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### Svalbard Science Forum



Kim Holmen and Jussi Paatero

The second Ny-Ålesund-Pallas Sodankylä
Atmospheric Researc Workshop
Ny-Ålesund, Svalbard, Norway

16-18 April 2007, Extended Abstracs



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# THE SECOND NY-ÅLESUND – PALLAS-SODANKYLÄ ATMOSPHERIC RESEARCH WORKSHOP NY-ÅLESUND, SVALBARD, NORWAY 16 – 18 APRIL 2007 – EXTENDED ABSTRACTS

KIM HOLMÉN AND JUSSI PAATERO

Norsk Polarinstitutt er Norges sentrale statsinstitusjon for kartlegging, miljøovervåking og forvaltningsrettet forskning i Arktis og Antarktis. Instituttet er faglig og strategisk rådgiver i miljøvernsaker i disse områdene og har forvaltningsmyndighet i norsk del av Antarktis.

The Norwegian Polar Institute is Norway's main institution for research, monitoring and topographic mapping in the Norwegian polar regions. The institute also advises Norwegian authorities on matters concerning polar environmental management.

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### TABLE OF CONTENTS

PREFACE	3
WORKSHOP PROGRAMME	5
LIST OF PARTICIPANTS	7
EXTENDED ABSTRACTS	8
Y. Viisanen et al.	9
J. Paatero et al	12
J. Hatakka et al	16
T. Laurila et al	18
J. Pulliainen et al.	20
V-M. Kerminen et al	22
A. Laaksonen et al	27
O. Meinander	28
O. Meinander et al.	31
U. Makkonen et al	36
S. Gagné et al.	40
H. Hakola et al	42
K. Pyy et al.	46
C. Ritter	50
R. Treffeisen et al.	54
T. Yamanouchi	55
A. Herber et al.	60
Y. J. Yoon et al.	61
A. Zinchenko	65
A. Zinchenko	66
A. M. Fjæraa	69
I Rurkhard at al	75

### THE SECOND NY-ÅLESUND – PALLAS-SODANKYLÄ ATMOSPHERIC RESEARCH WORKSHOP, NY-ÅLESUND, SVALBARD 16-18 APRIL 2007

#### **PREFACE**

In March 2004 an initiative was taken to communicate and stimulate the combination of the efforts in the two Global Atmospheric Watch stations in Ny-Ålesund, Svalbard and Pallas, Finland. This was pursued through a successful joint workshop held in Pallas-Sodankylä. It was agreed that such workshops should be repeated at regular intervals. The enhancement of the atmospheric observation programs in the Arctic is a high priority since there are few stations, and we know that the Arctic is particularly sensitive to climate change. Recently the international community has initiated an effort to build a sustainable Arctic observing network (SAON). SAON seeks to develop a set of recommendations on how to achieve long-term Arctic-wide observing activities that provide free, open and timely access to high quality data (obtained at the Earth's surface and from space) that will realize pan-Arctic and global value-added services and provide societal benefits. SAON promotes coordination, collaboration and communication among all parties to develop the recommendations and achieve a lasting legacy of International Polar Year 2007-2009.

In Svalbard The Research Council of Norway is pursuing the same coordination, collaboration and communication goals as put forth by SAON albeit for all science. Custodian of this pursuit under the auspices of the Research Council is Svalbard Science Forum (SSF). SSF organizes workshops to facilitate communication between scientists.

It was therefore timely to organize a workshop for atmospheric sciences in Ny-Ålesund. With support from SSF, The Finnish Meteorological Institute and the Norwegian Polar Institute the workshop attracted scientists from many of the active institutions in Ny-Ålesund and Pallas.

The present workshop report contains contributions from the attendees and provides a basis for continued discussions and cooperation. I want to thank all the attendees for their contributions and help in making this workshop successful. We are looking forward to the next meetings that our Finnish friends have offered to host in 2010. By then we expect SAON to have taken even stronger shape but also that the scientists that attended the present workshop have published many new papers stimulated by each other through the meetings and exchange.

I want to thank SSF, Kings Bay Ltd and above all the attendees for their valuable contributions that made this workshop possible.

Kim Holmén Norwegian Polar Institute The year 2007 was in many ways an exceptional year. The International Polar Year was started in March. The ice cover of the Arctic Ocean was at its record minimum in September (Comiso et al., GRL vol. 35, L01703). In Oslo in December the Intergovernmental Panel on Climate Change and the former U.S. vice-president Al Gore were awarded the Nobel Peace Prize "for their efforts to build up and disseminate greater knowledge about man-made climate change, and to lay the foundations for the measures that are needed to counteract such change". And in April The Second Ny-Ålesund – Pallas-Sodankylä Atmospheric Research Workshop took place at Ny-Ålesund, Svalbard, following the first workshop at Pallas, Finland, in 2004. Researchers from Norway, Finland, Sweden, Russia, Korea, Germany, and Japan gathered together to analyse, report and discuss various scientific questions concerning the atmosphere. We also had a possibility to visit the Mt. Zeppelin GAW Station offering a chance to see the technical solutions how to make atmospheric measurements in extreme environmental conditions. Last but not least, we had an opportunity to admire the magnificent view over Kongsfjorden both from the station and en route from the famous cable car that had "captured" the prime minister of Sweden somewhat earlier.

On behalf of the Finnish participants I would like to thank the workshop participants and local organizers, Norwegian Polar Institute, and the staff of Sverdrup Station and Kings Bay AS for the scientifically productive days, the excellent workshop arrengements, efficient logistical support and the warm hospitality at Ny-Ålesund. And I wish to meet all the members of the Ny-Ålesund atmospheric research community at Sodankylä, Finland, in the Third Ny-Ålesund – Pallas-Sodankylä Atmospheric Research Workshop in 2010.

Jussi Paatero Finnish Meteorological Institute

### WORKSHOP PROGRAMME

### **Monday 16 April**

14:30 – 14:40 Welcome, Goal of workshop. <u>K. Holmén</u> 14:40 – 15:00 The importance of long-term routine measurements. R. Treffeisen, R. Krejci, J. Ström, A.C. Engvall, A. Herber and L. Thomason 15:00 – 15:30 NIPR Atmospheric Science Activities in Svalbard, Arctic. T. Yamanouchi 15:30 – 15:50 Trace gas measurements at Pallas. J. Hatakka, M. Lallo and T. Laurila 15:50 – 16:20 Atmospheric observatory at Tiksi as a part of the IPY project IASOA. Y. Viisanen, J. Paatero, T. Uttal, T. Laurila, and H. Lihavainen 16:30 – 17:30 Dinner 18:00 Zeppelin visits for 9 - 15 persons

Tuesday 17 April		
08:30 - 08:50	Trends in light hydrocarbon (C2-C6) concentration measurements in background air in Finland. <u>H. Hakola</u> , T. Laurila and H. Hellén	
08:50 - 09:10	A summary of the most important halogenated greenhouse gases in the air at the Ny-Ålesund measuring station at Svalbard observed in the SOGE. <u>A.M. Fjæraa</u>	
09:10 - 09:30	Carbon dioxide and methane flux measurements in the GAW station of Pallas-Sodankylä. <u>T. Laurila</u> , J. Hatakka, M. Aurela, J-P. Tuovinen and Y. Viisanen	
09:30 – 10:00	Comparison and interpretation of the results of GHG measurements at Zappelin, Pallas, and Teriberka (Kola Peninsula, Russia). A.V. Zinchenko	
10:00 - 10:30	Coffee	
10:30 – 10:50	Total gaseous mercury in the air in southeastern Finland – First results. K. Pyy, J. Paatero and H. Hakola	
10:50 – 11:10	Seasonal variation of airborne lead-210 at Mt. Zeppelin GAW station, Svalbard, in 2001-2005. <u>J. Paatero</u> , J. Hatakka, M. Buyukay, K. Holmén and Y. Viisanen	

11:10 – 11:30 Studies of trace metal deposition in Lapland. <u>U. Makkonen</u>, J. Paatero,

11:30 – 11:50 The Korean atmospheric studies in Ny-Ålesund. Young Jun Yoon

K. Pyy and K. Stebel

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12:00 – 13:00	Lunch
13:00 – 13:20	Photo-oxidation of sulfur dioxide and aerosol nucleation in the atmosphere. <u>A. Laaksonen</u>
13:20 – 13:50	Ion-DMPS: a tool for atmospheric nucleation studies. S. Gagné, <u>L. Laakso</u> , V-M. Kerminen, T. Petäjä, P.P. Aalto and M. Kulmala
13:50 – 14:10	Tropospheric Lidar measurements at Spitsbergen. C. Ritter
14:10 – 14:40	Aerosol measurements at the Pallas GAW station. <u>V-M. Kerminen</u> , H. Lihavainen, M. Komppula, A. Hyvärinen, N. Kivekäs, V. Aaltonen and Y. Viisanen
14:40 – 15:00	About the Antarctic UV monitoring network and the snow UV-B albedo measurements in Sodankylä. <u>O. Meinander</u> , C. Torres, K. Lakkala, A. Redondas, et al.
15:00 – 15:30	Coffee
15:30 – 15:50	The Polar Aerosol Optical Depth Measurement Network. <u>A. Herber</u> , C. Tomasi, V. Vitale, R.S. Stone and T. Yamanouchi. (Presented by R. Treffeisen)
15:50 – 16:10	Sodankylä-Pallas Satellite Calibration and Validation Site: An Overview J. Pulliainen, E. Kyrö, T. Sukuvaara and R. Kivi
16:10	Zeppelin visits for 6 – 9 persons.

### Wednesday 18 April

- 08:00 08:20 Arctic smoke evaluation of an extreme air pollution event over Svalbard in the spring of 2006. <u>J.F. Burkhart</u>, A. Stohl, A.M. Fjæraa, C. Forster, C. Lunder, K. Tørseth, and K.E. Yttri
- 08:20-08:50 Shrews and contaminants at Pallas: background and research plans. <u>H. Henttonen</u>
- 08:50 09:10 Transport variations to Svalbard, implications for ecosystems regarding nutrients and contaminants. <u>K. Holmén</u>, A.C. Engvall, A. Hodson, I-T. Pedersen, G.W. Gabrielsen
- 09:10 09:30 Closing of workshop. K Holmén

### LIST OF PARTICIPANTS

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- 2. Jussi Paatero, Finnish Meteorological Institute (FMI), Finland
- 3. Juha Hatakka, Finnish Meteorological Institute (FMI), Finland
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- 20. Chris Lunder, Norwegian Institute for Air Research (NILU), Norway
- 21. John Burkhard, Norwegian Institute for Air Research (NILU), Norway
- 22. Christiane Hübner, Svalbard Science Forum (SSF), Norway
- 23. Kim Holmén, Norwegian Polar Institute (NPI), Norway

### **EXTENDED ABSTRACTS**

### ATMOSPHERIC OBSERVATORY AT TIKSI AS A PART OF THE IPY PROJECT IASOA

Yrjö Viisanen<sup>1</sup>, Jussi Paatero<sup>1</sup>, Taneil Uttal<sup>2</sup>, Tuomas Laurila<sup>1</sup>, Heikki Lihavainen<sup>1</sup> and Alexander Makshtas<sup>3</sup>

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### INTRODUCTION

The International Arctic Systems for Observing the Atmosphere (IASOA, www.IASOA.org) is an International Polar Year (IPY) Activity (#196) that was formally endorsed by the IPY committee in November 2005. IASOA is coordinated by the United States National Oceanic and Atmospheric Administration (NOAA). This activity seeks to enhance and integrate measurements of the Arctic atmosphere on an International scale by coordinating the activities of seven atmospheric observatories in Alaska, Canada, Greenland, Spitsbergen (Svalbard), Sweden, Finland and Russia. The IASOA key science questions focus on efforts to understand the processes driving Arctic weather and climate within the larger (marine, Cryosphere, biological and terrestrial) system; the primary activities involve combining the resources of intensive super-site observatories, distributed networks, campaigns and innovative technologies to provide a comprehensive Arctic observing system; and the primary mission is to understand change and the relative effects of natural and anthropogenic effects well enough for the global community to plan mitigation and adaptation strategies. A summary of the Finnish Meteorological Institute's (FMI) planned activities at Tiksi in Arctic Siberia, is presented in the following

### TIKSI OBSERVATORY

The NOAA with the support of the National Science Foundation (NSF) is currently upgrading the climate observatory near Tiksi, located close to the delta of Lena river in northern Siberia, in collaboration with the Russian authorities, e.g. Roshydromet and the government of the Republic of Sakha (Yakutia). The site is located on the shore of the Laptev Sea (71°35'N, 128°55'E). The research of the NOAA will focus mostly on climate processes involving the surface radiation balance as affected by clouds and aerosols, cloud microphysics, satellite validation, circulation patterns, snow and sea ice etc.

The work plan at Tiksi proposed by the FMI consists of four research topics of atmospheric observations which will compliment the other observations at the site. The FMI is planning to start research in cooperation with the other IASOA partners on four activities:

- Atmospheric concentrations of greenhouse gases, especially carbon dioxide and methane.
- Carbon dioxide and methane exchange between tundra ecosystems and the atmosphere,
- Aerosol properties relevant to radiative forcing, and
- Atmospheric concentrations of heavy metals and PAH compounds.

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### **OBJECTIVE**

Tiksi will efficiently fill the "black hole" of the atmospheric circumpolar monitoring activities in the vast area between northern Finland and Alaska. The site provides a unique opportunity to study the reasons and consequences of the melting permafrost during the warming of the Arctic Siberian climate.



**Fig. 1.** Location of Tiksi and other circumpolar atmospheric observatories (Map: UNEP/GRID Arendal).



Fig. 2. Candidate site for a clean air facility east of Roshydromet observatory at Tiksi.



Fig. 3. The town of Tiksi (population about 3000) 5 km northwest of the observatory.

### SEASONAL VARIATION OF AIRBORNE LEAD-210 AT Mt. ZEPPELIN GAW STATION, SVALBARD 2001-2005

Jussi Paatero<sup>1</sup>, Juha Hatakka<sup>1</sup>, Murat Buyukay<sup>1</sup>, Kim Holmén<sup>2</sup>, and Yrjö Viisanen<sup>1</sup>

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### INTRODUCTION

During the past three decades, there has been increasing interest in the presence of airborne pollutants in the Arctic region. However, the interpretation of the results has suffered from the lack of data concerning the transport and removal processes in the atmosphere.

In this project we have measured concentration of lead-210 in the air at Ny-Ålesund, Svalbard. The data on atmospheric <sup>210</sup>Pb can be used as a tracer to help to identify natural, e.g. due to the North Atlantic Oscillation (NAO), and anthropogenic variations in the transport behaviour of air masses and thus also air pollutants in the Arctic region.

Lead-210 is formed in the atmosphere from the radioactive noble gas radon-222 emanating from the Earth's crust. 99 % of the airborne <sup>222</sup>Rn originates from land and only 1 % from the sea (Baskaran et al., 1993). Owing to the long half-life (22 years) of <sup>210</sup>Pb, its removal from the atmosphere is governed by the different scavenging processes affecting the aerosol particles carrying it rather than radioactive decay.

