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Overview of Danish Contributions to Monitoring of SLCPs in the Arctic

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***Cover photo*: DMI staff deploying an ozone sonde in Thule**

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Content

1. Abstract

In the Tromsø Declaration (2009) the Arctic Council noted the role that shorter-lived climate forcers such as black carbon, methane and tropospheric ozone precursors may play in Arctic climate change, and recognized that reductions of emissions have the potential to slow the rate of Arctic snow, sea ice and ice sheet melting in the near-term.
Short lived climate pollutants/forcers (SLCPs) have in recent year become an area of attention, and several international initiatives and studies have underlined the potential climate and health benefits of reducing emissions of SLCPs.
Whereas reductions in anthropogenic sources of SLCPs warrant action within and outside of the Arctic region effects of such actions need to be monitored in the Arctic as to measure the effectiveness of mitigation at lower latitudes and as to support the development of our understanding of the role that the SLCPs play in Arctic climate change including a better understanding of trends, temporal and spatial distribution, transport mechanisms, atmospheric processes, chemical interactions, etc.
The monitoring of SLCPs in the Kingdoms arctic region (Greenland) is carried out by different institutions and for different purposes. In 2013 the Danish Environmental Protection Agency and the Danish Energy Agency decided to get an overview of the ongoing monitoring and research related activities on SLCPs in Greenland. Therefore, DEA has granted support from the Danish Corporation Program for Climate to the Danish Meteorological Institute to arrange a workshop for Danish institutions involved in SLCP monitoring. The purpose of the workshop was to create an overview of the monitoring activities and produce recommendations on a future Danish SLCP monitoring network in Greenland. The presentations from the workshop are summarized here while the recommendations are concluding the report.
The workshop and the report has been supported by the DANCEA programme (climate) administered by the Danish Energy Agency (DEA).
This report and its recommendations do not necessarily cover the views of the Danish Energy Agency and the Danish Environmental Protection Agency.

2. Resumé

This report presents an overview of Danish monitoring activities of short lived climate pollutants (SLCPs) in the Arctic. Main SLCPs include black carbon, tropospheric ozone and methane. These are all monitored at Danish stations in Greenland and results are reported in national and international programs such as the Arctic Monitoring and Assessment Programme, the World Meteorological Organization Global Atmospheric Watch or the global Network for the Detection of Atmospheric Composition Change. Apart from basic reporting the monitoring also serves as a basis for improved scientific understanding of climate related processes in the atmosphere.
Tropospheric ozone is the SLCP with the highest degree of monitoring over Greenland. Four different instruments are used at four monitoring stations. Tropospheric profiles are monitored at two stations while monitoring of surface ozone is performed at all stations. Surface based monitoring of black carbon is considered at one station while aerosol size distributions are considered at five stations and total columns at four stations. Surface based monitoring of methane is carried out at two stations while monitoring of methane profiles is considered at one station.
For the short term climate forcing of SLCPs the total tropospheric column is of importance and monitoring must consists of both surface and profile based observations. The geographical coverage in the Arctic is sparse and it is recommended in this report to investigate the usage of satellite data for monitoring SLCPs. Polar satellites carrying advanced instruments for monitoring of aerosol and trace gases have high spatial coverage near the pole. Such data is readily available and presents an opportunity to increase geographical coverage in a cost effective manner. Further recommendations include usage of already existing equipment currently not in operations and increased focus on existing measurement platforms. For instance it could be investigated whether already existing tropospheric soundings could be equipped with instruments for monitoring black carbon or further physic-chemical parameters could be monitored at already existing stations.

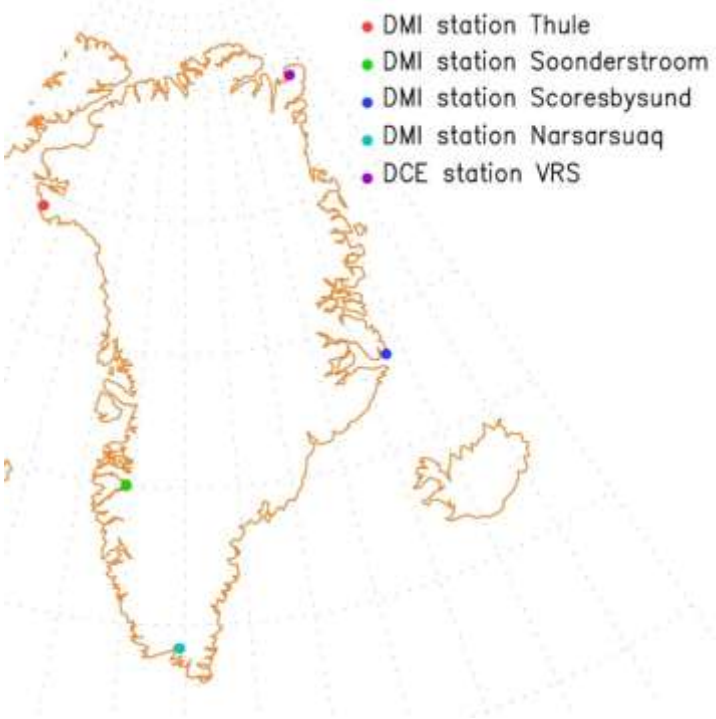
3. Introduction

This report is based on a workshop, supported by the Danish Energy Agency, on short lived climate pollutants (SLCPs) held at the Danish Meteorological Institute (DMI) in December 2013. The participants of the workshop have submitted extended abstracts which form the basis of the report. The purpose of the workshop was to generate an overview of Danish monitoring activities with respect to SLCPs in the Arctic.
SLCPs comprise a range of potent climate warming agents with atmospheric residence times much shorter than CO_2 and include methane, tropospheric ozone, black carbon and hydrofluorocarbons. The short residence time of SLCPs facilitate a fast response in radiative forcing and emission reductions leading to a 0.4°C global temperature reduction by 2050 have previously been suggested (*UNEP, 2011*). The effects of mitigating SLCPs are projected to be relevant mainly on short time scales and long lived species must be reduced in order to achieve lasting climate changes. CO_2 is generally accepted to be the most important longlived greenhouse gas and new results indicate that for some countries CO_2 is also the main climate warming substance on shorter timescales (*Miljødirektoratet, 2013*). In addition to their climate effects SLCP species also affect local air-quality and agricultural yield and significant co-benefits in terms of improved health and increased agricultural productivity is expected from emission reductions.
Monitoring of climatically and environmentally active species in the Arctic is important not only for policy-based reporting, but also for basic process-based understanding of the atmospheric system. In Denmark there are two Institutions with SLCP monitoring activities in the Arctic, the Danish Centre for Environment and Energy (DCE) at Aarhus University and the Danish Meteorological Institute (DMI). In addition the Geological Survey of Denmark and Greenland (GEUS) monitor properties of climate sensitive parameters on climate stations along the Greenland ice sheets.

