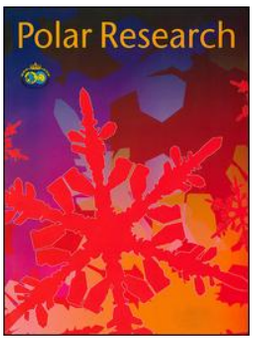

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# Effect of seasonal mesoscale and microscale meteorological conditions in Ny-Ålesund on results of monitoring of long-range transported pollution

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RESEARCH ARTICLE

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# Effect of seasonal mesoscale and microscale meteorological conditions in Ny-Ålesund on results of monitoring of long-range transported pollution

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**ABSTRACT**

Ny-Ålesund is an international research settlement where the thermodynamics and chemical composition of the air are monitored. The present work investigates the effects of micro-meteorological conditions, mesoscale dynamics and local air pollution on the data collected at two different locations around the village. Daily filter measurements of sulphur dioxide and non-sea salt sulphate from the temporary Ny-Ålesund station and permanent Zeppelin mountain station have been analysed along with meteorological data. The influence of different factors representing micrometeorological phenomena and local pollution from ships has been statistically investigated. Seasonal variation of the correlation between the data from Ny-Ålesund and Zeppelin stations is revealed, and the seasonal dependence of the relative contribution of different factors has been analysed. The median concentrations of $SO_4^{2-}$ measured in Ny-Ålesund increased significantly on days with temperature inversions in winter. In spring, concentrations of $SO_2$ and $SO_4^{2-}$ were higher than normal at both stations on days with temperature inversions, but lower on days with strong humidity inversions. In summer, local ship traffic affects the $SO_2$ data set from Ny-Ålesund, while no statistically significant influence on the Zeppelin data set has been observed. The pollution from ships has an effect on $SO_4^{2-}$ values at both stations; however, the concentrations in Ny-Ålesund were higher when local pollution accumulated close to the ground in days with strong humidity inversions.

**KEYWORDS**

Micrometeorology; air pollution; Arctic haze; atmospheric inversion; aerosol; sulphate

**ABBREVIATIONS**

ABL: atmospheric boundary layer; DMS: dimethyl sulphide; WRS: Wilcoxon rank sum

## Introduction

A small community on the north-west Coast of Spitsbergen island, in the Svalbard Archipelago, Ny-Ålesund is a place for fruitful international cooperation in connection with environmental monitoring. Data and knowledge exchange between researchers from 12 different nations leads to joint publications and improves our scientific understanding of various processes in a rapidly changing Arctic (Norwegian Polar Institute 2016). Specific attention is given to the study of the Arctic haze phenomena, aerosol of anthropogenic origin enriched in non-sea salt sulphate ($XSO_4^{2-}$) and transported over long distances from mid-latitudes to the Arctic during winter and spring (Quinn et al. 2007; Dekhtyareva et al. 2016; Ferrero et al. 2016). Ny-Ålesund is situated far from major industrial areas, and is therefore considered suitable for monitoring of long-range transported pollution. This study considers whether the data collected at different locations around the village show differences due to local environmental peculiarities of this site.

The settlement is in a mountainous coastal area near the narrow fjord Kongsfjorden, which has two glaciers at the one end and the Greenland Sea at the other (Fig. 1a). Thermally driven circulations, such as katabatic winds and sea–land breeze, channelling of mesoscale wind along the fjord and the shielding effect of surrounding mountains, are local topographically induced features (Esau & Repina 2012; Maturilli et al. 2013; Maturilli & Kayser 2016). Wind shear and turbulence, affecting the mixing and dilution of air pollutants, may be induced or suppressed by these features (Fisher 2002). Furthermore, temperature and humidity inversions often occur in the ABL as a result of surface cooling during winter and spring and are frequently observed in Svalbard (Vihma et al. 2011). Mixing processes are limited in the stable ABL. This leads to variation in humidity with altitude, and therefore affects the amount of water, condensation processes and particle growth (Stull 1988; Seinfeld & Pandis 2006). Consequently, different aerosol content appearing at different altitudes have been observed during several

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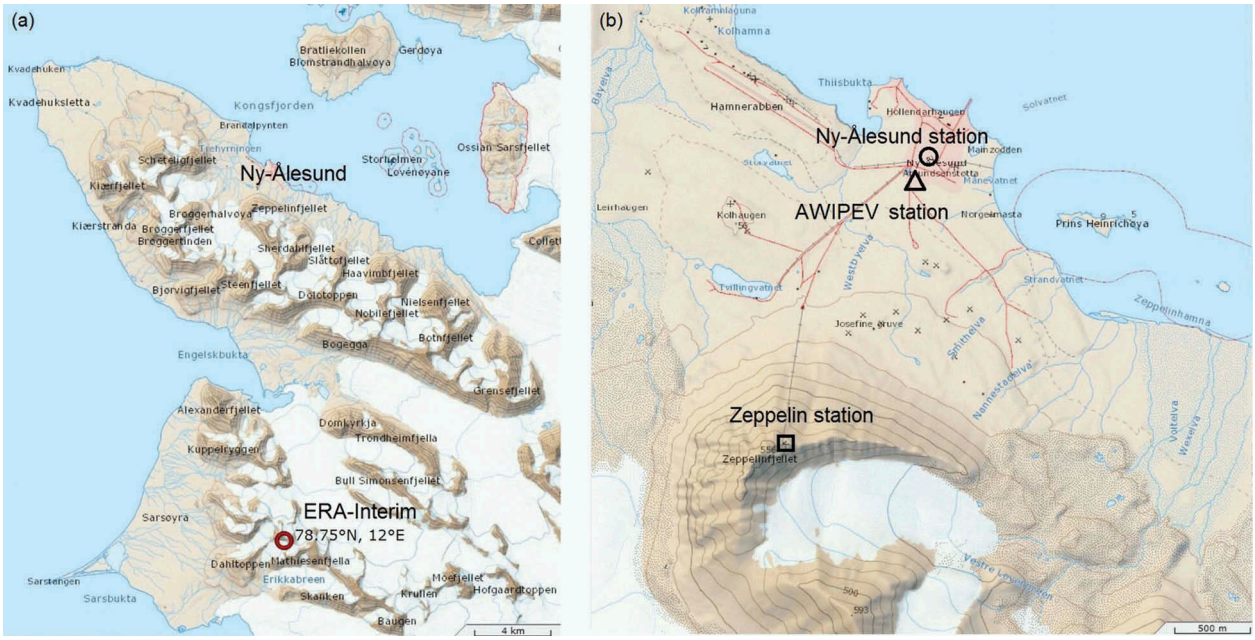


**Figure 1.** (a) The location of Ny-Ålesund on western Spitsbergen and the ERA-Interim data grid point (circled); (b) map of Ny-Ålesund, indicating the locations of the measurement stations.

field campaigns in Svalbard (Moroni et al. 2015; Ferrero et al. 2016). However, the humidity inversions are not always linked to the temperature inversions and may be associated with the unequal distribution of moisture with height in the air masses advected over the measurement site (Nygård et al. 2014).

Long-range transported pollution is dominant during all seasons of the year except summer, when instead ship traffic has been shown to be a significant local source of pollution (Eckhardt et al. 2013; Dekhtyareva et al. 2016). In summer, the mixing height of the ABL may increase, especially due to radiative heating of the surface leading to convection, and therefore local pollution may be transported aloft (Stull 1988). However, studies of wind climate in Kongsfjorden showed that despite this process, the thickness of local surface winds is lowest in summer, when it is estimated to be around 500 m (Esau & Repina 2012). Therefore, it is questionable to what extent convective mixing promotes the even distribution of pollutants within the local ABL. Pollution may also be trapped beneath an inversion layer and accumulate close to the ground, when the air above the measurement site descends and undergoes adiabatic compression and warming. Subsidence inversions may occur in the Arctic troposphere in summer as a response to diabatic heating and convection over land masses in the sub-Arctic (50°-55°N) after the springtime snowmelt (Matsumura et al. 2014). Another source of local pollution in Ny-Ålesund is the small power plant running on low-sulphur diesel year-round (Dekhtyareva et al. 2016). In order to prevent interference of local pollution in the monitoring of background air composition, the Zeppelin Observatory was established 2 km away from the settlement, on the top of Mount Zeppelin at 474 m a.s.l. (Fig. 1b; Braathen et al. 1990; Beine et al. 1996).

