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Umweltmineralogie
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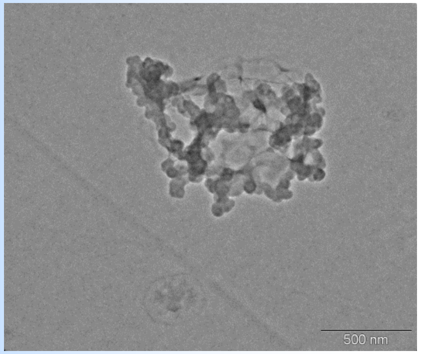
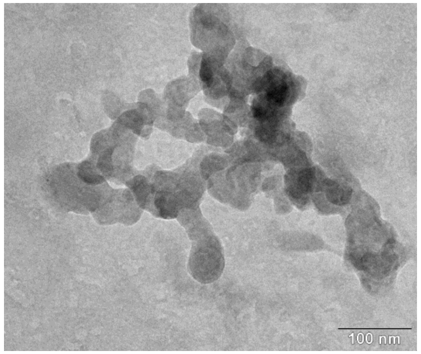
Source identification of individual soot agglomerates in Arctic air by transmission electron microscopy





coal burning Longyearbyen

cruise ship Ny Ålesund

Introduction

Soot (black carbon) influences the Arctic climate by (Quinn et al., 2011):

(a) direct forcing by absorption of solar radiation,

(b) reduction of albedo after deposition on snow,

(c) indirect and semi direct forcing by changing microphysical properties of clouds,

(d) radiative forcing outside the Arctic leading to changes in energy transport by the atmospere and oceans.

highest BC concentrations in late winter/early spring: 80 – 100 ng/m^3

lowest BC concentrations in summer/autumn: 5 – 10 ng/m^3

downward trend of annual mean BC of approximately 1 – 2 ng/m^3 per year (≈ 1989 – 2009)

main source region Northern Eurasia, but also contribution from South Asia

Contribution of different sources to BC in the Arctic are usually derived from

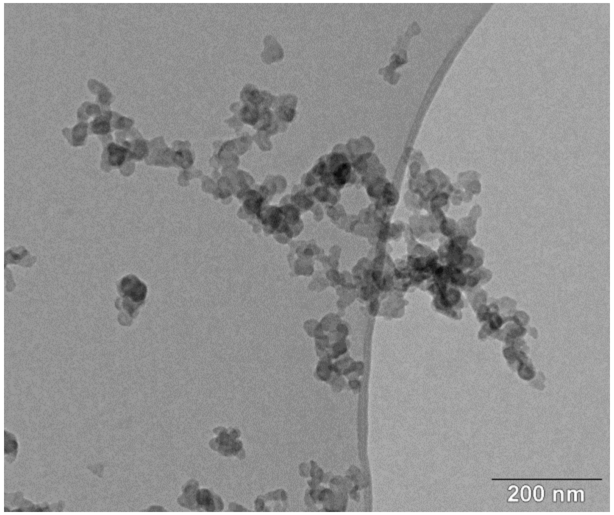
emission inventories **or** **chemical tracers measured at the receptor.**

In the latter case source identification usually based on bulk analysis.

In the present contribution, a single particle approach is elaborated:

(a) Properties of individual soot agglomerates

- **size of primary particles**
- **graphene sheet separation distance**
- **chemical composition of soot agglomerates**



(b) Particle groups externally mixed with soot

Sampling

location/source	code	date
ambient samples		
Longyearbyen settlement	A-LB	19.03.2010 22.03.2010
Ny Ålesund settlement	A-NA	11.03.2010
Ny Ålesund balloon	A-NAB	26.03.2014
Zeppelin Station, Ny Ålesund	A-ZS	07.10.2008 07.11.2008 22.11.2008
local sources		
Barentsburg downwind power plant	L-BBPP	27.03.2010
Longyearbyen downwind power plant	L-LBPP	22.03.2010 25.03.2010
Ny Ålesund downwind cruise ship	L-NACS	16.07.2007
Ny Ålesund diesel aggregate	L-NADA	26.03.2014
Ny Ålesund oil burning	L-NAOB	26.03.2014
Sveagruva diesel aggregate	L-SGDA	10.03.2014
other sources		
air plane	O-AP	09.04.2014
biomass burning (airborn sampling)	O-BB	22.01.2008 23.01.2008
diesel car	O-DC	26.04.2012