Monitoring activities at the DCE Villum Research Station/Station Nord (VRS) is part of the Arctic Monitoring and Assessment Programme (AMAP) assessment work (*http://www.amap.no/*) in part financed by the Danish Environmental Protection Agency (Danish EPA), Danish Cooperation for Environment in the Arctic (DANCEA) and results are part of AMAP and the European Monitoring and Assessment Programme (EMEP) network (*http://www.emep.int/*) with reporting to the AMAP/EMEP secretariats and where there regularly are joint efforts to produce Assessment reports to the Arctic Council and other stakeholders. Denmark is a party to the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) with the purpose of reducing air pollution including long range transported pollutants (www.unece.org/env/lrtap). Denmark has ratified the convention while Greenland only signed the convention. EMEP is a monitoring body within LRTAP. VRS is also part of the World Meteorological Organization (WMO) Global Atmospheric Watch (GAW) programme (*http://www.wmo.int/pages/prog/arep/gaw/gaw_home_en.html*). Furthermore, VRS is a member of INTERACT which is network that support foreigners stay at e.g. VRS.

Monitoring of ozone at DMI stations is carried out as a collaboration between the Danish EPA funded by DANCEA and DMI and serves the purpose of monitoring the recovery of the stratospheric ozone layer under climatically changing conditions. The monitoring is part of the Danish contribution to the Vienna convention for the protection of the ozone layer and DMI holds a position in the associated Ozone Research Managers Meeting which reports

monitoring efforts and results to the conference of the parties. These monitoring activities, however, also include monitoring of surface and tropospheric ozone, since ozone sondes start measuring at the surface upon deployment.
The DMI ozone monitoring stations are part of the global Network for the Detection of Atmospheric Composition Change (NDACC) and DMI is a member of NDACCs steering committee (*http://www.ndsc.ncep.noaa.gov/*). The measurements are reported to international databases under the WMO World Ozone and UV Data Centre (WOUDC), as well as GAW (*http://www.woudc.org/*). Ozone has been defined as an essential climate variable (UNFCCC) and the measurements are also part of the Global Climate Observing System (GCOS) (*http://gosic.org/gcos*). Monitoring at the DMI stations is also used as input to WMO's Global Framework for Climate Services (GFCS) (*http://www.gfcs-climate.org/*). The monitoring data will also be part of the AMAP expert group on black carbon and ozone assessments. Besides these more political agendas, the monitoring data are also used for calibration/validation of satellite data as well as basic research.
Monitoring SLCPs involves combining both environmental and climate related observations. From an environmental perspective it is the SLCP concentrations near the Arctic surface that are of concern. Both black carbon and ozone in the troposphere are known to impose health and ecosystem damage (*UNEP, 2011*) to vulnerable systems. From a short term climate perspective, however, it is the concentrations in the upper troposphere that are of importance. The long wave radiative forcing of ozone for example is greatest where the largest temperature difference between the surface and the troposphere exists.
The distribution of BC in the troposphere is uncertain as most observations are carried out at ground level. Only few observations of the vertical distribution exists and comparison with model results have uncovered large uncertainties (*AMAP 2011*).
Therefore, monitoring of SLCPs in the Arctic requires monitoring both within the boundary layer and in the free troposphere. For species with a longer lifetime such as methane columnar abundances may be more relevant.



Figur 0 Distribution of Danish SLCP measurement stations on Greenland

4. Properties of short lived climate pollutants

Black carbon (BC)

Following *Petzold et al., (2013)* BC is here used to qualitatively describe carbonaceous matter sharing certain physical properties such as strong absorption in the visible part of the spectrum, insolubility and thermal stability. Elemental (pure) carbon particles are a by-product of incomplete combustion (combustion with an oxygen deficit) of carbon rich fuels such as biomass, bio and fossil fuels. These particles initially cluster in fibre-like structures and a few hours after emission they have aged sufficiently through condensation of co-emitted species to a level that their physical characteristics have changed (*Bond et al., 2013*). Local sources include wind-blown emissions from coal piles, however, these particles are usually large and deposit fast due to gravitational settling. The largest component of BC in the Arctic is, therefore, currently believed to be smaller, aged particles transported into the Arctic from source regions mainly in Eurasia and North America. BC affects the short term Arctic climate through absorption of incoming radiation, influences on cloud radiative and microphysical properties, changes in albedo and enhanced melting of snow and ice upon deposition. A heating layer in the upper part of the troposphere may not be sufficient for heating the Arctic surface and sources closer to the Arctic may play a larger role for surface warming although emissions are not as large. These emissions are also more likely to be deposited on snow.

Organic carbon (OC)

The term organic carbon refers to the organic particle component that is not elemental. It is co-emitted with BC and can also be formed in the troposphere from the photooxidation of non-methane volatile organic compounds (NMVOCs). OC has less absorption properties than EC and is often also denoted brown carbon though it is often measured by a non-optical method.

Sulphur dioxide (SO_2) and sulphate

Sulphur dioxide is emitted during incomplete combustion of sulphur containing fuels such as coal and oil. SO_2 is by itself not a greenhouse gas but it is oxidized in the atmosphere and leading to the generation of secondary sulphuric acid and sulphate particles. These are known to reflect incoming short wave radiation and are therefore climatically active. Globally these particles have a cooling effect due to their ability to scatter sunlight but in the Arctic the situation is more complex. In the summer season scattering is believed to be the dominant effect but during the Arctic night it might have a small heating effect through scattering of the outgoing heat flux.

Methane (CH_4)

Methane is a well-mixed greenhouse gas with a chemical lifetime of about 8 years. Concentrations vary with latitude and decrease above the tropopause, however this variation only contributes 2% to the uncertainty in radiative forcing. The increase of methane in the atmosphere has caused the largest radiative forcing by any greenhouse gas after CO_2. Methane concentrations have grown as a result of human activities related to agriculture, including rice

cultivation and the keeping of ruminant livestock, coal mining, oil and gas production and distribution, biomass burning and municipal waste landfills.
Methane has a direct influence on Arctic climate but it has also a number of indirect effects through its importance for the oxidative capacity of the troposphere and including its role as an important precursor to the formation of tropospheric ozone and impacts on CO_2, being a source for stratospheric water vapour, important for the rate sulphate aerosol formation, and lifetimes of Hydrofluorocarbons and Hydrochlorofluorocarbons (HCFCs) (*IPCC, 2013*).