There is also a local biogenic source of sulphate in Ny-Ålesund. Sulphate is produced from oxidation of DMS, which is emitted by marine plankton (Keller et al. 1989; Seinfeld & Pandis 2006), and $SO_2$ is an intermediate product of this reaction (Yin et al. 1990). Previous studies have shown that the concentration of chlorophyll α increases in April—May, July and September in Kongsfjorden, suggesting multiple blooms of algae in the fjord (Seuthe et al. 2011). The occurrence of the blooms varies from year to year depending on sea-ice cover, dominating water masses in the fjord and the inflow of freshwater from glaciers and land (Hodal et al. 2012). Although there is a significant positive correlation between algal chlorophyll α and DMS concentration in seawater, the actual DMS concentration depends on the taxonomic composition of the plankton community and trophic interactions within it (Yoch 2002). Because the combination of sunlight and ice-free conditions favours increased DMS emissions, this marine source of biogenic sulphur may be considered significant only in late spring, summer and early autumn (Shikai et al. 2012; Levasseur 2013).

Taking into account these local micrometeorological features and sources of sulphur agents, we address the correlation of the daily concentrations of anthropogenic sulphur compounds measured on filter samples at the Zeppelin station (474 m a.s.l.) and in Ny-Ålesund

(8 m a.s.l.) and their seasonally dependent variations (Fig. 1b).

Chemical and meteorological data were used to test the following hypotheses. (1) Low-level temperature and humidity inversions during winter and spring prevent even mixing of pollutants with height and disturb correspondence between the Ny-Ålesund and Zeppelin station data sets. (2) If the wind direction is dissimilar at the two stations, we expect differences in the concentrations of pollutants as the air sampled at the two sites may have different origins, affecting the correlation between the measurements at sea level and on the mountain top. (3) Wind shear has a significant effect on the dilution of local pollution in the ABL and reduces differences in the measurements from the two stations. (4) Local summertime ship traffic has a strong impact on the Ny-Ålesund data set and induces deviations from the Zeppelin data set.

## Methods

### Study area and materials

Measurements of two key long-range transported sulphur compounds have been analysed in this work: sulphate, $SO_4^{2-}$ (particulate), and sulphur dioxide, $SO_2$ (gaseous). Daily filter samples were collected in Ny-Ålesund during the Monitoring of Local Air Quality in Ny-Ålesund project from 1 July 2008 until 31 December 2009 (Hermansen et al. 2011). A temporary measurement cabin, hereafter called the Ny-Ålesund station, (Fig. 1b), was installed in the middle of the settlement. Measurement results from the Zeppelin station (Fig. 1b) for the same period are available at the website http://ebas.nilu.no/. The sampling procedure was identical to the one used in the European Monitoring and Evaluation Programme (NILU 1996). No correction of filter sampling volume for temperature and pressure has been done, as only the inlets were placed outside while the measurement equipment was kept indoors at both sites, so the equipment was not subject to changes in the environmental parameters (NILU 1996). Only $SO_2$ and $XSO_4^{2-}$ data have been utilized in the present work. The data owner, the Norwegian Institute for Air Research, has performed a correction for sea salt. This institute also provided hourly temperature, pressure, relative humidity, wind speed and wind direction data obtained at the two stations. Previous studies have shown that the local wind measurements at the Zeppelin station are subject to a wind-shielding effect from nearby mountains, so these data cannot be used for comparison with measurement results from Ny-Ålesund (Dekhtyareva et al. 2016). Therefore, atmospheric stratification and local wind flows in the lowest atmosphere (0–500 m height) were studied using atmospheric radiosoundings performed by the Alfred Wegener Institute in Ny-Ålesund. Wind speed and direction, atmospheric temperature and relative humidity data were retrieved from measurements taken with the Vaisala RS92 radiosonde launched from the French–German AWIPEV station (Fig. 1b; Maturilli & Kayser 2016). The soundings provided by the Alfred Wegener Institute cover the whole measurement period except for one day, 8 December 2008. For days with more than one sounding available, the profile closest to the 12 UTC standard launch time has been chosen to maintain consistency.

Vihma et al. 2011 studied meteorological data at the pressure level of 850 hPa and at the surface to assess the influence of prevailing mesoscale meteorological situation on local wind flows and vertical stratification in the ABL. Similarly, we have analysed air temperature, specific humidity, wind speed and wind direction at these two vertical levels in Svalbard and at the point closest to the Zeppelin station (Fig. 1a). For this purpose, meteorological values for 12 UTC have been chosen from the global ERA-Interim reanalysis data set with a 0.75 × 0.75 degrees resolution, and no interpolation has been done (Dee et al. 2011).

### Data analysis

Vertical wind shear is identified as a change in wind direction and/or speed with altitude (Markowski & Richardson 2006). Directional and speed shears were analysed separately in the study reported here. In order to assess the influence of variation in wind direction with height on correlation between the data sets, the daily measurements for each season were divided into two categories: those with and those without a change in wind direction with height of more than 90 degrees. In each radiosonde profile, all measurement points with wind speed above 2 $ms^{-1}$ were defined to exclude cases with very weak winds, which may introduce ambiguity in the wind direction data (EPA 2000). Then the wind direction at each point was compared with values at points located higher in the profile, and the lowest height, when wind direction changes by more than 90 degrees, was defined. If this change happened in the lowermost 500 m, then the day was classified as a day with directional wind shear. The simulation done by Walcek (2002) with a vertical wind shear of 2.5 $ms^{-1}$ $km^{-1}$ shows the significant influence of higher wind speed above the surface on reducing the maximum concentrations and horizontal spreading of polluted air masses. Similarly, in our study, the effect of vertical speed shear was investigated by identifying two separate groups: one in which the wind speed increased by 1.25 $ms^{-1}$ or more per 500 m height and one where it was not observed.

**Table 1.** Meteorological phenomena observed in the radiosonde profiles in different seasons.

| Phenomena observed in the lowermost 500 m | Parameter | Season | | | |
|---|---|---|---|---|---|
| | | summer (n = 154) | autumn (n = 182) | winter (n = 121) | spring (n = 92) |
| Temperature inversion | Frequency of occurrence (%) | 36 | 51 | 66 | 60 |
| | Median inversion strength TIS (°C) | 0.80 | 0.95 | 0.90 | 1.10 |
| Humidity inversion | Frequency of occurrence (%) | 93 | 85 | 77 | 76 |
| | Median inversion strength QIS (g/kg) | 0.22 | 0.10 | 0.07 | 0.09 |
| Both temperature and humidity inversion | Frequency of occurrence (%) | 35 | 44 | 56 | 48 |
| Low wind speed conditions | Frequency of occurrence (%) | 50 | 16 | 13 | 23 |
| Low-level cloud | Frequency of occurrence (%) | 45 | 24 | 19 | 17 |

To assess whether air in the ABL is well mixed, moisture variation with altitude was estimated using relative humidity, air temperature and pressure retrieved from radiosoundings (Stull 1988). Formulas presented by Bolton (1980) and Wallace & Hobbs (2006) have been applied to calculate saturated vapour pressure and specific humidity, respectively.

In order to investigate the influence of temperature and humidity inversions on the correlation between the two data sets, a methodology for inversion detection identical to the one described by Vihma et al. (2011) was used. The height and temperature of the inversion base zTb and Tb, respectively, were defined at the point in the radiosonde vertical temperature profile where temperature begins to increase with height. The height and temperature of the level where temperature starts decreasing with height are defined as zTt and Tt, respectively. Similarly, using specific humidity profiles, height and humidity values at the humidity inversions' top, zQt and Qt, and bottom, zQb and Qb, were found. The temperature and humidity inversion strengths were calculated as the difference of these parameters at the top and bottom levels: TIS and QIS. Following Vihma et al. (2011), temperature and specific humidity changes of more than 0.3°C and 0.02 $g \cdot kg^{-1}$, respectively, through the levels with depth more than 10 m were defined as inversions.