Analysis

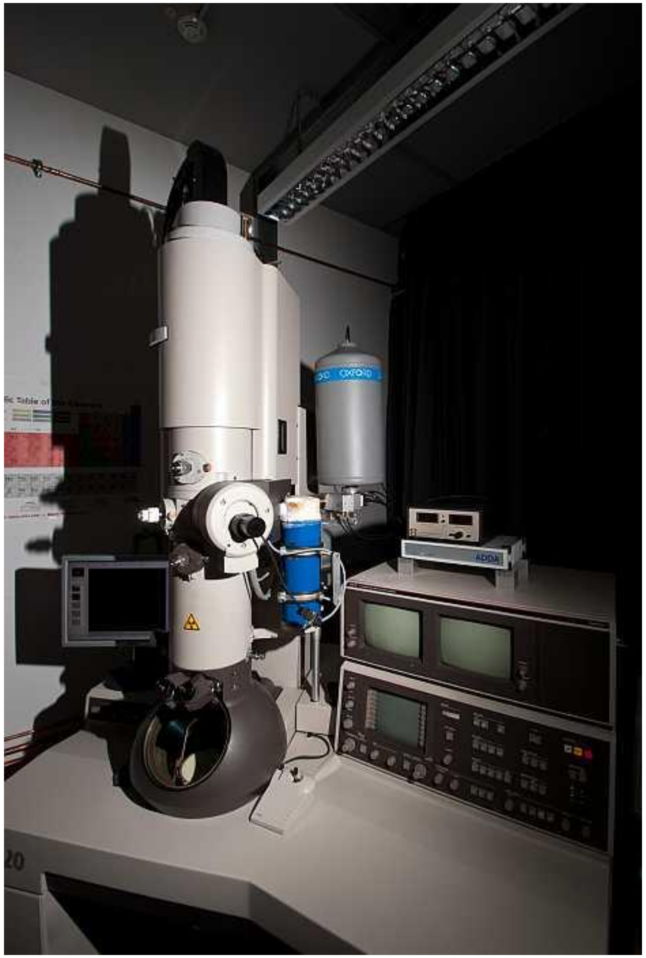
Sampling:

two-stage micro inertial cascade impactor

or

electrostatic precipitator

Ni or Cu grids covered with Formvar foil



Transmission electron microscopy:

Philips CM 20, LaB_6-cathode,

200 kV (≈ 0.23 nm point resolution)

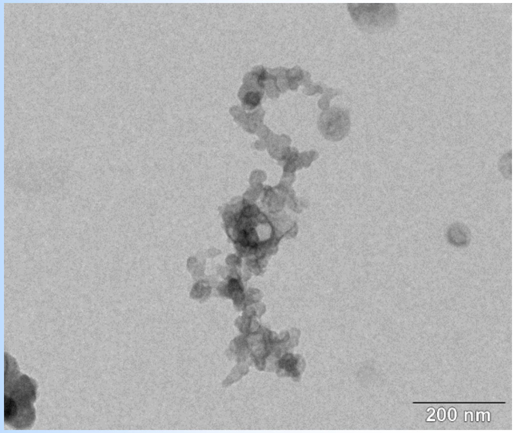
energy-dispersive X-ray microanalysis

Oxford Instruments (Max 80)

