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# Particulate and gaseous emissions of power generation at Svalbard (AtmoPart)

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Stephan Weinbruch<sup>1</sup>, Tatiana Drotikova<sup>2,3</sup>, Nathalie Benker<sup>1</sup>, Roland Kallenborn<sup>2,3</sup>

<sup>1</sup> Technical University of Darmstadt, Institute of Applied Geosciences, Darmstadt, Germany

<sup>2</sup> University Centre in Svalbard (UNIS), Longyearbyen, Svalbard

<sup>3</sup> Norwegian University of Life Sciences (UMB), Department of Chemistry, Biotechnology and Food Sciences (IKBM), Ås Norway

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## Abbreviation list

AMAP	Arctic Monitoring and Assessment Programme
C	Carbon
Cu	Copper
ESEM	Environmental Scanning Electron Microscopy
EDAX	Energy dispersive X-ray detector
EDX	Energy dispersive X-ray detector
Fe	Iron
Hg	Mercury
Na	Sodium
Ni	Nickel
O	Oxygen
P	Phosphorous
Pb	Lead
PAH	Polycyclic Aromatic Hydrocarbons
S	Sulphur
SEM	Scanning Electron Microscopy
SO <sub>4</sub> <sup>2-</sup>	Sulphate
SMV	Svalbard Miljøvernfond
TEM	Transmission Electron Microscopy
Zn	Zink

## Objectives

The AtmoPart initiative focused on characterisation and evaluation of the environmental impact by direct emissions of anthropogenic pollution originated from combustion of fossil fuel products (coal and gasoline products) on Svalbard.

The here reported results are meant as recommendation for suitable counter measures and remediation activities based upon science based surveillance of emissions from fossil fuel driven power plant installations on Svalbard (Barentsburg, Longyearbyen and Svea) as tools for regulative purposes.

## Introduction

Reliable electric power supply is an essential prerequisite for the very existence of isolated communities in the Arctic. In addition, electric power support is an important foundation of industrial activities under harsh Arctic conditions. However, a significant negative aspect of local power productions under these conditions is the expected environmental impact from direct pollution emissions into the Arctic atmosphere with potential impact both on the local Arctic ecosystems and the local populations..

The Svalbard electric power plants are driven on fossil fuels (Diesel or coal). The Longyearbyen, Svea and Barentsburg power plants are both serving municipal as well as industrial installations. Emissions from these power generation units are today confirmed as a major local pollution source on Svalbard (Aamaas et al., 2011, Jartun et al., 2009, Hicks and Isaksson, 2006, Rose et al., 2004, Rose, 1995). These emissions include a variety of different particulate (e.g. soot, fly ashes, lead) as well as gaseous species (e.g., trace metals, organic pollutants) which may have adverse effects on human health as well as on Arctic ecosystems in general (AMAP, 2008). For example, Schütze (Schütze, 2013) showed that approximately 20 – 40 % of particles with diameters  $\leq 0.5 \mu\text{m}$  emitted from coal burning in Longyearbyen contain soot, a component classified as carcinogenic in humans. Most of the soot agglomerates contain lead inclusions, another component of toxicological relevance. The large particle fraction (diameter  $> 0.5 \mu\text{m}$ ) emitted from the coal power plant in Longyearbyen predominantly (almost 100 %) consisted of silicate fly ashes. In Barentsburg, a similar picture emerges (Schütze, 2013): coal burning leads to emission of a large fraction of soot (about 30 %) for small particles (diameter  $\leq 0.5 \mu\text{m}$ ), and to silicate fly ashes (almost 100 %) for larger particles.

Volatile and particle associated organic pollutants from coal fired electric power plants are usually emitted at relative high temperatures directly into the receiving air. Along the ambient temperature gradient the heavier compounds (e.g. 5-8 ring PAHs) are deposited directly onto the ground or further transported particle associated. Several marker compounds are known directly associated with emissions from fossil fuel driven or coal fired power plants. Therefore we performed the AtmoPart initiative, funded by the Svalbard Environmental protection Fund (SMV) as a first study on the emission characterisation from relevant Svalbard power plants as a direct contribution to a general impact assessment of pollution profiles from Svalbard power plants.

## Material and methods

### Sampling

Particles were collected close to the emission sources with a two-stage micro inertial cascade impactor (50 % cut-off diameter: fine fraction 0.1 – 0.5  $\mu\text{m}$ , coarse fraction 0.5 – 10  $\mu\text{m}$ ) using either polished boron or TEM grids as substrate. The sampling locations as well as sampling dates are summarized in Table 1. Sampling time varied between 0.5 seconds and 5 minutes using a flow rate of 0.5 l/min.

**Table 1:** Sampling location and date

Location	Sampling site	Sampling date
Longyearbyen	Coal fired power plant	26.11.2014
Barentsburg	Coal fired power plant	28.11.2014
Svea	Diesel fired power plant	10.03.2014
Pyramiden	Diesel fired power plant	03. – 05.09.2014

### Electron microscopy

The particle samples were studied by scanning (SEM) and transmission electron microscopy (TEM). SEM was carried out with a field emission gun environmental scanning electron microscope (FEI ESEM Quanta 200 FEG, Eindhoven, The Netherlands) operated at 20 kV acceleration voltage. The particles were studied without coating in the low vacuum mode of the instrument ( $\approx 1.2$  mbar sample chamber pressure). The particle size (equivalent projected

area diameter) was obtained from secondary and backscattered electron images. The spatial resolution obtained with secondary electrons was approximately 10 nm, and 15 – 20 nm for backscattered electrons (both on atmospheric particles). The chemical composition (elements with  $Z \geq 5$ ) of the particles was measured with an energy-dispersive X-ray detector (EDAX, Tilburg, The Netherlands).