The seasonal $SO_2$ and $XSO_4^{2-}$ data sets from the Ny-Ålesund and the Zeppelin stations were divided into groups according to the absence or presence of the factor of interest: directional and wind speed shear, temperature inversion and/or humidity inversion, and local summertime pollution from ships. To assess the influence of these factors on the seasonal concentrations on filter samples from both stations, the WRS test was chosen, because it is more powerful for discrete samples and data from skewed distributions than the t-test (Krzywinski & Altman 2014). The WRS test checks whether two independent samples, grouped according to a specific factor, come from distributions with equal medians. In other words, if the hypothesis in this test is rejected at the 5% confidence level ($p < 0.05$), there is a statistically significant difference between the two samples, and the factor on the basis of which the data were grouped is recognized as being important.

Another relevant characteristic affecting the possibility of local pollution reaching the Zeppelin station in summer is the height of the mixed layer in the lowest atmosphere. The mixed layer has low variation in specific humidity, wind speed and wind direction (Stull 1988). According to Chernokulsky et al. (2017), mean total cloud cover for Svalbard is highest in summer and constitutes around 80%. To calculate the mixing height, a method combining information about the lapse rate, vertical variation of water content and mixing within clouds, was applied based on a three-step procedure (Wang & Wang 2014). First, mixing height $h_0$ was defined based on vertical gradients of potential temperature, relative and specific humidity and refractivity. Second, the location of a cloud was identified using relative humidity thresholds specific for altitude range from 0 to 2 km (table 1 in Wang & Wang 2014). Third, if a cloud base was lower than $h_0$ and there was a stable layer within the cloud, the consistent mixing layer height $h_{con}$ was set to the height of the sharpest inversion within this layer. The detailed procedure of determining $h_{con}$ is described by Wang & Wang (2014).

## Results and discussion

The seasonal variation of different meteorological phenomena in the lowermost 500 m is presented in Table 1. Temperature inversions were detected most often in winter, but the highest average temperature inversion strength was observed in spring. The rate of days when both temperature and humidity inversions were present is highest for winter. The strongest specific humidity inversions were detected in summer. In general, similarly to results reported by Nygård et al. (2014), humidity inversions were observed most of days, irrespective of the season. Hence, all days were divided into two groups of similar size within each season: (1) with no humidity inversion and humidity inversion with QIS below or equal to the seasonal median; and (2) with inversions with QIS above the seasonal median. This was done to statistically assess the influence of strong humidity inversions on filter measurements by using these

groups in the WRS test for $SO_2$ and $XSO_4^{2-}$ data from the Ny-Ålesund and Zeppelin stations. In low wind speed conditions, the dispersion of pollutants is controlled by a meandering horizontal flow and weak sporadic turbulence, and air stagnation may occur (Anfossi et al. 2004). Low wind speed conditions are defined here as when the median wind speed is below 2 $ms^{-1}$ in the lowest 500 m. The seasonal percentage of profiles with low wind speed conditions was highest in summer and reached its minimum in winter. This may indicate that in summer the measurements are potentially more affected by local processes, while the influence of cyclonic activity and advection is stronger in other seasons (Maturilli et al. 2013). Using the procedure described by Wang and Wang (2014), the cloud location in each radiosonde profile was identified. Low-level clouds were observed in nearly half of all summer days, while they were rarely detected in winter and spring. The frequency of occurrence of the phenomena described above indicate that there is a distinct difference between local micrometeorological conditions in summer and other seasons in Ny-Ålesund.

Figure 2 shows the seasonal data sets of $SO_2$ and $XSO_4^{2-}$ collected at the Ny-Ålesund and Zeppelin stations. Statistically significant positive correlations ($p < 0.05$) of $SO_2$ and $XSO_4^{2-}$ data sets between the stations are observed for all seasons except for summer $SO_2$ data, and values of Pearson correlation coefficient r are shown in each plot of the seasonal data. In order to explain dramatic seasonal variation in correspondence between the data sets from the Ny-Ålesund and Zeppelin stations, the WRS test was applied to $SO_2$ and $XSO_4^{2-}$ data from both stations.

Both $SO_2$ and $XSO_4^{2-}$ data sets show the weakest correlation in summer. According to the WRS test, several factors led to this: strong humidity inversions, insufficient vertical wind speed shear and local pollution from ships (Table 2).

The test results show that only $XSO_4^{2-}$ data from the Ny-Ålesund station had significantly higher median concentration for the days with strong humidity inversions than for the days with normal and no humidity inversion. The mesoscale meteorological situation for the days from both humidity inversion groups is shown in Fig. 3. The location of the isobars shows the area of local high pressure that may be linked to subsidence inversion (Fig. 3a). The mean sea-level pressure for the point closest to the Zeppelin station (Fig. 1a) was higher for the days with strong humidity inversion (1017 hPa) than for the days when strong humidity inversion was absent (1014 hPa). Indeed, there is a weak (r = 0.19), but statistically significant ($p = 0.02$), positive correlation between summer QIS and mean sea-level pressure. The mean wind speed was also lower for the first group (Fig. 3a) than for the second one (Fig. 3b). Meteorological observations at the Ny-Ålesund station show a similar picture as the reanalysis results: easterly winds with very low wind speed (1 $ms^{-1}$) prevailed during the days with strong humidity inversion. Taking into account the location of the station (Fig. 1b), these winds may bring local pollution from the ships anchored in the fjord and/or biogenic sulphur from Kongsfjorden. The presence of strong specific humidity inversion illustrates that pollutants were unevenly distributed with altitude. At the same time, light winds inhibited their removal from the ABL. This indicates that the air was more localized, and if any pollutants had been emitted close to the ground level, they might have persisted for a while.

The presence of gases that may become particle precursors, and enhanced photochemical oxidation, may lead to new particle formation (Seinfeld & Pandis 2006). The factor controlling dry deposition of particles is the particle size. Hygroscopic aerosol particles containing $XSO_4^{2-}$ absorb water vapour from the air, dissolve as humidity increases, and saturated droplets form. The diameter of the particles increases abruptly. If humidity increases further, the particle diameter and mass grows, and the particles may eventually be deposited (Orr et al. 1958). Therefore, the strong humidity inversions in summer may have a cleansing effect, decreasing aerosol concentration to the ambient level above the inversion. Indeed, when the strong humidity inversion was located below the level of the Zeppelin station, no effect of the pollution accumulation in the ABL was seen on the Zeppelin sulphate measurements, whereas a statistically significant influence was observed at the Ny-Ålesund station.

According to the test, the directional wind shear is not a statistically significant factor. However, wind speed shear was an important factor decreasing the median concentration of $SO_2$ and $XSO_4^{2-}$ at the Ny-Ålesund station in summer, while no shift of median in the Zeppelin $XSO_4^{2-}$ data was observed (Table 2). $SO_2$ is a moderately soluble gas, therefore oxidation to sulphate and dry deposition are the major pathways for $SO_2$ removal from the troposphere on non-cloudy days (Liang & Jacobson 1999; Seinfeld & Pandis 2006). The dry deposition velocity depends strongly on wind speed and turbulence strength. Indeed, the concentrations of $SO_2$ and $XSO_4^{2-}$ in Ny-Ålesund were almost two times lower when a vertical wind speed shear was observed. In the absence of wind speed shear, concentrations of both compounds increased dramatically in Ny-Ålesund, while no such effect was observed in the data from the Zeppelin station. Long-term studies based on the Ny-Ålesund radiosonde data record have shown that the ventilation within the ABL above Ny-Ålesund has decreased over the last two decades, as smaller wind