Ozone (O_3)

Tropospheric ozone is a secondary gas generated by chemical reactions between NO_x ($NO+NO_2$), volatile organic compounds and carbon monoxide in the presence of short wave radiation. NO_x and CO have mainly anthropogenic sources; due to burning of fossil fuels whereas about 90% NMVOC are emitted from biogenic sources.
O_3 is a greenhouse gas with both short wave and long wave absorption bands. Under normal conditions only small amounts of UV radiation penetrate the stratospheric ozone layer, which protects organisms and ecosystems from harmful effects. In the troposphere UV radiation is almost completely absorbed by ozone. Consequently, the short wave forcing is expected to be small but is of importance for the oxidative capacity of the troposphere (photolysis leads to the formation of OH which is the main oxidative radical in the troposphere). The long wave radiative forcing is expected to be largest in the upper troposphere where the temperature difference to the surface is greatest. Therefore, it is the upper tropospheric levels of O_3 that is expected to be of greatest importance from a climate perspective. Methane and carbon monoxide are therefore the most important O_3 precursors in terms of reducing short term climate forcing. It should be noted that the often stably stratified Arctic troposphere inhibits mixing and therefore radiative forcing at upper tropospheric levels does not necessarily imply surface warming.

Hydrofluorocarbons (HFC)

Hydrofluorocarbons are used as refrigerants, aerosol propellants, solvents, and fire retardants. The major emissions source of these compounds is their use as refrigerants e.g. in air conditioning systems. HFC's have no natural sources and only come from human-related activities. These chemicals were developed as a replacement for chlorofluorocarbons (CFCs) and HCFCs because they do not deplete the stratospheric ozone layer. However, they have large Global Warming Potentials (GWP), ranging from 140 to 14800 (HFC 23) on a 100 year scale (compared to the GWP of CO_2 which is normalized to 1). They have atmospheric lifetimes from 1 to 270 years and are thus mostly well-mixed in the atmosphere, spreading around the world after they're emitted. Fluorinated gases are removed from the atmosphere only when they are destroyed by sunlight in the upper atmosphere.

5. Measurements of SLCPs in the Arctic

This chapter gives an overview of measurements and techniques used for monitoring SLCPs at Danish measurement stations. Relevant species include O_3, particulate matter components such as BC and CH_4. Here we only consider the main SLCPs while other precursor species such as NO_x or SO_2 are not considered here. Such precursers are among the species measured at VRS; a full list of species measured at VRS may be found here: *http://envs.au.dk/en/research/facilities/villumresearchstation/* . An overview of monitoring at Danish stations is given in table 1 and the location of the stations is displayed in figure 0.

Station	Location	Species	Method	Surface/profile/Column	Organization	Period
VRS	81.60N;16.66W	O_3	API	+/-/-	DCE	1997-2002; 2006-present
Pituffik (Thule)	76.51N;68.74W	O_3	ORS	+/+/-	DMI	1991-present
Pituffik (Thule)	76.51N;68.74W	O_3	SAOZ	-/-/+	DMI	1990-present
Pituffik (Thule)	76.51N;68.74W	O_3	API	+/-/-	DMI	1996-2003
Kangerlussuaq (Sondrestrom)	67.01N; 50.65W	O_3	BS	-/-/+	DMI	1990-present
Kangerlussuaq (Sondrestrom)	67.01N; 50.65W	O_3	API	+/-/-	DMI	1995-1997
Ittoqqortoormiit (Scoresbysund)	70.48N; 21.95W	O_3	ORS	+/+/-	DMI	1999-present
Ittoqqortoormiit (Scoresbysund)	70.48N; 21.95W	O_3	SAOZ	-/-/+	DMI/CNRS	2007-present
Ittoqqortoormiit (Scoresbysund)	70.48N; 21.95W	O_3	API	+/-/-	DMI	1994-2000
VRS	81.60N;16.66W	BC	PSAP/MAAP	+/-/-	DCE	2008-present
VRS	81.60N;16.66W	EC/OC	EC/OC analyser	+/-/-	DCE	2009-2012
VRS	81.60N;16.66W	Particle number; size dist.	SMPS instrument	+/-/-	DCE	2010-present
Pituffik (Thule)	76.51N;68.74W	AOD	SR	-/-/+	DMI/NASA	2007-present
Kangerlussuaq (Sondrestrom)	67.01N; 50.65W	AOD	SR	-/-/+	DMI/NASA	2008-present
Ittoqqortoormiit (Scoresbysund)	70.48N; 21.95W	AOD	SR	-/-/+	DMI/NASA	2010-present
Narsarsuaq	61.16N; 45.44W	AOD	SR	-/-/+	DMI/NASA	2013-present
Pituffik (Thule)	76.51N;68.74W	CH_4	FTIR	+/+/-	DMI/NCAR	1999-present
VRS	81.60N;16.66W	CH_4	CRD	-/-/- Mast (3.6 m, 20 m, 40 m, 60 m and 80 m above ground level)	DCE	2012-present

Table 1 Overview of SLCP monitoring activities by Danish institutions. In the table method abbreviations refer to: API (advanced pollution instrument), ORS (ozone radiosounding), SAOZ (systeme d'analyse par observation zenithale), BS (brewer spectrometer), PSAP (particle soot absorption photometer), MAAP (multi angle absorption photometer), SR(spectral radiometer), FIR (Fourier Transform Infrared Spectroscopy), CRD (cavity ring down spectroscopy). AOD refers to aerosol optical depth while other abbreviations are standard. Note that ORS profiles also provide surface observations.

5.1 Monitoring of O_3

O_3 is monitored at four stations in Greenland. All of these monitor surface values while two of them also include soundings of the troposphere and stratosphere. Several instruments are used on the stations a brief description is given below.