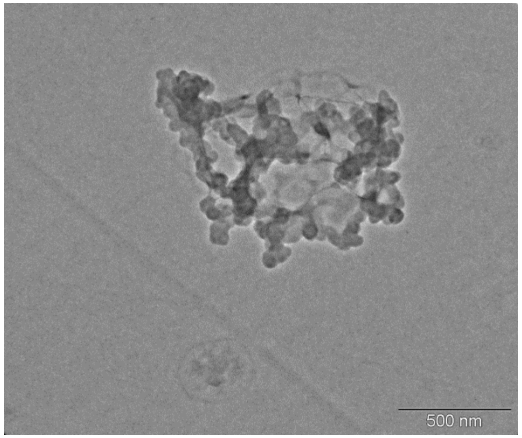
all elements with $Z \geq 5$

Results

chain-like or more compacted agglomerates of primary particles

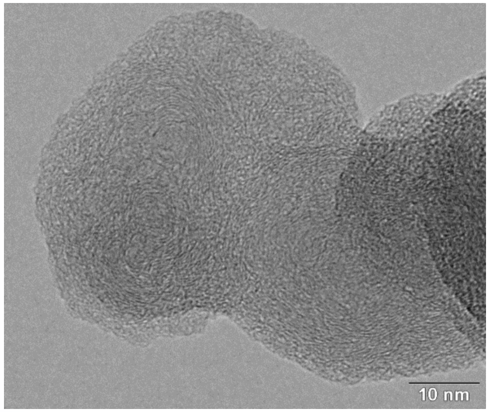


coal burning Barentsburg



coal burning Longyearbyen

typical onion shell structure of graphene layers



diesel car

Size and nanostructure of soot primary particles

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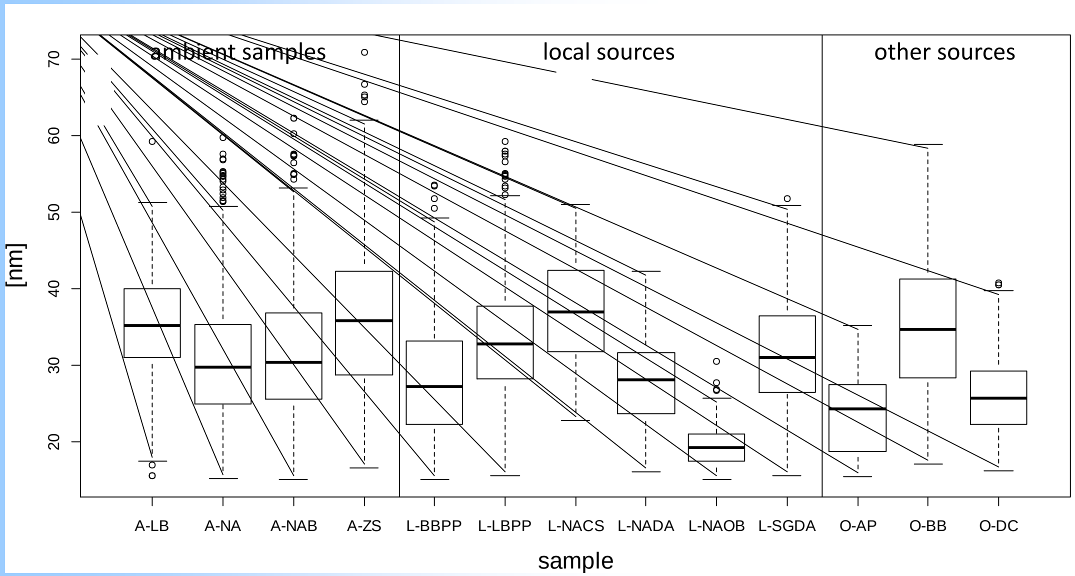
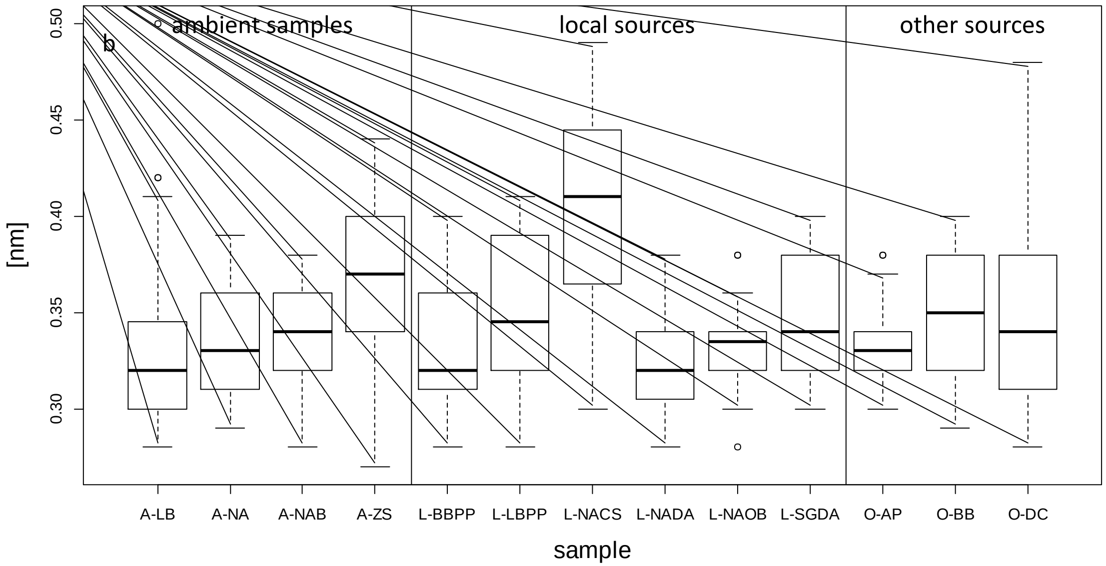
primary particle diameter