### **MATERIALS AND METHODS**

The sampling site was at Mt. Zeppelin Global Atmosphere Watch (GAW) station, Ny-Ålesund, (78°58′ N, 11°53′ E), on the western coast of Spizbergen (Fig. 1), the largest island in the Svalbard archipelago (NILU, 2005; WMO, 2005). The station is located 474 m above sea level.

High-volume aerosol particle samples have been collected onto glass fibre filters (Munktell MGA). Three samples per week have been collected with filter changes on Mondays, Wednesdays, and Fridays. The air flow is about 3000 m<sup>3</sup> per day. One out of 25 filters is left unexposed and is used as a field blank sample. The sampling programme was started in December 2000.

The exposed filters together with field blanks were assayed for <sup>210</sup>Pb six months after the sampling with an automatic alpha/beta gas-flow proportional counter instrument (Mattsson et al., 1996). The measurement is based on the alpha counting of the in-grown daughter nuclide <sup>210</sup>Po. Usually the one sigma standard deviation of the radioassay varies between 5 and 10 per cent.

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Fig. 1. Location of the Mt. Zeppelin Global Atmosphere Watch (GAW) station.



Fig. 2. On the left the inlet of the filter sampler. The red heating cable prevents the blocking of the inlet by snow and ice.

### RESULTS AND DISCUSSION

The observed <sup>210</sup>Pb activity concentrations present a clear seasonal variation with highest concentrations in winter. This is attributed to the small amount of precipitation, reduced air chemistry and stagnant mixing conditions in the troposphere during the Arctic night. These factors increase the aerosol residence time and thus the accumulation of <sup>210</sup>Pb into the air. The phenomenon is similar to the Arctic haze, accumulation of soot and sulphate particles into the Arctic atmosphere during the winter. The maximum concentrations are quite comparable between the High Arctic and northern Finland (Paatero et. al., 2003; Paatero and Hatakka, 2000).

The minimum <sup>210</sup>Pb activity concentrations occur in the High Arctic in summer when the continuous solar radiation induce efficient vertical mixing of the troposhere. Also the amount of precipitation, which causes wet deposition, and atmospheric chemistry induced by solar radiation, are at their seasonal maximum. This is in agreement with observations in northern Finland. However, the concentrations are even lower in the High Arctic.

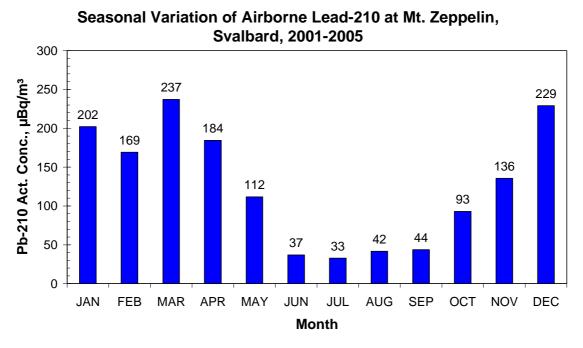


Fig. 3. Seasonal variation of airborne <sup>210</sup>Pb (μBq/m³) at Mt. Zeppelin GAW station, Svalbard 2001-2005.

### ACKNOWLEDGEMENTS

This work was started with the financial support of the Ny-Ålesund LSF Project, European Community – Access to Research Infrastructure action of the Improving Human Potential Programme. The authors are also indebted to Kings Bay AS for logistical support.

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WMO 2005. http://www.wmo.int.

### TRACE GAS MEASUREMENTS AT PALLAS

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### **INTRODUCTION**

The number of stations and components measured at Pallas continue to increase. A new wetland flux station, Lompolojänkkä, was established in 2005. At Sammaltunturi GAW station atmospheric hydrogen concentration measurements were started in 2006.

### **SAMMALTUNTURI**

The CO<sub>2</sub> measurements continue with a NDIR based system at Pallas. The system was updated to use LiCor-7000 analyser in 2006, and a new set of 6 cylinders of WMO/CCL standards were acquired in 2007. The whole time series for the station as daily means is depicted in fig. 1, showing ca. 2 ppm increase a year. Results agree quite well with flasks collected for NOAA's Cooperative Air Sampling Network since 2002, on the average FMI's results are 0.12 ppm lower (with FMI's scale based on standards from year 2000).

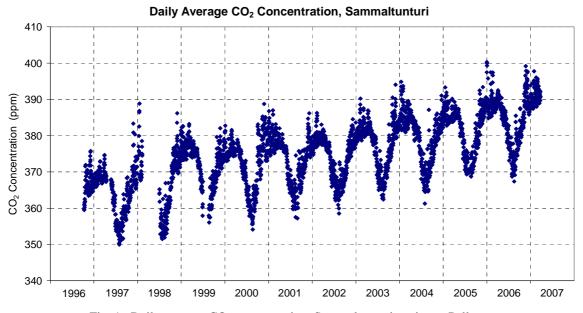


Fig. 1. Daily average CO<sub>2</sub> concentration, Sammaltunturi station at Pallas

The CH<sub>4</sub>, CO,  $N_2O$  and SF<sub>6</sub> measurement were started in 2004 at Sammaltunturi with a GC, and the system continues operating. A new set of 3 cylinders of WMO/CCL CH<sub>4</sub>/CO standards were obtained in 2005. Also SO<sub>2</sub>, NO<sub>X</sub> and ozone concentrations are measured at the station.

In connection with EUROHYDROS project a new instrument, Peak Laboratories Peak Performer 1, was installed in 2006 to measure atmospheric hydrogen concentration. It is based on RGD (Reduction Gas Detector), and one measurement takes 5 minutes to run. A working standard cylinder is measured as every third sample. A proper calibration for the system is still missing: working standard and instrument response is currently calibrated with a single 102 ppm (2 %) standard using a gas blender. This situation will improve during 2007.

### LOMPOLOJÄNKKÄ

Lompolojänkkä station lies in a small wetland (aapa mire) at an elevation of 270 m a.s.l., ca. 3 km NW from the forest flux site Kenttärova and 4 km NE from Sammaltunturi. Its measuring programme includes  $CO_2$ ,  $CH_4$  and ozone concentrations, and various hydro-meteorological parameters including e.g. air and soil temperatures, different radiation components and water table depth. The exchange of  $CO_2$  and  $CH_4$  between atmosphere and the ecosystem is measured continuously by eddy covariance (EC) method. Since 2006 the EC measurements have been complemented by flux measurements using automatic chambers. Lompolojänkkä mire is a level 3 NITROEUROPE site, and in 2007 the programme will be expanded to include  $N_2O$  concentration and flux measurements.

Hourly average methane concentrations from the Sammaltunturi and Lompolojänkkä stations are presented in fig. 2. Lompolojänkkä sample was taken at height 3 m above the ground (wetland). Wetland site concentrations reach much higher (ca. 4 ppm) values during summer if atmospheric mixing is weak, due to wetland being a source of methane. During wintertime the wetland site concentration follows much more closely to that measured at Sammaltunturi.

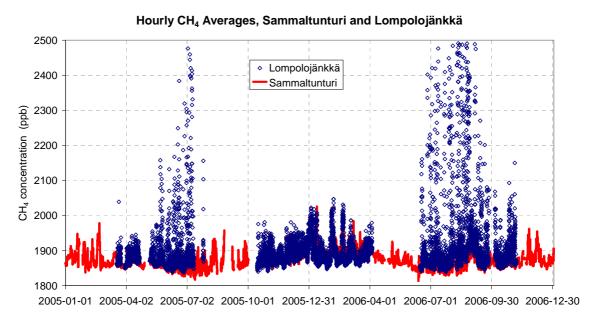


Fig. 2. Hourly CH<sub>4</sub> averages, Lompolojänkkä and Sammaltunturi

### CARBON DIOXIDE AND METHANE FLUX MEASUREMENTS AT THE GAW STATION OF PALLAS-SODANKYLÄ

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### **INTRODUCTION**

At the Finnish Pallas-Sodankylä GAW station, concentrations of trace gases, including carbon monoxide and methane, are measured on the top of an arctic mountain, Sammaltunturi. To improve our understanding how northern ecosystems act as sources and sinks of these trace gases and how they contribute to the concentration variations, we measure the biosphere-atmosphere exchange of these gases on an ecosystem scale using micrometeorological methods. Within the Pallas area, we run eddy-covariance measurements of CO<sub>2</sub> fluxes in a spruce forest at Kenttärova and CO<sub>2</sub> and CH<sub>4</sub> fluxes in a northern wetland (aapa mire). At the other node of the GAW station, Sodankylä, we measure CO<sub>2</sub> fluxes in a Scots pine forest. At all these sites, the micrometeorological flux measurement systems also record sensible heat and latent heat (evaporation) fluxes and a suite of meteorological parameters. In this presentation, we show examples of these fluxes together with trace gas concentrations at Sammaltunturi.

### **RESULTS**

The seasonal cycles of the tropospheric concentrations of CO<sub>2</sub> and CH<sub>4</sub> are most pronounced in the high northern latitudes. For CO<sub>2</sub> this is due to the terrestrial vegetation acting as a sink in summer and a source during the other seasons (Fig. 1). CH<sub>4</sub> concentrations are lowest in late summer, even though the CH<sub>4</sub> emissions from wetlands are highest in summer. This is due to the efficient photochemical sink in the atmosphere in summer.

The spruce forest of Kenttärova is a small source of CO<sub>2</sub> to the atmosphere in winter until late April, when air temperature exceeds 0 °C and photosynthesis commences (Figs. 1 and 2). The net uptake of carbon increases gradually in early summer in warm weather. The net fluxes to the forest are highest in July, about 0.4 mg CO<sub>2</sub> m<sup>-2</sup> s<sup>-1</sup> at noon. The concentrations of CO<sub>2</sub> begin to decline in late April, reaching the minimum in August. The nature of concentration variations is very different in winter and summer. In winter, the variations mainly reflect the long-range transport of CO<sub>2</sub> in changing air masses, while in summer the short-term diurnal variations constitute the dominant pattern.

In spring, global radiation provides an increasing source of heat to the surface (Fig. 2). First it is mostly converted to sensible heat flux to the atmosphere. After the snowmelt and commencement of photosynthesis, when stomatal gas exchange becomes effective, larger part of the available energy is used for evapotranspiration.

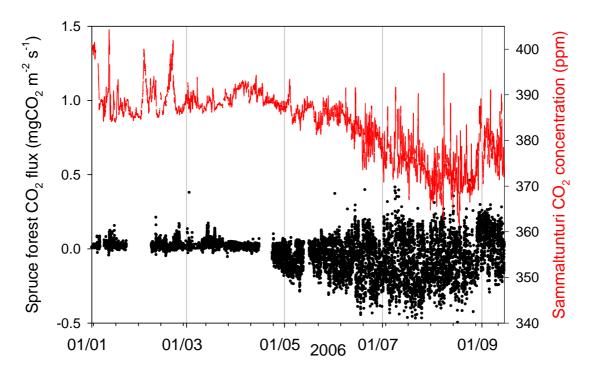


Fig. 1. Half-hourly CO<sub>2</sub> concentration at Sammaltunturi and CO<sub>2</sub> flux between the Kenttärova spruce forest and the atmosphere. Positive fluxes indicate a net CO<sub>2</sub> efflux from the forest.

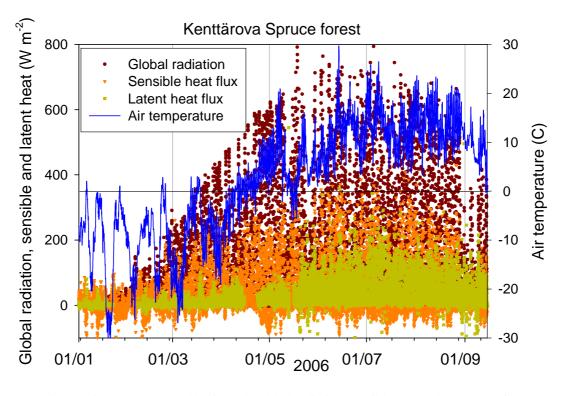


Fig. 2. Air temperature and half-hourly global radiation, sensible heat and latent heat fluxes at Kenttärova.

### SODANKYLÄ-PALLAS SATELLITE CALIBRATION AND VALIDATION SITE: AN OVERVIEW

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A major handicap in the utilization of Earth observation data has been the lack of reference and validation data with a high absolute accuracy and with a proper temporal and spatial coverage. The availability of *in situ* data has limited both the calibration of space-borne instruments and the development and validation of geo- and biophysical parameter retrieval algorithms. Moreover, the recent research indicates that in many cases the assimilation of satellite data with ground-based observations is a necessity in order to obtain accuracy characteristics required for operational or research end-use applications, such as climate trend analyses. Actually, the proper validation of satellite data retrieval algorithms can be considered as a continuous activity spanning over the lifetime of a single satellite mission and also covering the use of historical satellite data sets.

Dense ground-based observation networks monitoring atmospheric and surface environmental characteristics only exist in densely populated areas. However, in remote regions, such as arctic, sub-arctic and boreal zones of Eurasia and North America, the monitoring networks are typically sparse. In contrast to the general case, the Sodankylä-Pallas site is covered with dense weather, hydrological and environmental monitoring networks and specialized research stations. The Sodankylä-Pallas site is located in northern Finland north of the Arctic Circle and it is a good representative of boreal and sub-arctic Eurasian environment in a transition zone from marine to continental climate (a transition from marine to continental in the west to east direction). The site provides in situ monitoring and high spatial resolution land cover data sets that are not available for other regions north of the latitude of 60°. A special feature of the site is that it is the westernmost part of the Eurasian taiga belt that reaches close to the Pacific Ocean in its easternmost extent. As the Russian in situ environmental and climate monitoring network has declined since the early 90's, the Sodankylä-Pallas site provides data and a research infrastructure (available e.g. for measurement campaigns) that are not available elsewhere in that particular ecological and climate region.

The data sets available for the Sodankylä-Pallas region include the weather and atmospheric parameter monitoring data from the Finnish Meteorological Institute (FMI), land cover characteristics and hydrological monitoring and modelling data from the Finnish Environment Institute (SYKE), and selected data sets form other Finnish research institutes and universities. Intensive stations equipped with a large variety of atmospheric sampling, profiling and automatic surface parameter measurement systems are located near the town of Sodankylä (Arctic Research Centre of FMI with a permanent staff of around 30 persons), and at/in the vicinity of Pallas Mountain. Additional data sets are available from *in situ* and aerial monitoring campaigns.

The available data sets range from point-wise monitoring observations to regionally distributed information. The data sets are relevant for space-borne remote sensing instruments with a high or coarse spatial resolution, as well as for atmosphere or surface monitoring instruments. The available reference data also enables the analyses of mixed pixel effects that are highly relevant for the utilization of satellite observations with a coarse spatial resolution. Selected data sets from the Sodankylä-Pallas site are currently available from the web-service of FMI-ARC (<a href="http://fmiarc.fmi.fi">http://fmiarc.fmi.fi</a>). The data include e.g. vertical profiles of atmospheric constituents, such as ozone, columnar observations on various atmospheric characteristics, continuous *in situ* sampling of aerosol particles and *in situ* analyses of atmospheric gas composition and boreal forest canopy to atmosphere gas exchange characteristics. Also soil temperatures, soil moisture, leaf (needle) moisture, snow conditions (including snow depth) are continuously monitored.