Ozone radiosounding (ORS)

The ozone sonde is an electrochemical device containing two electrode chambers the anode chamber containing a saturated solution of potassium iodide while the cathode chamber contains 10 g/l potassium iodide. The device is put inside a Styrofoam box with a heater element to keep the solutions from freezing. A constant volume pump is drawing atmospheric air through the cathode chamber where sampled ozone reacts reacts (oxidizes) the iodide giving rise to an electric current in the external circuit and an ionic current through the salt bridge connecting the two electrode chambers. The electric current is proportional with the contained ozone which may then be calculated and transmitted to a ground receiver along with the other measured parameters (air pressure, air temperature, humidity, wind speed and direction and styrofoam box temperature). At best the sonde may reach an altitude around 35 km.

Saoz spectrometer (SAOZ)

The Saoz is recording spectra from 300-600 nm by viewing towards zenith when sun it setting/rising (for solar zenith angles at 86-91°) to get a large slant column. The recorded spectrum is compared to a reference spectrum. By differential spectral analysis the column of ozone may be calculated.

Advanced Pollution Instrument (API)

The API 400 is a low range ozone analyzer intended to measure ozone from 0-100 ppb. It is a point measurement where an air sample by means of a pump is continuously is drawn through an UV-lamp chamber. The absorption of UV by the sample is compared to the absorption of clean air drawn through an active carbon filter and the difference is proportional to the amount of ozone in the sample.

Brewer spectrometer (BS)

The Brewer instrument is a UV spectrometer able to track the sun and make different types of measurements towards the sun or towards zenith. The recorded values are a measure of the ozone and SO2 column respectively. The use of built in reference lamps (halogen and mercury) improves the accuracy of the instrument.

Examples of monitoring results

Figure 1 and 2 displays examples of surface and upper tropospheric monitoring results for O3 at VRS and Ittoqqortoormiit (Scoresbysund) respectively. The surface mixing ratio shows a clear seasonal variation. Each spring is characterized by a period where ozone mixing ratios are almost completely depleted, the so called ozone depletion events (ODE) (Skov et al., 2004). ODEs have been observed throughout the Arctic (Simpson et al., 2007, Steffen et al., 2008). During summer, autumn and winter the mixing ratio increases to about 40 ppb with maximum values near 50 ppb where after a period with ODEs is reached again.

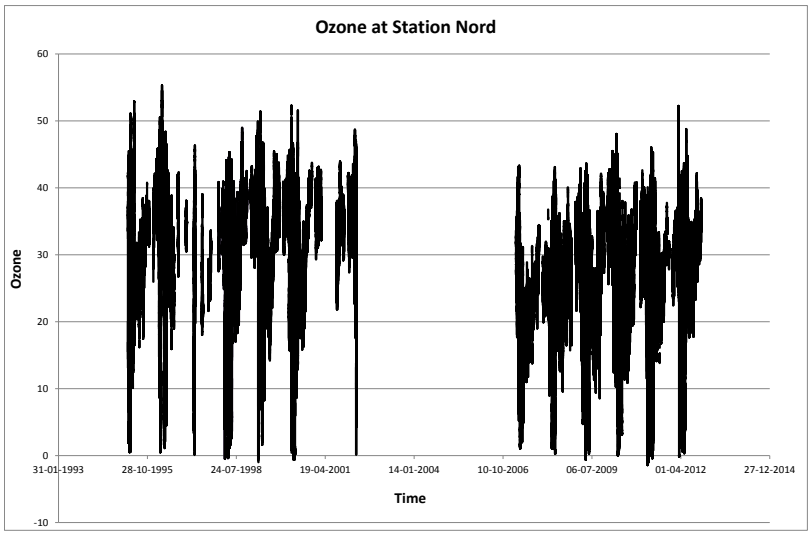


Figure 1 Surface ozone mixing ratio (ppb) as function of time at VRS.

The vertical distribution of O3 shows that there are regular downward propagating intrusions from the lower stratosphere into the upper troposphere. At these times the free tropospheric levels of O3 is controlled by O3 formed in the stratosphere.

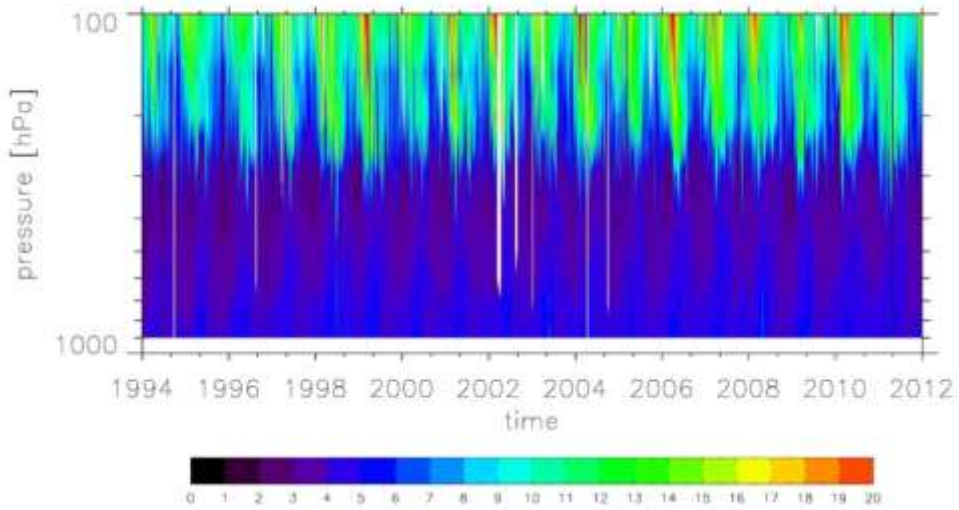


Figure 2 Vertical distribution of tropospheric ozone (mPa) measured at Ittoqqortoormiit (Scoresbysund) from 1994-2012. The tropopause is usually located around 230 hPa.

5.2 Monitoring of BC and aerosol optical depth (AOD)

Currently, BC is monitored at one station while AOD is measured at four stations (table 1). The AOD instruments at the Danish stations are owned by the National Aeronautics and Space Administration and are part of the aerosol robotic network (*http://aeronet.gsfc.nasa.gov/*).

Spectral Radiometer (SR)

The Aeronet is a sun photometer which measures sun and sky radiances at a number of fixed wavelengths through a number of filters. Two motors enable the instrument collimator to be directed at the sun or to perform almucantar, solar triplet and principal plane measurements. The wavelengths in question are 340, 380, 440, 500, 670, 870, 1020 and 1640 nm. The AOD value is calculated for each wavelength, and the inverted data is used for the calculation of aerosol particle distribution values.