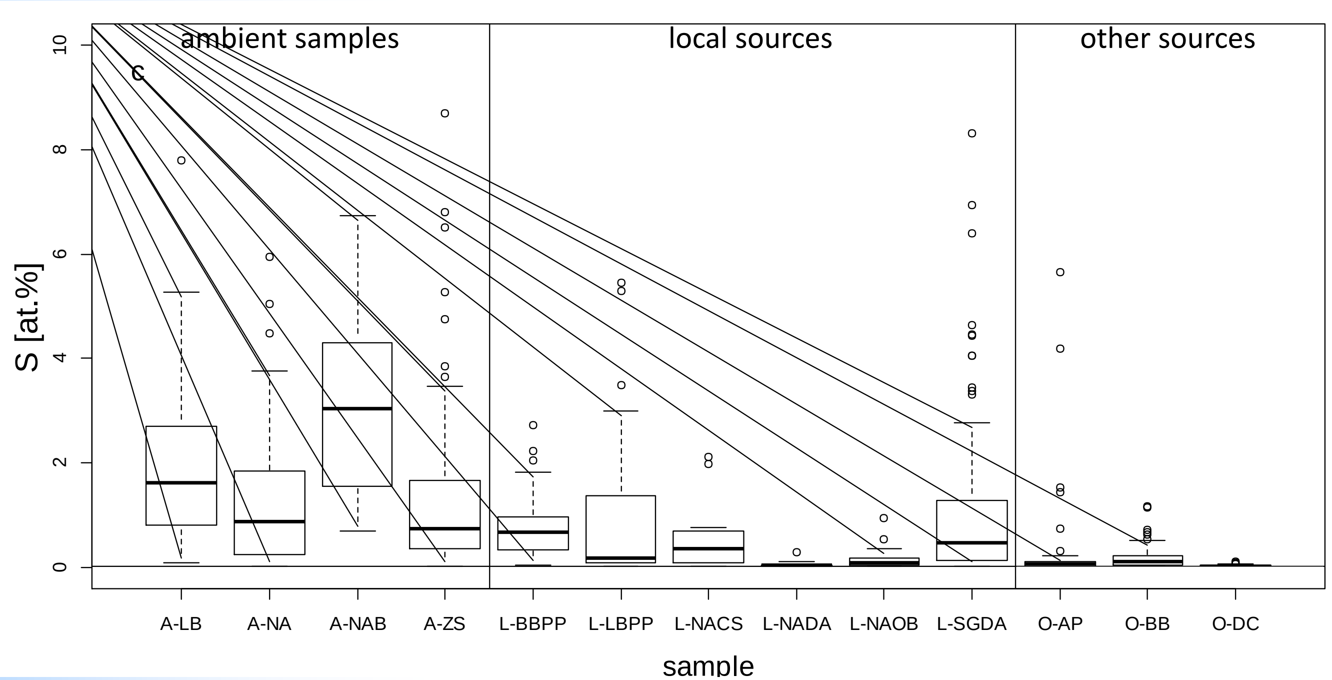
primary particle diameter <u>not</u> suited for source identification