The Sodankylä-Pallas satellite calibration and validation site is coordinated by the Arctic Research Centre of the Finnish Meteorological Institute (FMI-ARC) and the activities of the site are related e.g. to the Nordkalotten Satellite Evaluation cooperation Network (NorSEN), and to the Global Atmosphere Watch (GAW) network. NorSEN network operations are coordinated by the satellite data centre of FMI-ARC. Currently, FMI-ARC receives processes and delivers MODIS and OMI data from EOS Terra/Aqua satellites and EOS Aura satellite, respectively. During 2007 FMI-ARC starts the receiving, processing and archiving of near real-time UV-data from METOP, the polar orbiting satellite of EUMETSAT equipped e.g. with GOME2 ozone monitoring instrument (EUMETSAT O3SAF). EUMETSAT operations are now in the testing phase and a major METOP CAL-VAL campaign will be carried out in Sodankylä in summer 2007.

### AEROSOL MEASUREMENTS AT THE PALLAS GAW STATION

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### **BACKGROUND**

The Pallas GAW (Global Atmosphere Watch) station is located in the northern Finland and maintained by the Finnish Meteorological Institute (Hatakka et al., 2003). Aerosol measurements have been made principally at two sites in Pallas: Sammaltunturi (67°58'N, 24°07'E, 565 m above the sea level) and Matorova (68°00'N, 24°14'E, 340 m above the sea level). These two sites are located six kilometres apart from each other. The higher-altitude station, Sammaltunturi, is inside clouds during 10 % of the days, making it possible to conduct cloud microphysical measurements along with aerosol measurements (Komppula et al., 2005). The Matorova station is situated practically always below the cloud layer.

### MAIN FINDINGS

Aerosol particle number concentrations (diameter >10 nm) have been measured in Pallas since 1996 and particle number size distributions since 2000. Figure 1 shows a 10-year time series of measured total particle number concentrations. A clear seasonal cycle can be seen, with larger concentrations observed during the spring and summer (typically 1000-2000 particles cm<sup>-3</sup>) and substantially lower concentrations (down to a few tens particles cm<sup>-3</sup>) during the winter time. This pattern reminds somewhat that reported for Spitsbergen, in which the high particle number concentrations during the summer were ascribed to biogenic activities (Heintzenber and Leck, 1994). The seasonal cycle of Arctic haze, caused by the long-range transportation of anthropogenic pollution, is distinctively different with peak concentrations observed during the winter and early spring (e.g. Quinn et al., 2007).

The aerosol research in Pallas started from analyzing aerosol formation events. By such events we mean the nucleation of nanometer-size particles from precursor vapors in the atmosphere and their subsequent growth to larger sizes. Aerosol formation events are observed frequently in polar and Arctic air masses and they are taking place simultaneously at the two measurement sites in Pallas (Komppula et al., 2003a). The annual frequency distribution of the events was found to be bimodal, with the maxima in spring and autumn and slightly lower frequencies during the summer. Very few events are occurring during winter. Comparing our aerosol measurements to those made in

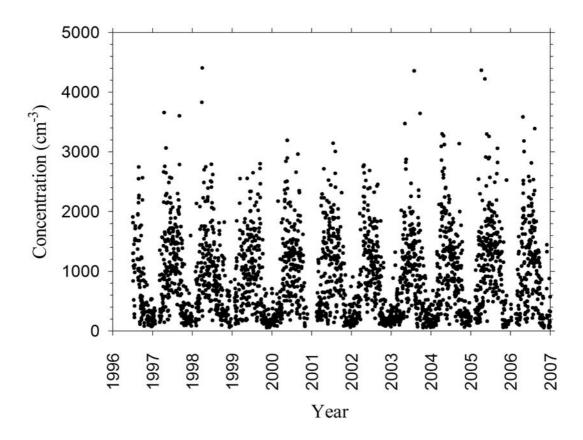


Fig. 1. Daily-average total particle number concentrations measured in Pallas.

Värriö (67°46'N, 29°35'E, 250 km from Pallas) revealed that aerosol formation covers in many cases a spatial scale of several hundreds kilometres in Northern Finland (Komppula et al. 2003b, 2006). A detailed analysis of the aerosol formations events at four Nordic stations showed many similarities, including the annual cycle of the events and the average particle growth rate of about 3 nm hour<sup>-1</sup> (Dal Maso et al., 2007). Compared with Northern Finland, events were found to be roughly twice more frequent in southern Finland and Sweden (about 50% of the classified days).

The above analysis suggests that atmospheric aerosol formation might influence the whole aerosol particle budget, and thereby aerosol climatic forcing, over the Nordic countries. By combining five years of aerosol measurement data from three stations (Pallas, Värriö and Hyytiälä), Tunved et al. (2006) showed that boreal forests in Northern Europe are able to maintain a relatively large natural aerosol particle population (1000-2000 particles cm<sup>-3</sup>) during the late spring to early fall period. These particles can be considered natural, since they seem to be formed via the oxidation and subsequent gas-to-particle conversion of terpenes emitted by the forests. The calculated mass increase of this natural particle population can be explained by the conversion of about 5-10% of the emitted terpenes into particulate matter (Tunved et al., 2006).

While particles formed in the atmosphere are probably too small to give a significant contribution to aerosol light scattering, they might act as cloud condensation nuclei (CCN) and modify thereby cloud properties. We made an investigation on this issue and found that atmospheric aerosol formation is, indeed, a potential source of new CCN over the Nordic countries (Lihavainen et al., 2003). In our later analysis, we found direct observational evidence that aerosol particles formed initially in the atmosphere may eventually participate into cloud droplet activation (Kerminen et al., 2005). The same analysis demonstrated that the radiative perturbation caused by the additional cloud droplets originating from atmospheric aerosol formation is large enough to warrant a further investigation of this issue.

Simultaneous measurements at Sammaltunturi and Matorova allow us to investigate size dependent activation of aerosol particles into cloud droplets during the periods when Sammalturi is inside clouds. By analyzing more than 40 individual cloud events, we found several associations between the aerosol population and corresponding cloud droplet population (Komppula et al., 2005). First, the average number concentration of cloud droplets increased with increasing aerosol particle number concentration (higher level of pollution), which is in line with the first indirect aerosol effect. Second, the fraction of activated aerosol particles was lower at higher pollution levels. Third, the effective activation diameter of particles increased with increasing level of pollution. In the cleanest air masses, the whole accumulation mode and a significant fraction of ulfrafine particles (<100 nm in diameter) were observed to activate into cloud droplets. This latter finding confirms that after their growth into sizes of 50-100 nm in diameter, aerosol particles formed in the atmosphere are able to modify cloud properties.

Our most recent measurements and analyses have demonstrates many interesting connections between atmospheric aerosol formation, aerosol number size distribution and clouds. For example, nanometer-size cluster ions that play a central role in aerosol formation were observed to be scavenged very effective by cloud droplets (Lihavainen et al., 2007). Furthermore, we showed that the cloud droplet number concentration can be related to the corresponding aerosol particle population using two very simple quantities: the total particle volume concentration and particle number-to-volume concentration ratio (Kivekäs et al., 2007).

We have also investigated aerosol optical properties in Pallas (Aaltonen et al., 2006). It was found that the aerosol scattering coefficient has a clear seasonal cycle with an autumn minimum and 4-5 times higher summer maximum. This is different from the seasonal cycle of the total particle number concentration, pointing toward different dominating sources for these two aerosol properties as measured in Pallas. A performed cluster analysis suggested that high values of the aerosol scattering coefficient were probably associated with anthropogenic sources in Russia, Eastern Europe, Great Britain and possibly Scandinavia. A comparison to simultaneously-measured aerosol number size distributions revealed that the scattering coefficient correlates strongly with the number concentration of accumulation mode particles. High nucleation mode particle number concentrations indicative of recent aerosol formation could only be observed in masses having a relatively low aerosol scattering coefficient.

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### PHOTO-OXIDATION OF SULFUR DIOXIDE AND AEROSOL NUCLEATION IN THE ATMOSPHERE

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Atmospheric nucleation is usually assumed to be controlled by sulfuric acid, whereas other molecular species possibly participating in the nucleation (such as water, ammonia and organics) are thought to mainly boost the nucleation rate, and to participate in the growth of the freshly formed particles. However, laboratory experiments first carried out already more than 25 years ago indicate that when SO<sub>2</sub>-water vapor mixture is irradiated with UV light, nucleation takes place at 3 orders of magnitude lower sulfuric acid concentrations than expected. These results, obtained by Friend et al. (*J. Phys. Chem.* 84, 2423-2436, 1980) were long overlooked. However, in 2005 Berndt et al. (*Science* 307, 698-700, 2005) obtained very similar results. Here we show that atmospheric nucleation rates recorded in Hyytiälä, Finland, during the 2003 QUEST field campaign, are in qualitatively good agreement with the laboratory experiments, and discuss the possible role of sulfur radicals in forming critical clusters.

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### SNOW UV-B ALBEDO MEASUREMENTS IN SODANKYLÄ

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### **INTRODUCTION**

Albedo of snow depends on the physical properties of snow, and varies according to wavelength. Albedo is often measured excluding the UV range. However, UV albedo of a snow covered surface is high and effects the downwelling radiation due to multiple reflections; important aspects for polar UV studies. Data of broadband albedo could also be empirically tranformed to UV albedo using simultaneous measurements. Therefore new snow UV-B albedo measurements were planned to be made in Sodankylä.

### MATERIALS AND METHODS

The measurements were started the 25<sup>th</sup> February 2007, including aspects of new and melting snow, as well as snow grain size. Two sensors of the UV-Biometer Model 501 from Solar Light Co. (SL501) with similar spectral and cosine responses were used, one placed upwards and the other downwards. SL-501 spectral response resembles the action spectrum for erythema, according to Commission Internationale de l'Eclairage (CIE). Wavelengths in the UV-B (280-310 nm) are most weighted. The data are logged at 1-minute-intervals. The intensity of the irradiance in Sodankylä in early spring is low (UVI<1), and the laboratory calibration of the sensors was found nonsatisfactory. New calibrated against each other, and also both of them against to SL-501 placed to the roof of the observatory. Using these calibration coefficients, snow UV albedo could be calculated. Both calibrations produced corresponding albedo results. The empirical calibration will be made again every two weeks until snow melt.



Fig. 1. UV-B albedo measurements in Sodankylä since 25<sup>th</sup> February 2007. (Photos Antti Aarva and Kaisa Lakkala).

### RESULTS AND DISCUSSION

As the calibration factors determined for other conditions produced albedo values of 150 %, the new empirical calibration factors were found satisfactory for the conditions (sun low and UVI<1). Using the roof sensor as a reference, the new calibration coefficients were determined (Fig 1).

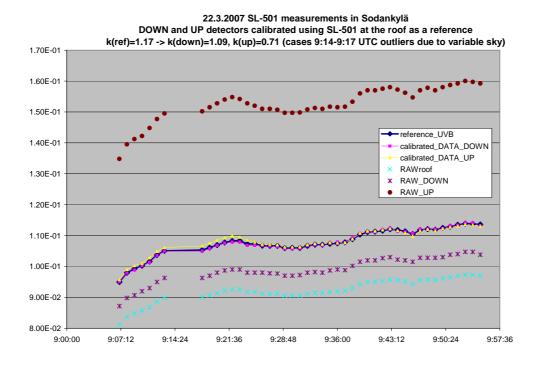


Fig. 1. The raw and calibrated signals of the SL-501 sensors, when both placed upwards side by side. Calibration using the third SL-501 sensor at the roof as a reference.

As the conversion of the raw signal into irradiance is linear, the calibration-factor-corrected raw data of upwelling and downwelling irradiance could be used for the calculation of the albedo. During a clear sky day (12.3.2007, Fig 2), the snow UV-B albedo (> 86 %) was found to have a low dependence on SZA (Fig 3).

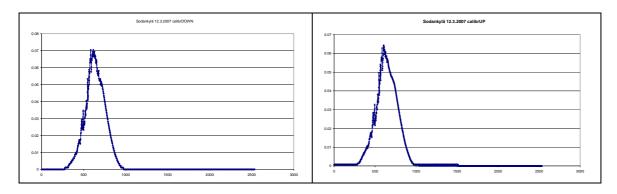


Fig. 2. The calibrated raw signals of the sensors a) upwards (left), and downwards (right).

#### Snow UV-B albedo 12.3.2007 Sodankylä (5 UTC - 15 UTC)

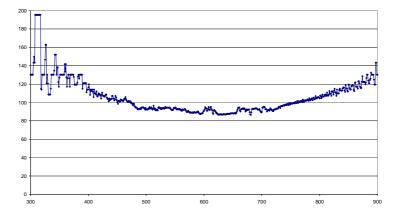


Fig. 3. The snow UV-B albedo in 12.3.2007 from 5 UTC to 15 UTC, with the minimum albedo of 86 %, indicated a low dependency on SZA.

Next more measured data will be analyzed, and data of new and melted snow as well as the grain size information provided by Anna Kontu, will be used, too. In addition it is planned to make some theoretical calculations on the albedo using a radiative transfer model Libradtran (Mayer and Kylling 2005) for the purpose. Snow albedo measurements at various scales (in a tower, in a helicopter) are planned for 2008, and multichannel as well as spectral measurement of snow albedo is planned to be carried out in 2008. A pilot project with the multichannel NILU-UV instrument (#102) is made in 2007, too.

### **ACKNOWLEDGEMENTS**

The snow UV-B albedo measurements would not have been possible without several people from FMI Kumpula and FMI Sodankylä. Thank you all. Especially I wish to thank Kaisa Lakkala and Antti Aarva, as well as Anna Kontu. The work is part of the FARPOCC (Finnish Antarctic Research Program on Climate Change) project financed by the Academy of Finland.

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### THE ANTARCTIC NILU-UV MONITORING NETWORK

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### **INTRODUCTION**

Severe ozone loss has been observed at high latitudes, especially above Antarctica, since early 1980's. This has led to a dramatic change in the natural cycle of the UV radiation penetrating the atmosphere. Surface ultraviolet radiation is related to climate change through changes in climate that affect the total ozone, cloud conditions, aerosols, as well as sea ice and snow cover.

In 1999, the Antarctic NILU-UV monitoring network was established by INM (Instituto Nacional de Meteorología, Spain) in collaboration with the FMI (Finnish Meteorological Institute), DNA-IAA (Dirección Nacional del Antártico – Instituto Antártico Argentino, Argentina) and CADIC (Centro Austral de Investigaciones Cientificas, Argentina).

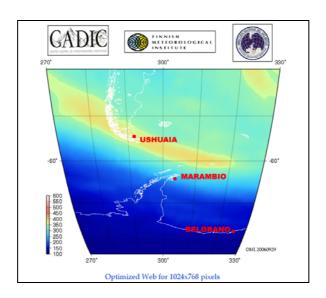


Fig. 1. The Antarctic NILU-UV network.

