IGACtivities

Newsletter

of the International Global Atmospheric Chemistry Project

A Note from the IGAC Co-chairs: Sandro Fuzzi and Phil Rasch

In a few months time IGAC will hold its 9th International Conference, "Atmospheric Chemistry at the Interfaces", in Cape Town, South Africa, joint with CACGP and WMO. The Conference will offer the opportunity to review the latest results within the whole range of activities relevant to atmospheric chemistry research. It will also highlight advancements in the joint activities with other atmosphere-related programs such as SPARC, iLEAPS, and SOLAS. More information on the conference can be found in a note at the end of this newsletter.

Over the last few years the IGAC SSC and IPO have made an effort to implement, with an Earth System perspective, interdisciplinary activities which also strengthen the collaboration between programs and researchers in different areas of expertise (e.g. modelling/experimental, meteorology/chemistry, etc.). To this end, IGAC is trying to stimulate research activities in scientific fields and regional areas where improvement of existing knowledge is deemed necessary.

In this issue, for example, a new effort to improve the existing representations of wet scavenging in models is proposed (*Rodriguez et al.*). We believe the improvement of wet deposition and scavenging parameterizations for large scale models is an area meriting more activity. These processes are crucial in influencing the distribution of many species in the atmosphere and they are also critical via interactions with other components of the earth system. Scavenging processes currently are being treated very simply in all large-scale chemistry and climate models. Numerous model inter-comparison efforts have shown that scavenging processes and their parameterizations are responsible for many of the differences seen in aerosol and gaseous constituent distributions in models, with consequent effects on our ability to model the earth system, in particular under a changing climate. Many improvements have taken place during the last decade in our understanding of the processes relevant to scavenging and cloud constituent interactions in general, and this knowledge needs to be incorporated into models. The article by Rodriquez et al outlines these issues and invites your participation in a workshop focused on the production of a research plan that will aid researchers and funding agencies in determining the next best steps to addressing this important problem.

It is interesting to consider this effort on the context of the historical research on scavenging and deposition processes, by contrasting what might be required in the future in order for the findings of deposition studies to be more easily incorporated into models. One example of current efforts at constraining deposition is documented in an article herein on long-term wet deposition chemistry measurements in Japan (*Hayashi et al.*). Another which is also relevant is the ongoing IGAC Task DEBITS (Deposition of Biogeochemically Important Trace Species; www.igac.noaa.gov/DEBITS.php).

A second activity that IGAC has actively encouraged and that is now taking off is a coordinated effort of atmospheric chemistry research in South America, a continent with very diverse chemical regimes in terms of sources, species, climate, meteorology and anthropogenic influence. Two contributions in this issue (*Gallardo et al.* and *Longo et al.*) show the wide range of opportunities for improving the observational capabilities, chemical weather forecasts and emission inventories in this area, as well as highlighting the high level of scientific capabilities of the local scientific community.

The last contribution in this issue concerns the POLARCAT project (*Stohl and Law*). POLARCAT is a large coordinated activity, jointly endorsed by IGAC, SPARC and iLEAPS, which has been accepted as one of the activities of the International Polar Year and which aims at assessing the fast climatic changes in the Arctic, a particularly sensitive region already strongly affected by man's activities. We would like to close by expressing our appreciation to Shaw Liu, our fellow Co-Chair who stepped down from the IGAC Steering Committee at the end of 2005. Shaw's enthusiastic and competent contribution to IGAC has been very valuable: first as a member of the SSC, and then as a Co-Chair. We continue to owe Shaw and Academia Sinica a big "Thank You!" also for continuing to support the production of this newsletter.

In this Issue:

A Note from the Co-Chairs

Science Features

Announcements

Wet deposition chemistry in Japan: Key feature of the Japanese Acid Deposition Survey from 1983 to 2002

Contributed by **Kentaro Hayashi** (kentaroh@affrc.go.jp), *National Institute for Agro-Environmental Sciences, 3-1-3 Kan-nondai, Tsukuba 305-8604, Japan*, **Izumi Noguchi**, *Hokkaido Institute of Environmental Sciences, Japan*, **Tsuyoshi Ohizumi**, *Acid Deposition and Oxidant Research Center, Japan,* **Masahide Aikawa**, *Hyogo Perfectural Institute of Public Health and Environmental Sciences, Japan,* **Moritsugu Kitamura**, *Ishikawa Prefectural Institute of Public Health and Environmental Science, Japan,* **Akira Takahashi**, *Central Research Institute of Electric Power Industry, Japan,* **Kazuhide Matsuda**, *Meisei University, Japan,* **YukiyaMinami**, *Ishikawa Prefectural University, Japan, and* **Hiroshi Hara,** *Tokyo University of Agriculture and Technology, Japan*

Introduction

Japan has implemented a monitoring program for a nationwide acid deposition survey under the auspices of Ministry of the Environment (MOE). This program, the Japanese Acid Deposition Survey (JADS), aims to detect the current and potential impacts of acid deposition in Japan. It should be noted that a variety of monitoring techniques were used during the 20-year period of JADS due to both technical and administrative reasons (Hara, 1993; Hara et al., 1995). JADS dates back to a seven-month measurement program in 1983 by the Environment Agency, the former MOE. From September 1983 to March 1984, precipitation samples were collected at 34 sites with either wet-only or bulk samplers. On the basis of the preparatory program, a newly organized monitoring network was started in April 1984 by locating both wet-only and bulk samplers at seven sites and bulk samplers at seven other sites in Japan. Event-basis samples were collected with wet-only samplers, whereas bulk samples were collected on a weekly basis. In April 1986, the network was expanded to include 15 additional stations with bulk samplers. This phase of the program was completed in March 1988. In April 1988, this program entered another five-year phase, Phase II, in which the network was highly modified. Samples were collected on a biweekly basis by wet-only samplers at 23 reorganized stations and at six off-shore islands in 1989. In Phase III, from April 1993 to March 1997, the distribution of the stations was reviewed to reorganize the network to install wet-only samplers at 48 stations. The Phase IV survey started in April 1998 with a network of 47 stations and ended in March 2001 with as many as 55 stations.

Quality control and quality assurance (QA/QC) procedures were applied to all measurements throughout all four Phases, using as a basis the electric charge balance and the conductivity balance, as had been applied in the precipitation chemistry community worldwide (e.g., Noguchi et al., 1995). In and after Phase III, data completeness standards were officially

introduced within the network. This was done by taking into consideration measurements that were missing for a variety of reasons in order to derive the annual and monthly means consistently with the data quality. Throughout all phases, precipitation samples were analyzed for electric conductivity and the commonly determined nine ionic species: H^+(pH), SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Ca^{2+}, K^+, Mg^{2+}, and Na^+. More details of the JADS methodology are found in the technical manual of the EANET program (Acid Deposition and Oxidant Research Center (ADORC), 2000).

All measurements up to 2002 were officially analyzed by an expert team of atmospheric and ecological scientists before the Report of Acid Deposition in Japan was submitted to the ministry (MOE, 2004). This paper addresses the issue of precipitation chemistry based on the results from the JADS measurements.
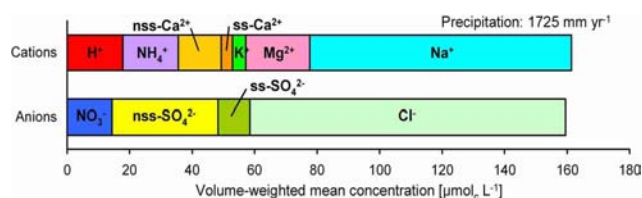

Figure 1. Mean composition of precipitation in Japan during the 1983-2002 period.

Mean composition of precipitation

The representative composition of the important ionic species in precipitation in Japan was derived from all the measurements during the survey period, 1983-2002. The data set used for the analysis was compiled and distributed by the Acid Deposition and Oxidant Research Center (ADORC, 2004). Stringent QA/QC procedures of data validity and data completeness were applied to the measurements at a total of 81 sites involved in the monitoring for at least one year during the survey period to yield a total of 554 year-site datasets. Information on the site can be found in earlier studies (Hara et al., 1990; Hara, 1993; Hara et al., 1995; Hayashi et al., 2005). The representative composition for Japan was calculated as the mean of each sites' annual values, which were calculated from the mean precipitation and volume-weighted mean concentration.

A feature of the precipitation composition in Japan was the large contribution of sea salt-derived ions, i.e., sodium (Na^+: 83.8 µmolc $^{-1}$) and chloride (Cl^-: 101 µmolc L^{-1}) ions, which accounts for 52% and 63% of the cation and anion sums, respectively (Fig.1). On average across the JADS sites, the ratios Na^+: Mg^{2+}: Cl^- = 0.83: 0.20: 1.00 for precipitation are close to the corresponding sea salt ratios of 0.85: 0.20: 1.00 for seawater. (Within various regions this ratio varied somewhat, where local sources added such species as HCl or Na^+ from soil dust). The sea salt contribution is explained by the geography, Japan being an archipelago located in the western Pacific. The non-sea salt sulfate (nss-SO_4^{2-}) concentration, 34.0 µmolc L^{-1}, is more than twice that of nitrate (NO_3^-), 14.3 µmolc L^{-1}, suggesting that the major acid

precursor for acid deposition in Japan is sulphur sulfur dioxide (SO_2). The sulfur dioxide or nss-sulfate is attributed to three sources in and around Japan: domestic anthropogenic, continental anthropogenic, and volcanic. The contributions of the domestic, continental, and volcanic sources to the nss-SO_4^{2-} deposited over Japan have been estimated to be 37-47%, 17-42%, and 11-45 %, respectively (Ichikawa, 1988).

Regarding reactive atmospheric nitrogen (Galloway and Cowling, 2002), the mean concentration of ammonium (NH_4^+), 17.9 $\mu mol_c L^{-1}$, is larger than that of NO_3^-, 14.3 $\mu mol_c L^{-1}$, where NH_4^+ and NO_3^- are interpreted to be due to a basic and an acidic species, ammonia (NH_3) and nitric acid (HNO_3), respectively. However, NH_3 acts as a univalent acid if included in the nitrification when deposited onto land and water surfaces. In terms of the potential acidity loading due to HNO_3 and NH_3, i.e., the sum of NH_4^+ and NO_3^-, 32.2 $\mu mol_c L^{-1}$, is on par with that of nss-SO_4^{2-}. An excess load of reactive nitrogen also has the potential to induce the status of eutrophication. Sources of ammonia in Japan are still poorly quantified, though the main sources are expected to be livestock excreta and chemical nitrogen fertilizers applied for agriculture. Vehicle exhaust and wastewater management may also contribute, particularly in urban areas.

A considerable amount of hydrogen ions (H^+) remains in Japanese precipitation after a high degree of neutralization of the original acidity with additional basic compounds. The concentration of the initial acidity, defined as the sum of nss-SO_4^{2-} and NO_3^-, was 48.3 $\mu mol_c L^{-1}$, whereas that of H+ was 17.7 $\mu mol_c L^{-1}$. This result indicated that 63 % of the initial acidity was neutralized by the coexisting base in precipitation. Based on the volume-weighted monthly means of the pH at each site during the survey period, the pH ranged from 4.60 as the first quartile to 5.15 as the third quartile with a median value of 4.80 (n = 7664).

Annual deposition

In order to assess the regional distribution of the measured species, Japan was divided into seven regions according to the characteristic sea or seacoast, as shown in Table 1: (1) Japan Sea–North (JS-N), (2) Japan Sea-Middle (JS-M), (3) Japan Sea-West (JS-W), (4) Pacific Ocean (PO), (5) Seto Inland Sea (SI), (6) East China Sea (EC), and (7) Southwestern Islands Sea (SW). The sites in each region were further classified into the three categories remote (Re), rural (Ru), and urban (U) (as defined by the Japan Ministry of the Environment), resulting in the 18 areas of interest. The annual mean deposition was assessed for each area (Table 1). We note that while some sites are classified as "remote", all JADS sites other than a few on offshore islands are close enough to domestic and overseas sources as to feel their influence. 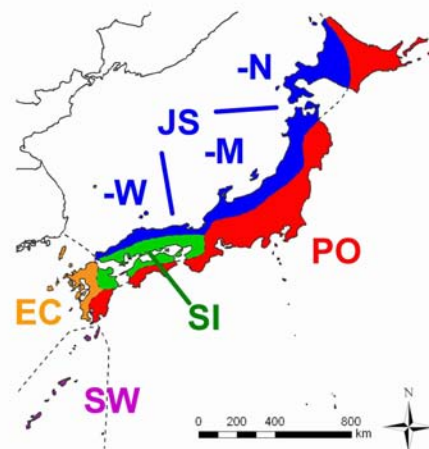In this sense the remote sites of JADS are not expected to be entirely free from air pollution. The wet deposition of nss-SO_4^{2-} in Japan was considerably larger than that in Europe and the United States. For example, the mean annual deposition values of nss-SO_4^{2-} from 2000-2002 in Europe and the United States was 11.7 5 mmol m^{-2} yr^{-1} (n = 262; CCC, 2005) and 9.5 mmol m^{-2} yr^{-1} (n = 705; NADP/NTN, 2005), respectively. On the other hand, that in Japan was 25.0 mmol m^{-2} yr^{-1} (n = 113; EANET, 2001; 2002; 2003), i.e., about twice that in Europe and the United States. Within Japan, a remarkably large deposition was noted in the JS-M, EC and SW regions (Table 1). In general, the deposition was high, not only in the urban sites but also in the rural and remote sites, and it remote sites, and it was correlated with the contributions of both local industrial emissions and long-range transported sulfur pollutants. Nitrate deposition did not

Region	C.	Prc. [mm yr-1]	Annual deposition [mmol m-2 yr-1]			Sites n
			nss-SO$_4^{2-}$	NO$_3^-$	NH$_4^+$	
JS-N	Re	720	10.1	8.8	13.7	1
	Ru	931	16.6	12.0	16.8	3
	U	974	20.3	11.8	20.6	1
JS-M	Re	1755	26.3	27.8	28.5	7
	Ru	1937	41.0	29.9	37.8	10
	U	1494	51.4	27.3	54.4	3
JS-W	Re	1348	19.0	21.6	22.4	1
	Ru	1806	30.6	22.3	28.1	1
	U	1559	23.0	25.6	23.9	3
PO	Re	2077	23.1	18.1	18.3	6
	Ru	1826	25.9	26.2	31.6	9
	U	1246	27.5	29.3	35.0	12
SI	Ru	1437	23.8	19.7	30.5	4
	U	1343	27.0	20.7	26.2	7
EC	Re	1743	25.5	23.9	31.3	2
	Ru	1859	33.7	18.8	34.0	2
	U	1676	36.1	24.2	36.1	3
SW	Re	3346	37.5	28.4	32.2	6
Japan	Re	2190	27.3	24.0	26.1	23
	Ru	1720	30.6	24.4	32.1	29
	U	1363	30.0	25.5	33.4	29

Region: JS-N: Northern part of the Japan Sea area, JS-M: Central part of the Japan Sea area, JS-W: Western part of the Japan Sea area, PO: The Pacific Ocean area, SI: The Seto Inland Sea area, EC: The East China Sea area, and SW: Southwestern Islands Sea area. C.; Re: Remote site, Ru: Rural site, and U: Urban site.

Table 1. Mean values of annual wet deposition in Japan during the 1983-2002 period.

vary remarkably from one area to the next in comparison with the nss-SO_4^{2-} deposition, although the NO_3^- deposition tended to be higher in the JS-M, PO, and SW regions. The ammonium deposition was very large in the JS-M, PO, and EC regions.

A general feature of the spatial distributions is that the deposition was commonly large in the JS-M and SW regions. Because the JS-M region faces the Asian continent and the SW region is a typically remote area of Japan, the large deposition in these regions suggests the long-range transportation of pollutants from China. In the JS-N region deposition was rather small for all species of interest, despite its location facing the continent.

This station is less influenced by the long-range transportation because of the different source-receptor relationships and reduced local pollution of the region, as given by Ohizumi (1999) and Hara et al. (2003).