graphene sheet separation distance



graphene sheet separation distance <u>not</u> suited for source identification

Minor element composition of soot

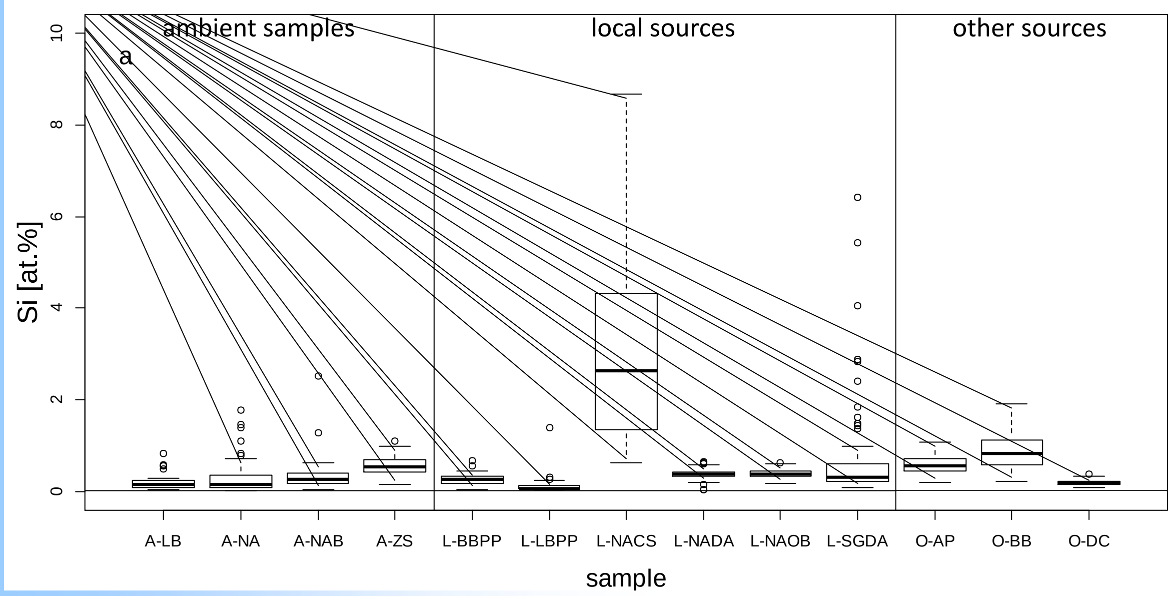


S, O and Na <u>not</u> used for source identification, as all ambient samples and all local source samples not directly collected at the stack contain secondary aerosol and/or (aged) sea salt

→ apparent concentration of these elements in soot are enhanced

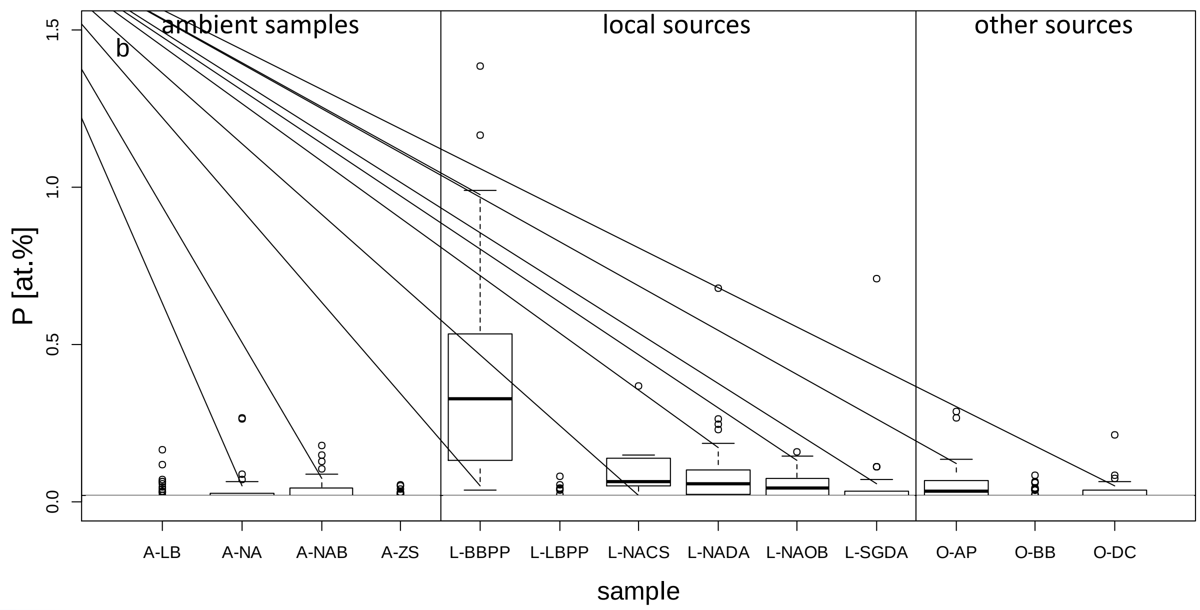
→ only minor elements in soot which not effected by the presence of other phases can be used for source identification