Particle Soot Absorption Photometer (PSAP) and the Multi Angle Absorption Photometer (MAAP) as well as EC/OC mesurements

The PSAP technique is based on the change in optical properties of a filter with deposited soot particles. Using Beers law the light absorption coefficient of the deposited particles may be related to the change in optical transmission of the filter. Uncertainty of PSAP results arise if light transmission is reduced due to the presence of scattering aerosol on the filter. The MAAP technique is also based on aerosol optical absorption but due to use of multi angles correction for scattering can be made. The BC content is determined through simultaneous measurements of absorption and scattering of the particles collected on the filter.

Examples of monitoring results

Figure 3 displays an example of measurements of BC at VRS. BC measurements contain signals from elemental carbon (EC) and from organic carbon (OC) but with EC dominating. Far from sources EC and OC have been chemically modified which also effects the optical properties of the particles or BC, see Quinn *et al.* (2011) for a further discussion. However, in order to get an appropriate description of the released soot more methods are needed. At VRS measurements of BC, EC and OC are thus performed using two optical methods along with off-line techniques. Measurements of BC in snow are performed in campaigns. In the figure a comparison of BC and EC measured with three alternative methods are shown.

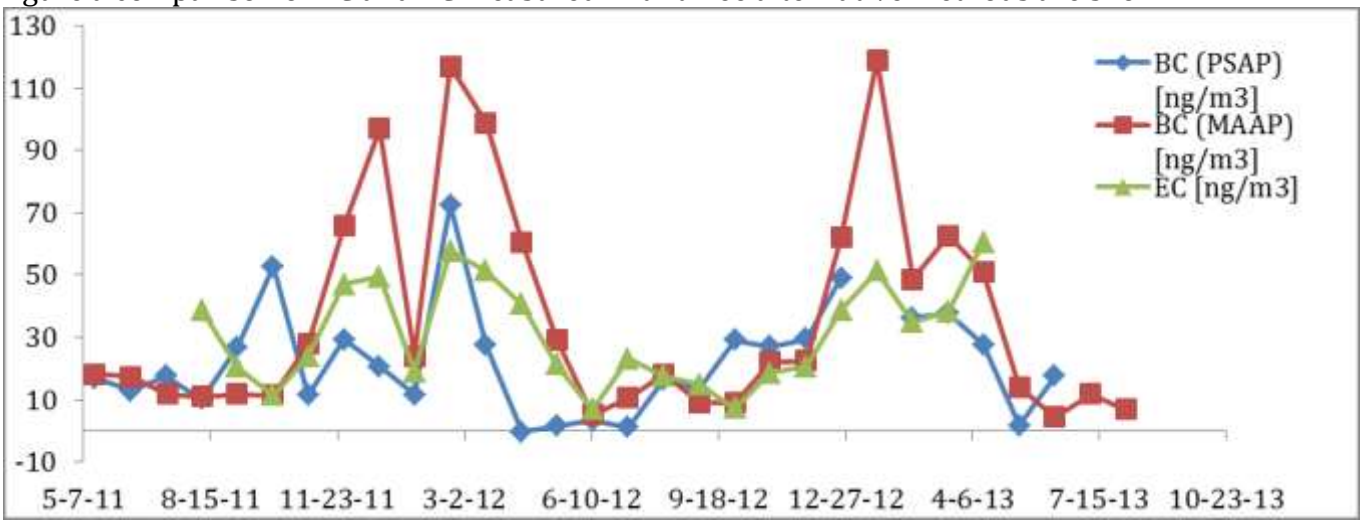


Figure 3 Monthly averaged mass concentration (ng m^{-3}) of BC ameasured with PSAP and MAAP at VRS. EC was measured offline.

The measurements using the three methods show the same general behaviour with very few exceptions and thus are strongly correlated. The ratio between EC and BC can be used to calibrate the PSAP and MAAP instruments. This calibration will be valid for VRS, but is site specific and is not necessary valid at other locations (*Massling et al. 2014*). An example of EC measurements in snow collected near VRS is given in table 2. The table suggests that it is possible to measure the low mass concentrations of EC in snow and that the concentration level varies between 0-2000 ng l^{-1}.

Location	EC (ng/l)	Range (ng/l)	Number of samples	Depth (cm)
Flyger's hut	521 ± 441	1-1754	7 (3 replicas)	5
Icebergs	483 ± 447	9-837	1 (3 replicas)	5
Knuth's fjeld	1159±1677	0-3082	1 (3 replicas)	5
The runway	1449±1047	274-2283	1 (3 replicas)	5
Blind	3±3	1-7	1 (3 replicas)	-

Table 2 ***Average EC concentrations (ng l-1) and standard deviations for snow samples at locations in the vicinity of VRS. Average blind concentrations (ng l-1) and number of observations are also indicated.***

Figure 4 displays an example of usage of the Aeronet data to extract particle size distributions. The figure shows that the particle size distributions are quite variable; the coarse mode peak around 10 µm, indicating natural primary particles, is seen to disappear after three hours while the accumulation and nucleation modes seem to persist. The existence of the smallest mode with a fraction of the particles less than 100 nm suggests local generation of secondary particles.