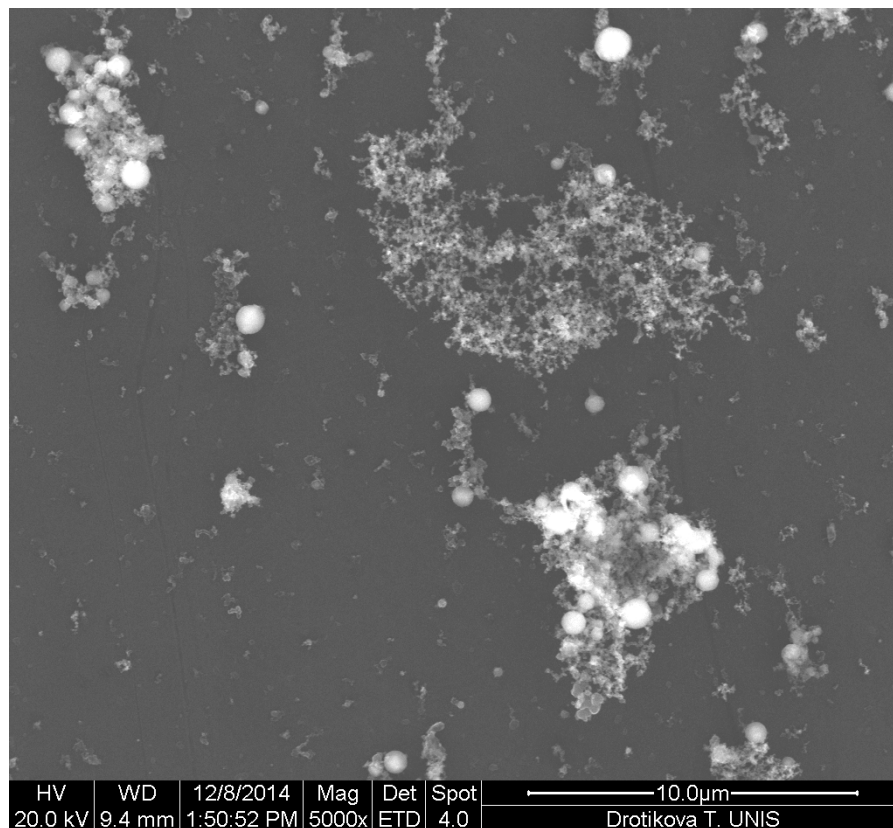
TEM was performed with two instruments: (a) FEI CM20 ST (FEI, Eindhoven, The Netherlands), and (b) Jeol 2100F (Jeol, Tokyo, Japan). Both instruments are operated at 200 kV acceleration voltage. The spatial resolution obtained was approximately 0.23 nm. Both microscopes are equipped with an energy-dispersive X-ray detector each (Oxford Instruments, Abingdon, UK). The particles were also studied without coating.

## Results

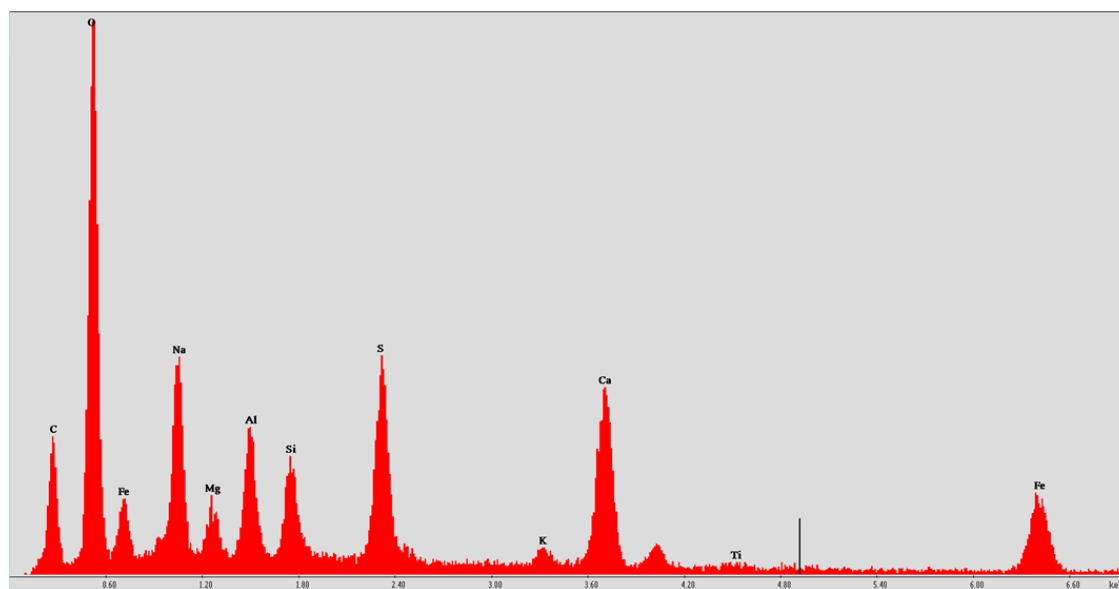
The particulate emissions of the coal power plant in Longyearbyen mainly consist of silicate fly ashes, soot and secondary aerosol. Due to the high source strength, most samples are overloaded preventing a precise estimation of the relative number abundance of the individual particle groups.

A typical example of a Longyearbyen sample is shown in Figure 1. Fly ashes are recognized by their spherical morphology. They have a silicate composition (Figure 2). Soot is present as chain-like or more compacted agglomerates (Figure 1) of primary particles (diameter 30 – 50 nanometer). Soot and fly ash particles are often internally mixed with secondary aerosol (mainly calcium sulphate). According to X-ray diffraction analysis of bulk samples collected at the stack of the power plant, gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) is the only sulphate phase observed.