Seasonal Changes in Monthly Deposition

The data were also analyzed to look at the seasonal (month-to-month) changes in deposition within each region. Sites

with more than three years of effective data were selected, and the monthly median deposition for each site was calculated. Furthermore, the monthly median depositions within each region were then compiled into a box-whisker plot. The data of the monthly deposition used for the analysis were not normalized for precipitation amount. The most dominant feature seen is the difference in the timing of the peak of the monthly deposition in the JS region versus the peak timing in the other regions (Fig.2). In the JS region, the monthly deposition increased in winter, while it increased in summer in the other regions. This tendency was observed among all the chemical species of interest, as discussed in detail for NO_3^- and NH_4^+ by Hayashi et al. (2005).

The monthly variation of the deposition is explained by the northwesterly winter monsoon in northeast Asia, where the airflow collides with the ridge of the mountains of the Main Island of Japan, resulting in precipitation along the Japan Sea coast. Northwesterly air parcels passing over the continent will transport the emissions from the area along the transport pathway, eventually resulting in elevated

resulting in lower pollutant deposition than in the other regions. However, it should be noted that while the amount of precipitation in the SI region is low relative to the other regions within Japan it is not low in a global context – i.e. the mean precipitation was about 1400 mm yr^{-1} in the SI region, while it was about 1700 mm yr^{-1} in the JS, PO, and EC regions and about 3300 mm yr^{-1} in the SW regions. In the EC and SW regions (map in Table 1), the volume of precipitation is relatively large, and considerably larger amounts of precipitation are added in association with the
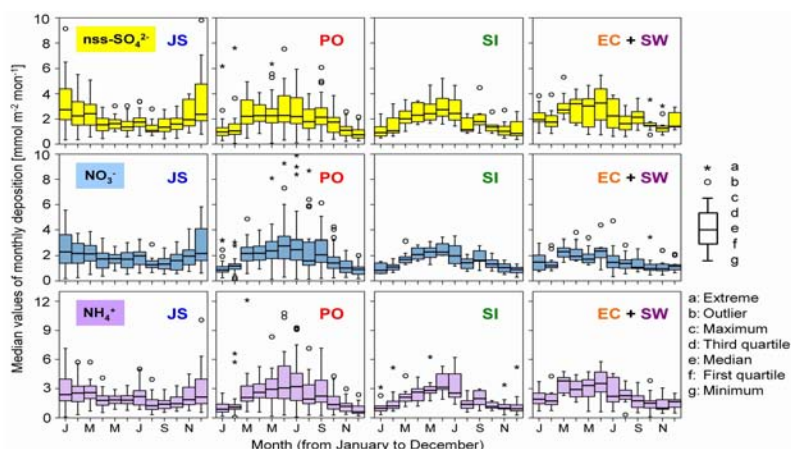

Figure 2. Seasonal changes in monthly wet deposition

a) Changes in monthly deposition [µmol m^{-2} mon^{-1} yr^{-1}]

Site	Area	nss-SO$_4^{2-}$	NO$_3^-$	NH$_4^+$	Period
TPP	JS	10	38	5	94-00; 94-02
WJM	JS	-160	-24	-127	94-00; 94-02
HPP	JS	-24	-36	41	94-00; 94-02
ECH	JS	-34	5	126	94-00; 94-02
KYS	JS	-108	16	-90	94-00; 94-02
TSM	EC	-102	111	105	94-00; 94-02
GTO	EC	116	64	-67	94-00; 94-02
YKS	SW	-80	166	51	94-00; 94-02

b) Change rate in monthly deposition [% yr^{-1}]

Site	Area	nss-SO$_4^{2-}$	NO$_3^-$	NH$_4^+$	Period
TPP	JS	0.8	2.6	0.4	94-00; 94-02
WJM	JS	-6.4	-0.8	-4.2	94-00; 94-02
HPP	JS	-1.5	-2.3	2.4	94-00; 94-02
ECH	JS	-1.3	0.1	3.5	94-00; 94-02
KYS	JS	-4.2	0.5	-2.8	94-00; 94-02
TSM	EC	-5.0	3.9	2.9	94-00; 94-02
GTO	EC	6.5	4.2	-3.6	94-00; 94-02
YKS	SW	-2.9	5.3	1.6	94-00; 94-02

Period: Until July, 2000, for nss-SO42- due to the eruption of Mt. Oyama in Miyakejima Island that started in August, 2000; Red value: A change rate of more than 1.0 % is considered to be on the rise; Black value: A change rate between -1.0-1.0 % is considered to be flat; Blue value: A change rate of less than -1.0 % is considered to be decreasing.

Table 2. Temporal trends in the monthly deposition at the eight remote sites in Japan

Baiu fronts and typhoons in the summer and autumn. The deposition in these regions is easily affected by the polluted air mass from the continent because of their close vicinity to the continent.

Temporal Trends of Monthly Deposition in Remote Sites

The trends in the monthly deposition of nss-SO$_4^{2-}$, NO$_3$, and NH$_4^+$ were evaluated for eight remote sites at which the same sampling techniques were used over more than six years until 2002 (Table 2). For nss-sulfate, the data in and after August 2000 were excluded from the analysis because of the emission of vast quantities of SO$_2$ from Mt. Oyama on Miyakejima Island after its eruption in August 2000.

Time-series of the monthly deposition data is ideally given as the sum of a linear long-term trend, seasonal variation, and error. Further, when doing the trend analysis we note that, for a given site and month, the deposition data are log-normally distributed (Hayashi et al., 2005). These considerations are consistent with a model (1) with a non-linear function of the logarithmically transformed deposition with time, where the function is the sum of the long-term trend and the seasonal variation with annual cycle. However, it is preferable for a

wet deposition of chemical species along the coast in wintertime. As the atmospheric water vapor passes over the mountainous ridge, it is deposited as precipitation with the so-called wash-out of pollutants in the atmosphere. Hence, the amount of rainfall (or precipitation) and the wet deposition of the pollutants steeply decrease during the winter months particularly in the PO region (map in Table 1) located in the lee of the mountainous ridge. In the SI region (map in Table 1), which is isolated from the surrounding regions by mountains on the northern and southern boundaries, the volume of precipitation is relatively less throughout the year,

model to have a smaller number of explanatory variables. Thus, a model (2) with a linear function of the logarithmically transformed deposition with time was also prepared. Furthermore, two more models were considered: (3) one with a non-linear function of the deposition with time, given as the sum of a long-term trend and a seasonal variation with annual cycle, and (4) one with a linear function of the deposition with time. The most appropriate model for the temporal trend, in terms of Akaike's Information Criterion (AIC) (Akaike, 1973), proved to be model (2). The linear term of the model was interpreted to express the temporal trend, which prompted us to calculate the average changes in monthly deposition per year (mmol m^{-2} month^{-1} yr^{-1}) and its rates (%) calculated relative to the monthly deposition in January 2004. Table 2 shows the temporal trends of the monthly deposition in the eight remote sites in Japan.

Nss-sulfate deposition decreased from 1994 until July 2000 (Table 2), which is ascribed to the decrease in SO2 emission from both domestic and continental sources. Then, as given by Noguchi et al. (2005), the nss-SO$_4^{2-}$ deposition peaked with the eruption of the Mt. Oyama volcano in August 2000. Deposition decreased in the post-eruption period of 2001-2002, though the magnitude of the deposition remained high in comparison to the pre-eruption levels. Thereafter, SO$_2$
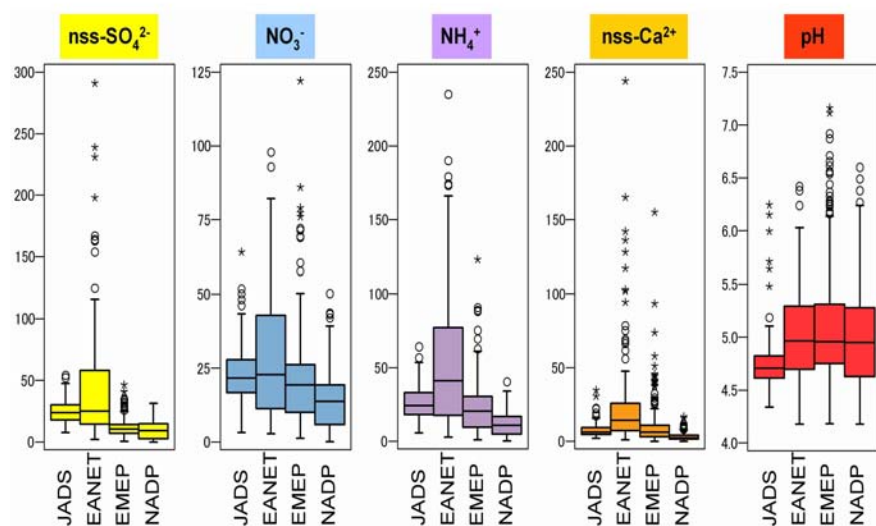

Figure 3. Comparison of the annual deposition in Japan, East Asia, Europe, and North America.

emission in China started to increase after 2003 (SEPA, 2005). This may have contributed to maintaining the relatively high levels of nss-SO$_4^{2-}$ deposition in Japan.

Nitrate deposition was on the rise at half of the remote sites. It is known that an oxidation rate of nitrogen dioxide (NO$_2$) into HNO$_3$ is tenfold higher than that of SO$_2$ into sulfuric acid (H$_2$SO$_4$) (Calvert and Stockwell, 1983). Therefore, the contribution of the domestic sources to the NO$_3^-$ deposition is generally higher than it is to nss-SO$_4^{2-}$ deposition. The non-decreasing trends apparently correspond to the fact that the emission reduction of NO$_2$ in Japan had not progressed compared to that of SO$_2$; hence, the atmospheric

concentrations of NO$_2$ in Japan have not remarkably improved but, rather, leveled off or improved only slightly (MOE, 2003). However, long-range transport also seems to have a considerable effect on the NO$_3^-$ deposition, particularly in the vicinity of the Asian continent; the increasing trends in deposition at the TSM, GTO, and YKS sites (Table 2) suggest the effect of increasing transportation from the Asian continent.

Ammonium deposition increased at some sites and decreased at others, showing no geographic pattern in the trends. This tendency reflected a large contribution of the local NH$_3$ sources to the NH$_4^+$ deposition because of the property of NH$_3$ of being highly water soluble and easily scavenged by precipitation. The relative contribution of long-range transport is, therefore, relatively low for this species. Only a part of the ammonium salts originated from the neutralization with NH$_3$ and acids in the atmosphere are transported over distances. However, the neutralization has the simultaneous effect of transporting acids over distances, which is particularly effective for acids with a high deposition velocity, such as HNO$_3$; NH$_3$ is also important in this sense.

Finally we note that because the deposition measurements are not normalized for total precipitation amount, trends in precipitation would affect deposition trends. However, trends in monthly-averaged precipitation were not large for any of the sites considered, and they varied in direction: Precipitation rate was flat at the WJM site, had a decreasing trend of 2 % yr^{-1} at the ECH site, and increased by 2-5 % yr^{-1} at the other sites.

Comparison with regional measurements in Asia, Europe, and the United States

Figure 3 shows a comparison of the JADS measurements of the atmospheric deposition of nss-SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, and non-sea salt calcium (nss-Ca$_2^+$) and of the precipitation pH with those from other regional measurements in the period 2000-2002. Comparisons are made to those in East Asia (EANET2001; 2002; 2003), Europe (EMEP; CCC, 2005), and the United States (NADP; NADP/ NTN, 2005).

In terms of the annual median deposition, the JADS deposition levels were higher than for EMEP (Europe) and NADP (U.S.) for nss-SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, and nss-Ca^{2+}. However, EANET (E. Asia) and JADS show comparable levels of annual median deposition for nss-SO$_4^{2-}$ and NO$_3^-$, and the JADS deposition levels were much lower than those for the E. Asian data for NH$_4^+$ and nss-Ca^{2+} (Fig. 3). Regarding acidity, the median pH of JADS was lower than those for the other regional networks (Figure 3). In the other countries in East Asia, very high concentrations of acids were largely neutralized by the high concentrations of basic compounds (i.e., NH$_4^+$ and nss-Ca^{2+}). In Japan, the degree of neutralization was relatively low because there were much lower concentrations of basic

compounds than there were of acidic compounds in the precipitation.

Conclusion

In general, Japanese soils are slightly acidic, mainly due to the long-term leaching of base cations induced by the pluvious climate, in which the extractable cations adsorbed onto the soils are replaced by H+ and aluminum ions ($Al_3{}^+$). Japanese ecosystems are considered to have acquired resistance properties against the naturally acidic conditions. The fact that no significant damage to ecosystems from acid deposition has been reported in Japan is perhaps ascribed to this natural tolerance. However, the deposition intensity of acids and related compounds in Japan has been observed to be as large as that in Europe and the United States, which warrants no optimism. In order to detect some of the symptoms of potential effects, the nationwide system of monitoring atmospheric deposition and relevant environmental elements should continue. The monitoring should focus on aspects of atmospheric deposition other than acidification. Nitrogen deposition, which can seriously affect the environment and even cause eutrophication, is an urgent concern.

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Improving the Representation of Wet Scavenging in Large-scale Models: A Call to the Scientific Community

Contributed by **Jose M. Rodriguez** (jrodriguez @hyperion. gsfc.nasa.gov), *University of Miami/NASA-Goddard Space Flight Center;* **Athanasios Nenes** (nenes@eas.gatech.edu), *School of Earth and Atmospheric Science, Georgia Institute of Technology;* **Daniel J. Jacob** (djacob@fas.harvard.edu), *Department of Earth and Planetary Science, Harvard University.*

Aqueous-phase processes play a crucial role in the chemical transformation and removal of constituents that are important in determining air quality, rain acidity, chemical reactions

involving other components, and radiative balance in the Earth's atmosphere. Oxidation of SO_2 to sulfate has been shown to occur primarily through cloud processes mediated by dissolved H_2O_2, O_3, and metallic ions (Seinfeld and Pandis, 1998). Sulfate production in turn contributes an important component of the tropospheric aerosol loading, and, along with dissolved nitrate, contributes to increases in rain acidity. Incorporation of soluble species, such as HNO_3, and H_2O_2, constitute a permanent sink of nitrogen oxides (NO_x), and hydrogen oxides (HO_x), which play the dominant role in ozone production in the troposphere. Such removal can occur by either in-cloud solution of these species, followed by downward transport in hydrometeors (rain out), or by below-cloud collection by rainfall ("washout"). Such wet scavenging processes also play a dominant role in the removal of a whole suite of tropospheric aerosols, thus determining regional and global aerosol loadings, and the magnitude of direct and indirect effects of aerosols (Seinfeld and Pandis, 1998).

Accurate representation of these processes in regional and global models is plagued by a host of difficulties due to the complex interaction of meteorological, microphysical, and chemical processes involved. Uncertainties still exist in the physics and chemistry of cloud formation and in the subsequent interaction of cloud droplets with gas molecules and interstitial aerosol. Even if the physics and chemistry of the aerosol cloud interactions were perfectly understood, the fact that these processes take place on unresolvable (or sub-grid) spatial scales in large-scale models introduces additional complications. In addition, it is difficult to test the accuracy of such representations in global models using atmospheric measurements. A concerted effort at evaluating the representation of wet and dry deposition processes was last carried out in 1995, under the auspices of World Climate Research Program (Rasch et al., 2000). This effort compared the modeled deposition of ^{210}Pb and sulfate by a host of different models with different degrees of sophistication in the representation of wet scavenging and dry deposition. In addition, model results were compared with available measurements of ^{210}Pb and sulfate deposition, as well as with measurements of the concentrations of these species at different altitudes. Although the model results agreed with each other (for example, showing a general agreement on the lifetime for sulfate aerosols in the lower troposphere) and they agreed with measurements in reproducing some features (such as the seasonal increase in sulfate burden in the summertime) there was a large spread in the modeled composition and deposition fields (by factors of 4 to 5 in some cases), in spite of the fact that emissions of sulfur and ^{222}Rn (the parent atom to ^{210}Pb) were prescribed in most cases. Simulations including a detailed treatment of SO_2 oxidation in clouds did better than those using a prescribed conversion time of 1.2 days, indicating that the proposed conversion time was underestimated. However, there was no further obvious connection between the level of sophistication of the wet scavenging representation and model performance.