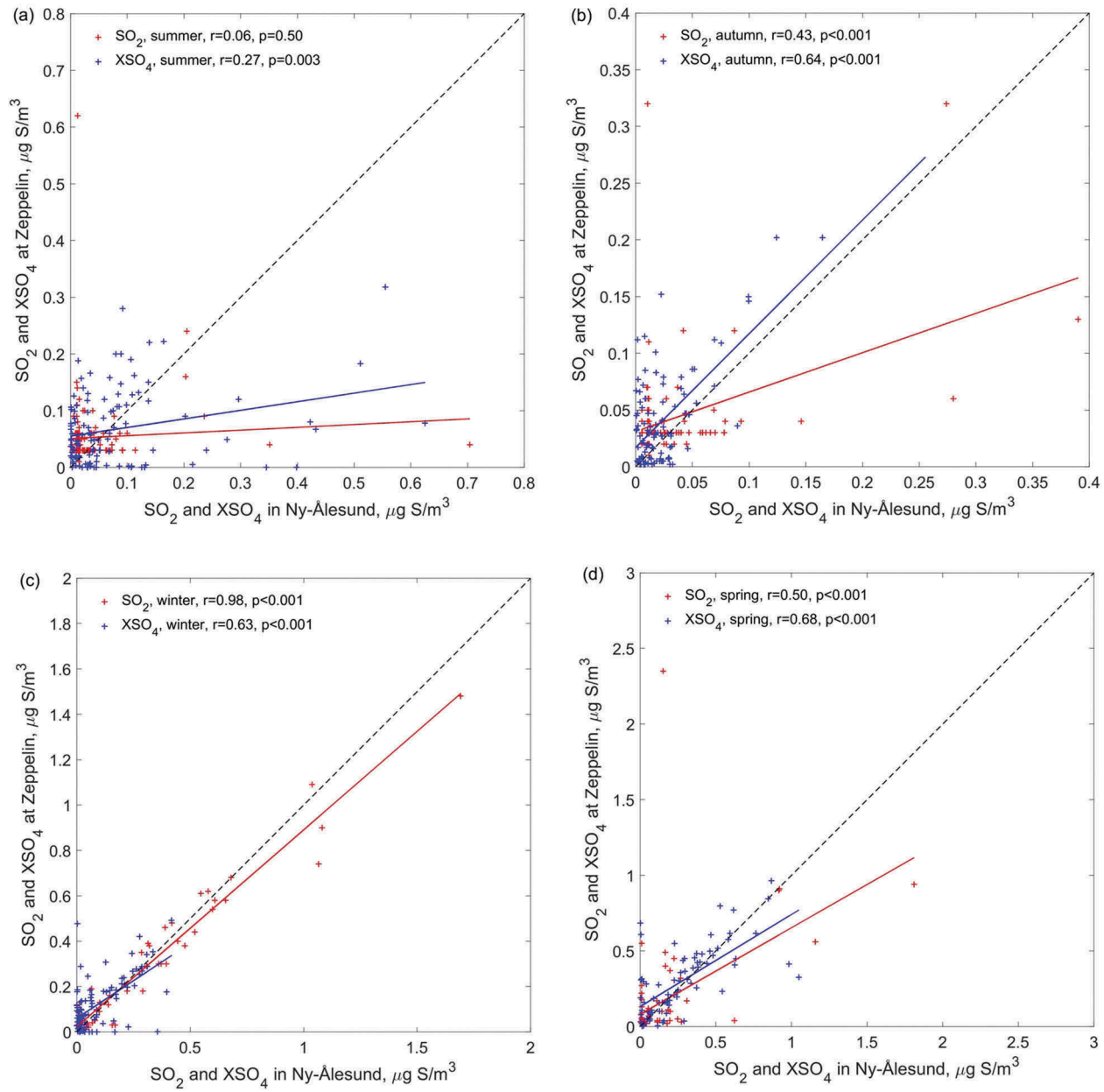


**Figure 2.** Seasonal $SO_2$ and $XSO_4^{2-}$ data from Ny-Ålesund (x axes) and Zeppelin station (y axes): (a) summer; (b) autumn; (c) winter; (d) spring.

**Table 2.** Significant results of WRS-test ($p < 0.05$).

| Season | Compound and station | Factor of influence | $p$ value | Median of the group where the factor of influence is absent (μgS·m$^{-3}$) | Median of the group where the factor of influence is present (μgS·m$^{-3}$) |
|---|---|---|---|---|---|
| Summer | $XSO_4^{2-}$, Ny-Ålesund | Vertical wind speed shear | < 0.01 | 0.0510 | 0.0235 |
| | | Strong humidity inversion | < 0.01 | 0.0296 | 0.0635 |
| | $SO_2$, Ny-Ålesund | Daily number of ship passengers above or equal to 100 | 0.02 | 0.0125 | 0.0220 |
| | | Vertical wind speed shear | < 0.01 | 0.0230 | 0.0133 |
| Autumn | $SO_2$, Ny-Ålesund | Temperature inversion | 0.01 | 0.0115 | 0.0120 |
| Winter | $XSO_4^{2-}$, Ny-Ålesund | Temperature inversion | 0.01 | 0.0292 | 0.0723 |
| Spring | $SO_2$, Ny-Ålesund | Temperature inversion | 0.03 | 0.0118 | 0.0350 |
| | $XSO_4^{2-}$, Ny-Ålesund | | 0.01 | 0.1618 | 0.2675 |
| | $SO_2$, Zeppelin | | < 0.01 | 0.0300 | 0.0400 |
| | $XSO_4^{2-}$, Zeppelin | | < 0.01 | 0.1390 | 0.3210 |
| | $XSO_4^{2-}$, Ny-Ålesund | Strong humidity inversion | < 0.01 | 0.2865 | 0.1218 |

velocities are being observed more frequently in all seasons (Maturilli & Kayser 2016). This implies that the conditions favourable for the accumulation of pollution in the ABL may occur more often.

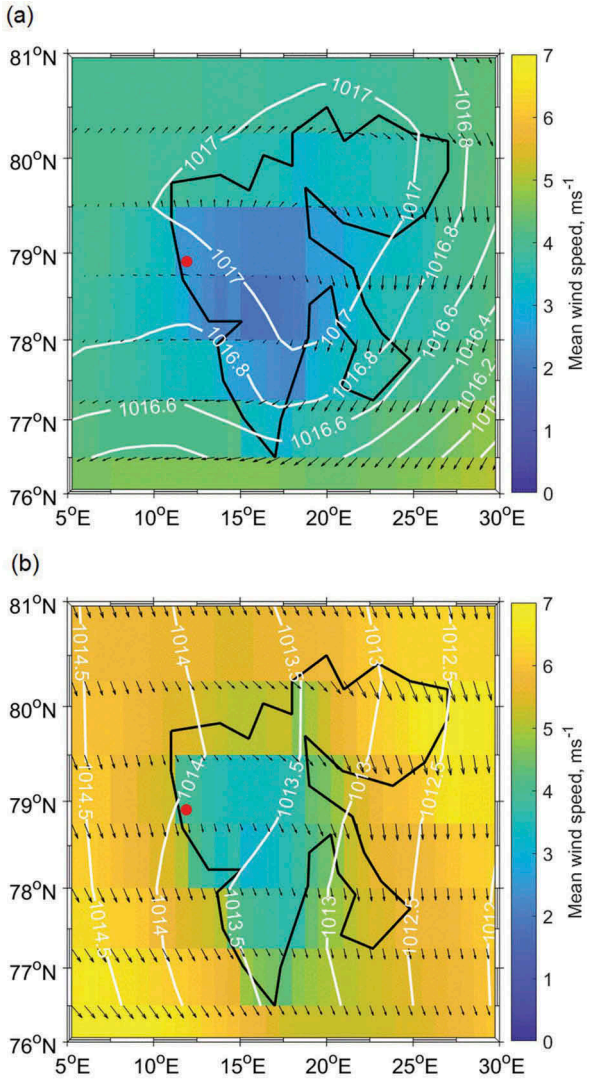


**Figure 3.** Summer mean wind speed in $m \cdot s^{-1}$ (colour scale), wind direction (black arrows with the length relative to the wind speed) and mean sea-level pressure in mbar (white lines) in the Svalbard area (black outline) and Ny-Ålesund (red dot), obtained from surface ERA-Interim data: (a) for days with strong humidity inversion; (b) for days with normal or no humidity inversion.

In order to assess influence of the ship traffic emissions on the measurements at both stations, all summer days were divided into two groups: one group with the daily number of people visiting Ny-Ålesund by ship above or equal to 100; and one group with the number of passengers below 100. The number of people is an indicator of ship size and, hence, the amount of emissions. In total, 66% of the summer data may have been impacted by pollution from the ships, since 101 out of 154 days of summer measurements belong to the first group. However, as Fig. 2 shows, summer $SO_2$ concentrations measured at both the Zeppelin and Ny-Ålesund stations were usually much lower than in winter and spring. There may be a few reasons for this. Firstly, long-range transported pollution prevailing in the Arctic in winter and spring decreases in summer because of the change in the position of the Arctic Front, which prevents effective south-to-north long-range air transport (AMAP 2006). Secondly, there are few local anthropogenic sources of air pollution in the Artic and some of them have intermittent emission rates, e.g., ship traffic (Dekhtyareva et al. 2016). Thirdly, the conversion rate of $SO_2$ to $SO_4^{2-}$ increases in summer, a process governed by two major mechanisms. The first mechanism is the oxidation of gaseous $SO_2$ by hydroxyl radical in clear-sky conditions. Studies have shown that the conversion rate increases with increasing temperature and relative humidity and is a function of Julian day (Meagher & Bailey 1983; Eatough et al. 1994). The second mechanism is the conversion of $SO_2$ to $SO_4^{2-}$ through various chemical reactions in the aqueous solutions in clouds and fog (Eatough et al. 1994; Seinfeld & Pandis 2006). Despite very low average summer concentrations of $SO_2$ measured at the Ny-Ålesund station, there was a noticeable impact of local pollution from ships on the Ny-Ålesund data set.