**Figure 1:** Secondary electron image of particles emitted from the coal power plant in Longyearbyen (see text for details).

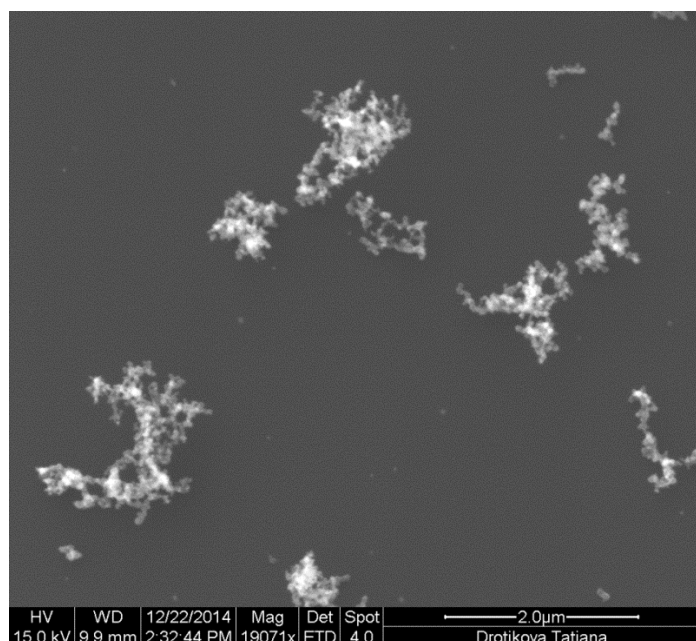


**Figure 2:** Typical X-ray spectrum of a silicate fly ash particle (from Longyearbyen) internally mixed with secondary aerosol, most likely calcium sulphate (gypsum).



Particles collected at the Diesel power station in **Pyramiden** predominantly consist of soot agglomerates (Figure 3). In addition, a few (less than 2 %) fly ash particles were encountered.

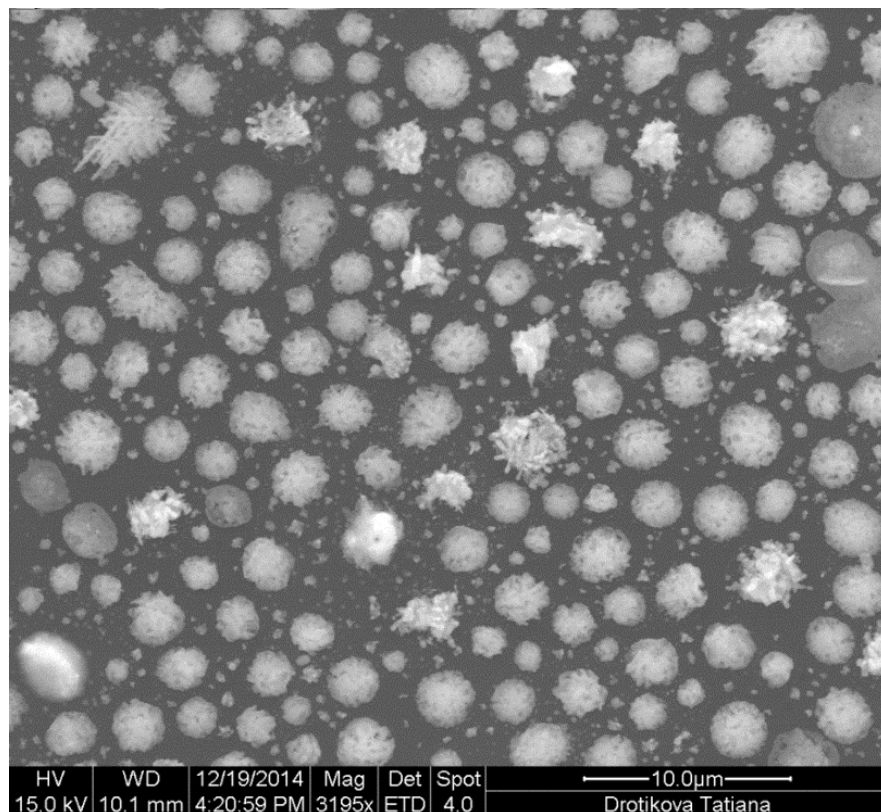
**Figure 3:** Secondary electron image of soot emitted from the Diesel power station in Pyramiden.