Recently, IGAC has identified the improvement of wet deposition and scavenging parameterizations for large scale models as an area meriting more activity (see this newsletter's introductory "Note from the Chairs"). The

catalyst for this increase in attention is 1) the improvement in understanding of the component processes gleaned from the vast number of laboratory studies, field measurements, and component modeling developments that have taken place in the past decade; and 2) a recognition within the community that these processes are very important to atmosphere chemistry and climate, but are treated very crudely in current large scale models. Evaluating the impact of the uncertainties in wet deposition on gas-phase concentrations and aerosol loadings has also been identified as an important area of research for NASA's Global Modeling Initiative (GMI; *http://gmi.gsfc.nasa.gov*). As a focus to this research priority, IGAC and GMI are planning to sponsor a workshop on wet deposition processes in large-scale models during 2007. The goals of this workshop would be to: a) summarize in a concerted manner our current knowledge of the wet scavenging processes and their representation in large scale models; b) determine the major uncertainties in the above representations and the impact of these uncertainties on modeling of air quality and climate; c) propose a coordinated modeling effort and recommend a strategy for producing the observations that would be required to reduce remaining uncertainties in the representation of wet scavenging.

The representation of wet deposition in large-scale models has a long history, ranging in sophistication from simple first-order removal of species (Logan et al., 1981), to more complex representations that include consideration of removal processes in convective updrafts, cumulous anvils, and large-scale cloud systems (Giogi and Chameides, 1985; Balkanski et al., 1993; Liu et al 2001.) These representations, for the most part, still rely on "tuned" parameters and assumptions for liquid water content, thermodynamic equilibrium, and cloud frequency,among others (Liu et al. 2001, Tost et al., 2006). Developments in cloud microphysical modeling can be incorporated into large-scale models to provide a more self-consistent representation of wet scavenging. There is also a need to better understand the retention efficiency of gases upon cloud freezing (Mari et al., 2000). These algorithms are also dependent on the model's representations of other sub-grid processes, such as convection and updraft velocities. A concerted effort to evaluate the impact of uncertainties in current representations of wet scavenging on atmospheric composition and aerosol loading is needed.

Efforts to improve the existing representations of wet-scavenging in global models would ideally take into consideration recent advances in laboratory experiments, atmospheric observations, and dynamical modeling, in order to develop physically-based representations that simulate well current observations and at the same time give us confidence in the model's response to different forcings. The purpose of this note is to suggest areas of discussion, and to call on the community's expertise in further defining the agenda, and, specifically, to solicit contributions to the workshop's organization. We can identify at this point the following themes for the workshop:

• *What are the uncertainties in the current parameterizations of wet scavenging in large-scale models?* The large spread in the results summarized by Rasch et al. (2000) were difficult to diagnose, due to the fact that participating models differed in their choice of meteorological fields, convective

parameterizations, advection algorithms, and chemistry. A concerted effort is needed to conduct parametric studies of wet scavenging representations that keep all other model components fixed. New techniques for sub-grid representation of other processes may be applicable to wet scavenging. Such studies would also point to information from other scientific disciplines (laboratory measurements, atmospheric measurements, small-scale process studies) that could constrain the large-scale simulations.

• *How do recent laboratory measurements help us in improving the parameterization of cloud activation and processing?* Although it is hard to completely reproduce the atmospheric conditions in the laboratory, important developments have occurred in our understanding of the activation of CCNs and ICNs, the required conditions of supersaturation, the dependence of activation on aerosol compostion, and hysterisis effects (e.g., Abbat et al., 2005; Archuleta et al., 2005; DeMott et al., 2003; Hori et al., 2003; Koop et al., 2000; Kreidenweis et al., 2006; Kumar et al., 2003; Raymond and Pandis, 2003).

• *How can cloud-resolving dynamical and microphysical models help us understand the couplings in the system in a way that will allow improvements in the sub-grid parametrizations in global models?* Two-dimensional and three-dimensional cloud-resolving models and Large Eddy Simulations (LES) have incorporated more complex processes in their formulation (e.g., Ackerman et al., 2004; Fridlind et al., 2005; Jiang et al., 2006; Yin et al., 2005). Parameterizations for cloud and ice activation have been developed that take into account the underlying physics and chemistry of cloud activation (e.g., Abdul-Razzak et al., 1998,2000; Fountoukis and Nenes, 2005; Karcher et al., 2006; Khvorostyanov and Curry, 2000,2004; Koop et al., 2000; Liu and Penner, 20005; Nenes and Seinfeld, 2003; Rissman et al., 2004). Some cloud models have also incorporated in-cloud aqueous processes to evaluate the transport, processing, and removal of gases and aerosols (e.g., Mari et al., 2000; Nenes et al., 2002; Xue and Feingold, 2004). Simplifications that invoke thermodynamic equilibrium need to be evaluated against more complete non-equilibrium representations.

• *How can recent field campaigns help us constrain existing uncertainties in cloud processes and their incorporation into large-scale models?* Coordinated measurement campaigns (e.g., CRYSTAL-FACE, CSTRIPE, ICARTT, MASE) have provided important constraints on cloud models and have produced information on water and ice cloud composition, supersaturation conditions for activation, and interactions with atmospheric constituents (e.g., Conant et al., 2003; Meskhidze et al., 2005; Fridlind et al., 2005; Fountoukis et al., *in review*)

• *What measurements exist, or are needed, in order to test the representation of wet deposition in large-scale models?* The GMI model has recently been used to evaluate uncertainties in modeled ^{210}Pb due to adoption of different meteorological fields (Considine et al., 2005), but this evaluation did not explore the whole parameter space of uncertainties in microphysical, meteorological, and sub-grid processes. Overall, the available data for testing modeled wet scavenging may not be sufficient to constrain the model assumptions. Well-defined experiments are needed to further constrain model simulations.

The ultimate goal of this workshop is to produce a research plan of action that will aid researchers and funding agencies in determining the next best steps to addressing this important problem. We thus encourage you to communicate to us your thoughts on the matter, as well as any specific roles/contributions you would like to make to the organization of this workshop. Interested parties will be contacted in the near future with specifics as to the time and location of this workshop, and possibly as well with information on financial support for participation where needed. Please send your comments to us via Sarah Doherty at the IGAC Seattle Core project office: *igac.seattle@ noaa.gov*.

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IGAC/WMO Workshop, Santiago De Chile, October 13-14 2005
From Chemical Weather Forecasts to Climate Change in South America: The Challenges and Opportunities of Integration and Collaboration

Contributed by **Laura Gallardo** (lgallard@dim.uchile.cl), *Centro de Modelamiento Matemático, Universidad de Chile, UMI CNRS 2087, Casilla 170-3, Santiago, Chile;* **Eugenio Sanhueza** (esanhuez@quimica.ivic.ve) *Laboratorio de Química Atmosférica, Centro de Química, IVIC. Apartado 21827, Caracas 1020A, Venezuela;* **Laura Dowidowski** (dawidows@cnea.gov.ar) *Grupo Monitoreo Ambiental (GMA), Unidad de Actividad Química (UAQ), Comisión Nacional de Energía Atómica (CNEA), Avenida General Paz 1499, B1650KNA San Martín, Argentina; and* **Karla Longo** (longo@cptec.inpe.br) *Centro de Previsão de Tempo e Estudos Climáticos – INPE, Rodovia Presidente Dutra, Km 40, SP-RJ, 12630-000, Cachoeira Paulista, SP, Brazil.*

Background

This past October, the Scientific Steering Committee (SSC) of IGAC held its annual meeting in Santiago de Chile, in coordination with a joint IGAC/WMO workshop "From Chemical Weather Forecasting to Climate Change in South America". This workshop brought together key people working on atmospheric chemistry research and monitoring in the Americas south of the Río Grande. The end goal was to promote collaborative international atmospheric chemistry

research and monitoring, building from existing projects and facilitating the formulation of new projects and collaborations. The long term atmospheric chemistry measurement program of the WMO's Global Atmosphere Watch (GAW) program in South America and nationally-based air pollution monitoring networks comprise the fundamental starting points for this effort, and their work to date was discussed during the meeting.

The two-day workshop was hosted by the Center for Mathematical Modeling at the University of Chile (*http://www.cmm.uchile.cl*). In addition to IGAC SSC and ex-officio members, thirty eight (38) scientists from the Americas, representing eight countries, attended the symposium. During the first day scoping talks were presented by leading scientists from IGAC, GAW and South American institutions. These talks were followed by discussion groups in which South American, IGAC, and GAW representatives participated. The resulting discussions made clear several areas in which international collaboration is both needed and feasible. These areas are: implementation, via connection of regional centers, of chemical weather forecast in South America; improvement and deployment of monitoring networks; and establishment and coordination of non-static emission inventories.

Chemical Weather Forecast In South America: A Coordination Effort

Over the last 20 years, the extensive biomass burning occurring in the Amazon basin has been a hot spot for atmospheric research in South America. The magnitude of the problem, the enormous challenges posed by it, and the sagacity of local scientists to convey global concern and collaboration led to an astonishing growth and development of local research capabilities in Brazil (e.g. see the accompanying article by Longo et al., as well as *http://lba.cptec.inpe.br*). Air quality degradation in the urban areas of South America -- home to 75% of the area's population -- presents scientific challenges as difficult as those being addressed with respect to biomass burning in the Amazon (Figure 1). We believe that a similarly focused approach to this issue could also result in improved research capabilities and, more important, in sound environmental strategies and a reduction in the detrimental impacts of urban emissions both locally and globally.

In contrast to the starting point for the studies of biomass burning in the 1980's, research on air quality in South American megacities would not start from scratch. Numerous and expensive efforts are already in place (e.g., several cities such as Lima, Santiago, and Sao Paulo already have implemented ambitious attainment plans). This is particularly true when referring to air quality modeling and its operational implementation "chemical weather forecasting". Since state-of-the-art chemistry-transport-deposition models are easy to access and required computational resources are affordable for most institutions (e.g., universities, weather offices, etc.), there are an increasing number of groups making air quality applications. In addition, the Brazilian Center for Weather Forecast and Climate (CPTEC) already has an operational numerical system for regional weather and air quality forecast (*http://www.cptec.inpe.br/meio_ambiente/; Freitas et al.,*

2005). This provides carbon monoxide, particulate material concentration and aerosol optical thickness from biomass burning and urban emissions in South America. The Brazilian team is now working to also include the tropospheric ozone forecast for the next biomass burning season. Finally, fast internet connections are already available (*http://www. redclara.net/en/index.htm*). making it possible to develop grid computing applications, particularly within the framework of atmospheric physical and chemical forecasts.

With these components as building blocks, the challenge is to coordinate the ongoing efforts and capabilities and to ensure a sound scientific basis for the applications. Of course this is not a trivial endeavor as it must face cultural and institutional difficulties as well as resource constraints in such a wide and diverse area of the world, where poverty and inequality has deep roots. However such an endeavor is feasible and would be beneficial both scientifically and economically not only for the countries in the region but for the whole globe. Fortunately things are moving forward at various levels, including political agreements (e.g.,*http://www.worldbank.org/wbi/airelimpio/*) and concrete projects.
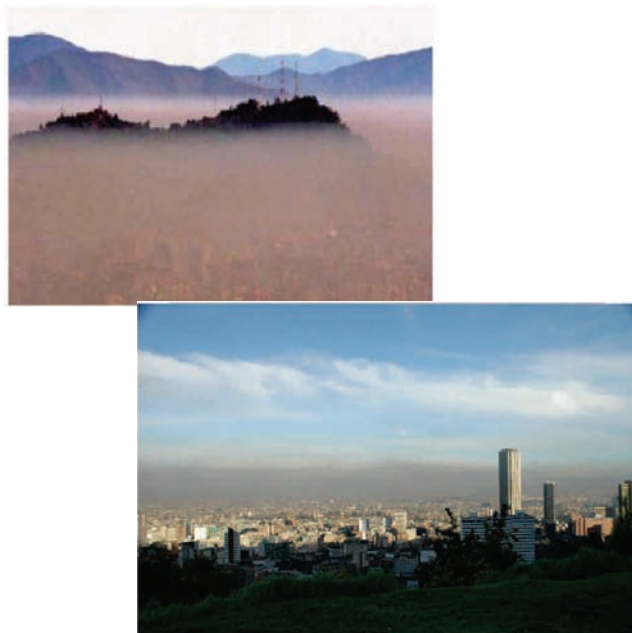

Figure 1. Pollution, trapped against the Andes in Santiago de Chile (top) and over Bogotà, Columbia, (bottom).

Among the latter, the WMO GAW Urban Research Meteorology and Environment (GURME) project (*http://www.wmo.ch/web/arep/gaw/urban.html*) has put resources into network and capacity building. Also, the Inter American Institute for Global Change (IAI, *http://www.iai.int/*) has financed a four year project that contains a pilot implementation of a chemical weather forecast network and tools for South American megacities, starting with the connection of CPTEC and the Chilean Weather Office for the purpose of operational chemical weather forecasting.

Altogether, the enormous challenge of air quality degradation in South American megacities offers an opportunity to improve and enhance hitherto disperse research capabilities, accelerate local know-how and capabilities, and to produce scientifically sound and policy-relevant knowledge and tools.

Observing The Changing Atmosphere Of Southern And Central America

Considering the extensive land area, diversity of ecosystems and climatic zones, and the strength and number of global change driving forces and vulnerabilities present in South and Central America, there is an appalling lack of observations of its changing atmosphere. Except in the Amazon region, the situation has not changed much in this respect since the early 1980's — i.e. even before the inception of IGAC. Nevertheless, a few key efforts have been made in this time. For instance, starting in the mid 1990's the Global Atmospheric Watch (GAW) program installed what is now up to ~20 regional stations in the area. Several of these have 10 years of high-quality data records (e.g., http://www.empa.ch/gaw/gawsis/). Considerable resources have also been invested by local authorities in urban areas and around industrial sources to monitor and characterize severe pollution problems.

The efforts referred to above, while constituting an excellent starting point, need updating and better coordination to really contribute to a better understanding of processes and to provide the basis for improved assessments. A common regional meteorological and chemical database could be of use for promoting coordination, providing quality assurance standards, and most important for making data available. Also, as there is an increasing need for and cost in instrument calibration and maintenance, more intensive use and sharing of local laboratory and analytical facilities is badly needed. Last but not least, the active involvement of researchers and graduate students in the region and elsewhere in the analysis of data collected in the various institutional observing networks must be strongly encouraged, for example, via small grant programs and dedicated workshops.

All in all, the slowly growing observational capabilities in Southern and Central America need to be reviewed, improved and better coordinated. Coordinating efforts and active involvement of researchers are badly needed and IGAC and GAW could play a crucial role in this matter.

Building Sound Emission Inventories For South America

Emission inventories are key tools for air quality management and are a starting point for the overall understanding of the processes leading to air quality degradation and ecological and climatic impacts. Developing emission inventories is an extremely complex process, which requires detailed statistics, process-level understanding, and continuous evaluation and updating (Granier et al, 2004). Furthermore, in order to make emission inventories fully useful and credible, their building process and results must comply with transparency, completeness, consistency, comparability, and accuracy requirements.

Hence, it is not surprising that South America lacks a sustained activity and record for building reliable emission inventories. The most coherent efforts have been developed under the IPCC, and in a few cities within the framework of the implementation of attainment plans (e.g., Santiago, Sao Paulo,

Bogotá; *http://www.dim.uchile.cl:8080/umesam/ emissions*). Unfortunately, these efforts have been largely decoupled from international networks and restricted to a few activity sectors – i.e. mainly urban and industrial energy use and transportation, leaving aside rural and natural emissions. In addition, the methodologies used are often poorly adapted to local and regional conditions increasing the uncertainty of the estimates.