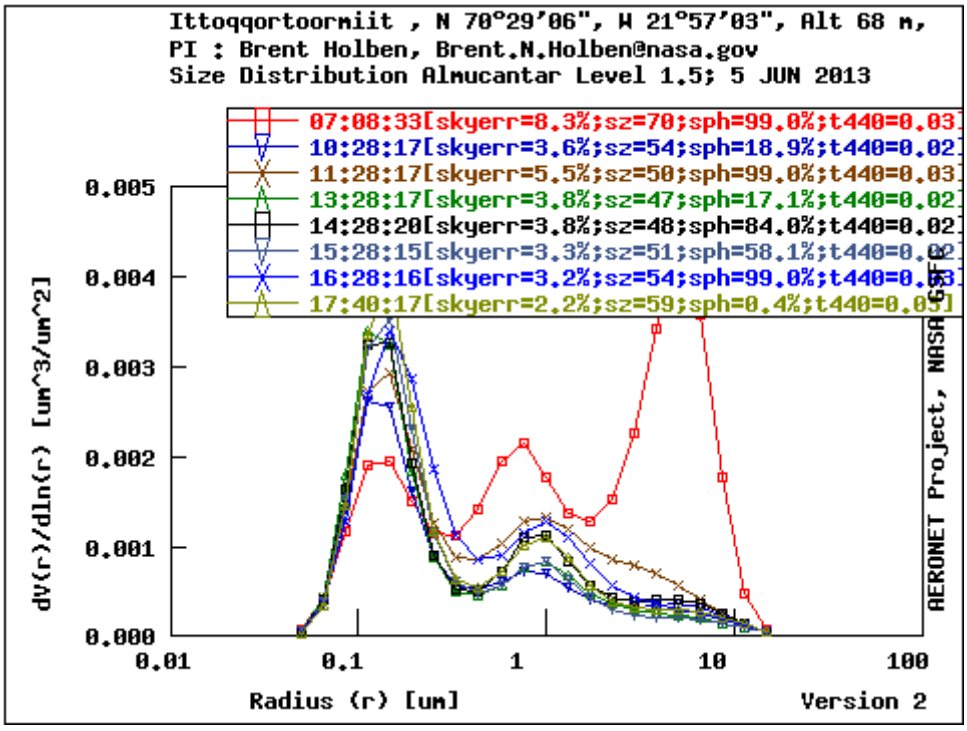
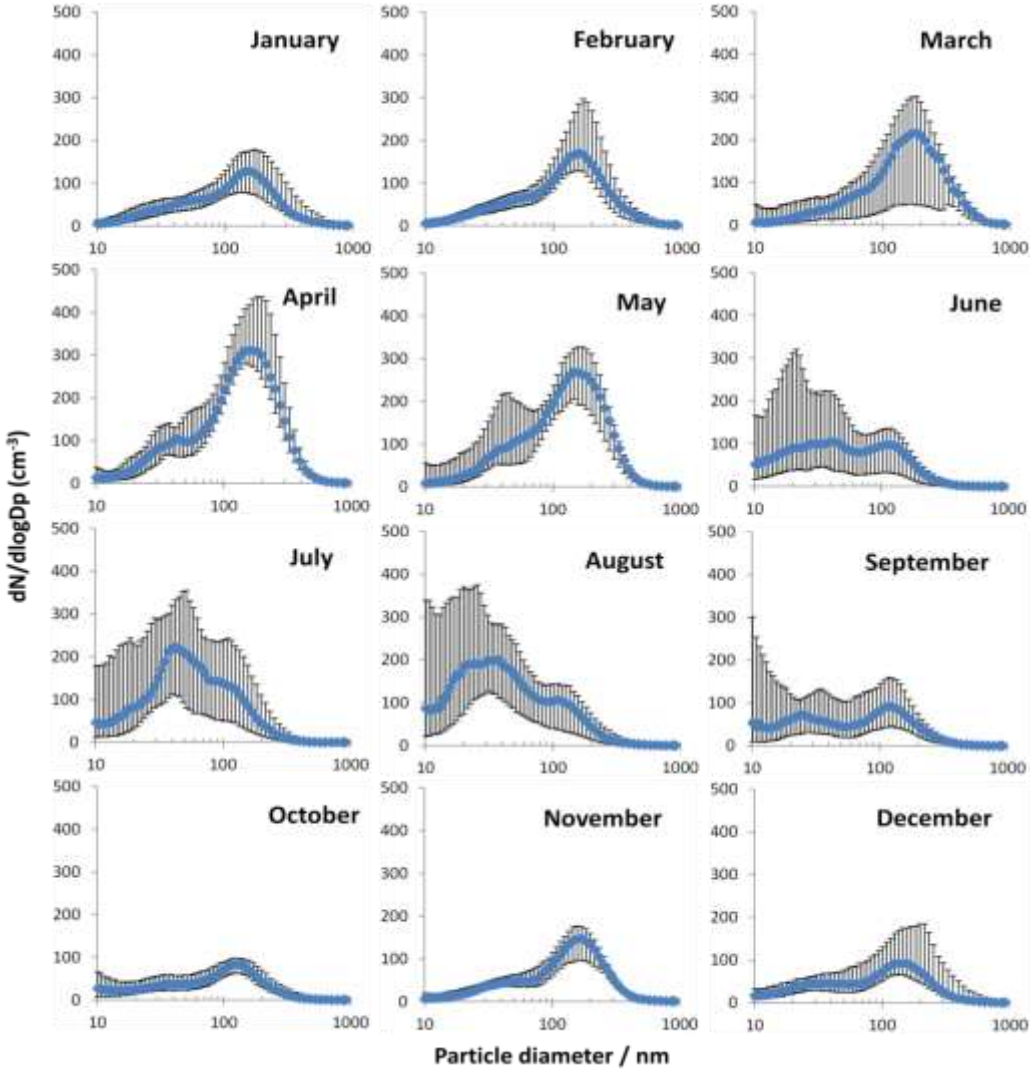


Figure 4 Aerosol size distributions at different times (0708, 1028, 1128, 1328, 1428, 1528, 1628, 1740 UTC) from inverted almucantar data at Ittoqqortoormiit (Scoresbysund) on 5 June 2013 at an altitude of 68 m.

Particle number size distributions in the submicrometer size range were measured at Villum Research Station (VRS) in Northeast Greenland at Station Nord using a Scanning Mobility Particle Sizer (SMPS). The measurements started in summer 2010 and are ongoing. In figure 5, the median particle number size distributions are displayed for each month in 2012.



Figur 5 Median monthly particle number size distributions (10 nm < particle diameter < 900 nm) measured at VRS in 2012

The particle number in the investigated size range, between 10 and 900 nm, reaches values up to a few hundred particles cm^{-3}. The shape of the particle number size distributions show a clear seasonal variation indicating different transportation patterns to the site and different emission patterns at VRS. A pronounced accumulation mode is observed during November – February and becomes more prominent from March to May representing the Arctic haze period at that site. This accumulation mode is expected to be linked to long-range transported air masses originating originated in Europe, Asia and North America. In contrast, the accumulation mode decreases in the summer months where an increased number of freshly nucleated particles are occurring on an irregular base. It is known that such nucleation events favourably occur in the presence of sunlight as the photo-oxidation of chemical species in the atmosphere enhances the concentration of available precursors that are needed for new particle formation. The nucleation events at VRS sometimes lasted several days.