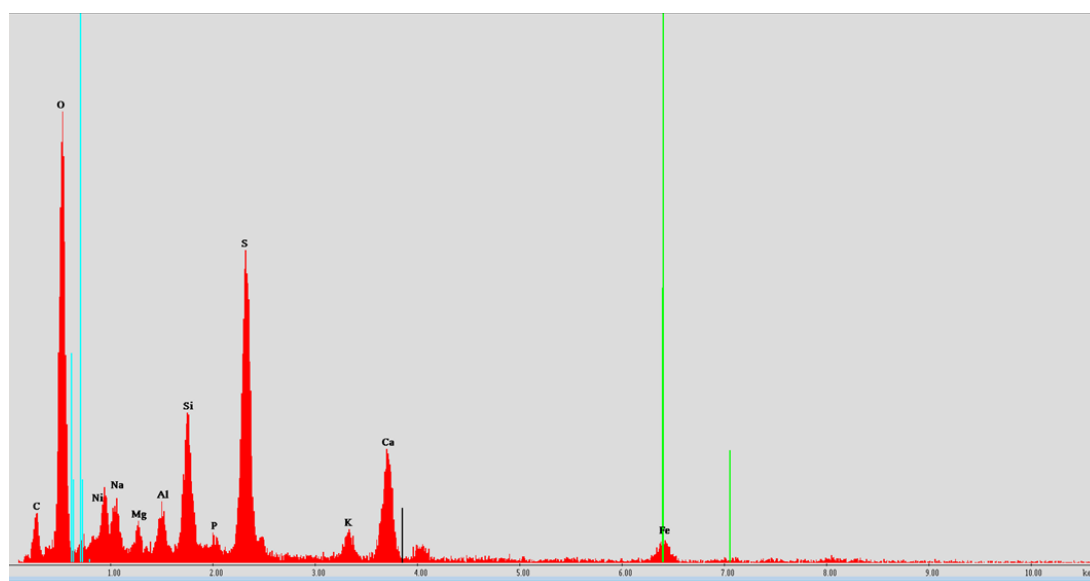
Samples from the plume of the coal power plant in **Barentsburg** are dominated by secondary aerosol particles (Figure 4), which are assumed to have condensed from the gaseous emissions of the power plant. They consist of sulfates (most likely gypsum) and may contain heterogeneous inclusions of silicates. A typical X-ray spectrum of the secondary aerosol particles encountered at Barentsburg is shown in Figure 5. The secondary particles often contain small (100nm diameter) inclusions of lead-rich particles (Figures 6 and 7). The abundance of external soot and silicate fly ashes is rather low (< 5 %). Some of the fly ash particle/secondary aerosol mixtures show very high phosphorous concentrations (Figure 8).

A second plume from the coal power plant sampled in Barentsburg is dominated by fly ash particles. In addition, soot (often mixed with secondary aerosol) occurs at minor abundance.

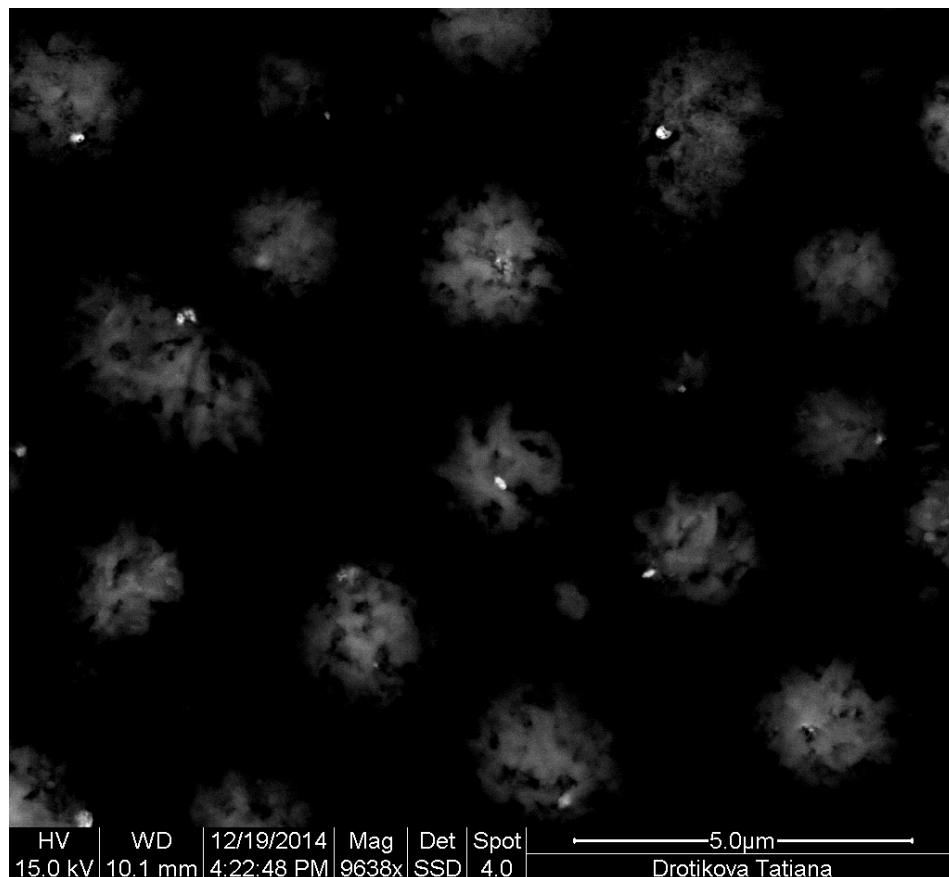
**Figure 4:** Secondary electron image of secondary aerosol particles found in the plume of the coal power plant in Barentsburg.



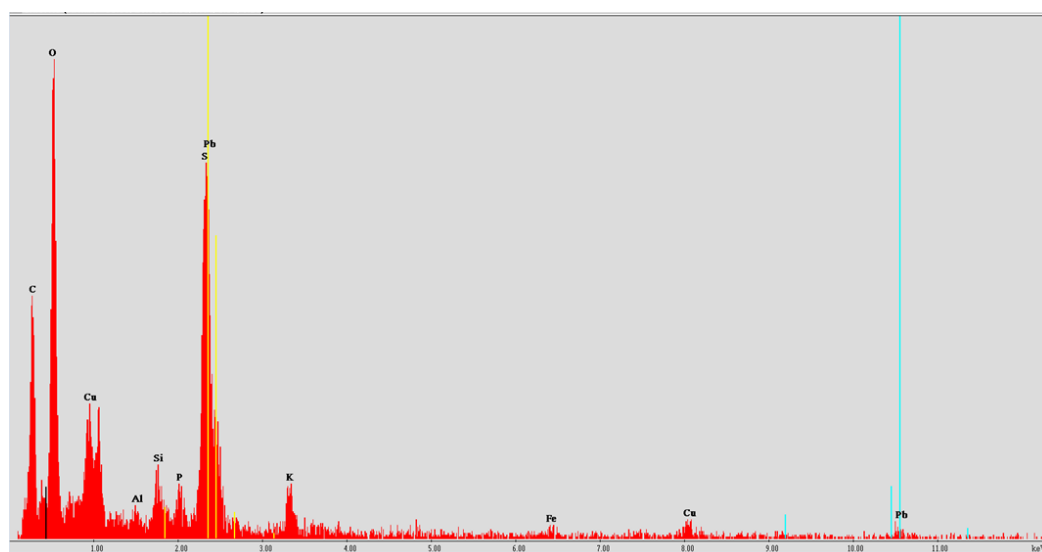
**Figure 5:** X-ray spectrum from secondary aerosol particles found in the plume of the coal power station at Barentsburg. Please note: The Ni peak stems from the substrate.