To improve the current situation it is necessary to promote coordination activities to reconcile global inventories with, when available, regional, country and city-specific data, and to put in place validation tools, including inverse modeling. To carry out these activities in the region, it would be important to establish a network involving not only members of the scientific sector but also technical staff from governmental agencies who are in charge of emission inventories, to assure meeting the interests of both groups while making efforts to harmonize the different approaches.

Summary

South America offers an extraordinary opportunity for the atmospheric research and monitoring needed to address issues of global change. Large contrasts in air quality exist between the cleanest air on Earth (as measured at the GAW Global station Ushuaia) and the extremely polluted air in major cities of the region. It has the most extensive and persistent stratocumulus deck of the world as well as deep convection over the Andes and pyrocumulus in the tropics. Such complexity constitutes itself a scientific challenge. However, except in Brazil, where there is a solid body of atmospheric chemistry research, the local capabilities in most South American countries are still sparse. Fortunately this is starting to change through collaborative efforts such as those promoted by IGAC and the Global Atmosphere Watch (GAW) program of WMO.

This opportunity is also a challenge for South and Central American researchers as it requires new ways and attitudes for driving research, crossing disciplinary and national borders, and defining new paradigms and organizational modes. We have to identify those key scientific questions that gather our capabilities, which are regionally and globally relevant, and that will enlarge our scientific bodies and its connections to decision-making and the international scientific community. Three such issues were identified during the Santiago meeting: chemical weather forecast, observing capabilities, and emission inventory building. These issues all have common requirements: international and regional coordination, networking and capacity building. The IGAC/WMO workshop this past October was a fruitful starting point to addressing these issues, and we anticipate ongoing efforts within GAW, IGAC and other relevant IGBP projects to attack the issues of chemical weather forecasting and climate change in the South American region.

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Numerical Modeling Developments Towards A System Suitable To A Real Time Air Quality Forecast And Climate Changes Studies In South America

Contributed by **Karla Longo** (longo@cptec.inpe.br) *and* **Saulo Freitas** (sfreitas@cptec.inpe.br), *Centro de Previsão de Tempo e Estudos Climáticos – INPE, Rodovia Presidente Dutra, Km 40, SP-RJ, 12630-000, Cachoeira Paulista, SP, Brazil,* **Maria Assunção Silva Dias** (assuncao@cptec.inpe.br), *Centro de Previsão de Tempo e Estudos Climáticos – INPE and Instituto de Astronomia, Geofísica e Ciências Atmosféricas, and* **Pedro Silva Dias** (pldsdias@master. iag.usp.br) *Instituto de Astronomia, Geofísica e Ciências Atmosféricas, SP, Rua do Matão, 05508-900,São Paulo, S. P., Brazil.*

Introduction and Background

The South American continent presents an extraordinary diversity of land use, vegetation and soil coverage. One area where this diversity is exemplified is the Rondonia state of SW Amazonia (Figure 1). As a consequence of the landscape variety, trace gases and aerosol emission sources are highly variable in space and time. Not only is the actual spatial emission pattern very complex but the time changes observed in the last decade have been immense. The annual deforestation area, just in the Brazilian Amazonia, ranged between 11,030 and 27,362 km^2 in the last 18 years for a total of 333,037 km^2 of deforestation according to the satellite monitoring program of INPE (The Brazilian National Institute for Space Research; *http://www.obt.inpe. br/prodes*)
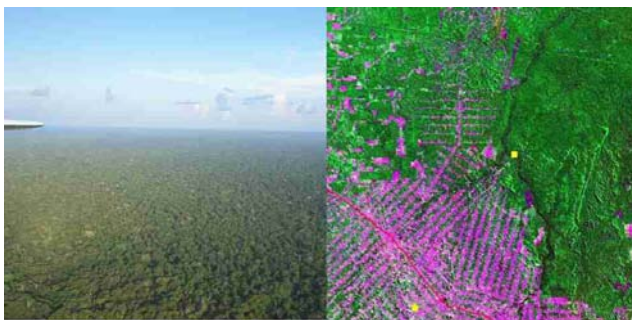

Figure 1. The Amazon "Green Ocean" (*courtesy M.O. Andreae*) stands in contrast with the fishbone deforestation pattern (*LANDSAT image, courtesy NASA*), both of which are found in the Brazilian Rondonia State.

Several hundred of thousands of vegetation fires are regularly detected in South America during the austral winter everyyear. This is driven by the fact that biomass burning is an agricultural practice often used for expanding the crop and livestock land. The fire activity affects mainly the forest and savanna biomes but it is also extensively used for the renewal of pasture and

agricultural areas. In spite of the undergoing degradation, vast areas of forest land still remains, comprising Brazil's so-called "Green Ocean" (Andreae et al, 2004). Biogenic emissions are the prime controller of atmospheric chemistry in the Amazon basin, except during the dry season. From July to October biomass burning emissions change the atmospheric composition at both the local and continental scale, with potential impact even in the global scale. The emission characteristics vary widely with the type of vegetation burned, not only in composition but also in terms of the height where the material is injected.

On a continental scale, a large contrast exists between pristine and extremely polluted areas due mainly to the vegetation fires emission but also to urban/industrial sources. Satellite observations have been detecting huge regional smoke plumes in South America covering approximately 4 to 5 million km^2 (Figure 2). Depending on the synoptic conditions, the regional smoke plume can invade pristine areas in the northern part
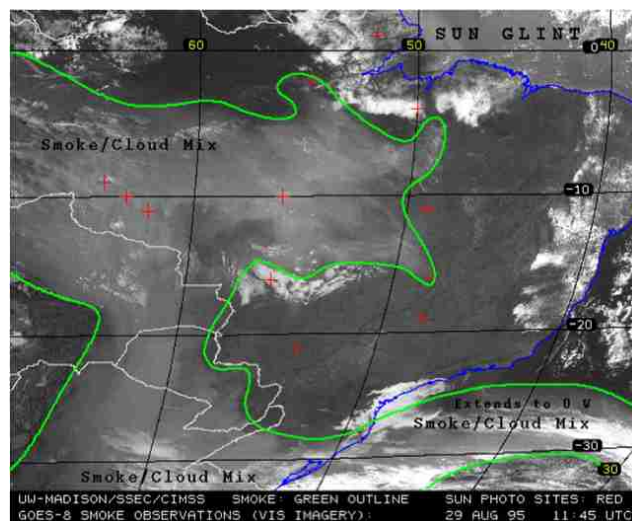

Figure 2. GOES-8 visible imagery with the smoke plume contour (green line) on 29 August 1995 from the UW/ SSEC/ CIMSS .

of the Amazon (Cordova et al., 2004) or it can be transported southward affecting highly urbanized and populated areas (Freitas et al., 2005). Recent studies point to the strong direct and indirect effect of the smoke aerosol on radiative balance and precipitation patterns (Andreae et al., 2004, Koren et al., 2004) and on photochemistry (Albuquerque et al., 2005).

At the local scale, South American mega-cities such as São Paulo and Santiago de Chile are facing severe air pollution problems associated with vehicular and industrial sources. In contrast, much smaller towns in the Amazon are highly affected by the smoke pollution, where the air quality index for tropospheric ozone and aerosol particle concentrations can be even worse than those observed in the polluted mega-cities.

The intrinsic complexity of the South American atmospheric air pollution problem associated with biomass burning represents a challenge to the modeling community. This environmental problem requires modeling of the integrated system, taking into account the inter-relationship between the biosphere and the atmosphere and between different time and spatial scales. On the other hand, from the operational

point of view, models must be simplified as much as possible for the real-time application of weather and air quality forecasts in view of the computational constraints.

Figure 3. Top: Pirocumulus over a deforestation fire and a smoke-polluted PBL in the Amazon. Bottom: Polluted PBL in São Paulo, and the town of Ji-Paraná (SW Amazonia) immersed in a thick smoke haze layer. (Courtesy M.O. Andreae and A. Castanho)

In this paper we describe the current status of an on-going effort at CPTEC-INPE (Center for Weather Forecast and Climate Studies; São Paulo, Brazil) to develop an operational air quality system applicable at the regional and local scale for the South American continent.

Biomass Burning Emission Estimation

Biomass burning emissions inventories have been developed for South America through a combination of remote-sensing products (land use and fire detection) and field observations of quantities such as emission and combustion factors and fuel loadings (Brazilian Fire Emission Model - BFEMO, Freitas et al., 2005). This methodology has been implemented in near real time (NRT) to provide daily emission estimates for the CPTEC/INPE air quality forecast system. Fire location is provided with a blending of WF_ABBA (Wild Fire Automated Biomass Burning Algorithm, Prins et al., 1998) and INPE *http://www.cptec.inpe.br/ queimadas* fire products. Figure 4 presents a comparison between the CPTEC/INPE methodology and two other biomass burning inventories: that derived from the TOMS aerosol index (Duncan et al., 2003) and that given by the Emission Database for Global Atmospheric Research, EDGAR 3.2 (*http://www.mnp.nl/ edgar; Olivier, 2002*), for August 2002. This inventory shows comparable total integrated emission estimation and introduces new features such as the diurnal variability and a higher spatial resolution, which is dictated by the atmospheric model. In addition, as the sources are spatially and temporally distributed and are assimilated on a daily basis, it is possible to improve the air quality forecast skill because the biomass burning emissions are regularly introduced in the atmosphere at the right time and position. This methodology is suitable for regional applications as well as for large-scale atmospheric transport models.
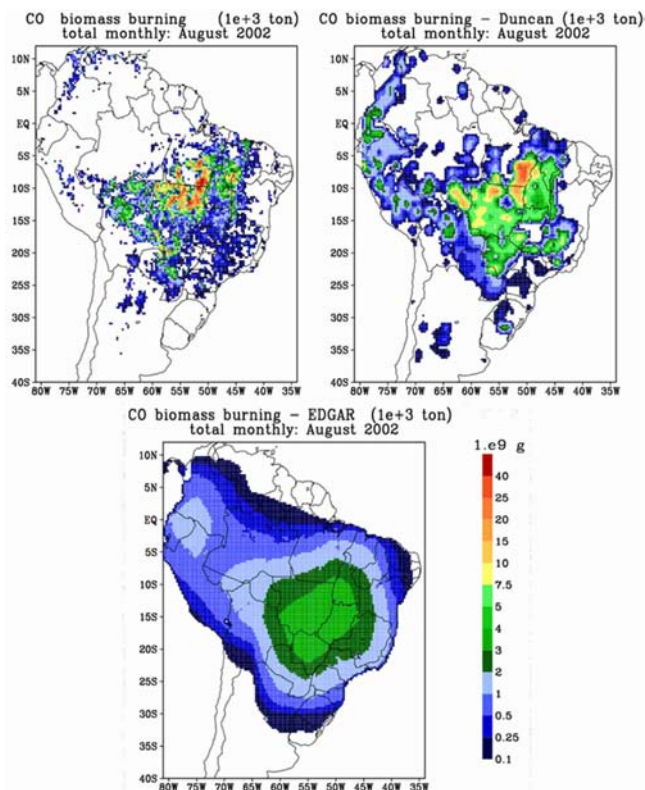

Figure 4. Carbon monoxide emission estimations from biomass burning for August 2002 are shown: (left) as given by the Brazilian Fire Emission Model (BFEMO; Freitas et al., 2005); (middle) as derived from the TOMS Aerosol Index (Duncan et al., 2003); and (bottom) as given in the EDGAR 3.2 emission inventory.

CATT-BRAMS: Numerical Tool Description And Validation

The atmospheric transport of biomass burning emissions in the South American and African continents is monitored year-round by a numerical simulation of atmospheric transport called CATT-BRAMS (Coupled Aerosol and Tracer Transport model to the Brazilian developments on Regional Atmospheric Modeling System; Freitas et. al., 2005). The BRAMS, *www.cptec.inpe.br/brams*, a Brazilian version of RAMS tuned for the tropics, is a multipurpose, numerical prediction model designed to simulate atmospheric circulations spanning from hemispheric scales down to the scale of large eddy simulations of the planetary boundary layer (Walko et al., 2000). CATT is an in-line Eulerian transport model fully consistent with the BRAMS dynamics and a coupled fire emission model. The sub–grid transport parameterizations include convective transport by shallow and deep convection, PBL turbulence with TKE closure, and a plume rise mechanism that uses the super-parameterization concept (Freitas et al., 2006, submitted). Dry deposition and sedimentation follows the resistance approach. Wet deposition of aerosols is coupled with the convective parameterization. Figure 5 illustrates several sub-grid atmospheric transport processes simulated by CATT-BRAMS. A radiation parameterization has also been implemented, which takes into account the interaction between aerosol particles and short and long wave radiation (Toon, et al., 1989). For the smoke aerosol, a dynamic model, derived from three years of optical properties retrievals from some

of the Amazonian AERONET sites measurements, is used (Procópio et al., 2003). Model simulations for the 2002 dry season were performed, and model results were compared with observational data and satellite retrievals. The model configuration was structured with 2 nested grids: a coarse grid with 140 km horizontal resolution covering the South American and African continents, and an inner grid with horizontal resolution of 35 km, covering the South American continent.

The model-derived aerosol optical thickness (AOT) can be compared to that derived from satellite sensors, such as from MODIS (Figure 6). Figure 6a shows the aerosol optical thickness (AOT) at 550 nm channel from MODIS (Moderate Resolution Imaging Spectroradiometer), combining TERRA and AQUA satellites observations on 27 August 2002. Values of AOT of order 2 ~ 3 were observed over the Brazilian Amazonia. Also evident is the large extension of the plume and its long range transport reaching Northern Argentina and crossing the South Atlantic Ocean. The black areas are locations not covered by the sensor or contaminated by clouds. The simulated AOT is shown in Figure 6b. The model and MODIS retrieval show quite good agreement and reveal how the South Atlantic anticyclonic circulation and approaching the cold front (not shown) control the dispersion and advection of smoke over South America and the South Atlantic Ocean. The model results depicted in the red box employed a 35 km resolution grid (versus a 140km grid used elsewhere), showing finer plumes that are compatible with those shown in the 10 km resolution MODIS retrieval.

Radiative forcing by the intense smoke aerosol loads (Figure 7) has a significant impact in the surface sensible and latent heat fluxes (not shown). As a result, in the model the mixed layer depth is significantly reduced and the triggering of clouds in the convective parameterization is reduced. Thus, non-linear effects are clearly produced by the radiative forcing. In addition, the vertical distribution of aerosols has a significant impact in the vertical stability of the atmosphere. Finally, these aerosols may also have a significant impact in the cloud microphysical processes. The inclusion of the cloud microphysical feedbacks is under development for the CATT-
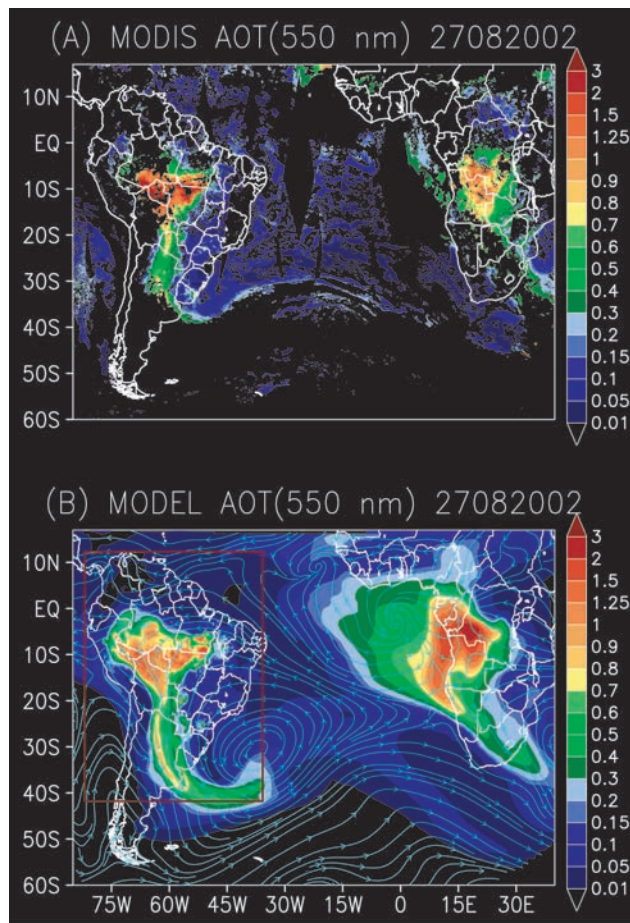

Figure 6. (A) Aerosol optical thickness (AOT) at 550nm channel from MODIS on 27AUG2002. (B) simulated AOT at 550 nm by CATT-BRAMS model.