The network consists of three multichannel moderate bandwidth filter NILU-UV radiometers at Belgrano II (77.52S, 34.37W), Marambio (64.14S, 56.37W), and Ushuaia (54.48S, 68.19W). The radiometers are continuously lamp-calibrated every second week. The central wavelengths for the five NILU-UV channels are 305, 312, 320, 340 and 350 nm with 10 nm FWHM. Since 1999, the reference NILU-UV #008 of the FMI travels between Marambio and Ushuaia during the sunny period of the year (October - May). After about one-week measurements at one site, the reference travels to the next site, until two to three solar comparisons at both sites are made. Lamp tests are made before and after each solar comparison to study the stability of the #008. At the end of the seasonal measurement period, the #008 returns to FMI for maintenance. Absolute calibration of each channel has been performed at least every second year by

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the manufacturer (NILU Products As, Norway) using methods described in Dahlback (1996). For the Belgrano station, the reference NILU-UV #023 of the INM has been applied. The calibration reference instrument is the SUV spectrometer of the National Science Foundation (NSF) at Ushuaia, whereby the results are directly linked into the NSF irradiance scale.

Here, the different methods to calculate the calibration coefficients are presented and discussed. Thereafter, results of the calibrated data as well as these data used for applications are given.

### MATERIALS AND METHODS

In addition to absolute calibration, producing calibration coefficients  $a_i$  (for more details see Dahlback 1996), we have used three different kinds of methods for the calibration of the Antarctic data. In the first method, by using the SUV radiometer NSF scale data, the calibration factors CF for Marambio and Ushuaia have been calculated (Lakkala et al. 2002) for each solar comparison as a three minutes average (t-1:t+1) of the ratio of the traveling reference (ref = #008) and the site NILU-UV (i = Marambio #011 or Ushuaia #012) for CIE (Commission Internationale de l'Eclairage) weighted UV dose rates or UV-B or UV-A doserates

$$CF = [ave(UVref, t-1:t+1)] / [ave(UVi, t-1:t+1)]$$
 (1)

The major benefit of this method is that it is easy to compute using the software we have been provided by the manufacturer. How ever, a drawback is that these calibration coefficients are variable according to SZA etc., as doserates are calculated prior to calculating calibration coefficients.

In the second method, the lamp test data is used to correct and calibrate the site instruments. For this purpose, the method described earlier in Torres et al. 2002 has been applied. The regular lamp test data performed every two weeks for the site instruments is used to calculate a correction polynomial for the data. The reference scale is the initial calibration performed by the manufacturer with all the instruments of the network at Izaña in 1999. The lamp test results are used to calculate a correction polynomial for the data, with the aim to maintain the three instruments at the same relative scale. The method tracks very well the relative changes of the channels giving good ozone calculations and promising results of CIE (Torres et al 2002).

The third way to calibrate lies on the fact that as the traveling reference #008, the #011 at Marambio and the #012 at Ushuaia, have filters originating from the same big filter batches, we can directly utilize the measured raw voltage values channel by channel to calculate channel specific calibration coefficients C prior to calculating any products (Meinander et al. 2003):

$$C_i = V_i / v_i \tag{2}$$

where i = channels from 1 to 5, V = raw voltage signal for the reference, and v = raw voltage signal for the NILU-UV to be calibrated. These calibration coefficients are more stable throughout the day than those calculated using dose rates. After this, the CIE and UVA and UVB calculations are performed to the data of the site instruments.

### RESULTS AND DISCUSSION

The results of the sky comparisons show that the drift of individual channels may be variable, and different from one channel to another. When only the first (1999) and the year 2006 intercomparison results of the values of  $C_i$  (=  $V_i$  /  $v_i$ ) are compared, the changes for Marambio are found to be as follows: -15 % for channel 1, -21 % for channel 2, -33% for channel 3, -61 % for channel 4, +4 % for channel 5, and +2 % for channel 6. In Ushuaia, the changes from 1999 to 2006 have been: -9 % for channel 1, -21.4 % for channel 2, -34.5 % for channel 3, -60.0 % for channel 4, 2.7 % for channel 5, -5.6 % for channel 6. In both stations, the channels 5 (350 nm) and 6 (PAR) have been the most stable ones, and channel 4 (340 nm) has suffered the biggest drift. These results on the calibration coefficients only indicate the changes of the raw channel values, on the basis of which the final results of integrated UV-A, UV-B, and erythemal irradiance are calculated with the help of the instrument specific absolute calibration factors. The effect of these changes has successfully been corrected from our results (Lakkala et al. 2005), but in case of increasing differences, changing of the filters has to considered, too.

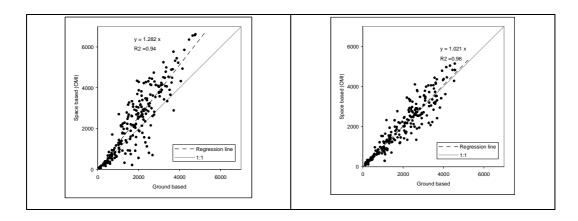


Fig. 2. Marambio (left) and Ushuaia (right) 2005/2006 erythemally weighted daily UV doses [J/m²], measured doses versus OMI data (adapted from Meinander et al. 2006).

Using lamp calibration data, the CIE daily dose results 2005/2006 have also been compared against OMI satellite UV data, as presented in Meinander et al. (2006), Figure 2. The agreement in Ushuaia was better than in Marambio, revealing possible problems in the OMI UV algorithm (albedo) and in separating snow and clouds, for example. Recently, the online data of the Antarctic network became available on the web, <a href="http://polarvortex.dyndns.org">http://polarvortex.dyndns.org</a>. The data were also used for the first time in the WMO Antarctic Ozone Bulletin 6/2007 (Figure 3). The Antarctic UV network work is also part of the IPY Oracle-O3 project (<a href="https://www.awi-potsdam.de/www-pot/atmo/ORACLE-O3">www.awi-potsdam.de/www-pot/atmo/ORACLE-O3</a>). Our future work includes homogenous processing and analysis of the whole data set since 1999, and utilizing these data for several publications now under preparation.

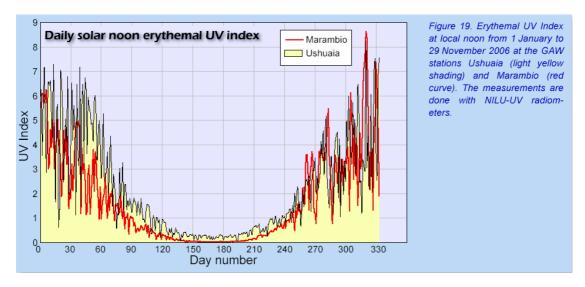


Fig. 3. The Antarctic NILU-UV data now available at <a href="http://polarvortex.dyndns.org">http://polarvortex.dyndns.org</a> were used for the first time for the WMO Antarctic Ozone Bulletin 6/2007.

### **ACKNOWLEDGEMENTS**

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# STUDIES OF TRACE METAL DEPOSITION IN LAPLAND

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# **INTRODUCTION**

Deposition of the trace elements has been studied at the Pallas GAW station, northern Finland, since 1995. In general, in Finland the amount of trace elements in precipitation decrease towards the north and the concentrations in Finnish Lapland are low. The most important source of heavy metals in the north is the mining and metallurgical industry at Kola peninsula. To study the environmental effects of the metal smelter at Nikel, northwestern Russia, a Finnish-Norwegian-Russian monitoring project at the Paz (Pasvik) River water shed was launched in 2003. As a part of this project the amount of metals deposited in the area was determined.

# **EXPERIMENTAL**

The precipitation samples were collected at Pallas, Matorova site, in Western Lapland, at Kevo in the northernmost Lapland, at Sevettijärvi in Eastern Lapland, and at Svanvik close to the Norwegian-Russian border (Figures 1 and 2). The samples were collected monthly in two parallel bulk collectors and acidified with nitric acid. The concentrations of trace elements (Al, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, V and Zn) were determined with inductively-coupled plasma – mass spectrometry.



Fig. 1. Map of the study area.



Fig. 2. Sevettijärvi air quality monitoring station.

# **RESULTS AND DISCUSSION**

The copper and nickel deposition is decreasing from east to west in the study area (Figure 3). The deposition values of metals related to the operations of the metallurgical industry at Nikel decrease two orders of magnitude within a range of 120 km.

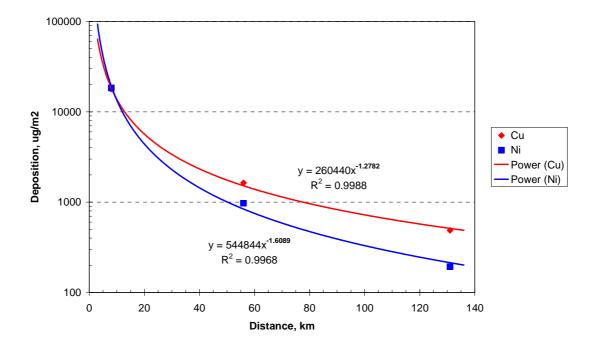


Fig. 3. The amount of copper and nickel deposition as a function of the distance from Nikel westwards.

The deposition of copper, nickel, lead and zinc are presented in Figure 4 and Table 1. The annual deposition values of nickel and copper are several times higher at Sevettijärvi compared to Kevo. On the other hand the deposition values of metals like lead and zink are rather similar at all sampling sites. These metals represent long-range transport from more remote industrial and populated regions. It is also evident that the smelter at Nikel emits considerable amounts of arsenic into the atmosphere.

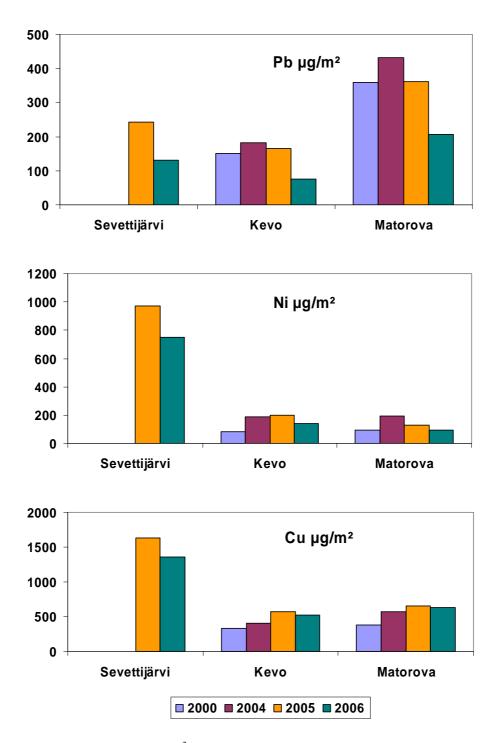


Fig. 4. Annual deposition (μg/m²) of lead, nickel and copper at Sevettijärvi, Kevo and Matorova.

Table 1. Deposition of copper, nickel, lead, zinc and arsenic at the studied sites in 2005.

						-
station	precipitation	Cu	Ni	Pb	Zn	As
	mm	μg/m²	μg/m²	μg/m²	μg/m²	μg/m²
Svanvik	421	24373	23244	774	2205	723
Sevettijärvi	356	1631	971	242	2208	70
Kevo	408	570	198	165	690	28
Matorova	648	650	129	362	1600	47

# **ACKNOWLEDGEMENTS**

The financial support of EU/Interreg IIIA Kolarctic fund managed by the Regional Council of Lapland is gratefully acknowledged.

# INVESTIGATING ION-INDUCED NUCLEATION WITH THE ION-DMPS

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Keywords: Particle formation and growth, Field measurements, Ion-induced nucleation, Ion-DMPS

Aerosol particle formation takes place frequently in the atmosphere (Kulmala et al., 2004). Despite great efforts, the exact nucleation mechanisms are not well known. Ion-induced nucleation is among the proposed mechanisms.

Information on the importance of neutral/ion-induced nucleation can be obtained with a special instrumental set-up called ion-DMPS (Laakso et al., 2007), which measures the charging state of atmospheric aerosol particles.

Charging state is the number of charged particles in a given sample divided by the measured number of particles corresponding to the steady-state charge distribution (equilibrium). In ion-DMPS this ratio is obtained with a neutralizer which can be switched either on or off. The size-range of our instrument is from 3 nm to 15 nm. The data presented here is from over one year measurement in Hyytiälä, Finland (Boreal forest).

Recently, Kerminen et al., (2007) developed a new method to calculate the contribution of ion-induced nucleation from the ion-DMPS-measurements. The resulting, simplified equation which can be used in the analysis has a following form:

$$S(d_p) = 1 - \frac{1}{kd_p} + \frac{(S_0 - 1)kd_0 + 1}{kd_p} \exp(-k(d_p - d_0))$$

where  $S(d^p)$  and  $S_0$  are the charging states at sizes  $d_p$  and  $d_0$ , where  $d_0$  is the diameter at which the new particle form. The parameter k is defined as:

$$k = \frac{\alpha N_{\pm}^{C}}{GR},$$

where  $\alpha$  is the recombination coefficient,  $N_{\pm}^{c}$  is the number of cluster ions and GR is the growth rate of particles. By fitting the equation to the ion-DMPS-data, we can estimate the contribution of ion-induced nucleation on total new particle formation rate.

Median initial charging state S0 at 1.5 nm during new particle formation events observed during one year with the ion-DMPS yielded values of 5.8 and 6.1 for negative and positive particles, respectively. Thus, in median, slightly less than 10% of the particle flux at this size resulted from ion-induced nucleation. However, the annual variation was large and in some cases the fraction reached slightly less than 40%.

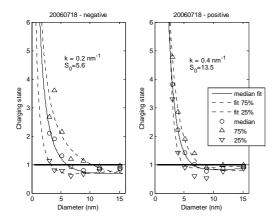


Fig. 1. Charging state as a function of diameter together with fitted values on the earlier equation.

Erkki Siivola and Pekka Pihkaka are acknowledged for his work on the Ion-DMPS. Veijo Hiltunen and Heikki Laakso are acknowledged for their efforts maintaining the instrument during the field measurements. This work was supported by Biosphere-Aerosol-Cloud-Climate Interactions (BACCI).

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# TRENDS IN LIGHT HYDROCARBON (C<sub>2</sub>-C<sub>6</sub>) CONCENTRATION MEASUREMENTS IN BACKGROUND AIR IN FINLAND

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# **INTRODUCTION**

Light molecular weight hydrocarbons (C<sub>2</sub>-C<sub>6</sub>) are common compounds in the atmosphere. The main source of light hydrocarbons is the usage of fossil fuel. They are emitted into the air as a result of petrol exhaust and evaporation, stationary combustion, gas leaks, solvent use, etc. Some light hydrocarbons also have natural sources (e.g. isoprene, ethene, propene). In the atmosphere, reaction with the hydroxyl radical is the main sink reaction for all hydrocarbons, but reaction with ozone is also an important sink for alkenes (Atkinson 1994). The reaction with hydroxyl radicals can result in ozone production when enough nitrogen oxide is present. Ozone formation takes place on various space and time scales. The fast-reacting compounds react close to their source areas, resulting in high ozone concentrations downwind, but slowly-reacting compounds can be transported far, particularly in winter when the amount of light, and hence hydroxyl radicals, is limited. During winter the VOCs accumulate in the northern latitudes, their concentration reaches a maximum during January-February and starts declining during spring when days get longer. Ozone formation can then occur in clean areas, and high ozone concentrations have been measured at high latitudes during spring (Laurila, 1999). Ozone concentrations have increased in Finland in the years 1990-2000, although a decreasing trend is observed in the other Nordic countries (Laurila et al., 2004; Solberg et al., 2005). The present study describes the concentration changes of non-methane hydrocarbons (C2-C6) at two background stations in Finland, Pallas and Utö, in the years 1994-2006. The trends have earlier been studied by Hakola et al., 2006.