Mean $SO_2$ values measured at the Ny-Ålesund station for the days in the first group were almost three times higher (0.05 $\mu gS \cdot m^{-3}$) than for days from the second group (0.02 $\mu gS \cdot m^{-3}$), while the Zeppelin data showed almost no difference for these two groups (0.05 $\mu gS \cdot m^{-3}$ vs 0.06 $\mu gS \cdot m^{-3}$). This finding is supported by the WRS test that showed that median $SO_2$ value for the first group of days in Ny-Ålesund measurements was significantly higher than for the second one, while no difference was found for $SO_2$ measurements at the Zeppelin station (Table 2).

Mean $XSO_4^{2-}$ values at the Ny-Ålesund and Zeppelin stations for the first group of days (≥ 100 passengers) were 50% higher (0.09 $\mu gS \cdot m^{-3}$) than for the second one (< 100 passengers; 0.06 $\mu gS \cdot m^{-3}$). This finding corresponds well with a previous study showing that at the Zeppelin station there was an 55% increase in the number of particles with diameters characteristic of aged ship plumes, due to local pollution from ships (Dekhtyareva et al. 2016). However, the WRS test did not show a significant difference in medians for the two groups, which indicates that the sulphate measurements were infrequently affected by local pollution, but sufficiently to influence the mean value, which was increased on account of a few high concentration values.

Total daily number of passengers, indicating the size of ships, shows moderate ($r = 0.38$), but significant ($p < 0.001$), positive correlation with $SO_2$ concentration on filters in Ny-Ålesund, while no significant correlation between these parameters has been found for the Zeppelin data set. In contrast, both the Zeppelin and Ny-Ålesund $XSO_4^{2-}$ data sets show significant ($p = 0.03$), but weak positive correlations ($r = 0.20$) with number of passengers. A possible explanation to this is proximity to the emission source. Whether the ships were docked at the pier or anchored in the fjord, they were closer to the

measurement station in Ny-Ålesund than to the Zeppelin station. The proximity to the source of emission is important because the oxidation from $SO_2$ to $XSO_4^{2-}$ in aqueous and gas phases are major pathways for $SO_2$ removal during the summertime (AMAP 2006), and therefore pollution emitted by ships may contribute to the total sulphate concentration but not to the sulphur dioxide measured at the Zeppelin station.

One may conclude that emissions from ships have an impact on both data sets in a different manner. Sulphur agents emitted by ships reach Zeppelin mostly in the oxidized form of $XSO_4^{2-}$, while the influence on measurements in the village is directly seen on the $SO_2$ data set.

Median and mean consistent mixing layer heights were 546 m and 657 m, respectively. However, only 47% of all profiles showed a mixing height higher than 474 m. This finding supports the original argument for positioning of the Zeppelin station at the mountain top in order to prevent the influence of local pollution on measurements (Braathen et al. 1990). Furthermore, during days with little vertical mixing and strong humidity inversions below the Zeppelin station the local pollution does not reach the station, while higher concentrations were observed in the filter samples in Ny-Ålesund.

An examination of two small sub-groups of summer days serves as an example. In both groups, the mixing height was lower than 474 m and the daily number of ship passengers exceeded 100. The only parameter that was different is the presence of strong humidity inversions in the first group (28 days) and the absence of them in the second one (21 days). On the days in the first group, a slightly lower average wind speed was observed in Ny-Ålesund than on the days in the second group (1.6 $ms^{-1}$ vs 2.1 $ms^{-1}$). The percentage of days when no cloud base was detected and when the cloud base was identified above the Zeppelin station was 39% and 18%, respectively, in the first group of days, and 20% and 12% in the second one. Mean $XSO_4^{2-}$ concentrations in the first group were 0.13 $\mu gS \cdot m^{-3}$ and 0.09 $\mu gS \cdot m^{-3}$ at the Ny-Ålesund and Zeppelin stations, respectively. Mean $XSO_4^{2-}$ concentrations in the second group were 0.07 $\mu gS \cdot m^{-3}$ and 0.09 $\mu gS \cdot m^{-3}$ at the Ny-Ålesund and Zeppelin stations, respectively. In presence of a strong humidity inversion in the lowest 500 m of the ABL, concentrations in Ny-Ålesund increased, while at the Zeppelin station the average concentrations for both groups, with and without strong humidity inversion, were identical.

The processes affecting concentration distribution of aerosols are complex and depend on the particle size and parameters of the atmospheric turbulent boundary layer and the temperature inversion layer if the inversion is present (Elperin et al. 2007). For example, thermal diffusion increases near-surface concentration of coarse particles ($PM_{10}$) in inversely stratified flows over elevated terrain, while for gases and fine particles ($PM_{2.5}$) the effect is small (Sofiev et al. 2009). The shipping emissions increase concentrations of $SO_2$, $PM_{2.5}$ and $PM_{10}$ (Viana et al. 2014). $PM_{2.5}$ and gases remain in the air for a longer time after emission and may be transported over long distances from the source, while $PM_{10}$ have limited spatial distribution and are often deposited downwind of the pollution sources. At the same time, sulphate may be present in both $PM_{2.5}$ and $PM_{10}$ (Chan et al. 1997), while $SO_2$ oxidizes and contributes to the $XSO_4^{2-}$ concentration as well. Moreover, the processes affecting aerosol population cannot be thoroughly investigated without studying the evolution of vertical aerosol profiles (Kupiszewski et al. 2013). However, no particulate matter or vertical aerosol profiles data are available for the current study period, and the time resolution of the filter samples used in this study is too coarse to discuss the aerosol and gas processes in detail.

The WRS test showed that only $SO_2$ measurements in Ny-Ålesund were influenced by temperature inversions in autumn (Table 2). However, the medians for both groups were very low and quite similar. This is due to the generally very low concentration during autumn. No other statistically significant impact was revealed in autumn.

In winter, according to the WRS test, temperature inversions become an important factor increasing the median concentration in the Ny-Ålesund $XSO_4^{2-}$ data set, but not affecting the Zeppelin station (Table 2). Change of wind direction with height and humidity inversions did not affect either of the data sets. This can be explained by low specific humidity variation with height (average standard deviation of specific humidity is 0.07 $g \cdot kg^{-1}$ in the lowest 500 m in winter versus 0.18 $g \cdot kg^{-1}$ in summer), low median QIS, low number of days with directional wind shear and high seasonal average median profile wind speed within the lowest 500 m (5.7 $ms^{-1}$).