BRAMS model. Model results have been also compared with direct observations. Trace gases and aerosol particles concentration (Dp < 2.5 m or PM 2.5) measurements near the surface were conducted at the ABRACOS (Anglo-Brazilian Amazonian Climate Observation Study; *http://www.cptec.inpe. br/abracos)* pasture site Fazenda Nossa Senhora at 10.75° S, 62.37° W, and were made from aircraft in various locations in the Rondonia state during the SMOCC campaign (Smoke Aerosols, Clouds, Rainfall and Climate; Andreae et al., 2004) from September 10 to November 5, 2002. Figure 8 shows the model-calculated PM2.5 concentrations and the SMOCC observation time series. The daily mean and the standard deviations of the mean (STD) of the observed PM2.5 are shown. The observed concentration exhibits strong variability, with the peak values exceeding 200 μg•m^{-3}. This variability mainly indicates the proximity of fires to the measurement site; the plumes are intense and still have not been significantly dispersed. The model results show quite good agreement with measurements; i.e. the model values are within the 1-sigma range of the observations.
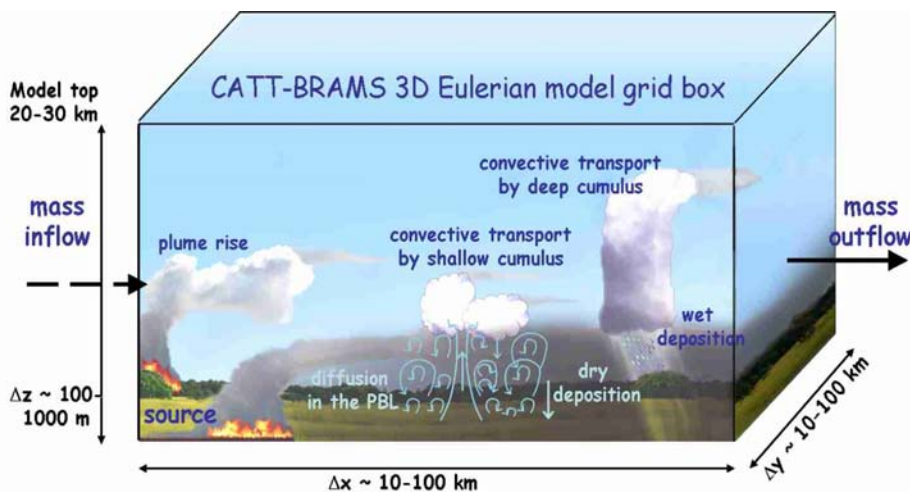

Figure 5. Several sub-grid processes involved in gases/aerosols transport and simulated by CATT-BRAMS system.
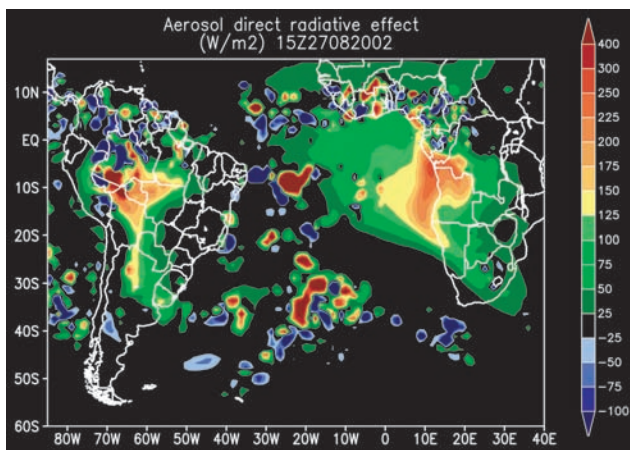
Figure 7. Impact of the inclusion of the direct aerosol radiative effect on the surface short wave radiation for 27 August 2002 at 15:00UTC.

The data shown in Figures 8 and 9 employed the Brazilian Fire Emission Model (BFEMO) described earlier. CATT-BRAMS simulations were also conducted using emissions sources as given by the EDGAR inventory and as derived from the TOMS Aerosol Index (i.e. Duncan et al., 2003). The resulting runs were used to generate model profiles of carbon monoxide (a tracer in this case for biomass burning) for comparison with aircraft measurements (Figure 9). Model CO concentration with the BFEMO emission was able to properly represent the mean observed CO profile from the surface to the low troposphere (4 ~ 4.5 km height, the typical maximum altitude reached by SMOCC aircraft). The simulated CO concentration using the EDGAR and Duncan et al. emissions underestimates the observed concentrations and presents a non-realistic vertical structure.

These results suggest the need for a near-real-time emission estimate with very high spatial and temporal resolution, as is given by the BFEMO model.

Concluding Remarks

The outstanding impact of biomass burning pollution in the South American continent has lead to the development of an operational system for air quality and weather forecasting that is now based on CATT-BRAMS at CPTEC-INPE. On a regular basis, forecasts of aerosol and carbon monoxide concentrations, as well as weather products, are produced and made available to the public in an open homepage *http://www.cptec.inpe.br/meio_ambiente.* For the next burning season, the inclusion of a tropospheric ozone forecast is planned.

An ongoing comparison of model results and MODIS and AERONET retrievals has just started and has been essential for the model validation as well as in the identification of the model current limitations. This has been and will be a focus of the model development efforts and also is a first step towards the aerosol and chemistry data assimilation.

From the weather forecast point of view, model results have shown the impact of aerosols on forecasts of near-surface properties and energy balance as well as on precipitation

fields. This reinforces the idea that aerosols must be included in weather and climate models in regions heavily affected by smoke fire emissions.
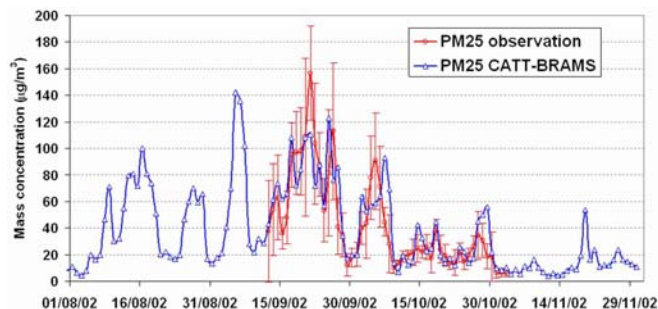

Figure 8 . Time series showing a comparison of surface PM2.5 ($\mu g \cdot m^{-3}$) as observed (red) and modeled (blue). The measurements are daily averages centered at 12:00UTC. The error bars are the standard deviations of the mean values. The model results are presented as instantaneous values at 12:00UTC.
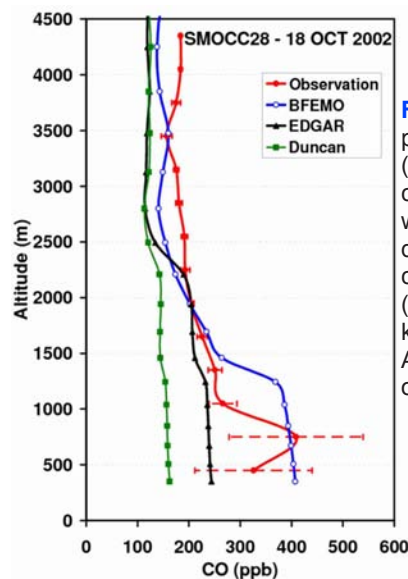

Figure 9. CO vertical profiles from the model (blue, black and green colors) as compared with observed CO (red) during SMOCC campaign flight 28 (conducted at about 100 km Northeastern ABRACOS pasture site) on 18 October 2002.

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Polar Study using Aircraft, Remote Sensing, Surface Measurements and Models, of Climate, Chemistry, Aerosols, and Transport (POLARCAT)

An International Polar Year (IPY) Activity, jointly endorsed by IGAC, SPARC and iLEAPS

Contributed by **Andreas Stohl** (ast@nilu.no) *Norsk institutt for luftforskning, P.O. Box 100, Instituttveien 18, 2027 Kjeller, Norway &* **Kathy Law** (kathy.law@aero.jussieu.fr) *IPSL Service, Aéronomie Boite 102, Université Pierre et Marie Curie, 4 Place Jussieu, 75252 Paris Cedex 05, France.*

In 2005, POLARCAT was endorsed as one of several International Polar Year (IPY) activities. It has also been endorsed as a new IGAC Task as well as being endorsed by IGBP's iLEAPS Project (Integrated Land-Ecosystem Atmosphere Process Study) and by WCRP's SPARC Project (Stratospheric Processes and their Role in Climate). The last International Polar Year took place in 1882-83 and was followed by the International Geophysical Year in 1957-58. Both produced a significant increase in our understanding

of the polar regions. The next IPY will take place from March, 2007 – March, 2009, allowing for research in both polar regions during all seasons. The main POLARCAT activities are planned to take place in this timeframe. As such, the exact research platforms/stations and instruments that will be involved are still being finalized. The following describes the scientific background, key research questions, and framework implementation plan for the POLARCAT study.

Updated information will be posted on the project web page towards the end of 2006, when it is anticipated that concrete plans regarding specific platforms and measurements will be in place: *http://zerdoz.nilu.no/~andreas/POLARCAT*
The following article is based on the POLARCAT white paper document, a full version of which is also available on the web page above.

Introduction

Climate change is proceeding fastest at the high latitudes of the Arctic. Surface air temperatures in the Arctic have increased more than the global average over the past few decades (Houghton et al., 2001). Precipitation and river discharges into the Arctic Ocean have also increased (Wu et al., 2005), whereas sea ice extent has dropped dramatically (Parkinson et al., 1999). The enhanced freshwater input is suspected to have already freshened the Arctic Ocean and deep North Atlantic Ocean (Dickson et al., 2002; Curry et al., 2003). If this trend continues long enough, the present thermohaline circulation could eventually collapse, with serious worldwide consequences. This makes the Arctic a region where a better understanding of the processes leading to climate change is most urgently needed. While some of the changes are related to the global increase of the long-lived greenhouse gases, many processes causing them are specific to the Arctic. The POLAR CAT study aims to improve our understanding of the role that tropospheric chemistry, aerosols, and transport play in these processes.

Because of its remoteness, the Arctic troposphere was long believed to be extremely clean. However, just before the last International Geophysical Year in 1957/58, pilots flying over the Canadian and Alaskan Arctic discovered a strange haze (Greenaway, 1950; Mitchell, 1957), which significantly decreased visibility. This so-called Arctic Haze is a recurring phenomenon that since then has been observed every winter and spring. It is now known to be the result of long-range transport of anthropogenic pollution mostly from Europe and western Asia. While it is clear that deposition of some species associated with Arctic Haze can significantly impact Arctic ecosystems (Macdonald et al., 2005), the climate impact of Arctic Haze is still under discussion. Radiative effects of aerosols, both direct and indirect, can be quite different in the Arctic compared to elsewhere. Due to the high surface albedo of snow and ice, even weakly absorbing aerosol layers can heat the Earth/atmosphere system (Pueschel and Kinne, 1995). Furthermore, infrared emissions from the haze can heat the surface during the polar night, and during spring when the solar zenith angle is still large (MacCracken et al., 1986). These effects clearly need further study.

In addition, satellite imagery (Figure 1) shows that the Arctic can also be affected by pollution transport in summer, when

forest fires are prevalent in the boreal region and are a strong high-latitude source of black carbon (Lavoue et al., 2000). As the boreal zone is warming, the frequency of fires appears to be increasing (Stocks et al., 1998). Smoke from the fires can travel over continental (Wotawa and Trainer, 2000), intercontinental (Forster et al., 2001), and even hemispheric (Damoah et al., 2004) distances. It has also been found recently that boreal forest fire smoke can penetrate deeply into the stratosphere (Fromm et al., 2005), where residence times could be long enough to have a significant impact on polar stratospheric ozone loss. Smoke aloft heats the atmosphere but cools the surface (Robock, 1991). However, black carbon particles can also be deposited and can significantly decrease the albedo of snow and ice surfaces (Hansen and Nazarenko, 2004). The enhanced absorption of solar energy could possibly contribute strongly to the melting of Arctic land and sea ice. However, no data exists yet to quantify this effect. A model study has also suggested that growing emissions in East Asia may increase the soot deposition in the Arctic (Koch and Hansen, 2005). This impact needs quantification based on observations and further model studies.

Several chemical phenomena were discovered recently that are unique to the Arctic troposphere. Both ozone and mercury can be almost instantaneously and completely removed near the time of polar sunrise (Oltmans, 1981; Barrie et al., 1988; Schroeder et al., 1998) as a result of catalytic bromine chemistry. Satellite measurements show the existence of high total columns of BrO at the time of the ozone and mercury depletion events (Wagner and Platt, 1998; Frieß et al., 2004).
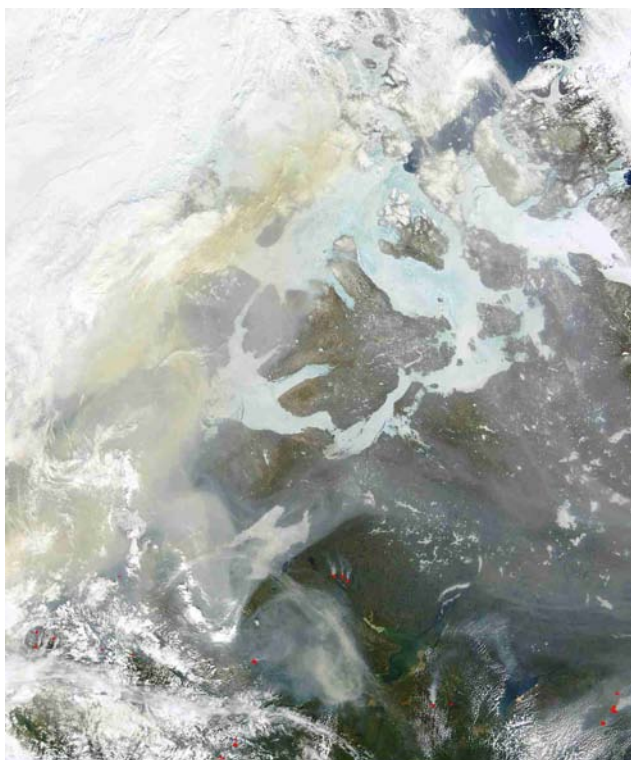

Figure 1. Fire and Ice: MODIS image of smoke from forest fires burning in Alaska and the Yukon, traveling into the Arctic over ice-covered areas, in July 2004. (Image courtesy NASA-GSFC).

However, the origin of the bromine has not been clarified yet. Furthermore, it is not known whether the bromine is located exclusively near the surface, or whether it can also exist throughout the free troposphere, with possibly large consequences for the chemistry of the Arctic atmosphere. Another recent discovery is the flux of nitrogen oxides from the snow pack into the Arctic boundary layer (Honrath et al., 1999). A unique feature at the cold temperatures of the Arctic troposphere is that most of the reactive nitrogen is stored in organic forms (e.g., peroxy acetyl nitrate, PAN) (Singh et al., 1992). However, if exported to warmer regions of the troposphere, PAN is easily decomposed to produce nitrogen oxides and lead to ozone formation. All of the phenomena described above are strongly influenced by the coupling of surface exchange processes, vertical transport, unique Arctic air chemistry, and import from and export to midlatitude regions. This coupling cannot be studied at a single site or by a single platform but instead needs a broad approach using measurements at the surface, aboard ships and aircraft and from satellites, and models as integrative tools, such as suggested by POLARCAT.