# **EXPERIMENTAL**

The air samples are collected into evacuated stainless steel canisters (0.85 l) twice a week, two canisters at a time, using a Teflon membrane pump.  $C_2$ - $C_6$  hydrocarbons are analyzed from the canister samples using a gas chromatograph equipped with a flame ionization detector (FID) and an  $Al_2O_3/KCl$  PLOT column (50 m, i.d. 0.32 mm). Prior to analysis, samples are passed through a stainless steel tube (10cm\*1/4") filled with  $K_2CO_3$  and NaOH in order to dry them. Air samples are concentrated in two liquid nitrogen traps. The first trap is a stainless steel loop (1/8"\*125cm) filled with glass beads, while the other one is a capillary trap. Calibration is performed using a gas-phase standard from the NPL (National Physical Laboratory, UK) including 30 hydrocarbons at concentration levels of 1-10 ppb. A calibration standard and a blank sample are run about once a month. The detection limits for individual compounds vary from 3 to 11 ppt. The precision of the analysis was evaluated by sampling two canisters each time, analyzing both of them and calculating the standard deviation of the two analyses. The mean standard deviation varies from 4% (ethane) to 33 % (i-hexane), being less than 20 % for most of the compounds.

The linear trends in the VOC concentrations were estimated by using the nonparametric Mann-Kendall significance test for the existence of the trend together with the nonparametric Sen's method for the magnitude of the trend (Gilbert, 1987; Salmi et al., 2002). With Mann-Kendall we test the null hypothesis of no trend against the alternative hypothesis that there is an increasing or decreasing monotonic trend as described in Salmi et al. (2002). The significance level of 0.001 means that there is a 0.1% probability that the observation values are from a random distribution and with that probability a mistake is made when rejecting a hypothesis of no trend.

# **RESULTS**

During the course of the thierteen years of VOC measurements, VOC emissions have decreased in Europe. According to the emission data reported to the Convention on Long-Range Transboundary Air Pollution (CLRTAP), non-methane hydrocarbon emissions have decreased 35 % between the years 1988 and 2001 in Europe (Vestreng, 2003). A decreasing trend in the ambient concentrations would also be expected and it has been observed in many locations in Central Europe. For example at two EMEP sites, Waldhof and Košetice, the median VOC concentrations have decreased about 20-50% during the years 1993-2000 (Solberg et al., 2002). In the same report no trend was found for the Finnish sites Utö and Pallas. Also, in the Netherlands, compounds emitted mainly from traffic (acetylene and ethene) showed a downward trend of about 50% during the years 1981-1999 (Roemer et al., 2001).

Table 1 shows the Sen's slope estimates together with the significances and the changes as percentages. Most of the observed trends are not significant due to the limited amount of data.

Ethane concentrations at both sites seem to be increasing, 6 % at Pallas and 5 % on Utö over 13 years. The main sources are related to energy production and particularly natural gas production and use (Hellén et al., 2006). The positive trend is observed both in summer and in winter.

Utö data shows that concentrations of butanes, pentanes, and ethyne are decreasing by one third during the past decade. The decrease of the concentrations of the more reactive compounds is a result of the emission reductions, this decline is of the same magnitude as reported emission reductions of traffic which is the main source of these compounds to the atmosphere. The Pallas data show more variable trends of these compounds especially in summer that may be due to low concentrations and thus greater uncertainty of measurements.

Benzene concentrations have decreased on Utö due to catalytic converters and also due to lower benzene content in the fuel, but not clearly at Pallas. Wood combustion has been estimated to be the main local source of benzene in Finland (Hellén et al., 2006) and therefore a decline of the benzene concentration would not be expected.

Propene and ethene are also important constituents from wood combustion, but it is unlikely that the wood burning alone could explain the significant increases in propene (77%) and ethene (11%) concentrations at Pallas. The increase is most significant in summer. The increase could be at least partly due to biogenic sources since also isoprene concentrations have increased. On Utö ethene concentrations have been decreasing while propene has been increasing particularly in summer. The only

declining trend is observed at Utö in winter where we may expect anthropogenic influences being of dominant.

In summary, the trends of anthropogenic species which have long lifetimes in the atmosphere seem to be increasing in remote sites, but more reactive ones show decreasing trends which are roughly in accordance with emission reductions from traffic sources. This reduction of concentrations is more evident at the Baltic Sea site which is closer to main European emission areas. The data suggest that propene and ethene concentrations during summer are increasing, probably due to biogenic emissions because increase occurs during summer and traffic related species which have similar lifetimes are decreasing.

The most significant uncertainties of this analysis are the relatively short observation period to reach statistically significant trends and the long-term stability of laboratory analyses at these very low concentration levels.

*Table 1*: The Sen's estimator for the true slope of linear trend. The significance of a trend is implicated by Mann-Kendall test (\*\*\* 0.001, \*\*0.01, \*0.05 and +0.1 level of significance). The relative total change is indicated as a percentage relative to the first year.

	Pallas	Pallas	Pallas	Pallas	Pallas	Pallas
	year	year	summer	summer	winter	winter
	Sen's	R (%)	Sen's	R (%)	Sen's	R (%)
Ethane	7.35	6	7.88	13	32.5	19
Ethene	1.66	11	** 3.21	67	4.69	14
Propane	-3.91	-7	+ 2.93	32	5.2	5
Propene	** 1.46	77	* 1.29	68	1.12	47
Ethyne	+ -5.03	-16	0.88	14	-4.39	-8
i-Butane	1.39	14	0.71	60	3.67	18
n-Butane	-3.4	-16	-0.15	-5	-6.43	-15
i-Pentane	-0.17	-2	+ 1.32	167	-2.1	-12
n-Pentane	-1.26	-20	* 0.12	9	-1.98	-15
2/3-Methyl-						
pentane	0.14	6	+ 0.56	1306	-0.68	-12
n-Hexane	-0.37	-20	+ 0.22	167	-0.78	-18
isoprene	* 1.03	168	2.19	97	0.01	
benzene	-0.37	-4	0.72	29	1.4	9

	Utö	Utö	Utö	Utö	Utö	Utö
	year	year	summer	summer	winter	winter
	Sen's	R (%)	Sen's	R (%)	Sen's	R (%)
Ethane	6.05	5	10.26	15	25.78	14
Ethene	-10.38	-27	-0.52	-4	-11.13	-17
Propane	-6.02	-10	-2.02	-11	-0.87	-1
Propene	0.05	1	0.92	31	-0.67	-9
Ethyne	*-12.44	-26	**-3.32	-23	-21.56	-28
i-Butane	-1.64	-12	-0.69	-16	1.78	7
n-Butane	*-10.11	-34	+ -4.14	-48	*-18.98	-34
i-Pentane	-2.77	-22	-1.77	-30	*-8.33	-33
n-Pentane	-2.43	-27	-1.04	-25	-3.35	-20
2/3-Methyl-						
pentane	-0.93	-22	0.07	5	-1.71	-22
n-Hexane	-0.83	-28	-0.43	-30	-1.27	-24
isoprene						
benzene	-0.991	-8	+ -3.15	-41	-2.384	-11

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# TOTAL GASEOUS MERCURY IN THE AIR IN SOUTH-EASTERN FINLAND - FIRST RESULTS

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# INTRODUCTION

Extensive knowledge of mercury concentrations in Finland is not available at the moment. Until now, the only recent mercury measurements have been conducted at Pallas by Swedish Environmental Research Institute IVL. These samples have been collected manually twice a week for 24 h each. Knowledge of short term variation in the total gaseous mercury (TGM) concentration in Finland is lacking. In the near future, background measurements of short time resolution TGM will be performed at three stations with different surroundings. These stations are Pallas (67°58'N, 24°07'E), Virolahti (60°32'N, 27°41'E) and Ähtäri (62°35'N, 24°12'E). At these locations, precipitation samples will also be collected and analysed in the laboratory. These measurements fulfill the monitoring requirements of the EU air quality directives.

In this study, the first results from a south-eastern station Virolahti are presented. A special emphasis is given for the occasions where elevated levels of TGM have been observed. These events have been found to be due to influence of anthropogenic sources (air masses coming from highly industrialized areas) as well as natural sources (forest fires).

# **EXPERIMENTAL**

The Virolahti site is an EMEP station situated near the Russian border in south-eastern Finland. The station is located a few kilometres from the sea. Land use surrounding the site is agricultural with areas of fragmented forest. The measurements were started in June 2006 and have been conducted since with the exception of short breaks due to power failures or maintenance operations.

TGM measurements were performed with an automated instrument, Tekran 2537A. This monitor is based on CVAFS and it enables continuous data acquisition through the use of two gold cartridges in turn. The sampling resolution was 5 min and sample flow 1.5 l min<sup>-1</sup>. The sampling line was made of Teflon. A 0.2 µm PTFE filter (diameter 47 mm) protected the cartridges against contamination by particulate matter. The instrument was calibrated every 25 h using an internal permeation source.



Fig. 1. EMEP air quality monitoring station at Virolahti, southeastern Finland.

# **RESULTS AND DISCUSSION**

TGM concentrations at Virolahti site during the studied period July-December 2006 are shown in Fig.2. The values are presented as 24 h averages. The average concentration is 1.35 ng m<sup>-3</sup>. This value agrees with the background concentrations measured earlier in northern Europe<sup>1</sup> although is slightly low. This might be due to subtle passivation of the sampling cartridges. Since 1996, TGM has also been measured at the Pallas background station in northern Finland in cooperation with the Swedish Environmental Research Institute IVL. The range of the annual average at Pallas was 1.30-1.50 ng m<sup>-3</sup> in 1996-2004 (data: Ingvar Wängberg, IVL, Sweden).

The highest concentrations were observed in mid-August when the air masses were coming from east. During that time, some extensive forest fires were occurring in Russia near the Finnish border. The 5-min. concentrations exceeded 3 ng m<sup>-3</sup> several times while the hourly averages just reached this level, see Fig. 3. In the figure, data on hourly PM2.5 is also presented. The PM2.5 and TGM peaks coincide surprisingly well.

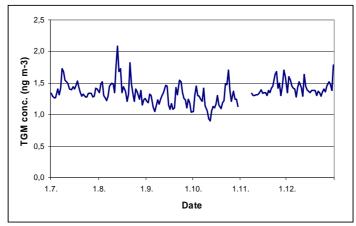


Fig. 2. 24 h average TGM concentration at Virolahti in July-December 2006

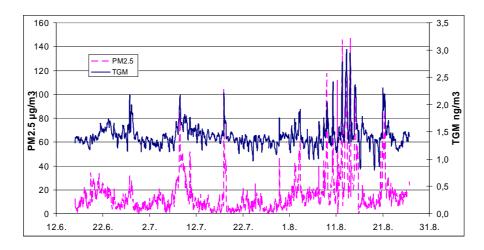


Fig. 3. PM2.5 mass (data: Pia Anttila, FMI) and TGM hourly averages at Virolahti in summer 2006.

Elevated levels of mercury are also seen occasionally due to air masses coming from the highly industrialized areas in Europe. In Fig. 4, 120 h backward trajectories calculated using the HYSPLIT model<sup>2</sup> are shown for the event on the 8th of July. The figure shows a typical trajectory picture for higher TGM concentrations.

Also some rather low levels of TGM were observed during the studied period. The backward trajectories showed that the air masses with low TGM concentration were usually arriving from the Arctic Ocean or the North Atlantic via Scandinavia. Also during the small morning hours, the TGM concentrations were fairly often lower than during the daytime. This was most evident for the summer months.

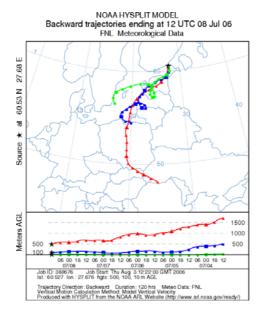


Fig. 4. 120h backward trajectories at three different heights for the 8th of July episode calculated with the NOAA/HYSPLIT model.

# **ACKNOWLEDGEMENTS**

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# TROPOSPHERIC LIDAR MEASUREMENTS AT SPITSBERGEN

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# STATIONARY LIDAR KARL AT THE NDACC STATION

The KARL (Koldewey Aerosol Raman Lidar) is firmly integrated into the NDACC station of the AWIPEV research facility. Its purpose is the observation of tropospheric aerosol and water vapour. Technically KARL consists of a Nd:Yag laser which emits light pulses at 3 different colours, at 355nm, 532nm and 1064nm with about 10W power each. Light which is backscattered at molecules, aerosol or clouds is collected by 2 telescope mirrors, a 30cm mirror for altitudes above 2km and a 10cm mirror for altitudes from 500m to 3km. Photomultiplier record this backscattered light, such that with the time lag between the firing of the laser and the detection event the height of the recorded atmospheric layer can be derived. Apart from the 3 laser wavelengths inelastic scattered light (Raman effect) is additionally recorded at 387nm and 607nm (from N<sub>2</sub> molecules) and at 407nm and 660nm (from H<sub>2</sub>O molecules). Height profiles of the extinction coefficients can be derived out of the inelastic scattered N<sub>2</sub> colours and therewith the backscatter coefficient profiles from the 3 elastic scattered colours (355nm, 532nm and 1064nm).

Additionally KARL records 532nm profiles in a polarisation state perpendicular to the laser. Spherical (liquid) particles do not shift the state of polarisation, contrary to crystals. So the state of aggregation can be derived from KARL, next to water vapour profiles and 5 independent optical properties (3 backscatter coefficients and 2 extinction coefficients).

KARL has a field of view of 0.83mrad. Measurements cannot be performed when thick clouds are present or precipitation occurs (multiple scattering).

# Aerosol measurements

Apart from Arctic Haze several other aerosol events have been detected by KARL in the recent years, mainly in the free troposphere. "Fig.1" gives an example of a forest fire event which occurred on June, 23<sup>rd</sup>, 2003 over Ny Ålesund. This is an example of a Siberian forest fire which was observed by several lidar stations all around the northern hemisphere.