In spring, in accordance with the test, temperature inversions affect $SO_2$ and $XSO_4^{2-}$ data sets from both stations. The samples from the group of days with temperature inversions had significantly higher median values of both compounds than from the group without temperature inversions (Table 2). Average air temperature at 850 hPa was −15°C and −12°C in the first and second groups of days, respectively. The orientation of the wind velocity vectors and the location of the isobars in Fig. 4a show that there was a horizontal advection of colder air masses from the east–north-east in the first group of days, with higher concentrations. The air flow from the Arctic Ocean to the north-west of Svalbard prevailed for

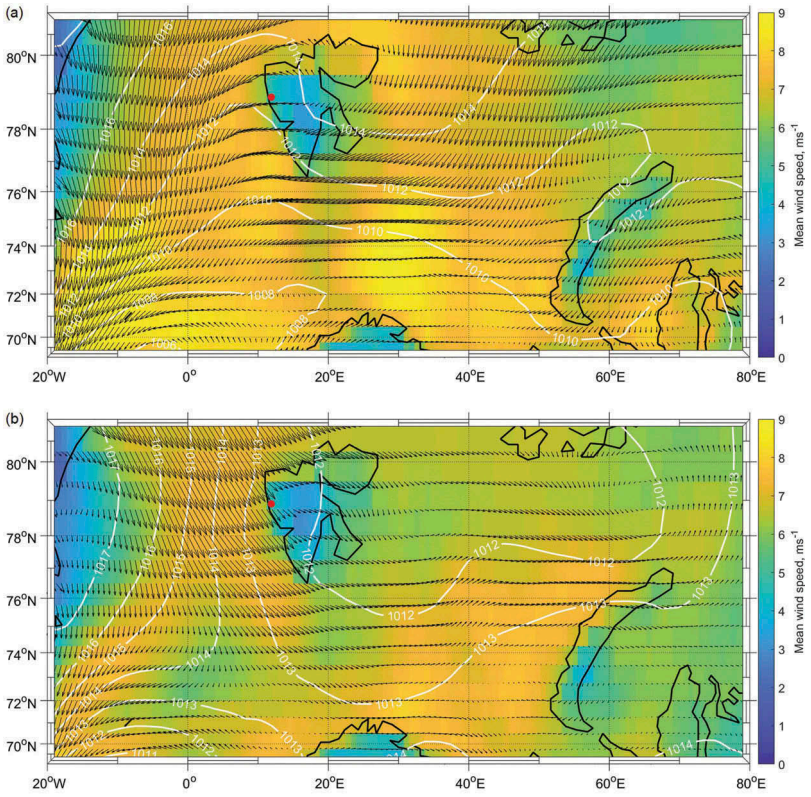


**Figure 4.** Spring mean wind speed in $m \cdot s^{-1}$ (colour scale), wind direction (black arrows with the length relative to the wind speed) and mean sea-level pressure in mbar (white lines) in the Greenland and Barents seas, obtained from surface ERA-Interim data: (a) for days with temperature inversion; (b) for days without temperature inversion.

**Table 3.** Mean spring concentrations in daily filter samples at the Ny-Ålesund and Zeppelin stations, $\mu gS \cdot m^{-3}$. The values in boldface correspond to significant results of the WRS test shown in Table 2.

| Measured compound and station | Seasonal | Change in wind direction | No change in wind direction | Temperature inversion | No Temperature inversion | Strong humidity inversion | Normal or no humidity inversion |
|---|---|---|---|---|---|---|---|
| $SO_2$, Ny-Ålesund | 0.124 | 0.087 | 0.141 | **0.175** | **0.031** | 0.042 | 0.177 |
| $XSO_4^{2-}$, Ny-Ålesund | 0.270 | 0.334 | 0.239 | **0.326** | **0.169** | **0.194** | **0.320** |
| $SO_2$, Zeppelin | 0.146 | 0.133 | 0.152 | **0.200** | **0.058** | 0.082 | 0.185 |
| $XSO_4^{2-}$, Zeppelin | 0.297 | 0.356 | 0.269 | **0.363** | **0.195** | 0.264 | 0.318 |

the second group of days, with lower concentrations (Fig. 4b).

The mean spring concentrations on daily filter samples from the Ny-Ålesund and Zeppelin stations are shown in Table 3. The values for the groups for which the WRS test detected significant difference in medians are indicated. The mean spring $SO_2$ and $XSO_4^{2-}$ values changed dramatically at both the Ny-Ålesund and Zeppelin stations, depending on the presence or absence of strong humidity inversions. Mean spring $SO_2$ values for days with strong humidity inversions were four times and two times lower at the Ny-Ålesund station and at the Zeppelin station, respectively, than mean $SO_2$ values for the days when no strong humidity inversions were observed. Mean spring $XSO_4^{2-}$ values for the first group of days were 65% and 17% lower at the Ny-Ålesund and Zeppelin stations, respectively, than the mean value for the days without strong humidity inversions. However, the magnitude of the reduction of the $SO_2$ and $XSO_4^{2-}$ concentrations suggests that these conditions affect measurements at the Ny-Ålesund station more than at the Zeppelin station. Indeed, according to the test, the median $XSO_4^{2-}$ values in the Ny-Ålesund station data are significantly lower for the group of days with strong humidity inversions, while no difference was found in the data from the Zeppelin station. Strong humidity inversions have the opposite effect on filter measurement results to the one from temperature inversions. This may be explained by the different origin of air masses. Strong humidity inversions were observed when the wind direction at 850 hPa was from the south and west and the average air temperature at that pressure level was −11.3°C, while for the days with no strong humidity inversions it was −15°C. Figure 5a shows that there was a transport of warmer and more humid air from the Atlantic Ocean south of Svalbard during the days with strong humidity inversions. Prevailing weather conditions at 850 hPa for the days with strong humidity inversions correspond very well with the ones described by Vihma et al. (2011), who characterized the effect of

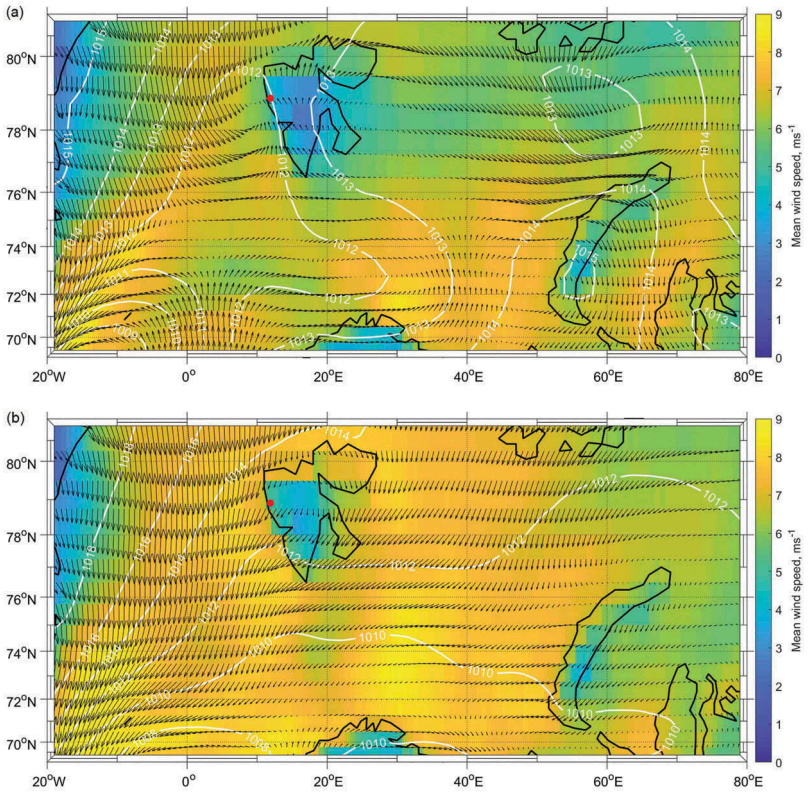


**Figure 5.** Spring mean wind speed in m·s$^{-1}$ (colour scale), wind direction (black arrows with the length relative to the wind speed) and mean sea level pressure in mbar (white lines) in the Greenland and Barents seas, obtained from surface ERA-Interim data: (a) for days with strong humidity inversion; (b) for days without strong humidity inversion.

warm and humid air masses from the marine sector (200–290°) on the formation of humidity inversions. In contrast, the location of isobars in Fig. 5b for the group without strong humidity inversions was similar to the one for the group of days with temperature inversions (Fig. 4a), indicating the transport of colder air masses with higher concentrations of sulphur compounds from the east. Moreover, the temperature at the level of 850 hPa correlates differently with temperature and humidity inversion strengths in spring. There is a statistically significant negative correlation between TIS (r = −0.30, $p$ = 0.004) and the air temperature at 850 hPa, while the correlation with QIS is positive (r = 0.26, $p$ = 0.01). This indicates that strong humidity inversions are caused by the horizontal transport of warmer air masses and not by the radiative cooling of the surface layer. Lower concentration of $XSO_4^{2-}$ in Ny-Ålesund than at the Zeppelin station may be explained by the fact that the air masses are of marine nature, and the deposition velocities above the air–water interface may be slightly higher compared with deposition to dry surfaces because of growth of hygroscopic particles in the humid boundary layer (Seinfeld & Pandis 2006).