Minor element composition of soot

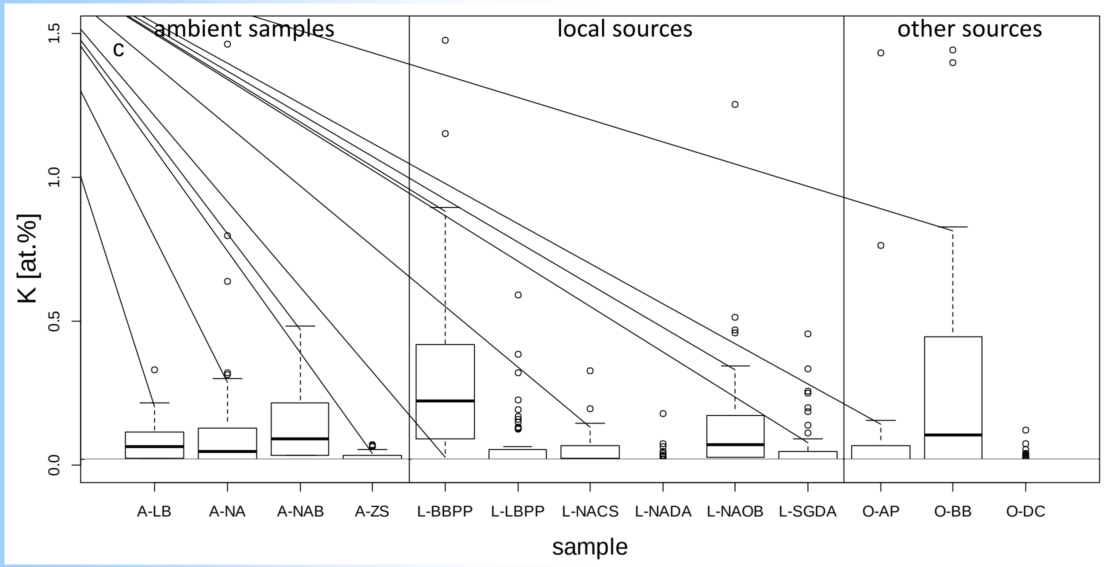


Si is a good marker for ship emissions

P is a good marker for coal burning at Barentsburg

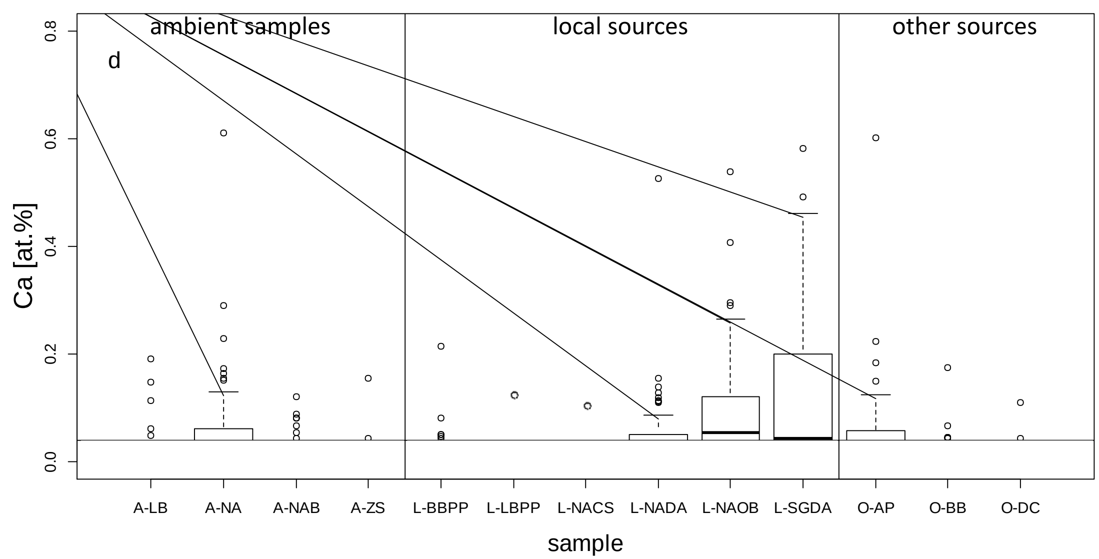