# Backscatter profiles June, 23rd, 2003

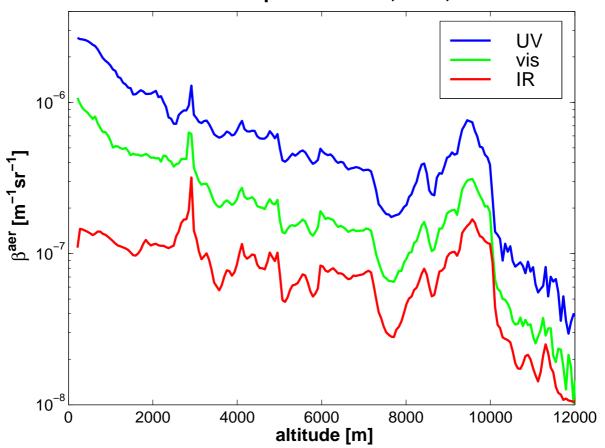


Fig. 1. Backscatter profiles of the wavelengths 355nm, 532nm and 1064nm. Two aerosol layers in 2.7km and 9km altitude are present. Lidar data have been averaged over 2 hours in time and 60m in altitude.

Several aerosol layers are visible in "Fig.1" which were, on that day, remarkable stable in time. Air trajectories confirmed a long-range advection of biomass burning aerosol from forest fires. Due to relatively low values of the volume depolarisation (<3%) for this kind of aerosol, Mie theory of scattering should provide a suitable approximation and an estimation of the size distribution out of the lidar data becomes possible. In the following the aerosol peak at 2.7km altitude shall be examined further.

In a first step an appropriate index of refraction which represents the lidar data in an optimal way is searched. "Fig.2" shows a typical result of this step. Lidar data depend on backscatter and extinction, but, unfortunately not directly on absorption. Hence, the retrieval of the imaginary part is principally challenging. An independent measurement of the absorption in one of the applied wavelengths would reduce the insecurities of this step significantly.

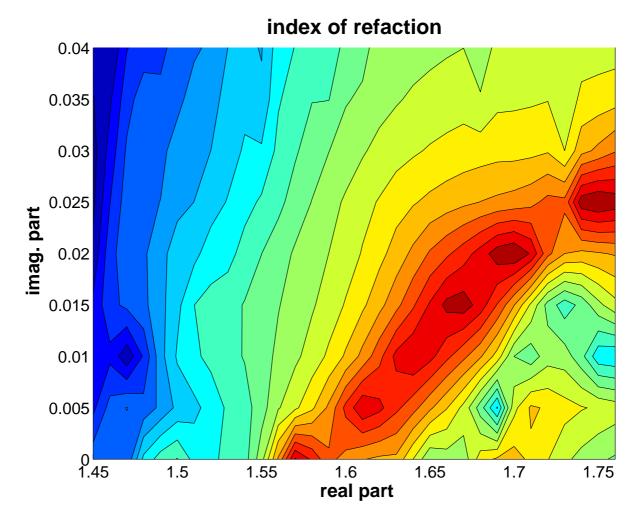


Fig. 2. Probability distribution that the index of refraction matches to the recorded backscatter and extinction values. Red values correspond to high, blue to low probabilities. Frequently not a single maximum is visible.

Here in the example of "Fig.2" an index of refraction of n=1.67+i 0.017 was chosen, which is close to the center of gravity of the stripe in the figure.

Having been able to estimate the index of refraction (n) one can invert the Fredholm integral equation

$$\alpha(\lambda)/\beta(\lambda) = \int_{r_{\min}}^{r_{\max}} Q^{ext/backscat.}(\lambda, n, r) \cdot N(r) \cdot dr$$

Where Q are the Mie efficiencies for extinction ( $\alpha$ ) and backscatter ( $\beta$ ) and  $\lambda$  denotes the wavelength. N(r) is the size distribution of aerosol. For the reason of a less pronounced singularity at r-0, however, not the size distribution N(r), but the volume distribution

$$V(r) = \frac{4\pi}{3} \cdot N(r)$$
 will be inverted. The result is presented in "Fig.3".

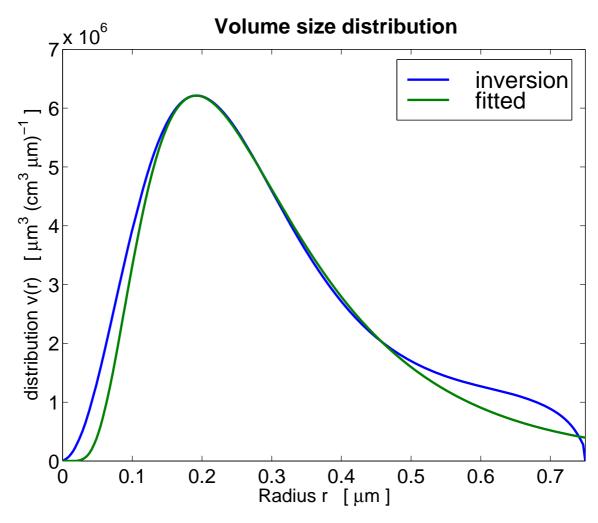


Fig. 3. The inverted aerosol size distribution function is given in blue. Green is a log-normal fit to the accumulation mode. As can be seen, these accumulation mode particles are responsible for the majority of the backscatter and extinction values.

According to the green curve in "Fig.3" the aerosol peak in 2.7km altitude can be best explained by a log-normal distribution with the parameters: effective radius =  $0.23\mu m$ , number of particles =  $116cm^{-3}$  and  $\sigma = 1.79$ . This accumulation mode is responsible for the majority of the recorded lidar signal, but the presence of a nucleation mode seems possible, because the one-modal log-normal distribution underestimates the particles smaller than  $0.1\mu m$ . However, due to the small interaction of nucleation mode particles with visible light their precise detection is principally challenging.

# AIRBORNE MOBILE AEROSOL LIDAR (AMALI)

For mobile operation in the Polar aircraft of Alfred Wegener Institute, a mobile lidar can be used, which emits light in 532nm and 355nm. For the visible wavelength an additional detection in a perpendicular polarisation detection is provided. This lidar is eye-save after less than 3km distance, so it can be operated from a plane in nadir configuration in that altitude. In previous ASTAR campaigns the orographic effects of aerosol load have been measured by AMALi which will be shown.

# THE IMPORTANCE OF LONG-TERM ROUTINE MEASUREMENTS: 14 YEARS OF HUMIDITY OBSERVATIONS IN THE ARCTIC TROPOSPHERE OVER NY-ÅLESUND, SVALBARD

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With this contribution we like to demonstrate the importance of long-term routine measurements. Water vapour is an important component in the radiative balance of the polar atmosphere. We present a study covering fourteen-years of data of tropopsheric humidity profiles measured with standard radiosondes at Ny-Ålesund (78° 55' N 11° 52' E) during the period from 1991 to 2005. It is well known that relative humidity measurements are less reliable at cold temperatures when measured with standard radiosondes. The data were corrected for errors and used to determine key characteristic features of the vertical and temporal RH evolution in the Arctic troposphere over Ny-Ålesund. We present frequency occurrence of ice-supersaturation layers in the troposphere, their vertical span, temperature and statistical distribution. Supersaturation with respect to ice shows a clear seasonal behaviour. In winter (October – February) it occurred in 22 % of all cases and less frequently in spring (March – May 13 %), and summer (June – September, 10%).

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# NIPR ATMOSPHERIC SCIENCE ACTIVITIES IN SVALBARD, ARCTIC

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# **INTRODUCTION**

The National Institute of Polar Research (NIPR) started observations of atmospheric parameters at Ny-Ålesund (78°55'N, 11°56'E), Svalbard using Rabben Observatory close to the air strip in 1991, based on the bi-polar standpoint with the Antarctic observation at Syowa Station (69°00'S, 39°35'E). Since then, several parameters have continued to be measured at the site, such as greenhouse gases, ozone, aerosols, clouds and precipitations with the support by Norwegian Polar Institute (NPI). Founded on these ground-based observations, airborne observations were planned to obtain vertical distribution of aerosols, radiation and so on, together with balloon borne observations. International cooperative campaigns have been conducted as ASTAR (Arctic Study of Tropospheric Aerosol and Radiation) 2000, 2004, (2007) and AAMP (Arctic Airborne Measurement Program) 2002, following AAMP 98 carried out in 1998. Longyearbyen airport (78°N, 15°E) was used as a base of flight operations. In order to see air-sea exchange (source and sink) of CO<sub>2</sub>, shipborne observations of pCO<sub>2</sub> were also conducted during 1992 and 2001 in the Greenland Sea and the Barents Sea on board r/v Lance and others. Related to the International Polar Year (IPY) 2007–2008, several activities are to be made by Japanese scientists in IPY projects.

Atmospheric science activities in Svalbard by NIPR are reviewed in the following.

# GROUND-BASED OBSERVATIONS AT NY-ÅLESUND

Measurements of greenhouse gases, CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, CO, SF<sub>6</sub> and isotopes of CO<sub>2</sub> and CH<sub>4</sub>, have been conducted through analyzing weekly sampled air at Rabben. Also, insitu measurements of surface meteorology and surface ozone concentration are continued. Temporal variations of atmospheric CO<sub>2</sub> concentration and carbon isotope ratio of CO<sub>2</sub> were reported (Morimoto et al., 2001), temporal variations of atmospheric CH<sub>4</sub> concentration and carbon isotope ratio of CH<sub>4</sub> were shown, and contribution of wetland and biomass burning as the source of CH<sub>4</sub> was discussed (Morimoto et al., 2006). Sudden ozone depletion was frequently found in the data of the surface ozone in Spring (Wessel et al., 1998).

Remote sensing of aerosols and clouds has been continued using a Micro-pulse Lidar (MPL) and sky radiometer, and in-situ measurements of aerosols were conducted using optical particle counters, integrating nephelometer, absorption photometer and several kinds of samplers. Fine structures of clouds and their temporal change were observed by MPL and statistical features of the cloud base height were investigated (Shiobara et al., 2003). Overpass experiment for ground varidation of the ICESat (Ice, Cloud and land Elevation Satellite)/ GLAS (Geoscience Laser Altimeter System) atmospheric measurements were performed in 2003 and 2004, and comparisons of lidar measurements from space-borne GLAS and ground-based MPL were made (Shiobara et al., 2006).

Chemical compositions of aerosols were shown from the analyzed data of samples and effects of transformation and transportation were discussed (eg., Hara et al., 1999).

Cloud and precipitation were measured using a vertical pointing radar, precipitation particles were measured by POSS and atmospheric water vapor was monitored by a microwave radiometer up to 2004. Seasonal variations of the precipitations/ clouds were investigated from these data and cloud coverage and vertical distribution were also discussed (Wada et al., 1996).

Stratospheric lidar measurement was carried out sporadically during winter months by Nagoya and Fukuoka University and very special feature of the vertical distribution of polar stratospheric clouds (PSCs) as "sandwich structure" was found related to ozone destruction (Shibata et al., 1999). In order to confirm these PSCs distributions, aerosol sonde measurements using OPC sonde were also conducted (Hayashi et al., 1998). Moreover, ozone profiles in the upper stratosphere were measured by Tohoku University with Alfred Wegener Institute for Polar and Marine Research (AWI) using a UV sensor on board a light-weight high-altitude balloon (Okano et al., 1996).

Characteristics of radiation budget at Ny-Ålesund were analyzed using the data obtained by NPI, and comparative discussion was made with the data from Syowa Station, Antarctica (Yamanouchi and Orbaek, 1995).

# AIRBORNE CAMPAIGNS

The Arctic Airborne Measurement Program (AAMP) 98 was planned to investigate the transport, exchange and transformation processes of greenhouse gases and aerosols in the Arctic atmosphere. An instrumented aircraft, Gulfstream II (G-II) was flown from Nagoya, Japan, through Barrow, Alaska to Longyearbyen, Svalbard passing over the Arctic Ocean in 5-14 March 1998 (Shiobara, et al., 1999).

Series of airborne observations were carried out under the research project "Variations of atmospheric constituents and their climatic impact in the Arctic (FY1999-2004)" (Spec. Sci. Res. Prog. 11208201). The project has been conducted to clarify the variation of greenhouse gases, aerosols, and clouds in the Arctic troposphere and stratosphere; to explain the transport and transformation processes, source and sink; to compare with the Antarctic and evaluate the radiative effect and then the climate impact.

- (1) **ASTAR 2000** (Arctic Study of Tropospheric Aerosols and Radiation): In collaboration with the AWI together with support by other institutions, coordinated airborne and ground-based observations of aerosols (Arctic haze) and radiation were carried out in the Svalbard area through March and April, 2000 (Yamanouchi et al., 2005). AWI aircraft Polar 4 (Dornier 228) based on Longyearbyen was used to measure vertical distributions of aerosols and radiation, while remote sensing, sonde observations and sampling were conducted on the ground at Ny-Ålesund (eg., Hara et al., 2003). In addition, SAGE-II satellite observations were compared (Thomason et al., 2003), and the radiative forcing of aerosols over a wide area was evaluated by incorporating into an Arctic regional climate model (HIRHAM; Treffeisen et al., 2005).
- (2) **AAMP 02** (Arctic Airborne Measurement Program): In connection to AAMP 98 similar campaign of airborne observation using jet plane (G-II) was carried out in March 2002 with long range stratosphere flights over the Arctic Ocean and local profiling flights in the vicinity of Svalbard. Research objectives were to elucidate spatial distribution, long-

range transport and transformation of greenhouse gases and aerosols, related to stratosphere-troposphere exchange and polar vortex; optical properties of aerosols and their radiative forcing; the structure of atmospheric disturbance, especially of polar low, (Yamanouchi et al., 2003; Morimoto et al., 2003; Treffeisen et al., 2006).

(3) **ASTAR 2004** (Arctic Study of Tropospheric Aerosols, Clouds and Radiation): With the initiative of AWI, another coordinated airborne campaign of aerosols and clouds were carried out through May and June 2004. Two AWI aircrafts Polar 2 and Polar 4 full equipped with instruments were used to measure mainly clouds and aerosols, respectively. In comparison to ASTAR 2000, observation term was chosen to cover the transitional season between spring and summer, the end of the Arctic haze season and start of clean summer air, and interactions of cloud and aerosols (indirect effect) were tabled as the second objective. Characterization of washout processes of aerosols were expected.

# AIR-SEA CO<sub>2</sub> EXCHANGE

In order to elucidate the seasonal and interannual variations of oceanic  $CO_2$  uptake in the Greenland Sea and the Barents Sea, the partial pressure of  $CO_2$  in the surface ocean (p $CO_2$ ) was measured in all season between 1992 and 2001 (Aoki et al., 1996). A large seasonal variation in p $CO_2$  was found with a sudden decrease in April, May and June from about 300 to 220  $\Box$  atm due to the marine biological activities in spring. However, due to another large seasonal variation of the wind speed, both seasonal variations compensated each other and rather constant  $CO_2$  uptake was seen. The annual  $CO_2$  uptake was estimated to be  $0.005 +_{\_} 0.020$  GtC/yr (Nakaoka et al., 2006), which is one of the maximum among world oceans.