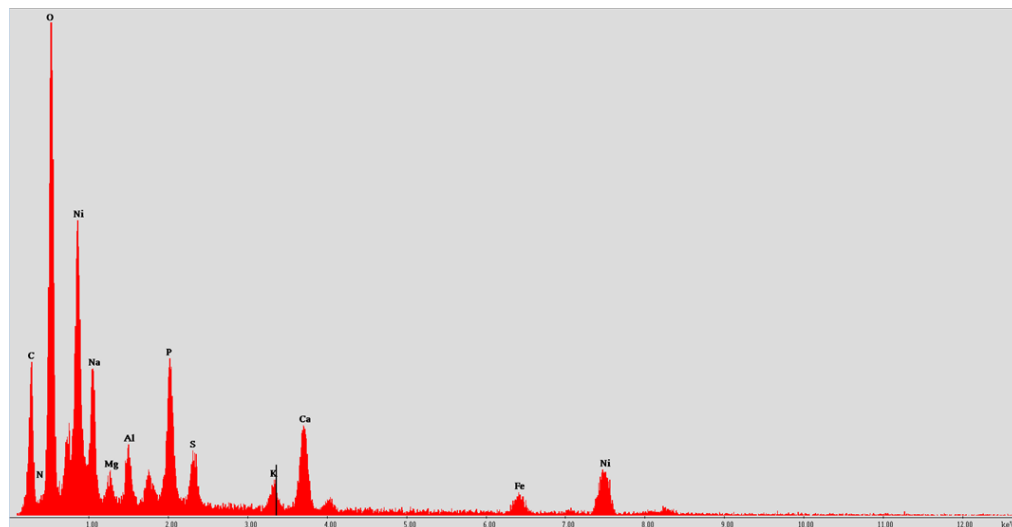
**Figure 6:** Secondary electron image of secondary aerosol particles in Barentsburg containing small heterogeneous Pb-rich inclusions (bright inclusions).



**Figure 7:** X-ray spectrum of Pb-rich inclusions in secondary aerosol particles (Barentsburg).

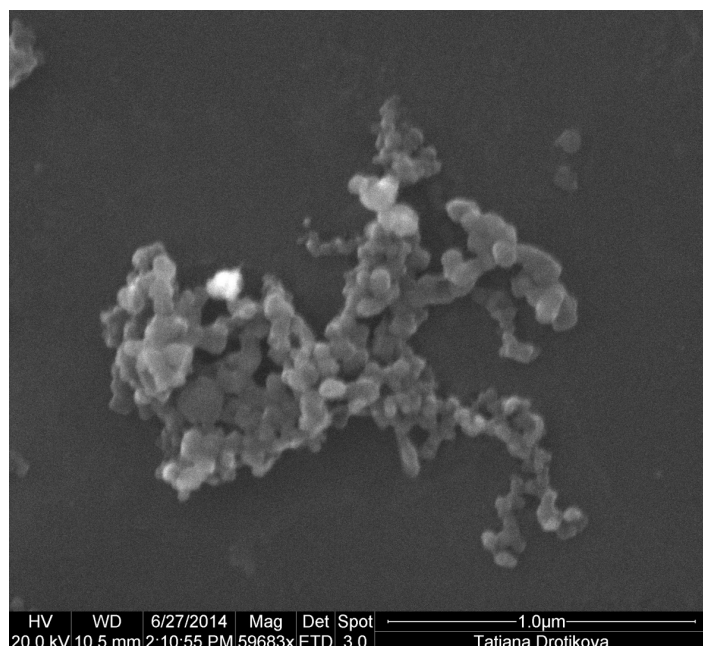


**Figure 8:** X-ray spectrum of fly ash/secondary aerosol mixtures having very high phosphorous concentrations (Barentsburg). Please note: The Ni peaks stem from the substrate.