Minor element composition of soot

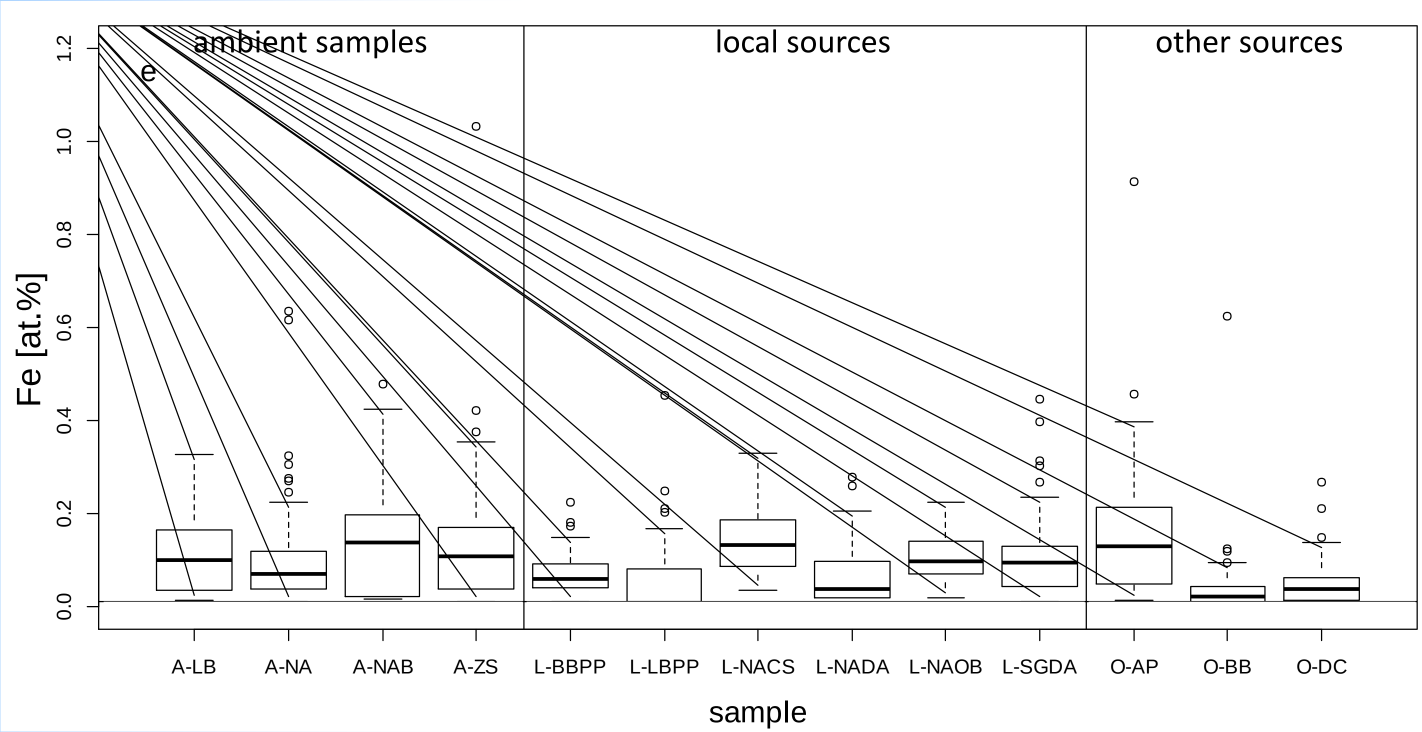


K is considerably enhanced in biomass burning soot and soot from coal burning in Barentsburg