A main weakness in this study is the filter-sampling method, which affects the number of values below the detection limit, and consequently the correlation between the data sets. As Aas et al. (2007) stated, the reference European Monitoring and Evaluation Programme method, based on the use of potassium hydroxide-impregnated filters for $SO_2$ detection, is not well suited to monitoring low background concentrations because of its high detection limit. The seasonal percentage of $SO_2$ values below the detection limit ($L_d$) in the data sets at both stations is very high irrespective of the season, while the percentage is lower in the $XSO_4^{2-}$ data set (Table 4). The rate of missing data in the Ny-Ålesund station data set is higher than in the Zeppelin data set in all seasons.

Another uncertainty is related to the fact that only one radiosounding per filter measurement was utilized in the data analysis. This gives the information about the atmospheric stratification and vertical wind profile at the time of launching (generally at 12 UTC), but does not describe the profile evolution during the day. However, as one can see in Fig. 6 the lowest correlation between daily averaged temperature measurements at the Ny-Ålesund and Zeppelin stations were observed during summer. This points to a higher rate of non-linearity in interrelation between temperatures at the Ny-Ålesund and Zeppelin stations and, consequently, reflects complexity of the processes in the ABL. In general, the atmospheric lapse rate is formed under the combination of convection and radiation processes (Wallace & Hobbs 2006); however, we have seen from the high variation of specific humidity and frequently observed directional wind shear and clouds in

**Table 4.** Characteristics of the filter data sets.

| Station | Characteristic | Compound | Season | | | |
|---|---|---|---|---|---|---|
| | | | summer (n = 154) | autumn (n = 182) | winter (n = 121) | spring (n = 92) |
| Ny-Ålesund | Rate of values below $L_d$ (%) | $SO_2$ | 45 | 42 | 45 | 43 |
| | | $XSO_4^{2-}$ | 31 | 41 | 35 | 10 |
| Zeppelin | Rate of values below $L_d$ (%) | $SO_2$ | 45 | 54 | 40 | 49 |
| | | $XSO_4^{2-}$ | 21 | 32 | 17 | 14 |
| Ny-Ålesund | Rate of missing data (%) | $SO_2$ and $XSO_4^{2-}$ | 21 | 32 | 17 | 14 |
| Zeppelin | Rate of missing data (%) | $SO_2$ and $XSO_4^{2-}$ | 1 | 14 | 6 | 3 |

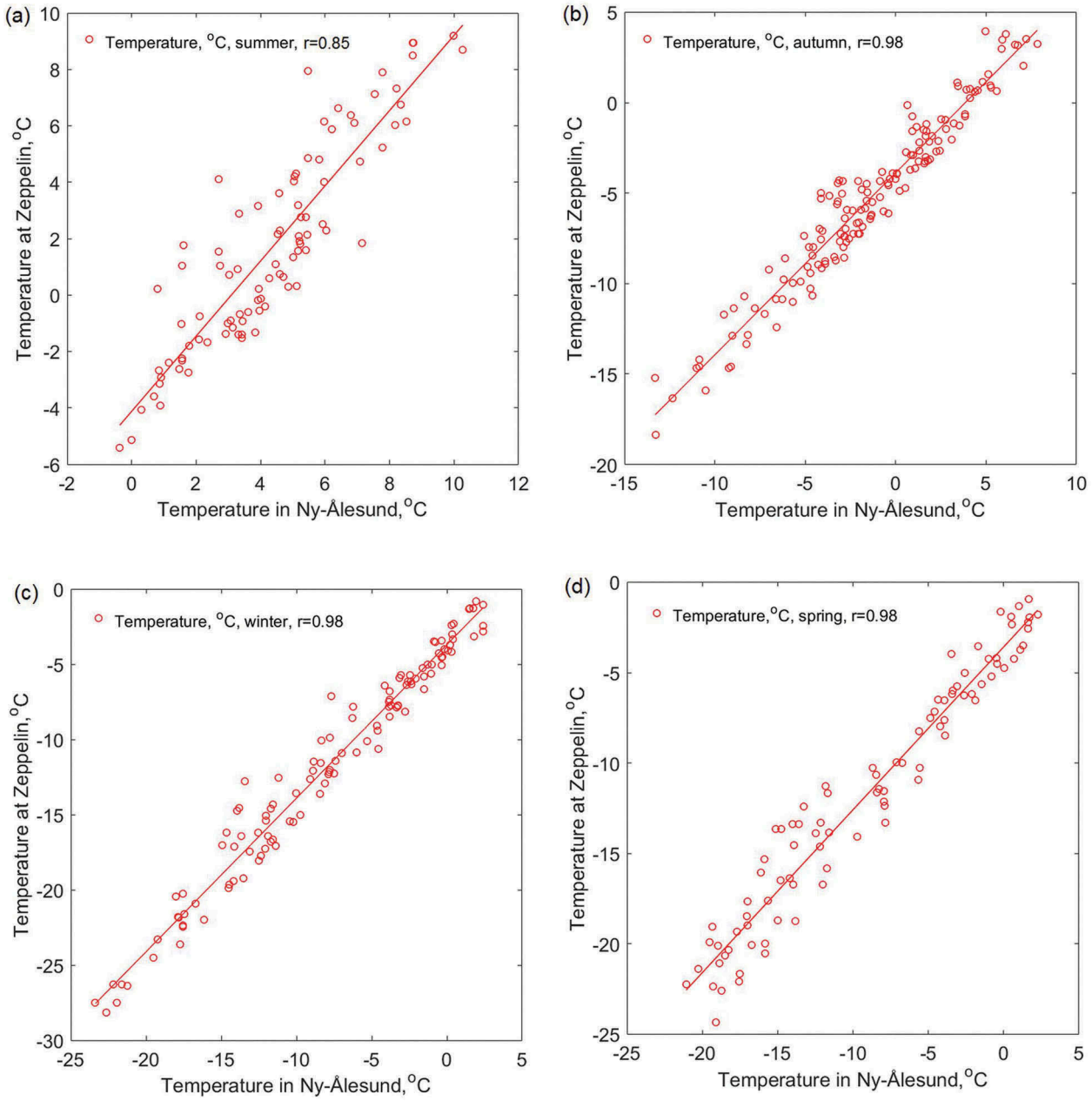


**Figure 6.** Seasonal plots of daily averaged air temperature measured at the Ny-Ålesund and Zeppelin stations: (a) summer; (b) autumn; (c) winter; (d) spring.

summer profiles that there have been air flows with quite different characteristics in the lowest 500 m. Interaction of these flows and radiation processes within the clouds may influence the correlation of daily temperature measured at the two stations, so the characteristics of the profiles are not only instantaneous features of the ABL, but are important for the lapse rate formation throughout the day in summer.

The findings presented in this paper may be relevant for planning future fieldwork campaigns and comparing modelling results with measurements

done in a region with complex topography, such as the Ny-Ålesund area. Knowledge of the micrometeorological conditions of the study area is crucial if one wants to eliminate local effects by choosing the right location for the station or to contrast results from already existing stations situated close to each other. For comparison of historical data with modelling results, one needs to investigate local meteorological factors that may affect measurement results and choose the model resolution that correctly represents these factors. A study by Mölders et al. (2011) has shown that the combined chemical and meteorological model WRF-Chem with a resolution of 4 km tends to underestimate the inversion strengths, presents biases of temperature and wind speed and determines incorrect wind direction at low wind speeds. Furthermore, it has been stated that errors in modelled temperatures may lead to erroneous modelled concentrations of aerosols (Mölders et al. 2011). As there is a trade-off between the resolution and the computational time in the model, it is useful to know if the measurement results from the station are subject to a significant impact of local pollution and/or micrometeorological factors. This can be checked using the statistical methods applied in this article.

## Conclusion

The correlation of daily sulphur dioxide ($SO_2$) and non-sea salt sulphate ($XSO_4^{2-}$) data sets from the Ny-Ålesund station and the Zeppelin mountain station has a large seasonal variation. No significant correlation between the $SO_2$ data sets has been observed in the summer data, while a very strong correlation is present in the winter data, with autumn and spring showing intermediate to moderate values of Pearson correlation coefficient. Although the correlation between the $XSO_4^{2-}$ data sets is significant for all times of year, it is much lower for the summer data compared to other seasons.

The first hypothesis, stating the significance of the effect of temperature and humidity inversions on the correspondence between the data sets from the Ny-Ålesund and Zeppelin stations, is supported. In winter, concentration of $XSO_4^{2-}$ at the Ny-Ålesund station is significantly higher in the days with temperature inversions, while in spring this effect is seen in data sets from both stations. Local meteorological conditions on the days with strong humidity inversions reduce and increase the sulphate concentration at the Ny-Ålesund station in spring and summer, respectively.