POLARCAT will execute a series of aircraft experiments at different times of the year in order to follow pollution plumes of different origin as they are transported into the Arctic and observe the chemistry, aerosol processes, and radiation effects of these plumes. It will also observe the atmospheric composition in relatively cleaner regions outside major plumes. The experiments will also take advantage of the long residence times of pollutants in the stably stratified Arctic atmosphere to study ageing processes by targeting air masses that have spent considerable time in the Arctic. The Arctic will, thus, also serve as a natural laboratory for investigating processes that cannot be studied elsewhere in such isolation. Measurements performed on a ship will investigate processes occurring in the lowest part of the troposphere, such as spring-time tropospheric ozone depletion events. In addition to the aircraft and shipboard experiments, satellite remote sensing data and surface measurements in the Arctic will be utilized. The wide range of surface measurements and ground-based remote sensing measurements (e.g., Notholt et al., 1997) that will take place as part of POLARCAT will also provide important information on the seasonal evolution of trace gases, aerosols and soluble species in rain and snow over the IPY timeframe. The combined analysis of these longer-term datasets, many of which will continue after IPY, will allow the campaign data to be put into a wider context. The aircraft campaign data, through vertical profiling, will also aide the interpretation of surface observations of trace constituents and precipitation chemistry, as well as ice core and firn measurements, by linking the surface with the boundary layer and free troposphere. Likewise, aircraft data profiles or lidar data will be used to validate satellite data. Models of differing complexity will be used to test our understanding of Arctic processes against the measurement data sets. These range from box models up to global or regional scale chemistry-aerosol-climate models.

Some of the processes that will be studied by POLARCAT in the Arctic are also operating in the Antarctic. However, the Arctic is influenced to a much larger extent by anthropogenic and biomass burning pollution sources.

Therefore, POLARCAT will concentrate its field activities in the Arctic. Model studies, though, will also consider the Antarctic, and will compare the situations in both polar regions.

The overall goal of POLARCAT is to study the *role of long-range poleward transport of aerosols and trace gases for climate change in the Arctic*. Specific areas of research related to the overarching goal fall into four categories:

• Transport processes

• Aerosol radiative effects in the Arctic

• Boreal forest fires and their effects on the Arctic

• Composition and chemistry of the Arctic troposphere

Each of these are discussed in detail below, followed by a framework discussion of the research implementation plan.

Transport Processes

Seasonal aspects of pollution transport.

Field studies have established that the Arctic haze phenomenon occurs regulary in winter and maximizes in early spring, with the number and depth of the haze layers increasing with the season (Scheuer et al, 2003). In the 1970s it became clear that the haze was of anthropogenic origin (Rahn et al., 1977), and in the 1980s the Arctic Haze was traced back to sources located predominantly in northern Eurasia (Barrie, 1986).

The haze phenomenon is a result of the special meteorological situation in the Arctic in the winter. During winter, air in the lower troposphere over the Arctic is partially isolated from the rest of the atmosphere by a transport barrier. Potential temperature at the ground becomes extremely low within the Arctic. This leads to an extremely stable vertical stratification (Bradley et al., 1992), which reduces turbulent exchange and, thus, dry deposition. The low water vapour content also makes wet removal very inefficient, leading to a very long atmospheric lifetime of aerosols and other pollutants in the Arctic. The low surface temperatures also enhance the latitudinal temperature contrast. This means that surfaces of constant potential temperature form closed domes over the Arctic bounded by the "polar front" (Carlson, 1981; Iversen, 1984; Barrie, 1986). Air can only cross these surfaces and escape the "polar dome" if there are waves on the polar front resulting in equator-ward excursions of air and associated heating from the warmer underlying surface. Air can enter the polar cap where it experiences significant diabatic cooling close to the polar front (Klonecki et al, 2003). There is a preferred entry route into the polar cap from Europe, associated partly with the extreme sea-land temperature contrast on the western seaboard of Eurasia (Rahn, 1981). Furthermore, because Europe is located at relatively high latitudes, a significant fraction of the European emissions can actually be injected directly into the polar cap, especially when wave activity takes the polar front relatively far south over Europe. Therefore, near the surface, Arctic Haze is primarily caused by emissions in Europe and northwestern Asia (Eckhardt et al., 2003; Figure 2).
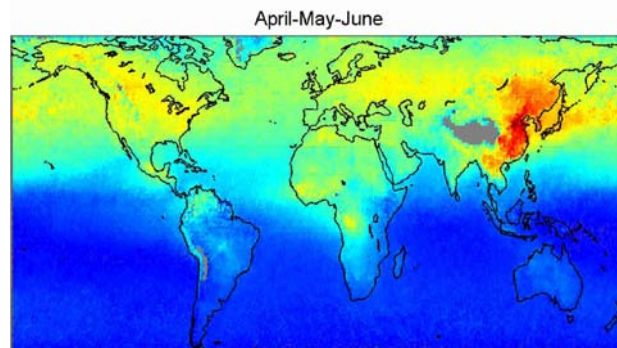
Figure 2. This composite image depicts the MOPITT CO measurements at an altitude of 850 hPa. The seasonal plot shows the observations averaged over 4 years for April-May-June. In the Northern hemisphere, most of the pollution is associated with urban activity and fires, such as those frequently observed over Alaska and Siberia (Clerbaux et al., 2004).

Since black carbon (BC) emissions in south Asia are increasing, Koch and Hansen (2005) suggested that currently emissions from South Asia, together with biomass burning emissions, are the dominant source of BC in the Arctic. Emissions in south Asia occur at much more southerly latitudes (and, thus, higher potential temperatures) and they tend to be lofted over the North Pacific stormtrack if transported north (Stohl, 2001; Stohl et al.,2002). Upon arrival in the Arctic, they should be located at relatively high altitudes. However, according to Koch and Hansen's (2005) model calculations, Asian BC rivals European BC even at the surface. But it is not entirely clear how, in their model, south Asian air masses are cooled sufficiently to reach the Arctic lower troposphere in winter, although there is generally cooling and slow sinking motion over the Arctic itself, and pollutant layers aloft are slowly entrained into the Arctic boundary layer.

During the breakdown of the polar cap as polar night ends, outbreaks of Arctic air are most common across the Labrador Sea and along the coasts of Greenland (Honrath et al, 1996). The pollution that has built up over winter is then released to the mid-latitudes (Penkett et al, 1993). It has been speculated that this pulsed release of ozone precursors from the Arctic could lead to the observed spring-time ozone maximum at middle latitudes (Penkett and Price, 1986). Arctic Haze can also be exported to the middle latitudes (Heintzenberg et al., 2003) and it is assumed that polar air masses influence mid-latitude particle formation (Nilsson et al., 2001; Kulmala et al., 2003).

Inter-annual variability of pollution transport pathways into the Arctic.

In the NH, especially during the Arctic Haze season in winter and early spring, the most prominent and recurrent pattern of atmospheric variability is the North Atlantic Oscillation (NAO) (Hurrell, 1995). Oscillations between high and low NAO phases produce large changes in the Arctic wind field, surface air temperature, precipitation, river runoff, ocean currents, sea ice, and biological responses (see Macdonald et al., 2005, for a recent review). Especially in the 1980s, there

was a strong upward trend in the NAO, which was associated with a substantial decrease in the Arctic sea ice cover. Transport of anthropogenic emissions from Europe, North America and Asia into the Arctic is significantly enhanced under positive NAO conditions, as can be seen both in model calculations and in Arctic observations (Eckhardt et al., 2003; Duncan and Bey, 2004). Transport of emissions from Europe is particularly enhanced under positive NAO conditions (Figure 3).

Changes in transport pathways to the Arctic associated with climate change.

In climate change simulations, many models indicate a poleward shift of the Atlantic stormtrack between the 2071-2100 and 1961-1990 climates. Over the last century poleward shifts in the stormtrack have been highly correlated with positive NAO phases (IPCC, 2001). Ulbrich and Christoph (1999) examined the stormtrack and NAO in a 300 yr control run and 240 yr scenario run of a coupled model (ECHAM4 +OPYC3). They found that stormtrack activity increased over northwestern Europe in the scenario run, but the increase in NAO index was barely significant. The implication of the study is that the winter stormtrack is likely to shift polewards and downstream towards Scandinavia with ramifications for increased pollution transport into the Arctic. Also associated with this shift would be increased heat transport and decreased sea-ice extent. Possibilities exist for climate feedbacks involving sea-ice cover and albedo changes over ice/snowpack associated with enhanced deposition of black carbon.

Stratosphere-troposphere exchange.

The low tropopause elevation and its weak expression during the winter, combined with the large-scale downward transport in the stratosphere at high latitudes, suggest that the Arctic troposphere may be strongly influenced by injections of stratospheric air. Intensive sampling campaigns have documented distinct episodes of stratosphere-to-troposphere transport (STT), but mainly in the North American region (e.g. TOPSE-2000). Results indicate that STT is probably the dominant source of O_3 and HNO_3 in the winter, and remains significant through the year. However, it appears that photochemical production of O_3 becomes the major source in late spring and into summer (e.g., Browell et al., 2003; Mauzerall et al., 1996; Wang et al., 2003). In contrast, a year-long study at Alert confirmed that STT was significant even at the surface throughout the year (Dibb et al., 1994). Careful quantitative assessment of this transport term remains mandatory for estimating the ozone winter depletion by halogen species and spring photochemical production from measurements of the ozone seasonal cycle. Using water vapor isotope measurements above the Arctic tropopause in Scandinavia, Zahn (2001) showed that there is a 1-2 km thick layer where upwelling air from the troposphere mixes with downwelling air from the stratospheric vortex. A better characterization of the chemical composition of this layer and its dynamical coupling with the troposphere or the stratosphere is still an open issue.

POLARCAT objectives related to transport processes.

• Quantification of the residence times of (polluted) air masses in the Arctic polar dome, and their seasonal dependence, using transport models validated with Lagrangian balloon data.

• Determination of the gradients in the chemical composition of the troposphere at the edge of the polar dome and how it relates to mixing with mid-latitude air masses.

• Study of the transport pathways of pollution from south Asia into the Arctic free troposphere and boundary layer, using models, satellite and aircraft measurements.
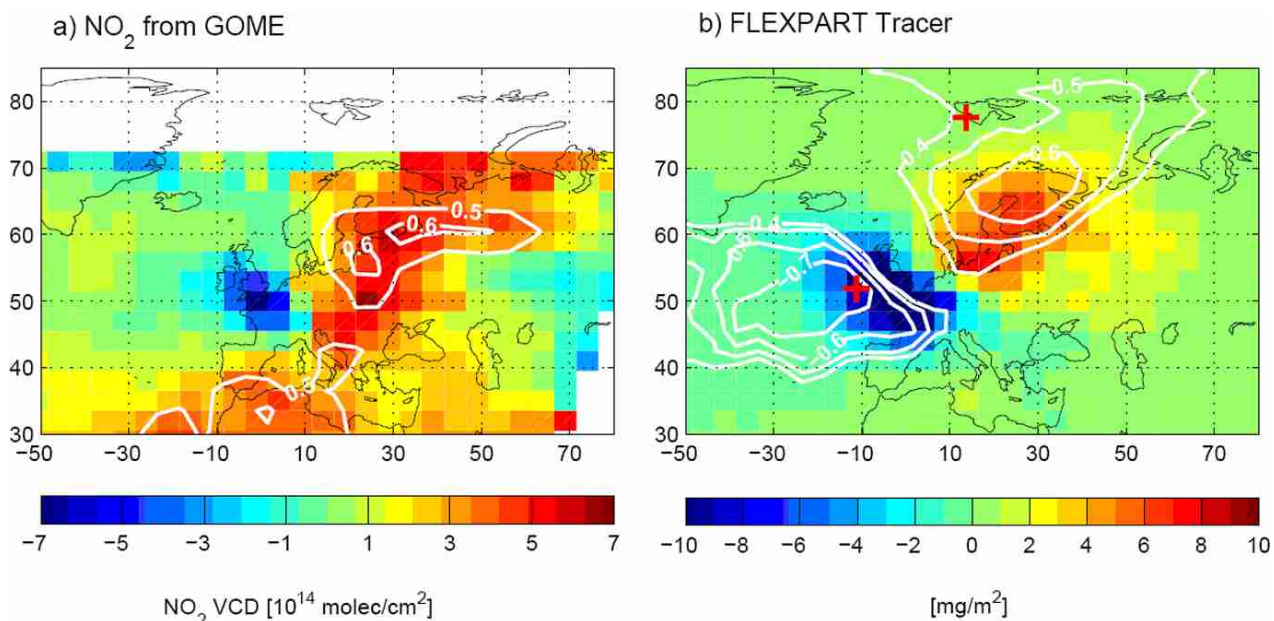

a) NO$_2$ from GOME   b) FLEXPART Tracer

NO$_2$ VCD [10^{14} molec/cm^2]   [mg/m^2]

Figure 3. Comparison of observed and simulated NAO signal in pollution transport from Europe (from Eckhardt et al., 2003). Map of residual NO$_2$ vertical columns [10^{14} molecules cm^{-2} retrieved from GOME satellite observations for NAO$^+$ minus NAO – composites during seven (1996–2002) winters (a). Same but in mg m^{-2} for a simulated European emission tracer with a 1-day lifetime (b). Superimposed as white lines are the correlation coefficients with the NAO index.
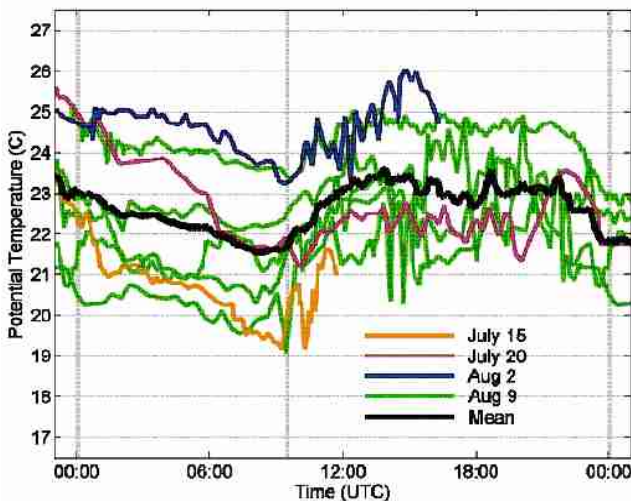
Figure 4. Air temperature observed over eight days of constant-altitude Lagrangian balloon flights in the New York City plume during the ICARTT experiment in summer 2004. The magnitude, direction, and regularity of the daily cycle as well as the stability of the marine atmosphere where the balloons were flying suggest that the heating and cooling was due to radiative processes in the polluted air mass. Lagrangian balloons flying for several days or weeks in the Arctic could help quantify in-situ heating rates.

• Determination of the vertical layering of pollution from different source regions, including its seasonal variation.

• Exploration of the horizontal and vertical structure and extent of Arctic Haze, and how it changes seasonally, using aircraft and surface measurements.

• In-situ quantification of net heating rates in polluted air masses in the Arctic, using Lagrangian balloons (see Figure 4). Measurement of the vertical temperature stratification in the remote Arctic using repeated vertical soundings by Lagrangian balloons.

• Study of the interannual variability in pollution transport pathways and associated removal mechanisms (e.g., wet deposition) into the Arctic.

• Investigation of the likely change of transport processes in a future climate, using climate model predictions. •

• Quantification of transport from the stratosphere on the tropospheric chemistry, including deposition of nitrogen species to the snowpack, using models and surface measurements of tracers.

Aerosol Radiative Effects in the Arctic

Occurrence and optical properties of Arctic aerosol and pollutant haze.