Ca is higer in soot from oil burning in Ny Ålesund and from the diesel power plant in Sveagruva



Minor element composition of soot

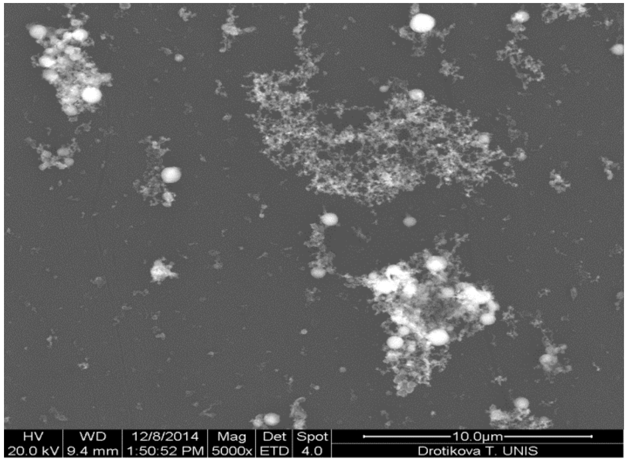


Fe concentrations are rather constant. Lowest values observed for coal coal burning in Longyearbyen, diesel power plant in Ny Ålesund, biomass burning, diesel car.

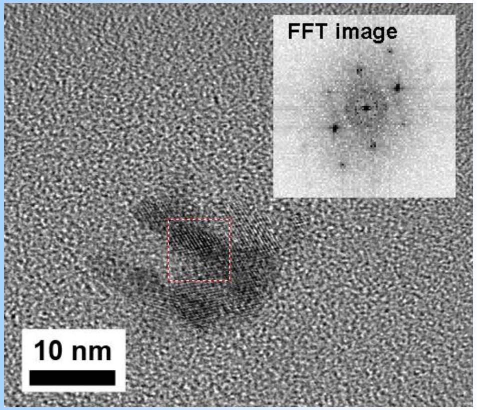
Particle groups externally mixed with soot

coal burning samples (Barentsburg, Longyearbyen) contain a significant fraction of fly ashes (this is also true for coal burning in general)

fly ashes were found in all ambient samples on Svalbard



coal burning Longyearbyen



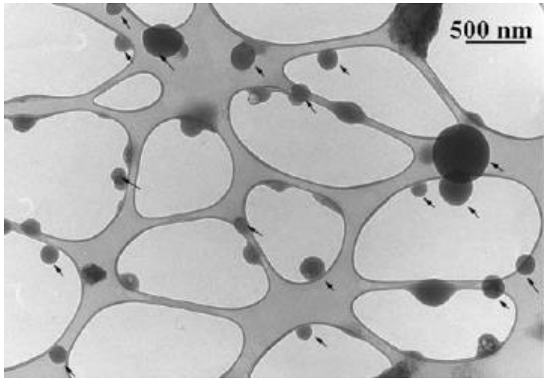
diesel aggregate Sveagruva

emissions from the diesel aggregate in Sveagruva contain small (≈ 10 – 30 nm) metacinnabar (cubic HgS) particles

not observed in ambient samples on Svalbard

biomass/wood burning always leads to emission of tar balls

not observed in ambient samples on Svalbard



Biomass burning, Mozambique; (Pósfai et al., 2003)

Sources of soot in ambient samples

source	contribution	criteria
coal burning Barentsburg	excluded	P content
coal burning Longyearbyen	likely in Longyearbyen excluded for Zeppelin Station	fly ashes Si content
long-range transport of coal burning emissions	likely	fly ashes
ship emissions	excluded	Si content, other tracers (V, Ni)
biomass/wood burning	excluded	K content, tar balls
diesel aggregate Sveagruva	excluded	HgS particles
oil burning Ny Ålesund	possible	no criterion found
diesel aggregate Ny Ålesund	possible	no criterion found
diesel engines	minor contribution possible	Fe content
aircraft emissions	possible	no criterion found

Further work

ice core samples

→ differences between the wood burning, coal burning and oil burning era

sources of EC in urban environments

→ wood burning versus diesel cars

carbonaceous particles in stratospheric samples

→ not enough material for bulk analysis available