The second hypothesis has been rejected, since the directional shear appeared to be an insignificant factor, while the third hypotheses about the influence of wind speed shear on the correspondence of measurements is relevant only for the summer season. In the absence of the wind speed shear, local pollution affects Ny-Ålesund sulphur dioxide and sulphate data set more strongly.

The fourth hypothesis, stating the effect of local summertime emissions from ship traffic on the correspondence between the data sets from the two stations, is partially supported. Local summertime ship traffic has a strong impact on the $SO_2$ Ny-Ålesund data set, while there is no statistically significant effect on the Zeppelin data set. The $XSO_4^{2-}$ data sets at both stations are impacted by pollution from ships, but the influence on the Ny-Ålesund data set is more pronounced on days with strong humidity inversions.

The findings presented here are highly important for planning joint monitoring campaigns and the exchange and comparison of measurement results in Ny-Ålesund. Both anthropogenic factors, such as local pollution in summer, and natural ones, namely local circulation patterns and variation of temperature and humidity in the ABL, affect the correspondence between the data sets collected at the locations within the range of 2 km. Furthermore, the nature of the sulphur species studied here determines which factors affected the concentration of the measured compounds at the two stations.

The environmental phenomena described here still represent a challenge for modellers and need to be taken into account when comparing modelling results with in situ measurements taken at different heights in an area with complex topography.

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## Disclosure statement

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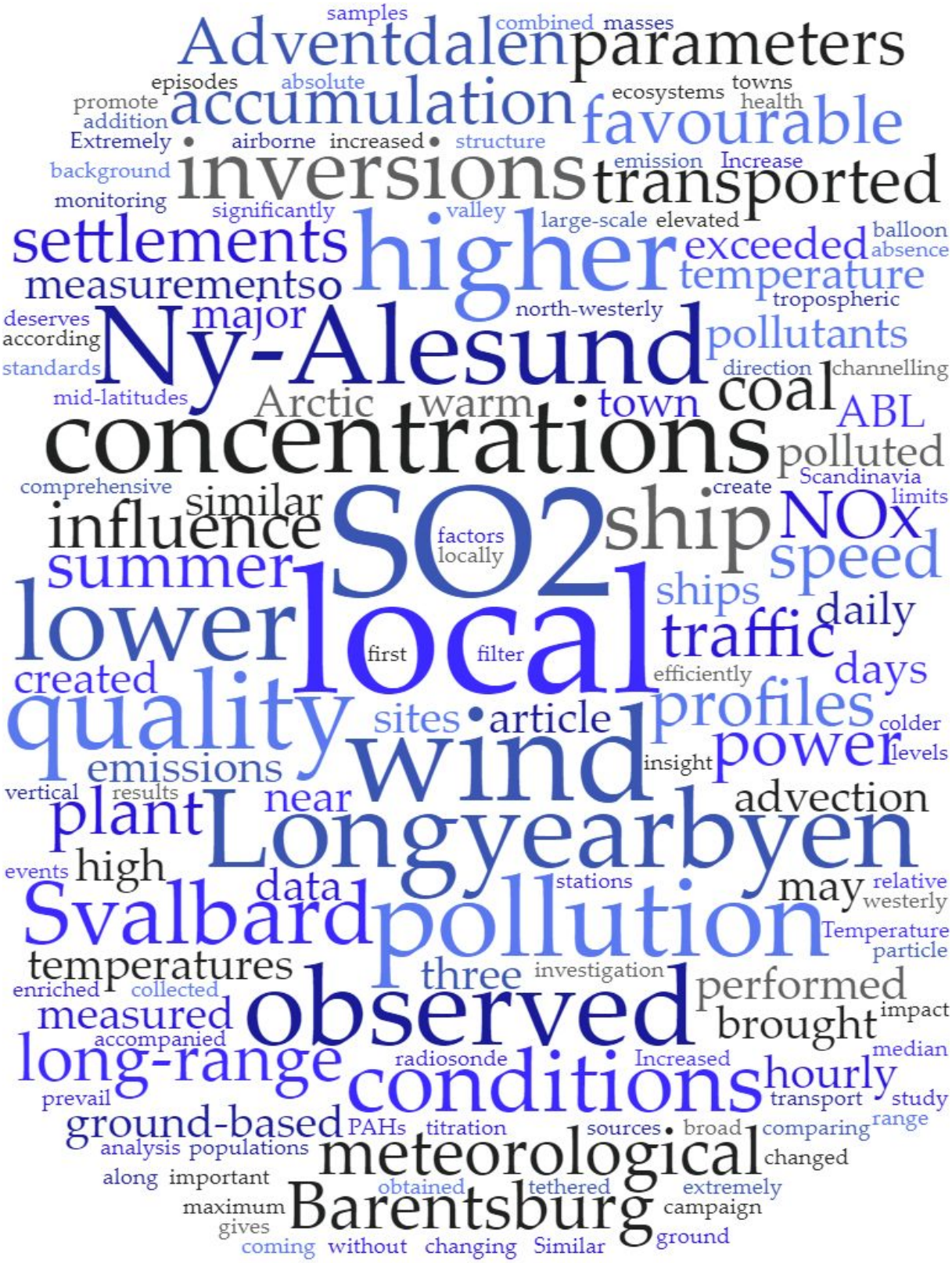
**Paper III**


temperature
Ny-Ålesund
meteorological
revealed
correlated
inversions
settlements
measurements
significant
magnitude
temporary
Svalbard
depend
prevailing
large-scale
analysed
contrast
direction
fresh
reveal
elevated
variable
identify
Adventdalen
correlation
source
harbour
Measurement
stations-network
common
daytime
results
summer
power
increasing
highly
three
stations
ships
nitrogen
time
weakly
cases
local
ozone
study
data
likely
wind
pollutant
pollution
calmer
measurement
stationary
determines
along
percent
sensors
new
channelled
tropospheric
near
projects
titration
station
mean
effect
rarely
quality
daily
field
ship
region
atmospheric
plant
days
situation
various
case
route
air
topographical
higher
hence
occur
first
diurnal
season
reduce
way
ratio
lower
traffic
located
presence
accumulation
features
often
low
circulation
masses
highest
since
diesel
pattern
special
correlate
value
investigate
thus
colder
needs
sondes
sites
observatory
difference
expected
independent
values
Local
ERA5
recommended
high
speed
impact
show
reanalysis
sources
measured
nighttime
fieldwork
different
dependent
soundings
campaign
close
weekly
compared
long-range
investigation
research
radiosonde
ground-based
evaluate
NO2/NOx
oxides
concentrations
combined
identified
contributing
controlled
valley
Zeppelin
conditions
therefore
hourly
snowmobile
depleted
performed
enriched
dominant
transport
influence
Longyearbyen
promote
background
change
intensity
downwind
synoptic
observed
strongly

**Paper IV**


samples
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masses
Adventdalen
parameters
episodes
absolute
accumulation
ecosystems
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promote
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favourable
Extremely
airborne
increased
structure
background
emission
Increase
inversions
transported
monitoring
significantly
valley
large-scale
elevated
balloon
settlements
higher
exceeded
absence
temperature
measurementso
tropospheric
deserves
according
major
north-westerly
Ny-Alesund
pollutants
standards
direction
channelling
mid-latitudes
Arctic
warm
town
coal
ABL
concentrations
polluted
Scandinavia
comprehensive
create
limits
similar
SO2
ship
NOx
influence
factors
locally
speed
summer
ships
local
daily
lower
traffic
first
filter
efficiently
days
created
quality
sites
article
profiles
colder
power
levels
insight
emissions
wind
vertical
results
near
advection
plant
Longyearbyen
events
high
data
stations
may
relative
Svalbard
pollution
westerly
Temperature
particle
temperatures
three
investigation
performed
enriched
collected
measured
brought
impact
accompanied
long-range
radiosonde
Increased
median
conditions
hourly
prevail
transport
study
ground-based
PAHs
titration
sources
broad
comparing
range
analysis
populations
meteorological
changed
along
important
obtained
tethered
extremely
maximum
Barentsburg
campaign
gives
coming
without
changing
Similar
ground