5.3 Monitoring of CH_4

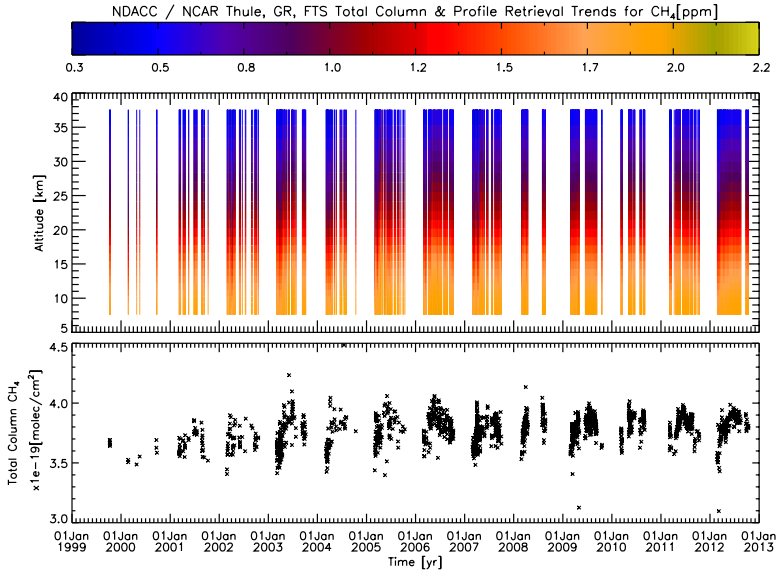
CH_4 is currently monitored at two stations in Greenland (table 1). On one station surface measurements are performed and on the other tropospheric as well as stratospheric profiles are performed.

Fourier Transform Infrared Spectroscopy (FTIR)

The basic principal behind FTIR is the following: incident radiation hits a beamsplitter where half of the light is reflected to a movable mirror and the other half is transmitted and hits a stationary mirror. When the two beams of light returns from the mirrors to the beamsplitter the light is recombined and directed towards a detector. By moving the movable mirror a so called interferogram is recorded consisting of the relative intensity versus optical path difference. By means of the matemathical method Fourier Transform the interferogram may be converted to a spectrum, ie the time domain interferogram is converted to a frequency domain spectrum.

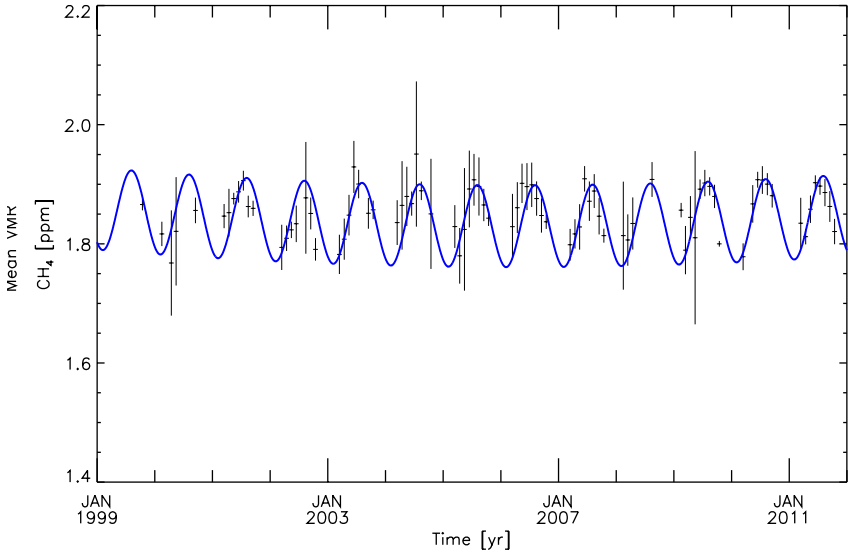
Examples of monitoring results

Figure 6 displays CH_4 vertical distributions as a function of time. The figure shows that the largest part of the CH_4 mass resides in the troposphere likely due to the existence of natural emission sources.



Figur 6 Vertical distribution of CH_4 (ppm) in the upper troposphere and stratosphere as a function of time from 1999-2913 (upper plot) along with total CH_4 column burden (molec cm^{-3}; lower plot). Measured at Pituffik (Thule).

The trend of tropospheric monthly mean CH_4 at Pituffik (Thule) may be calculated by fitting a sinusoidal finction to the data (figure 7). Sparse data early in the time series may skew the early trends, though an increase in the mid-2000's is evident and the linear trend at 1 Jan 2012 is calculated to be +1.05%/yr.



Figur 7 Monthly mean tropospheric CH_4 observations (ppm; horizontal lines) from Pituffik (Thule) with bars indicating ± 1 standard deviation. The blue line is a sinusoidal best fit.

At VRS measurements of both CH_4 and CO_2, using with cavity ring down spectroscopy, are carried out at several heights on a tall mast (3.6 m, 20 m, 40 m, 60 m and 80 m above ground level). The mixing ratio of CH_4 varies from 1.88 ppm to 1.93 ppm. It increases when the wind originates from the north, which is shown in figure 8, and a corresponding variation between CH_4, CO_2 and NO_x is found. The corresponding variations indicate a common source, which could be a distant and anthropogenic.