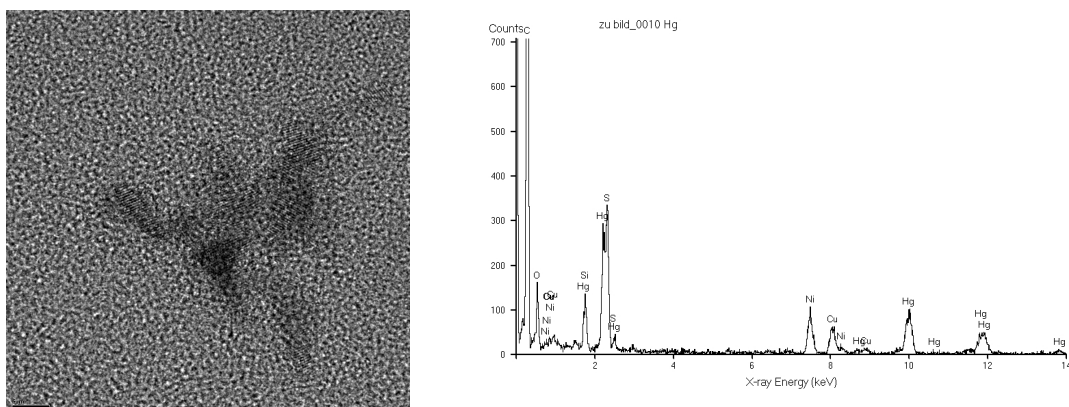


Samples from the Diesel station at **Svea** are dominated by soot often covered by secondary aerosol (Figure 9). Fly ashes (silicates and iron-rich particles) occur as minor component. Occasionally Pb-rich particles are found as external particles and internally mixed with soot and with secondary aerosol. In Svea, a large number of small ( $\approx 10$  nm) Hg-rich particles was found, which are stable under the electron beam.

**Figure 9:** Secondary electron image of soot from Svea (the bright particle is Pb-rich).



**Figure 10:** High-resolution TEM image and X-ray spectrum of a Hg-rich particle encountered in the plume of the Diesel power at Svea. Please note: The Ni peaks stem from the substrate.



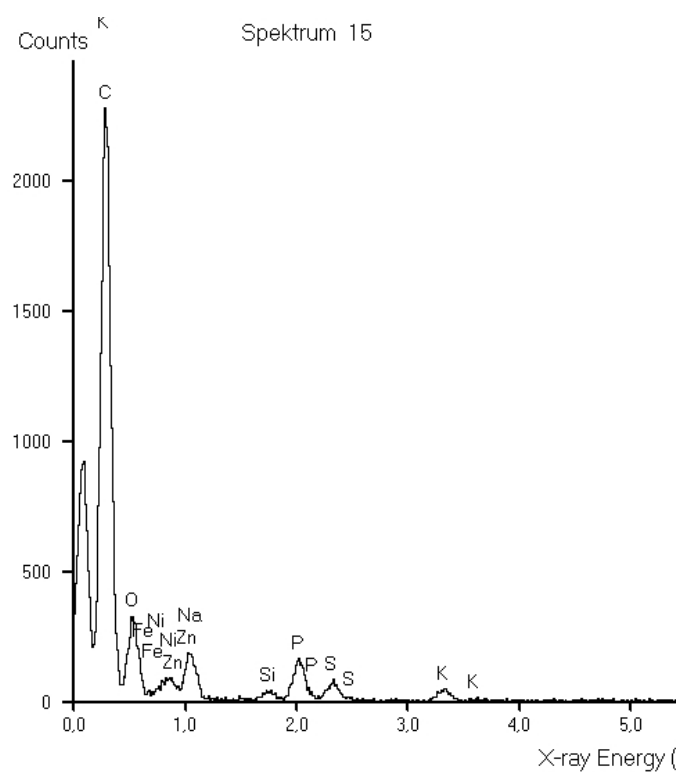
The most important findings of scanning electron microscopy are summarized in Table 2. Fly ashes, soot and secondary aerosol are the major particle groups emitted from coal burning in Longyearbyen and Barentsburg, soot the major particle group emitted from Diesel aggregates in Svea and Pyramiden. Other toxicologically relevant particle groups include Pb-rich particles from coal burning in Barentsburg and Diesel emissions in Svea as well as Hg-rich particles from Diesel emissions in Svea.

**Table 2:** Summary of scanning electron microscopy results

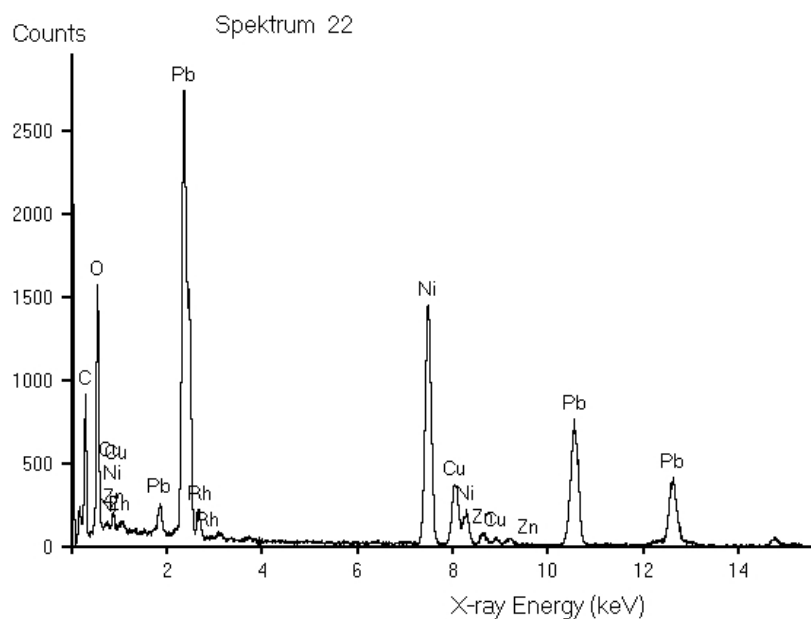
Location	major particle groups ( $\geq 20$ % relative number abundance)	minor particle groups (1 – 10 % relative number abundance)	additional observations
Longyearbyen (coal burning)	fly ashes soot secondary aerosol		
Barentsburg (coal burning Plume 1)	secondary aerosol	fly ashes soot	frequent Pb-rich inclusions, high P contents
Barentsburg (coal burning Plume 2)	fly ashes	soot secondary aerosol	
Svea (Diesel station)	soot	fly ashes	frequent Pb-rich particles/inclusions Hg-rich particles
Pyramiden (Diesel station)	soot	fly ashes	

Soot agglomerates from the two coal burning power stations were studied in more detail by high-resolution TEM. There seem to be pronounced differences in the chemical composition of individual soot agglomerates from Longyearbyen and Barentsburg. Approximately 80 % of the soot agglomerates (26 out of 32) from Barentsburg show phosphorous as minor constituent (Figure 11). In contrast P-containing soot was only found in 1 out of 70 individual soot agglomerates from Longyearbyen. In Longyearbyen a higher abundance of Pb inclusions within soot agglomerates (Figure 12) was found compared to Barentsburg.