# IPY 2007-2008

Endorsed projects of IPY 2007-2008 in which Japanese scientists are active are ORACLE-O3 (Ozone layer and UV radiation in a changing climate evaluated during IPY), POLAR-AOD (the Polar aerosol optical depth measurement network project), POLARCAT, IASOA (International Arctic Systems for Observing the Atmosphere) and HIAA (Hydrological Impact of Arctic Aerosols). Related to ORACLE-O3, the second ozonesonde network observation (Antarcitc Match) is to be made at 9 stations around Antarctica including Syowa Station with FTIR measurements of minor constituents acting on the ozone hole chemistry and PSCs in 2007. Under POLAR-AOD, the first field campaign for the AOD measurement radiometer inter-comparison was carried out in March 2006 at Ny-Alesund. Twelve institutions from 9 countries participated in the campaign along with their own radiometers. Pre IPY activity of Japan-German airborne aerosol measurements was successfully accomplished around Syowa and Neumayer Stations, Antarctica in 2006/07 summer season, and ASTAR 2007 has been just finalized around Svalbard with Polar 2 of AWI and Falcon of DLR, as a part of POLARCAT. In HIAA project AUV flight observation of heat flux and sea ice features is planned to be made over the Arctic sea ice area from Ny-Alesund airport.

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# THE POLAR AEROSOL OPTICAL DEPTH MEASUREMENT NETWORK PROJECT (POLAR-AOD-IPY)

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Despite the important role that aerosols play in modulating the radiation budget of the Polar regions, our knowledge of their physical and radiative properties, horizontal and vertical distributions and temporal variability remain inadequate. Surface-observing stations are few and far between, and although providing greater coverage satellite retrievals of low AOD are prone to error when made over highly reflective surfaces. The POLAR-AOD project aims to characterize the means, variability and trends of the climate-forcing properties of aerosols in Polar Regions. It is designated as International Polar Year (IPY) Project # 171. The primary objective is to establish a bipolar network of observatories to obtain data needed to quantify aerosol physical and radiative properties at high latitudes, including seasonal background concentrations inferred from aerosol optical depth AOD measurements, spectral characterizations, and patterns of natural and anthropogenic processes that influence the radiative balance of the surface and atmosphere. Activities will involve numerous cooperative institutes, including 40 research groups from 22 countries. Measurements at Arctic and Antarctic stations will be carried out with the logistical and financial support of established national programs. Data archiving and management, inter calibration efforts and coordination of research activities will be developed primarily by ISAC-CNR - Italy. During IPY 15 stations in the Arctic (five of them located in Ny-Alesund) and 16 stations in Antarctica will supply data to be archived and analyzed by the participating institutes. An effort to quantify direct and indirect climate forcing by polar aerosols will be made through a set of closure experiments using observations in conjunction with model calculation and satellite data. POLAR-AOD will coordinate its activities with other IPY projects as part of the International Arctic System for Observing the Atmosphere (IASOA). Future POLAR-AOD observations will complement ongoing Baseline Surface Radiation Network (BSRN) measurements made at several Arctic and Antarctic stations. In the frame of this project a first field inter-comparison campaign was carried out at the Japanese Rabben station in Ny Alesund March 25 to April 5, 2006.

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# AEROSOL-CLIMATE RESEARCH PROGRAM BY KOREA POLAR RESEARCH INSTITUTE

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# INTRODUCTION

Aerosols affect the global radiation budget (Houghton, et al., 2001), directly through scattering incoming solar radiation (Charlson, et al., 1992), and indirectly through modulation of cloud albedo (Twomey, 1974). Polar region can provide an ideal place to investigate the relationship between atmospheric composition change and climate. Korea Polar Research Institute (KOPRI) has launched an aerosol-climate research program at it's research stations in the Antarctica and Arctic since 2006. In this report, we report brief summary of KOPRI's research program and secondary aerosol formation events observed at the King Sejong Station, Antarctica.

# **EXPERIMENTS**

King Sejong Station, Antarctica

Secondary particle formation events are frequently observed over coastal environments (Yoon et al., 2006, O'Dowd, et al., 2002), the marine boundary layer (Clarke, et al., 1998), boreal forests (Makela, et al., 1997), Antarctica (Koponen, et al., 2003), and Arctic areas (Wiedensohler, et al., 1996). A bank of Condensation Particle Counters (CPC) with different cut-off diameters (TSI CPC model 3025: D > 3nm and 3010: D > 10 nm) and an Optical Particle Counter (OPC, Grimm model 1.108) were deployed at the King Sejong station (KSJ, 62.22 °S, 58.78 °W) during austral summer (6 Dec. 2005 - 7 Jan. 2006). Meteorological data, irradiance, air temperature, wind speeds and direction, atmospheric pressure, were obtained from the KSJ meteorological observation tower. The KSJ station is located in Barton Peninsula, King George Island (Figure 1). The station is facing Marian Cove and the sampling location is about 500 m from the shore.



Fig.1. Location of the King Sejong Station, Antarctica.

Particle formation events were observed 18 days out of 33 days (55%) during the measurement period. During the events the total particle number concentration increased from  $200 - 300 \text{ cm}^{-3}$  up to  $50,000 \text{ cm}^{-3}$ , and the nucleation lasted for more than 4-5 hours depending on the availability of the direct solar radiation. Figure 2 shows an example of particle formation event, on 1 January 2006.

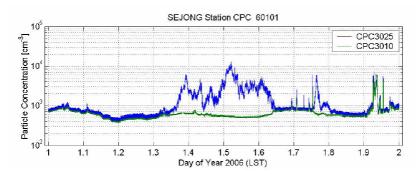


Fig. 2. Particle formation event observed at the King Sejong Station, 1 January 2006...

For the day, the air mass originated from the Antarctic continent and traveled over the biologically fertile open ocean before arriving at the sampling site. The particle number concentrations from both CPCs were below 500 cm<sup>-3</sup> before the formation events. The event started around 4 am, as soon as direct solar radiation was available, and lasted for several hours. Under clean marine air mass condition, nucleation events were observed only when the direct solar radiation is available, implying that photochemical reactions of the precursor gases are required to induce the observed events. When air mass traveled over biologically fertile coastal regions, nucleation occurred not only under clear sky days, but also under cloudy conditions, showing higher number concentrations for cloud-free days. The sources of precursor gases for the secondary particle formation at the Antarctic coastal areas (during austral summer) are thought to be marine biota derived sulfur compounds. To test this hypothesis, long-term measurements of particle formation characteristics are being made.

# Dasan Station, Ny-Ålesund

**Ongoing project:** A Cloud Condensation Nuclei Counter was installed at the Zeppelin station, Ny-Alesund to investigate long-short term variation of aerosol activation into cloud droplet size. The CCN counter was installed at the Zeppelin station on the 20<sup>th</sup> March 2007, and it's first look-at data showed that about 80 % of particles can grow into CCN size at super-satulation of 0.2 to 1% (i.e. Figure 3). These results are obtained from an unusual clean air-mass spring, as a result, a continuous measurement of CCN in parallel with other physico-chemical monitoring program, such as number concentration, absorption, aerosol size distribution, are now being deployed at the Zeppelin station.

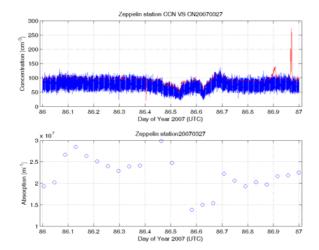


Fig. 3. Upper plate: Daily CCN concentration (blue line) and CN data for D > 10 nm (red line). Lower plate: Aerosol absorption data on the  $27^{th}$  March 2007.

**Future plan**: It has been hypothesized that the marine biota and the atmosphere have a feedback through a contribution of biogenic sulfur containing gases to the aerosol formation and growth. Though numerous attempts have been made to prove or challenge this hypothesis during last decades, a quantitative projection of the relationship has not been achieved. In August 2007, The DMS(g) measurements will be made from the Zeppelin station, by the Pohang University of Science and Technology (POSTECH) team in a collaborative manner with the KOPRI. Two important scientific issues can be addressed by continuous atmospheric measurements of DMS in Ny-Alesund. First, the atmospheric DMS variations above Ny-Alesund likely reflect variations in the DMS-producing marine phytoplankton such as Emiliania huxleyi in the North Atlantic. Second, long-term measurements of atmospheric DMS will also provide an insight into how seasonal or interannual variations in the sea ice cover affect the oceanic release of DMS into the atmosphere.

# ACKNOWLEDGMENTS

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# COMPARISON AND INTERPRETATION OF THE RESULTS OF GHG MEASUREMENTS AT ZAPPELIN, PALLAS, AND TERIBERKA (KOLA PENINSULA, RUSSIA)

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Timeseries of measured CO<sub>2</sub> mixing ratio at Zeppelin, Pallas, and Teriberka have been fitted by the lineally increasing harmonic function. Comparison of detrended average seasonal cycles for these stations gives an opportunity to distinguish the influence of the local forest ecosystem upon atmospheric CO<sub>2</sub> concentration measured at Pallas from the long-range transported influences. Next the divinations of the measured CO<sub>2</sub> from the fitted curves have been analyzed by means of trajectory analysis.

The negative divinations at Teriberka are often connected with the trajectories which have come to Kola Peninsula from the north sector. It is most likely that such divinations are the results of marine sink of CO<sub>2</sub>. This suggestion supports by comparison with background level at Zeppelin in the days when the back trajectories before coming to Zeppelin passed over the Arctic.

The advantage of using Teriberka as the sampling point and Zeppelin as the reference point for studding the marine sink of CO<sub>2</sub> is due the fact that in spring and summer air transport pathways from the Arctic to continent are more common. At some occasion the measured records displays negative divinations when trajectories have come to Teriberka from the continent (from SW). This may be the result of photosynthesis of terrestrial plants.

The positive divinations are often connected with the trajectories coming to Teriberca from SE (the West Siberia direction). The trajectory analysis itself can offer explanation only of timing of the observed concentrations anomalies. To calculate the flux across the air surface boundary we have been developing an inverse technique. This technique is based on combination of trajectory model and calculation of GHG vertical profile transformation in a moving air mass.

# COMPARISON AND INTERPRETATION OF GHG MEASUREMENTS AT ZEPPELIN, PALLAS, AND TERIBERKA (KOLA PENINSULA, RUSSIA)

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# **INTRODUCTION**

Humanity faces global warming. The most fast climate change is taking place in the Arctic and it is especially important to take into account the future climate projections while designing infrastructure in this region. Though the global warming is a reality there are many uncertainties of climate change even in the near future. The carbon cycle has not been revealed completely. Future investigations of atmospheric carbon souses and sinks, especially in Arctic, are required. The Barents See region deserves enhanced study since it includes active oceanic sources and sinks of CO<sub>2</sub>, increasing excavation of natural gas and oil, and large industrial activities.

# **MATERIALS**

The Zeppelinfjellet on Spitsbergen and Pallas in northern Finland are WMO/GAW stations that monitor atmospheric trace gases and aerosols in the Barents See region. In Russia there is the weekly flask monitoring station at Kola Peninsula (Teriberka 69° 12' N, 35° 06' E) where monitoring of CO<sub>2</sub> has taken place since 1988 and CH<sub>4</sub> since 1999 (WMO WDCGG, 2006). The Russian station is closer to the GHG emission regions and great benefit would be achieved through a combine interpretation of its data with Zeppelin and Pallas.

# **RESULTS**

Fig. 1 shows the comparison of the average seasonal cycles of  $CO_2$  for 3 stations: Zeppelin, Pallas, and Teriberka.

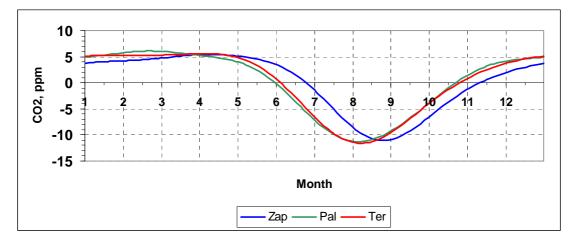


Figure 1. Comparison of the average annual cycles of  $CO_2$  for 3 monitoring stations in the Barents See region: Zeppelin (Norway), Pallas (Finland), and Teriberka (Russia). Annual amplitudes are as follows: Zep -16.5 ppm, Pal -17.4 ppm, Ter -17.1 ppm.

The seasonal cycles have been obtained from the least square fitting of monitoring data from which the long-term trends were subtracted. The main difference of the seasonal cycles between these stations is the lag in time at Zeppelin from Pallas and Teriberka. By comparison of seasonal cycles it will be possible to estimate the rate of air masses exchange between middle latitudes and the Arctic. Proximity of the seasonal cycles at Pallas and Teriberka is a result of fast rate of exchange along longitudes.

Next the deviations of the measured CO<sub>2</sub> from the fitted curves have been analyzed by means of trajectory analysis. Trajectory models were successfully applied for interpreting measurements at this region (*Aalto et al.*, 2003; *Eneroth, K., Holmen, K.*, 2004). We calculated the backward air mass trajectories by means of HYSPLIT (NOAA) model (*Draxler, R.R., Rolph, G.D.*, 2003). The negative divinations at Teriberka are often connected with the trajectories, which have come to Kola Peninsula from the north sector (see Figure 2, for example).

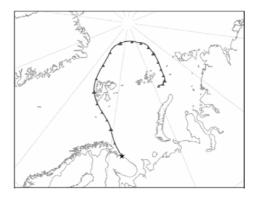


Fig. 2 A plot of 5-day HYSPLIT air parcel backward trajectory arriving at the sampling point Teriberka at 15 hours LT 11.02.2001. The trajectory arrived to Teriberka over the sea and is associated with negative anomaly in CO<sub>2</sub>. The decrease of CO<sub>2</sub> in the pathway from Zeppelin to Teriberka is 4.5 ppm.

It is likely that such divinations are the results of marine sink of CO<sub>2</sub>. This suggestion is supported by comparison with background level at Zeppelin in the days when the back trajectories before coming to Zeppelin passed over the Arctic. The advantage of using Teriberka as the sampling point and Zeppelin as the reference point for studding the marine sink of CO<sub>2</sub> is due the fact that in spring and summer air transport pathways from the Arctic to continent are more common. At some occasion the measured records in summer displays negative divinations when trajectories have come to Teriberka from the continent (from SW). This may be the result of photosynthesis of terrestrial plants. The positive divinations are often connected with the trajectories coming to Teriberca from SE (the West Siberia direction).

The trajectory analysis itself can offer explanation only of timing of the observed concentrations anomalies. To calculate the flux across the air surface boundary we have been developing an inverse technique. This technique is based on combination of trajectory model and calculation of GHG vertical profile transformation in a moving air mass.

# **CONCLUSION**

Joint analysis of measurement data from several observations sites in Barents region can give synergetic effect and support the international research work concerning climate active spaces in the atmosphere. Teriberka is a coastal station located in a good position for estimation of marine  $CO_2$  uptake in Barents Sea by means of its comparison with Zeppelin. Disadvantages of present monitoring at Teriberka are as follows: flask sampling doesn't provide means for monitoring day-to-day variability, there are no observations of aerosols.

# **ACKNOWLEDGEMENT**

The author gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and READY website (http://www.arl.noaa.gov/ready.html) used in this publication.

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# **SOGE**

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### INTRODUCTION

NILU is responsible for the scientific programmes at the Zeppelin Station and coordinating the scientific activities undertaken by NILU and other institutions, as well as a number of international research groups' campaigns. Stockholm University (SU) cooperates closely with NILU in developing the scientific activities and programmes at the station. The Zeppelin Station is owned and operated by Norwegian Polar Institute The monitoring and research programmes address several issues, such as: climate change, arctic stratospheric ozone layer depletion, global distribution of toxic pollutants, distribution of radioactive contaminants.