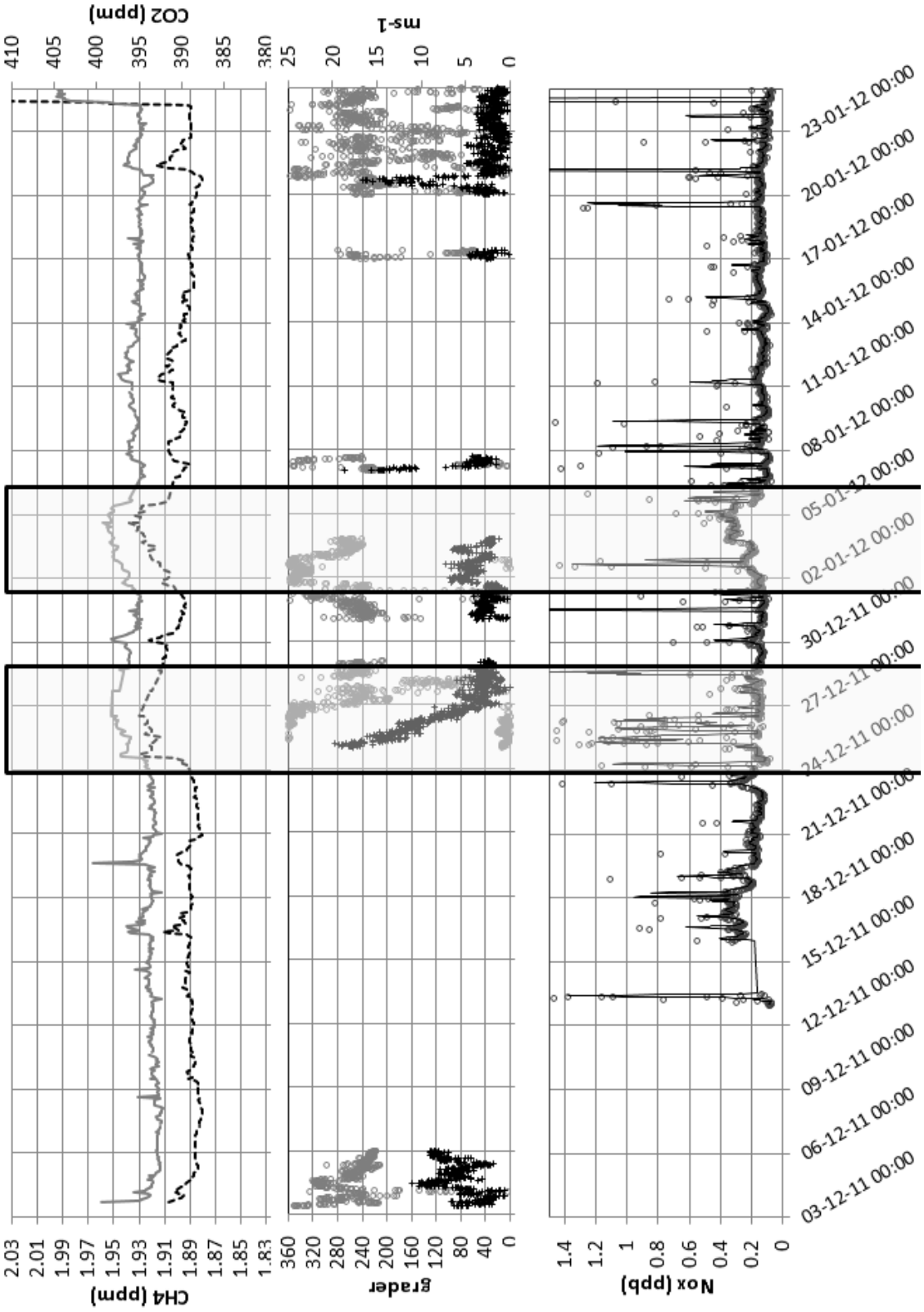


Figure 8 *The top figure shows the concentration of CH_4 (dotted line) and CO_2 in ppm. The figure in the middle shows the wind direction (o) and the wind speed (+) and the figure at the bottom shows the concentration of NOx (circles are half hour mean values and the line is running mean). Only data from December 2011 and January 2012 are shown.*

5.4 Usage of other data types in monitoring SLCPs

Ice core data

Ice core data has previously been used to evaluate BC amounts in connection with precipitation processes (*Painter et al., 2013*). The ability to extract information on BC or other SLCPs for historical periods almost up to current time enables interpretation of monitoring data in a wider perspective. For example *McConnell et al. (2007)* found that industrial emissions from 1850 to about 1950 caused a sevenfold increase in ice core BC concentration. Similarly, *Bauer et al. (2013)* found an eightfold BC concentration increase in the beginning of the twentieth century. Such information is also valuable in establishing general trends in tropospheric SLCP amounts.

Satellite data

The geographical coverage of the Danish monitoring stations is limited to Greenland and in particular there are no observations over sea. Valuable information for both policy relevant reporting and scientific understanding could be gained by utilising already available satellite observations. Polar orbiting satellites have densely packed tracks near the poles with more overpasses over a given point than at lower latitudes and often carry instruments suitable for characterizing tropospheric aerosols and trace gases. Early satellites with aerosol capabilities such as Terra (using the MISR instrument) observe aerosol optical thickness, however, newer satellites such as the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) (*http://www-calipso.larc.nasa.gov/*) (launched April 2006 and still functional March 2014), which is part of the A-train (*http://atrain.nasa.gov/*) satellite set have more advanced instruments for detection of aerosol and trace gases. By using lidar measurements in combination with analysis of the the Aeronet data CALIPSO distinguishes between six types of aerosol, where polluted continental, biomass burning, desert dust and polluted dust (mixture of dust, smoke and anthropogenic aerosol) aerosol are available over continental areas and corresponding classifications are available over marine areas. By combining the lidar measurements with spectral data from other satellite based instruments vertical profiles of aerosol concentrations and properties can be derived. Other satellites and instruments, such as the Moderate resolution Imaging Spectroradiometer on e.g. the Terra and Aqua satellites, are also available and more advanced instruments are planned for the future. The satellites do not only provide increased spatial coverage for benefitting monitoring, but also provide a convenient base for studying aerosol and trace gas processes, such as aerosol cloud interactions, when combining with satellite cloud observations or transport pathways into the Arctic.

6. Recommendations on the Danish SLCP monitoring network

Quantification of climate effects in the Arctic require a monitoring strategy which includes combined measurements of both surface and profile based monitoring of the most relevant SLCPs. Currently, only O_3 is monitored at both the surface and upper tropospheric levels and this is also the species with the best geographic coverage. BC is one of the central SLCPs due to its ability to absorb incoming shortwave radiation. Furthermore, it is well known that BC is involved in cloud processes, thus being further deposited on snow and ice covered surfaces

changing the surface albedo and leading to accelerated melting. Monitoring of BC with higher spatial coverage both at the surface and aloft is generally required to improve monitoring and the role of BC is Arctic climate change. An effective measure for BC monitoring including column based information would be to equip the Danish O_3 radiosondes with BC measurements. It is believed that it would be more valuable (in terms of scientific progress and cost benefit) to equip already existing measurement stations with instrumentation to monitor more relevant species than it would be to expand the geographical coverage of the measurements. This is because large geographical coverage is expensive and is most easily achieved through international collaborations such as NDACC Aeronet, AMAP or WMO-GAW. In addition to BC and ozone, other physico-chemical parameters of sub-micrometer aerosol are relevant to evaluate the fate and impact of long-range transported particles in the Arctic. Here, the scattering potential and the detailed chemical composition are key parameters to enhance our understanding of SLCPs and their role for climate in the Arctic.
For monitoring of SLCPs the synergy between surface and upper air measurements is obvious and it is here suggested to investigate and employ already existing unused measurement equipment. Furthermore, we recommend considering the potential in using satellite data to expand the data coverage. Finally it is important to have the needs of models in consideration both climate and chemical/physical transport models so data can support model validation and development.

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