**Figure 11:** X-ray spectrum of an soot agglomerate from Barentsburg with P as minor element.



**Figure 12:** X-ray spectrum of a soot agglomerate from Longyearbyen containing Pb-rich inclusions.



## Discussion

Particulate emissions from coal burning found a wide attention in the literature, as they may contribute significantly to urban air pollution (He et al., 2006, Huang et al., 2010, Thurston et al., 2011, Frey et al., 2014). Compared to bulk chemical analysis, studies on individual particles emitted from coal burning are rare and focus mainly on fly ashes (e.g., Ramsden and Shibaoka, 1967; Gieré et al. 2003; Chen et al., 2004, 2005).

Fly ashes are generally the most abundant component among particulate emissions from coal burning. They may be even used as fingerprint for a coal combustion component after long range transport (Ramsden and Shibaoka, 1982, Mamane and Dzubay, 1986, Mamane et al., 1986, Weinbruch et al., 2010). Also in both coal power plants investigated in this study, fly ash particles are the dominating primary particle group.

The high abundance of soot particles in coal burning emissions is important to note, as this component has severe impact on the atmospheric radiation system (Lary et al., 1997, Pueschel, 1996, Pueschel and Kinne, 1995, Horvath, 1993), as well as on human health (Wierzbicka et al., 2014, Peng et al., 2012, Rissler et al., 2012, Liu et al., 2010, Comstock, 1998). The Diesel power station in Svea also emits large quantities of soot particles. However, it was not intended in our study to determine the source strength. Instead, we focus on the

presence of minor constituents with potential toxic effects for humans as well as potential adverse effects for the ecosystem. Pb-rich particles were observed for coal burning in Longyearbyen and Barentsburg, Hg-rich particles for the Diesel power station in Svea. From bulk chemical trace element measurements it is well known that both elements are emitted in fossil fuel burning (Fabianska et al., 2014, Tan et al., 2013, Fonseca et al., 2013, Rose et al., 2012, Kochubovski, 2010, Ahmad et al., 2009, Demirbas, 2005, Asokan et al., 2005, Premuzic et al., 1994). However, emission of Hg particles from Diesel stations was – to the best of our knowledge – not reported before. Mercury (Hg) associated with individual carbonaceous particles was observed in coal fires from Eastern Kentucky (Silva et al., 2011). In these measurements, however, Hg occurred encapsulated within carbon nanotubes. In contrast, individual Hg-rich particles were observed in our study.

The different chemical composition of individual soot agglomerates from coal burning in Longyearbyen and Barentsburg determined by high-resolution TEM offers the possibility to establish a fingerprint for source apportionment. During spring, autumn and winter, chain-like as well as compact soot agglomerates were observed at the Arctic background station Zeppelin Mountain in Ny Ålesund (Weinbruch et al., 2012). As the abundance of soot agglomerates was not associated with distinct air mass back-trajectories clusters, it was suspected by these authors that local sources on Svalbard may be important. However, coal burning in Barentsburg can be certainly ruled out as source, due the lack of detectable P concentrations within the soot agglomerates. Coal burning in Longyearbyen may still be a source of soot agglomerates at the Zeppelin station, as no definite elemental finger print for soot from Longyearbyen was found. The higher abundance of Pb inclusions within soot observed in Longyearbyen does not provide an unambiguous answer as such inclusions were also observed at the Zeppelin station, albeit at significantly lower abundance.

## **Perspectives and conclusions**

The characteristic composition of direct emissions from fossil fuel driven power plants in Barentsburg, Svea, Longyearbyen and Pyramiden (Svalbard, Norway) were comprehensively investigated. For this study, a variety of electron microscopic as well as spectroscopic methods were applied for a complete characterisation of the emission samples. Both, transmission electron microscopy (TEM) as well as environmental scanning electron microscopy (ESEM) equipped with energy-dispersive X-ray detectors (EDX) were used for

elucidation of element composition. The sampling of direct emissions was performed in the period between March (Svea) to September 2014 (Pyramiden).

The ESEM characterised particle emissions from the Longyearbyen power plant consist mainly of silicate fly ashes, soot and a variety of secondary aerosols. The emissions from the Pyramiden diesel driven power generator consists mainly of soot agglomerates. The Barentsburg emissions are dominated by fly ashes or secondary aerosol particles. High phosphorous levels (P) were found in Barentsburg fly ash and soot emissions. The particulate emission from the Svea diesel driven power plant is dominated by soot. In Svea, occasionally Pb enriched particles were found. A large number of nano-size particles were found to be enriched in mercury (Hg).

Particle emissions from the coal driven power plants (Longyearbyen and Barentsburg) were also characterised with high resolution TEM. Marked differences between the Barentsburg and Longyearbyen emissions were found. Around 80% of the soot agglomerates from Barentsburg showed phosphorous inclusions (as minor constituents). In Longyearbyen a high abundance of Pb inclusions were associated with soot agglomerates (compared to the Barentsburg samples).