As already described, each winter through spring a sulfate-rich, persistent haze is observed in the Arctic. During early February, significant enhancements in sulfate aerosol are confined near the surface (< 2 km) as long-range transport from northern Eurasia occurs along low level, sinking

isentropes (Klonecki et al. 2003). As the haze season progresses, enhanced sulfate occurs at higher altitudes (up to at least 8 km). Since vertical mixing is prohibited by the persistent low-level inversion (Kahl, 1990), the higher altitude haze layers are thought to be due to transport into the Arctic along vertically higher isentropes tracing back to increasingly warmer surface source regions in northern Eurasia. During early April, sulfate layers below 3 km begin to dissipate due to the beginning of solar heating and resulting mixing near the surface. However, more stable isentropic transport continues at higher altitudes. By the end of May, both the lower and higher altitude sulfate enhancements are significantly decreased due to the continued break-up of the inversion and return of wet deposition. Pollutant particles within the Arctic Haze are well-aged with a mass median diameter of about 0.2 µm or less; as such, they are very efficient at scattering solar radiation. The haze also is weakly absorbing due to the presence of black carbon. The result is a noticeable reduction in visibility, to a few kilometers or less. Model calculations suggest that the "weak" absorption has significant climatic influences when the dark colored haze spreads out over the highly reflecting snow and ice pack of the Arctic. In turn, the highly reflecting surface enhances aerosol-radiative interactions due to multiple scattering between the surface and the haze.

The seasonality and trends of Arctic Haze are clearly seen in time series data of light absorption and scattering by aerosols. Both aerosol scattering and optical depth (AOD) measurements at Barrow showed a maximum in 1982 followed by a factor of two decrease between 1982 and 1992 (Bodhaine and Dutton, 1993; Figure 5). A combination of a reduction in the pollution aerosol output by Eastern Europe and the former Soviet Union, and stricter pollution controls in Western Europe most likely contributed to the decrease. However, from 1997 to 2005 there has been a significant increasing trend (Quinn et al., 2005). Similarly, light absorption at Barrow indicates an overall decrease between 1988 and 2005 but an increase for both March and April between 1997 and 2005. These results support the hypothesis that increasing black carbon emissions from southern Asia may be impacting the Arctic (Koch and Hansen, 2004). AOD at Barrow and Ny Ålesund, Spitsbergen also appears to be increasing although the trends do not match in time (Herber et al., 2002). The
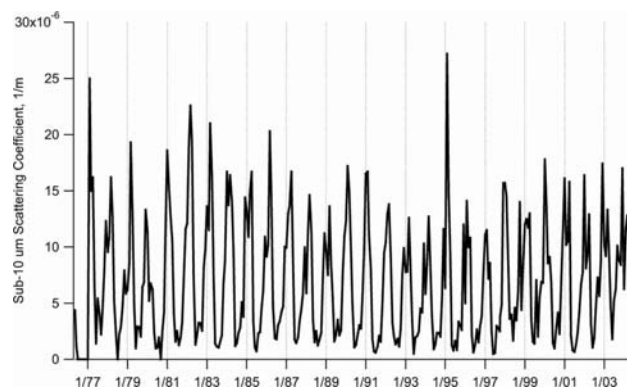

Figure 5. Monthly averaged light scattering at 550 nm by sub-10 micron aerosol from a surface station at Barrow, Alaska, showing a pronounced annual increase in aerosol scattering in March to April. Data made available by NOAA CMDL.

apparent changing trends in aerosol burdens and associated optical properties in the Arctic provide impetus for further investigation into the causes and impacts.

While almost all trace atmospheric constituents in the Arctic boundary layer, including aerosol mass, reach minimum concentrations during summer, resulting also in the seasonal minimum of AOD, the number concentration of aerosol particles reaches a maximum that time of the year. Most likely, in situ formation of new particles causes this maximum, but the mechanism is an ongoing matter of discussion. Leckand Bigg (1999) hypothesized that organic films in the surface water of open ice leads provides a source of new particles in the Arctic summer atmosphere. Ström et al. (2003) showed that there is a very strong relation between the amount of solar radiation reaching the Arctic surface and the number density of aerosol particles, suggesting that photochemistry could be the key process in the formation of the new particles. Year-round aerosol size distribution measurements at Ny Ålesund revealed a prevailing accumulation mode (150 nm) in spring followed by a dominating nucleation mode (30 nm) in the summer (Ström et al., 2003). The transition between the two regimes occurred within a few days. This distinct seasonal change also is seen in the upper troposphere (Treffeisen et al., 2005).

Climate effects of pollutant haze – direct effects.

Absorption and scattering of radiation by aerosols directly affect the radiation balance of the Arctic. This region is thought to be particularly sensitive to changes in radiative fluxes because of the small amount of solar energy normally absorbed in the polar regions. Arctic Haze is present as a layer of light absorbing material over a highly reflective ice/snow surface. Several early calculations using 1-D radiative transfer models estimated that the diurnally averaged atmospheric warming due to the aerosol layer ranged between 2 and 20 W/m^2 with a corresponding depletion of the solar flux at the surface of 0.2 to 6 W/m^2 (e.g., Leighton, 1983; Blanchet and List; 1987, Shaw et al., 1993). These estimates agreed with direct measurements from wideband sun photometers (Mendonca et al., 1981). Heating rates of about 0.1 to 0.2 K/day were measured by Valero et al. (1989) during AGASP (Arctic Gas and Aerosol Sampling Program) II and by Treffeisen et al. (2005) during the ASTAR 2000 campaign in Svalbard. The AASE (Airborne Arctic Stratospheric Expedition) II flights in winter of 1992 revealed soot-contaminated Arctic aerosols at altitudes of 1.5 km. Pueschel and Kinne (1995) calculated that this layer of aerosols could heat the earth-atmosphere system above surfaces of high solar albedo (ice/snow) even for single scattering albedos as high as 0.98. Hence, a modest amount of black carbon in the haze layers can result in a measurable contribution to diabatic heating.

MacCracken et al. (1986) estimated that the cooling of the surface due to absorption of solar radiation by the haze layers could be compensated by infrared emission from the atmosphere to the surface. During the dark winter, infrared emissions from the haze may heat the surface if deliquescent sulfate salts grow and become cloud droplets or ice crystals thereby enhancing their impact in the longwave. In addition, since the haze is present throughout the Arctic night, the integrated effect may modify the radiative budget.

Climate effects of pollutant haze – indirect effects.

The indirect effect of aerosol particles on irradiances in the Arctic results from the impact of aerosol particles on the microphysical properties of clouds. Enhanced aerosol particle concentrations may be increasing solar cloud albedo of Arctic stratus and stratocumulus by increasing the number concentration of cloud condensation nuclei and thereby decreasing the average size of cloud droplets (provided the liquid water content in the clouds remains constant; Twomey, 1977), resulting in more radiation being reflected back to space (Albrecht, 1989; Twomey, 1991). The normally low aerosol number concentrations in the Arctic results in a large percentage of particles activating during cloud formation (e.g. Komppula et al., 2005). Hence, changes in aerosol properties are likely to have a significant impact on microphysical and optical cloud properties. As the cloud droplet number concentration increases, cloud droplet size decreases which reduces drizzle formation and increases cloud coverage and lifetime (Hobbs and Rangno, 1998). Garrett et al. (2004) has shown that low-level Arctic clouds are highly sensitive to particles that undergo long range transport during the winter and early spring. The sensitivity was detected as higher cloud droplet number concentrations and smaller cloud droplet effective radii compared to summertime clouds exposed to particles nucleated in the Arctic from local biogenic sources. The most significant effect of the change in cloud properties due to Arctic Haze may be on cloud emissivity. A decrease in droplet effective radius in these optically thin clouds will increase the infrared optical depth and thus the infrared emissivity (Curry and Herman, 1985; Garrett et al., 2002). The result is expected to be an increase in downwelling infrared irradiances from the cloud and an increase in the rate of spring-time snow pack melting (Zhang et al., 1996).

According to observations during the SHEBA experiment, supercooled cloud droplets are common in the Arctic even at temperatures of -20°C or lower (Curry et al., 1996). The sulfate-containing pollution aerosol within Arctic Haze is thought to impact ice nucleation. Models estimate that aerosols containing sulfuric acid produce fewer ice nuclei than nearly insoluble aerosols (Blanchet and Girard, 1995). Measurements corroborate this finding. Borys (1989) reported that Arctic Haze aerosol had lower ice nuclei (IN) concentrations, a lower IN to total aerosol fraction, and slower ice nucleation rates than aerosol from the remote unpolluted troposphere. The reduction in ice nuclei leads to a decrease in the ice crystal number concentration and an increase in the mean size of ice crystals (Girard et al., 2005). As a result, the sedimentation and precipitation rates of ice crystals increase leading to an increase in the lower troposphere dehydration rate and a decrease in the downwelling infrared irradiances from the cloud. Using a 1-D simulation and observations from Alert, Girard et al. (2005) found that a cloud radiative forcing of -9 W/m^2 may occur locally from the enhanced dehydration rate produced by sulfate aerosol. The mechanism that decreases IN concentrations in the presence of sulfuric acid aerosol is unknown and warrants further research. If this mechanism applies to much of the Arctic, it could explain the cooling tendency in the eastern high Arctic during winter.

Because of the combination of the static stability of the Arctic atmosphere, the persistence of low level clouds, and the relatively long lifetime of aerosol particles during the haze season, the impact of aerosols on cloud microphysical and optical properties may be larger in the Arctic than elsewhere on Earth (Garrett et al., 2004). The winter/spring occurrence of Arctic Haze events allows the study of anthropogenic influences against a very clean atmospheric background. In other regions of the globe, a reliable distinction between natural and anthropogenic effects is more difficult. In this sense, the Arctic is a natural laboratory to study the anthropogenic portion of the aerosol-cloud-radiation interactions.

Climate effects of pollutant haze – surface.

Surface albedo affects the magnitude and sign of climate forcing by aerosol particles. Absorbing soot deposited to the surface via wet and dry deposition impacts the surface radiation budget by enhancing absorption of solar radiation at the surface (Warren and Wiscombe, 1980). Clarke and Noone (1985) found a 1 to 3 % reduction in snow albedo due to deposited BC with another factor of 3 reduction as the snow ages and BC becomes more concentrated. Hansen and Nazarenko (2004) have estimated that soot contamination of snow in the Arctic and the corresponding decrease in surface albedo yields a positive hemispheric radiative forcing of +0.3 W/m^2. The resulting warming may lead to the melting of ice and may be contributing to earlier snowmelts on tundra in Siberia, Alaska, Canada, and Scandinavia (Foster et al., 1992). New techniques to measure surface albedo from airborne platforms have been developed recently (Wendisch et al., 2004).

Clearly, the radiative impacts of pollutant aerosol particles in the Arctic are quite complex. Multiple feedbacks between aerosols, clouds, radiation, sea ice, and vertical and horizontal transport processes complicate a comprehensive picture as do potentially competing effects of direct and indirect forcing. As a result, the magnitude and sign of the forcing are not yet well understood in this region. Technological advances made in the past decade have provided us with new tools to further improve our understanding of the Arctic Haze phenomenon. These advances include modern aircraft payloads, model calculations of long-range pollutant transport, and new spaceborne observational methods. With these new tools, we are well posed to re-visit the Arctic and address questions of aerosol effects that cover large horizontal and vertical scales.

POLARCAT objectives related to aerosol radiative effects in the Arctic.

• Improvement of knowledge on the sources, evolution and removal of the tropospheric aerosol particles in the Arctic Haze season.

• Determination of the vertical distribution of chemical, physical and optical properties of Arctic aerosol particles.

• Characterization of direct radiative effects (solar and terrestrial) within pollution layers in the Arctic.

• Investigation of the interactions of aerosols with clouds and their impact on radiative forcing based on observations, experiments and model studies.

• Detailed in situ observations of microphysical and optical properties of Arctic clouds including particularly the ice phase (mixed-phase clouds).

• Determination of the role of aerosols as ice nuclei.

• Characterization of albedo changes of snow and ice surfaces and the resulting solar radiative effects due to the deposition of black carbon from anthropogenic and biomass burning sources.

• Validation of aerosol and cloud products of space observations from polar orbital satellites (Aqua-Train, i.e. CALIPSO, CloudSat, etc.)

Boreal Forest Fires & their Effects on the Arctic

Climate change and boreal forest fires.

Observed and forecast impacts of climate change are greatest at northern latitudes and over land, particularly over the more continental regions of Canada, Russia and Alaska. These are areas where large fires have been common since the last Ice Age, and recent research (e.g. Stocks et al. 1998; Flannigan et al. 2003) indicates that more frequent and severe fires are expected as the climate changes. This will have a significant impact on the age class structure and carbon budget of the boreal/Arctic zone in particular and the globe in general. Boreal fires consume large quantities of fuel and spread quickly, creating high energy release rates that are often sustained for long burning periods. This frequently results in convection columns with strong vertical development that reach beyond the tropopause. Long-range smoke transport from large boreal fires is already common, with smoke loads from Siberian fires often reinforcing smoke from North American fires. This phenomenon is expected to become even more common with more frequent and severe fires in the future, increasing the likelihood that smoke from boreal fires will provide a positive feedback to climate change (Kurz et al. 1994).

Long-range transport of boreal forest fire emissions.

Large amounts of smoke and trace gases emitted by boreal forest fires can be subject to considerable vertical and horizontal transport (e.g. Stocks and Flannigan, 1987; Siebert et al., 2000; Fromm et al., 2005). Fire emissions can travel over continental (Wotawa and Trainer, 2000), intercontinental (Forster et al., 2001; Honrath et al., 2004), and even hemispheric (Damoah et al., 2004) distances. Recent satellite observations and lidar measurements observed substantial amounts of forest fire smoke in the tropopause region and lower stratosphere at high latitudes and in the Arctic region (e.g. Waibel et al., 2000; Fromm et al., 2000; Damoah et al, 2004). Especially over snow/ice surfaces the short-wave reflectivity to space can be considerably reduced by forest fire smoke, which may have important implications for the radiative energy budget in the polar region (Hsu et al., 1999). Episodically, the fires also pollute large regions in the lower troposphere at high latitudes (Forster et al., 2001), but unfortunately few measurements exist for the Arctic free troposphere (e.g., during the ABLE 3A and 3B campaigns;

Figure 6. Picture of a pyro-cumulonimbus located at 58ºN, 126ºW on 27th June at 21 UTC. The picture was taken from a commercial airliner cruising at about 10 km (Picture courtesy of Noriyuki Todo of Japan airlines). Note the smoky nature of the cloud.

Harriss et al., 1992 & 1994, Shipham et al., 1992) or the surface (e.g., Yli-Tuomi et al., 2003). There is clear evidence for deposition of ammonia from biomass burning sources in Arctic ice core measurements (Whitlow et al., 1994). Presumably, the deposition of substances like soot from such fires may decrease the albedo of ice and snow and lead to enhanced melting of Arctic glaciers and sea ice (Kim et al., 2005). However, to date no data exist to reliably establish such a connection.

Pyro-convection.

It has recently been discovered that the transport of biomass burning emissions may also be transported into the lower stratosphere through an explosive combination of intense forest fires and extreme convection (see Figure 6). This "pyroCb" source of stratospheric injection has been observed remotely (lidar, balloon sounding, and satellite solar occultation) (e.g. Fromm et al., 2000; Fromm and Servranckx, 2003) and in-situ (e.g.Jost et al., 2004). Although boreal fire research scientists have reported forest fire convection columns above 13 kilometres in height (e.g. Stocks and Flannigan 1987), recent publications and unpublished data paint an emerging picture of the pyroCb phenomenon as a recurring one, with hemispherical impact (e.g. Fromm et al., 2005). UTLS enhancements, attributable to pyroCb, of aerosols, carbon monoxide, ozone, and acetonitrile have all been observed. While observations clearly

show that deep upward transport of biomass burning emissions into the upper troposphere and lower stratosphere is frequent (e.g., Nedelec et al., 2005), the mechanisms are poorly understood. Factors that could enhance convective uplift over the fires are the heat and water vapour released by the fire, microphysical cloud processes (Andreae et al., 2004), and probably radiation absorption by soot particles above the cloud tops and in the stratosphere.

The highest altitude where forest fire smoke was observed in situ was 17km (remote sensing observations exist even at higher altitudes), several kilometers above the tropopause and at potential temperatures greater than 380K (Jost et al., 2004), thus in a region that is commonly referred to as the stratospheric overworld. The chemical impact of the forest fire emissions at such high altitudes is unknown. Both efficient ozone formation as well as severe ozone destruction are possible scenarios. Furthermore, the stratospheric residence time of aerosols may in fact be long enough to affect stratospheric ozone depletion during the following winter/spring.