SOGE is an integrated System for Observation of halogenated Greenhouse gases in Europe. A combination of observations and modeling is used to estimate regional emissions as well as global trends and impact on climate and the ozone layer. In situ observations at four background stations in Europe forms the back bone of SOGE. A wide range of halogenated greenhouse gases are measured in-situ at these four background measurement stations. The four stations are Mt. Zeppelin at Svalbard in arctic Norway, Mace Head in Ireland, Jungfraujoch in Switzerland and Mt.Cimone in Italy. Measurements are performed with high frequency by the use of automated gas chromatographs with mass spectrometry detectors. Measurement data from the years 2001 - 2003 are analyzed for pollution events. Trends in background concentrations were also analyzed.

The gases have a wide variety of applications such as refrigeration, foam blowing and fire extinction [Midgley and McMulloc, 1999]. Regarding their environmental impact, these substances can further be divided into two main groups. The first group consists of halocarbons responsible for the depletion of stratospheric ozone, i.e. CFCs, HCFCs and long lived chlorinated solvents. The substances are regulated in the Montreal Protocol and are due to be phased out worldwide. The second group combines halocarbons with fluorine as the only halogen i.e. HFCs. These substances do not have a direct effect on stratospheric ozone, but contribute to global warming and where therefore included into the Kyoto Protocol.

# **INSTRUMENT**

The instrument is a fully automated adsorption/desorption sampling device (ADS) coupled with an automatic gas chromatograph with a mass spectrometric detector (GC-MS). The system provides 6 air samples during 24 hours. The instrument is the same instrument as the ones located at the SOGE stations Mace Head and Jungfraujoch and all the AGAGE (Advanced Global Atmospheric Gases Experiment) sites. The four sites within the SOGE project are using calibration tanks, which are pressurized simultaneously at Mace Head and then calibrated to AGAGE scale. The instrument

currently monitors more than 20 compounds, including CFCs, HFCs, HCFCs, halons and a range of other halogenated species.

# DETERMINATION OF BACKGROUND DATA

Based on the daily mean concentrations, an algorithm is selected to find the values assumed as clean background air. If at least 75% of the trajectories within +/- 12 h of the sampling day are coming from a so called clean sector (Arctic or Atlantic air masses) one can assume the air for that specific day to be non polluted. The remaining trajectories from either European, Russian or North-American sector are removed before calculating the background. We find that most of the spikes, i.e. assumed polluted events, are removed during the filtering. One can assume that the chosen filter is a good method for finding mean background concentrations for further work.

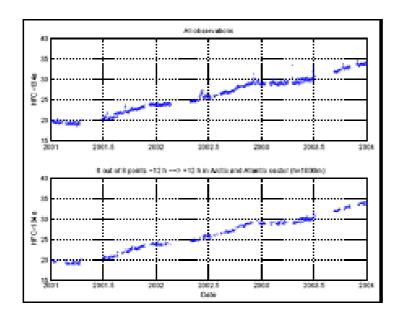


Figure 3.1: Finding the mean background data, HFC-134a. Upper panel shows all daily observations, lower panel shows data selected for background concentration calculations.

# **CURVE FITTING**

The distinct annual cycles in concentrations can be explained at least qualitatively in terms of distances from sources (largely northern latitude), intensity and seasonality of the global circulation. For some components, like Methyl Chloroform and other components with atmospheric lifetime shorter than a few years, the seasonal cycle in their reaction with OH is also used to explain the annual cycles.

The compounds that have a strong seasonal variation in the concentration, due to the reaction with OH radicals in the atmosphere, will fit the harmonic curve better than the other compounds discussed here.

For the gases that have a sudden change in the growth rate, described by a step function, the harmonic function with its typically cyclic period does not fit the data very well.

A close look at the observations reveals that the fluctuations on a scale less than one year are less pronounced than the yearly ones. On the other hand, variations on longer timescales are important. To take this into account, we introduce Legendre polynominals.

$$f(t) = a + b N P1(t/N-1) + (1/3)d N^2 P2(t/N-1) + (1/8)e N^3 P3(t/N-1) + c1 cos(2t) + s1 sin(2t)$$

The observed f can be expressed as functions of time measured from the 2N-year interval of interest. The coefficient a defines the average mole fraction, b defines the trend in the mole fraction and d defines the acceleration in the trend. The c and s define the annual cycle in mole fractions. N is the mid point of the period of investigation. The polynomial P3 (with coefficient e) is small, but added to better t the full data set. The detrending of the baseline by harmonics allows the generation of the seasonal cycle of each measured compound per year of data, which is then aggregated to develop an annual averaged seasonal cycle for each compound. The cycles for the long lived components result from a combination of seasonal cycles (if any) in their release (mainly from NH mid-latitudes) and seasonal cycles in their transport. For the components with shorter atmospheric lifetime, e.g. weeks/months, the annual cycles in their atmospheric destruction by OH (summer maximum and winter minimum) are superimposed on their source induced and transport induced cycles.

Examples of use of the Legendre polynomial fit function in combination with harmonic functions are showed in the lower panel in the figure below It is easily seen that this method fits both regular and irregular trends in the different gases.

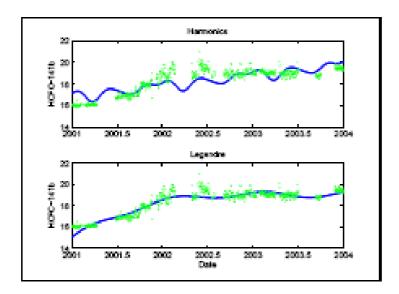


Figure 3.4: Upper panel shows the background data plotted as a harmonic function overloaded the daily means for HCFC-141b. Lower panel shows the background data as a function of Legendre polynomials overloaded the daily means for HCFC-141b.

# **TRENDS**

The monitoring at the Zeppelin Mountain Station in 2001 - 2003 revealed that the upward trend of most greenhouse gases in the atmosphere continued. The highest percent increase was the 25% rise of the concentrations of HFC-125 and HFC-134a, which is even more than the 20% rise measured the year before. [SFT-report, 2003] This rise is probably a consequence of the phasing-out of CFCs controlled by the Montreal Protocol, as HFCs have been introduced as substitutes for HCFCs and CFCs. Even though global emissions of CFCs have been reduced considerably over the last few years, only small reductions in the concentrations of CFCs have been registered so far and some CFC species are even still growing in concentration. This is likely due to the fact that these gases have very long lifetimes in the atmosphere.

Updated ground-based measurements of chlorflouorocarbons (CFCs) show that surface mixing ratios of CFC-113 peaked around 1996 and have been decreasing slowly thereafter. The decrease noted for CFC-11 mixing ratios in 1996 has continued [Montzka et al., 1999; Prinn et al., 2000].

In 2001 - 2003, the tropospheric mean mixing ratio of CFC-11 at Mt.Zeppelin is about 264 ppt. For the period, the trend in CFC-11 seems to be increasing with a growth rate about 3.46 ppt/year, but the increase in the trend is negative, -2.83 ppt/year2. The mixing ratio of CFC-11 was at its highest in spring 2002 and decreased in late 2002 and 2003. Updated measurements of global hydrochlorofluorocarbons (HCFCs) indicate that global mixing ratios of the three most abundant HCFCs continue to increase in the atmosphere, owing to sustained emissions [Montzka and Fraser, 2002]. HCFCs make up about 6% of the total 2003 chlorine burden in the atmosphere.

The global emissions of HCFC-141b were strongly reduced from 2000 to 2003. Measurements at Mt.Zeppelin show that the mean mixing ratio of HCFC-141b rose from about 17 ppt in 2001 to nearly 19 ppt in 2002, compared to a rise of only 0.1 ppt to 19.1 ppt in 2003. The growth rate from 2001 to 2003 is 1.93 ppt/year and decreasing at a rate of -0.93 ppt/year2. The rate of increase seen for HCFC-141b may also be shown for HCFC-142b. In 2001 the yearly growth rate was exponentially, and it slowed significantly during 2002 - 2003. Mean mixing ratio of HCFC-142b in 2001 - 2003 is 15.6 ppt. With a current, slightly decreasing, growth rate of 0.8 ppt/year for 2001 - 2003 one can assume the concentration to be stable at the same level for some years from now. The rate of growth in the HFC-125, HFC-134a and HFC-152a have increased over the period of SOGE observations (2001 - 2003). All HFC mixing ratios are at their highest at the end of the 2001 - 2003 period.

For the 2001 - 2003 period, the trend in HFC-134a seems is increasing at a rate of 4.99 ppt/year, and the increase in the trend is positive, 0.17 ppt/year2. The mean mixing ratio of HFC-134a were 26 ppt. Measurements suggest a mean mixing ratio at Mt.Zeppelin for HFC-152a of 3.4 ppt in 2001 - 2003, and a nearly constant growth rate from 2001 to 2003 at 0.59 ppt/year.

Compound	2001-2003 mean	b,trend	d,change in trend	s	С
CFC-11	264	3.46	-2.83	2.82	-0.02
CFC-113	82.0	1.19	-0.19	0.33	-0.25
CFC-115	8.37	0.24	-0.05	0	0
HCFC-22	167	5.77	-0.57	-0.02	0.89
HCFC-123	1.12	-0.05	0.01	0.03	-0.04
HCFC-124	1.67	0.14	-0.02	0.02	0.02
HCFC-141b	18.2	1.93	-0.93	0.19	0.19
HCFC-142b	15.6	0.82	-0.1	0.12	0.05
HFC-125	2.60	0.95	-0.90	0.02	0.18
HFC-134a	26.1	4.99	0.17	-0.4	0.5
HFC-152a	3.39	0.59	0.03	0.33	0.12
H-1211	4.47	0.09	-0.01	0	0
H-1301	3.14	0.20	0.08	-0.02	0
CH3 Cl	515	20.9	-0.1	56.8	14.3
CH3 Br	8.85	0.40	-0.82	0.46	0.38
CH3 I	0.66	0.05	0.15	-0.08	-0.27
CH2 Cl2	30.5	-0.15	1.15	6.12	1.89
DCE	0.66	0.12	0.04	0.25	0.18
MC	34	-3.42	1.00	-0.39	-0.22
CT	93.9	1.36	-1.10	0.41	-2.75

Once a major source of chlorine to the stratosphere, Methyl Chloroforms chlorine burden is now less than the major CFCs and HCFC-22. MC and HCFC-22 made up about 14 percent of the total chlorine burden in the atmosphere in 2003. The Methyl Chloroform mixing ratio at Mt.Zeppelin has been declining exponentially since 2001 because of the rapid drop in emissions to low levels; global mixing ratios in 2000 were less than one-half of the peak observed in 1992. [Montzka et al. 2002] The rapid decline in emissions of Methyl Chloroform and its relatively short lifetime, have together resulted in a rapidly decreasing mixing ratio during the period of investigation. The mean concentration of Methyl Chloroform is 34 ppt and the growth rate for 2001 - 2003 is -3.43 ppt/year and increasing at a rate of 1.00 ppt/year2. The rate of decline observed for Methyl Chloroform (and chlorine from Methyl Chloroform) at Mt. Zeppelin during 2003 was about two-thirds of what it was in 2001.

# **IDENTIFICATION OF EPISODES**

The observed polluted events at Mt.Zeppelin are rare and small in amplitude, mainly due to long distance from urban source areas. Identification of polluted episodes in this thesis are used to evaluate European and Russian emissions, which can be found by using backward trajectories. 10 days backward trajectories from the European Center for Mediumrange Weather Forecasts (ECMWF) have been used to investigate the major transport pathways into the region. During 2001 - 2003 there are six main events that seem to be able to compare for more of the compounds.

For both HFC-134a and HFC-152a we find that almost all of the clearly seen events come from European. The relatively new fluorocarbon and CFC-11 replacement HFC-134a (in use since 1990) is at a high rate represented from emissions from vehicles, concentrated in the US and Europe with 200 million vehicles each in the year 2000. The known event in May 2002 is easily seen for both HFC-134a and HFC-152a, with peak concentrations about 50% higher than the estimated background. We do also see an event in December 2002, with trajectories from European sector.

# **USE OF DATA**

By using a combination of air mass trajectories and the sector information, we observe that days with elevated concentration levels in most cases are associated with air from a non-Arctic sector.

For all components the individual daily values are naturally located below and above the monthly mean. Episodes can only be identified when the amplitude is large compared to the noise from the instrument. The pollution events do also contain essential information about regional (European and Russian) emissions.

Most of the events occur during the northern hemisphere fall, winter and spring. Out of 1095 possible days for pollution events, we find polluted episodes in 19 - 23 days in European sector, and 13 - 19 days in Russian sector. The numbers are based on averages for all the components.

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# ARCTIC SMOKE – EVALUATION OF AN EXTREME AIR POLLUTION EVENT OVER SVALBARD IN THE SPRING OF 2006

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During a glaciological campaign in the spring of 2006 abnormally warm conditions set new temperature records for the European Arctic. During this period a polluted air mass was efficiently transported into the high Arctic over Svalbard. During this warm period, smoke from agricultural fires in Eastern Europe intruded into the European Arctic and caused the most severe air pollution episodes ever recorded there.

This paper confirms that biomass burning (BB) was indeed the source of the observed air pollution, studies the transport of the smoke into the Arctic, and presents an overview of the observations taken during the episode. Fire detections from the MODIS instruments aboard the Aqua and Terra satellites were used to estimate the BB emissions. The FLEXPART particle dispersion model was used to show that the smoke was transported to Spitsbergen and Iceland, which was confirmed by MODIS retrievals of the aerosol optical depth (AOD) and AIRS retrievals of carbon monoxide (CO) total columns. Concentrations of halocarbons, carbon dioxide and CO, as well as levoglucosan and potassium, measured at Zeppelin mountain near Ny Ålesund, were used to further corroborate the BB source of the smoke at Spitsbergen. The ozone (O3) and CO concentrations were the highest ever observed at the Zeppelin station, and gaseous elemental mercury was also elevated. A new O3 record was also set at a station on Iceland. The smoke was strongly absorbing – black carbon concentrations were the highest ever recorded at Zeppelin – and strongly perturbed the radiation transmission in the atmosphere: aerosol optical depths were the highest ever measured at Ny Ålesund.

We furthermore discuss the aerosol chemical composition, obtained from filter samples, as well as the aerosol size distribution during the smoke event. Photographs show that the snow at a glacier on Spitsbergen became discolored during the episode and, thus, the snow albedo was reduced. Samples of this polluted snow contained strongly elevated levels of potassium, sulphate, nitrate and ammonium ions, thus relating the discoloration to the deposition of the smoke aerosols.

This paper shows that, to date, BB has been underestimated as a source of aerosol and air pollution for the Arctic, relative to emissions from fossil fuel combustion. Given its significant impact on air quality over large spatial scales and on radiative processes, the practice of agricultural waste burning should be banned in the future.

# 08 KORTRAPPORT/BRIEF REPORT SERIES

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