Fly ashes are usually the most abundant particulate constituents from coal burning processes. They are therefore often considered as fingerprints for coal based combustion for the elucidation of atmospheric long-range transport. However, the high abundance of soot particles in coal burning emissions is important to note, as this component has obviously considerable impact on atmospheric radiation. The large diesel driven power station in Svea also emits large quantities of soot particles with considerable Hg-rich particles. Similar particulate mercury emissions as shown for the Svea power station were never reported before. However, earlier bulk chemical studies showed that Hg emission may originate from fossil fuel burning.

The here presented characteristic particle emission profiles allow a first estimation of potential local sources as contribution to the source apportionment of atmospheric long-range transport processes and events into the region. In background measurements from the Zeppelins atmospheric research station in Ny-Ålesund soot agglomerates were identified (2011) with no distinct association to air mass trajectory clusters. Thus, local sources were considered as potential source. Since no elevated phosphorous levels in the soot agglomerates are identified in the Zeppelin samples, Barentsburg can, thus, be ruled out as potential source.

## Recommendations and future priorities

The presented results clearly confirm the scientific strength of the here chosen methods. Single particle characterisation as performed here will give valuable science based information on characteristic fingerprints and local emission profiles in appropriately collected atmospheric samples. In combination with bulk measurements routinely performed during long-term monitoring activities (as already done on Svalbard for many years), these methods will provide useful additional information for a comprehensive source apportionment and the contribution of local contamination on the background patterns identified during the continuous national atmospheric monitoring in Svalbard. Similar investigations should be considered as valuable additional inform for risk evaluation and impact assessment of local emissions for the health and well being of the local populations as well as the local Arctic environment.

## Norsk Sammendrag

Partikulært utslipp fra kraftstasjonene i Longyearbyen, Barentsburg, Svea og Pyramiden ble omfattende undersøkt ved hjelp av elektronmikroskopiske og spektroskopiske metoder. For denne undersøkelsen ble transmisjons elektronmikroskop (TEM) og scanning elektronmikroskop (ESEM) brukt i kombinasjon med energidisperse røntgen detektorer (EDX). Prøvetaking for denne undersøkelsen ble gjennomført i perioden fra mars 2014 (Svea) til September 2014 (Pyramiden). ESEM analyser viser at utslipp fra Longyearbyen kraftstasjon inneholder mest silikat baserte flyveaske "fly ashes", sot og en del sekundære aerosoler. Emisjoner fra dieselaggregatet i Pyramiden derimot består primært av sot holdige partikler. Utslipp fra Barentsburg kraftstasjonen er dominert av flyveaske og sekundære partikler. Flyveaske fra Barentsburg er karakterisert av høy fosfor innhold. Det direkte utslippet fra diesel aggregatet i Svea er derimot dominert av sot. Noen partikler i Svea prøvene inneholder bly (Pb) men en stor andel av analysert partikulært materialet inneholder også kvikksølv (Hg).

Høy oppløselig TEM analysene bekrefter igjen steds spesifikke sammensetninger for begge kullfyrte kraftstasjonene (Longyearbyen og Barentsburg). Mer en 80% av sot partiklene fra Barentsburg inneholder fosfor som en karakteristisk men mindre komponent. I Longyearbyen derimot ble det funnet mange bly-inklusjoner (Pb) i sot partiklene sammenlignet med Barentsburg.

Vanligvis dominerer flyveaske i partikulæret utslipp fra kullforbrenning. Disse er derfor ofte betraktet som typiske kjennetegn for kullforbrenning og relater utslipp i atmosfæren. Derfor er det overraskende at andelen av sot partiklene i våre undersøkelser er høy. Sot har også en betydelig effekt på strålingsregime i atmosfæren. Også Svea diesel aggregatet slipper ut en høy andel av sot i atmosfæren som inneholder overraskende høye mengder av kvikksølv (Hg). Utslipp av kvikksølv partikler har aldri blitt påvist før i lignende målinger.

Denne studien viser at en slik karakterisering av partikkel utslipp fra primærkildene (kraftstasjoner, industri-anlegg etc) kan bidra med viktig informasjon for å karakterisere utslippsmønstre og utslippsegenskaper. Denne typen av informasjon er komplementært til data som produseres gjennom de atmosfæriske overvåkingsprogrammene både i Longyearbyen og Ny-Ålesund (Zeppelin stasjon). I en lignende undersøkelse i 2011 ble partikler samlet fra Zeppelin stasjonen (Ny-Ålesund). I denne tidligere undersøkelsen ble også sot partikler regelmessig funnet. Disse kunne ikke festes til en bestemt luftretning (bestemt gjennom luftmasse trajektorie beregninger). Dermed ble det konkludert at sot kan ha en lokal opprinnelse i disse partikkel prøvene. Siden det ikke ble påvist fosfor i Zeppelin partikkelprøvene kan dermed Barentsburg ekskluderes som mulig kilde for sot funnet i Zeppelinprøvene fra 2011.

## **Anbefalinger**

Denne undersøkelse viser at enkelt partikkel bestemmelse kan bidra med viktig vitenskapelig informasjon i vurderinger av kilder og opprinnelsen av forurensete luftmassene. I kombinasjon med standard luftovervåkingsmetoder (som gjennomføres på Svalbard allerede siden 90 tallet) kan denne type av måling brukes for nøyaktig kilde identifisering og mønster analyser. Vi anbefaler derfor at lignende undersøkelse for kartlegging av partikulært utslipp fra alle potensielle kilder inkludert industri og turisme og bosetninger gjennomføres i kombinasjon med allerede etablerte overvåkingsprogrammer i Ny-Ålesund og Longyearbyen.

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