Due to their proximity to the Arctic, their large source strength, and the special processes accompanying them as described above, boreal forest fires need special attention during POLARCAT. For this, an integrated study using low- as well as high-flying aircraft, satellite measurements, and models is needed. The launches of the AURA, CALIPSO and METOP satellites come at an opportune point to observe these processes and also provide critical data needed for the flight planning. In addition, satellite data like TOMS aerosol index, solar/lunar occultation profiles (e.g. POAM III, SAGE II and III, and GOMOS), MODIS imagery, MOPITT, IASI, ACE, NOAA POES and GOES imagers will be used to provide forecast guidance for potential fire blowup conditions worldwide, enabling also targeted operating modes for certain satellite instruments.

POLARCAT objectives related to boreal forest fires.

• Comprehensive study of the impact of boreal forest fire emissions on the chemical composition of the Arctic troposphere.

• Study of the pathways of boreal forest fire plumes into the Arctic, particularly regarding the plume altitudes.

• Quantification of the impact of the deposition of soot from forest fires on the surface albedo of snow and ice surfaces, and investigation of the link with Arctic sea ice and glacier retreat.

• Investigation of the contribution of pyroCb aerosol injections to the stratospheric background aerosol concentrations in the Arctic, in particular during volcanically-quiescent periods.

• Determination of the residence times of aerosols in the Arctic stratosphere, in particular whether forest fire aerosols canremain in the stratosphere long enough to play a role in winter/spring ozone depletion.

• Study of the fates and effects of chemical compounds injected into the stratosphere by pyroCbs, including their role for ozone formation and ozone depletion.

Composition and Chemistry of the Arctic Troposphere

The polar troposphere is a unique environment within the earth's atmospheric system. Its uniqueness stems from generally cold temperatures, a prolonged period of darkness followed by a period of continuous light, underlying snow and ice, and a low tropopause above. While there are virtually no anthropogenic pollution sources within the Arctic itself, as has been discussed it is impacted by emissions from many of the world's largest industrial regions (in particular northern Eurasia and eastern Asia; e.g., Rahn et al., 1977; Barrie, 1986; Koch and Hansen, 2005) and, in summer, "pollution" from boreal forest fires (e.g., Mauzerall et al., 1996; Dibb et al., 1996). The composition of the Arctic troposphere is further influenced by snow to air exchange of key trace chemicals followed by homogeneous and heterogeneous reactions (e.g., Honrath et al., 1999; Dibb and Arsenault, 2002; Dibb et al., 2002).

Atmospheric chemistry research in the Arctic has tended to come in waves targeting largely separate questions, most notably: a) the origin and characteristics Arctic Haze, with measurements starting in the late 1950s; b) the cause of severe ozone depletion events (ODEs) in the boundary layer over the Arctic Ocean, which were first noticed in the early 1980's (Oltmans, 1981; Barrie et al., 1988); and c) investigations into stratospheric ozone depletion following the discovery of the stratospheric ozone hole over Antarctica.
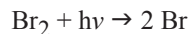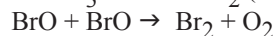
Recent years have seen tremendous advances in in-situ measurement capability and satellite observations of tropospheric composition. IPY offers a unique and timely opportunity for a coordinated and integrated international experiment to explore the chemistry of the entire arctic troposphere and its impacts on global chemistry and climate. Several unique phenomena have been identified in the arctic troposphere that need further systematic and coordinated investigation. Salient among these are: (1) causes of surface ozone and mercury depletion events; (2) the likely presence and role of halogen free radicals; (3) presence of atmospheric reservoirs of reactive nitrogen and their influence on ozone chemistry; (4) emissions of OVOC and NO_X from ice surfaces; (5) influences of stratospheric intrusions; and (6) investigations of glacial ice cores to understand past atmospheric composition and recent human impacts.

Surface ozone and mercury depletion.

As noted above, the discovery of ODEs in the Arctic atmospheric boundary layer near the time of polar sunrise (Oltmans, 1981, Barrie et al., 1988) sparked one of the waves of interest in Arctic tropospheric chemistry. An equally surprising discovery was that gaseous elementary mercury (Hg) appeared to undergo depletion in concert with ozone/HO_X/NO_X raising the specter of a potential major contamination of the Arctic biosphere (Schroeder et al., 1998; Figure 7).

It has been postulated that this phenomenon is a result of the following gas-phase bromine atom chain reactions:

$$Br + O_3 \rightarrow BrO + O_2 \text{ (x2)}$$
$$BrO + BrO \rightarrow Br_2 + O_2$$
$$Br_2 + h\nu \rightarrow 2\ Br$$
$$\overline{}$$
$$2O_3 \rightarrow 3O_2 \text{ (net)}$$

A mechanism suggested to cause the observed sudden BrO enhancements in the marine boundary layer is the autocatalytic release of BrO involving heterogeneous reactions on sea-salt surfaces (Tang and McConnel, 1996; Vogt et al., 1996). Substantial concentrations of BrO have been observed in both the Arctic and the Antarctic (Wagner and Platt, 1998, Frieß et al., 2004). The efficiency of this cycle is limited by conversion of Br to the non-radical reservoir species. Models have been developed to explain the role of bromine and iodine chemistry in ozone and Hg depletions (Calvert and Lindberg, 2003).

More recently, studies have pointed out the importance of polar snow as a source of ambient nitrogen oxides (NO_X) (Honrath et al., 1999) and of precursors of hydrogen oxide radicals (HO_X) such as HCHO during spring (Sumner and Shepson, 1999). Snowpack photochemistry has been identified as a likely cause of large interstitial-air and ambient HCHO concentrations, and was shown to result in NO_X concentrations in interstitial-air up to an order of magnitude larger than ambient levels, consistent with the presence of an unexpected diurnal cycle in ambient-air NO_X. In short, the Arctic boundary layer in spring time is influenced by industrial pollution, halogen chemistry, and ice driven NO_X and OVOC intrusions at the same time. To date, attempts to develop a coherent mechanistic explanation for these depletion processes on the basis of known gas-phase chemistry has not been successful and heterogeneous processes involving snow and aerosol
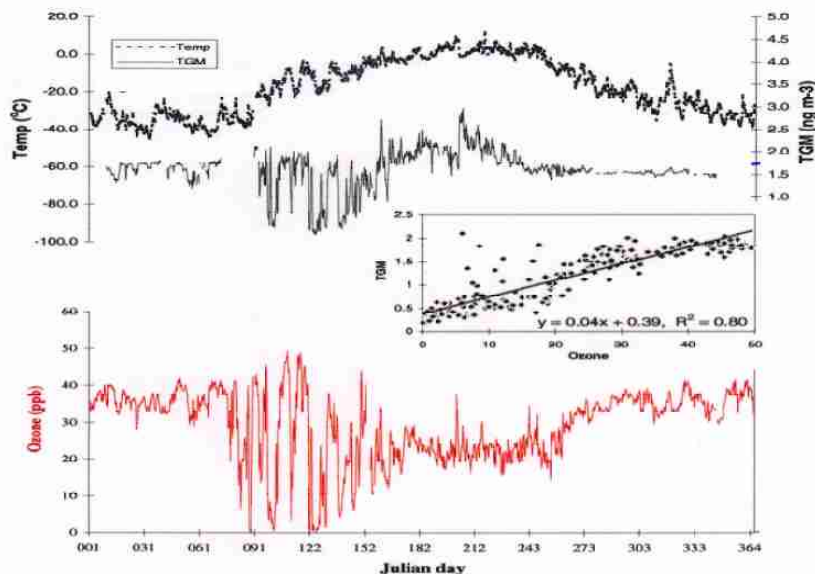

Figure 7. Simultaneous mercury and ozone depletion at Alert (82ºN) (Schroeder et al. 1998).

particles are likely implicated. Observational data are critically needed to analyze the coupled evolution of BrOx/ClOx/IO-NOx-HOx-O$_3$ chemistry during O$_3$ and Hg depletion events in the Arctic spring. ODEs and other aspects of snow and ice surface/atmosphere interactions and chemistry are also the subject of another IGAC Task, Air Ice Chemical Interactions (AICI; IGACtivities No. 29, Dec. 2003). This activity has also spawned an overarching IPY activity, AICI-IPY. POLARCAT and AICI-IPY will, as such, encompass synergistic and coordinated activities.

Halogens in the free troposphere.

Little attention has been paid to the possibility that reactive halogens may have a significant impact in the Arctic free troposphere. Recent field campaigns at Summit, Greenland (72ºN, 38ºW, 3.1 km asl) suggest that this may indeed be the case. High levels of peroxy radicals (HO$_2$+RO$_2$) were measured consistent with photochemical theory given observed mixing ratios of precursors. However, OH levels were significantly elevated compared to steady state model simulations and previous measurements at South Pole (Huey et al., 2004a, 2004b; Sjosted et al., 2005). Observed values of the (HO$_2$+RO$_2$)/OH ratio were generally 4 – 5 times lower than expected from theory. This disagreement was greatly accentuated during periods of high wind, when observed values of the (HO$_2$ +RO$_2$)/OH ratio were more than an order of magnitude lower than model estimates. These observations lead to the hypothesis that halogen chemistry may be responsible for much of the observed disturbance in HO$_X$ partitioning at Summit.

Satellite observations (GOME and SCHIMACHY) suggest that the atmospheric column of BrO above central Greenland during summer is often on the order of 3-5x10^{13} mol cm^{-2} (Figure 8) which would yield mixing ratios near 20 ppt if most of the BrO were in a 1 km deep boundary layer above the ice sheet (Richter et al., 1998; Wagner and Platt, 1998). Although substantial concentrations of BrO may be near the surface there is reason to believe that this BrO is distributed throughout the Arctic troposphere. Measurements of large perturbations in hydrocarbon ratios, enhanced soluble gas phase bromine (Dibb, unpublished data; Evans et al., 2003; Ridley et al., 2003) and ozone depletion (Peterson and Honrath, 2001; Helmig et al., 2005) in air filling the pore spaces of the snowpack indicate that halogen activation may proceed via heterogeneous reactions on ice crystal surfaces similar to those observed during polar sunrise at lower elevations in the Arctic. A small reactive halogen flux from the snow pack into overlying air could account for persistently elevated OH throughout summer. The impact on photochemistry in the free troposphere above sunlit snow may be significant if snow-impacted boundary-layer air is vertically mixed upward.

Reactive nitrogen, hydrogen, and ozone in the free troposphere.

As has been stated above, the composition of the Arctic free troposphere and its linkages with the surface below and the stratosphere above have not been extensively studied. The ABLE-3A and 3B (Harris et al., 1992; 1994) campaigns did study O$_3$ chemistry but were restricted to middle troposphere
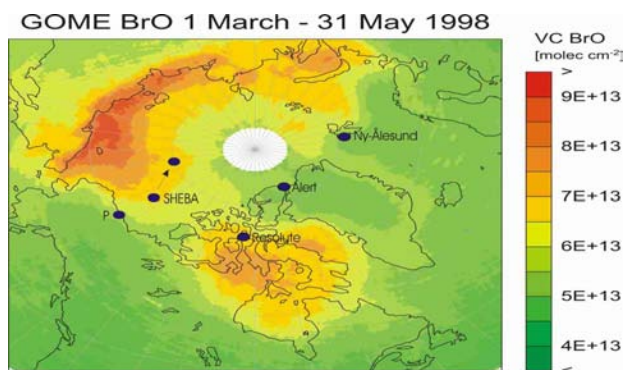
Figure 8. Tropospheric BrO column derived from the GOME satellite.

altitudes due to platform limitations and were performed at a time when suitable instrumentation to measure many key species (e.g. free radicals) was unavailable. As noted earlier, one of the primary ABLE 3 objectives was to constrain biosphere/atmosphere exchange, hence there was a lower troposphere focus. A more recent effort was in TOPSE during Feb-March 2000 with an altitude limitation similar to the ABLE 3 experiments but improved instrumentation (Atlas et al., 2003). In this campaign, determining the impact of stratosphere-to-troposphere transport (STT) on the oxidative capacity of the troposphere was one of the key objectives. It should be noted that STT was found to be significant during both the summertime and the spring. However, model calculations constrained by the ABLE 3 and TOPSE data sets found that photochemical production appeared to be the dominant source of O3 in the Arctic troposphere from late spring into summer.

The free troposphere of the Arctic is greatly influenced by fires in summer and Eurasian outflow in spring. Figure 9 shows an example of the role of PAN in the free troposphere where 80% of all reactive nitrogen is tied in this form. This is a unique feature of a very cold atmosphere where the organic forms of reactive nitrogen are highly stabilized
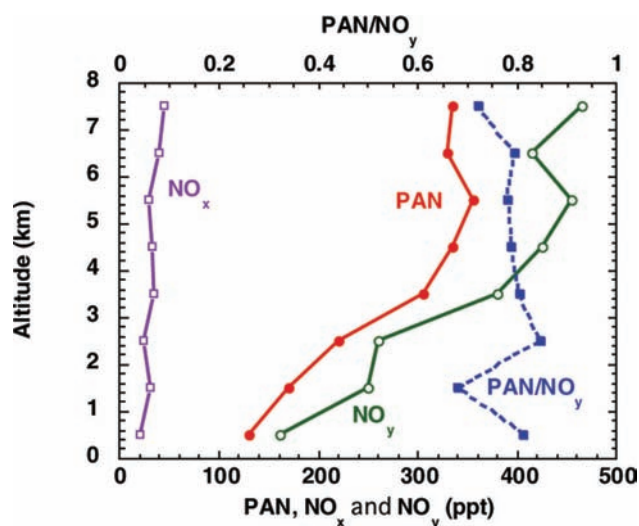
Figure 9. PAN, NO$_X$ and total reactive nitrogen (NO$_y$) in the Arctic troposphere (50-80 N) in May. Median mixing ratios as measured during TOPSE (see Atlas et al, 2003).

(Singh et al., 1992). However, PAN is easily decomposed to produce NO_x and influence O_3 chemistry in other regions of the troposphere. There is a clear need to perform field missions that can cover the entire Arctic troposphere in at least two seasons with a capability to measure the O_3-HO_x-NO_x- halogen cycle including precursors and aerosols.

POLARCAT objectives related to tropospheric composition and chemistry.

The objective is to investigate the composition and chemistry of the entire Arctic troposphere in two seasons (spring and summer) with the goal of understanding the reactive nitrogen,reactive hydrogen, reactive halogen, and ozone cycles. An integrated approach that links surface, free tropospheric, and satellite observations with models of chemistry and climate is envisioned. The intensive sampling will quantify the relative importance of transport to and from the free troposphere, impact of halogen/OVOC/NO_x formation in the snowpack on the free troposphere, and the role of Arctic reservoir species on the global troposphere. Specific objectives are:

• Determination of the chemical composition of the entire Arctic troposphere in two seasons (spring and summer) using airborne, satellite, and surface platforms at a level of detail not hitherto possible. Improved understanding of the O_3/NO_x/HO_x chemistry in the Arctic troposphere.

• Investigation of the role of halogen atom chemistry in the Arctic boundary layer and the free troposphere.

• Improved understanding of the impact of snow pack emissions of NO_x, OVOC, and halogens on the Arctic troposphere.Validation of satellite observations of tropospheric composition.

• Improved knowledge of sources impacting the Arctic troposphere.

• An assessment of the impact of pollution transport to the Arctic on chemistry and climate based on an integrated analysis of data collected during IPY 2007/8 campaigns using 3-D models.

POLARCAT Research Framework

The POLARCAT IPY activity is still in the implementation planning phase. As such, the detailed list of measurement that will be made and the exact models/modelling groups that will be involved is still being determined. As such, we describe below the implementation framework for the project. The role of models and satellite measurements are discussed explicitly in the context of their role for mission planning and data interpretation, integration and synthesis. The specific list of surface and aircraft-based measurements that will be made is in flux, so only a preliminary list is given. Parties interested in participating in the POLARCAT activity should contact Andreas Stohl (*ast@nilu.no*) after referring to the POLARCAT website for the latest planning information (*http://zardoz.nilu.no/~andreas/POLARCAT/*).
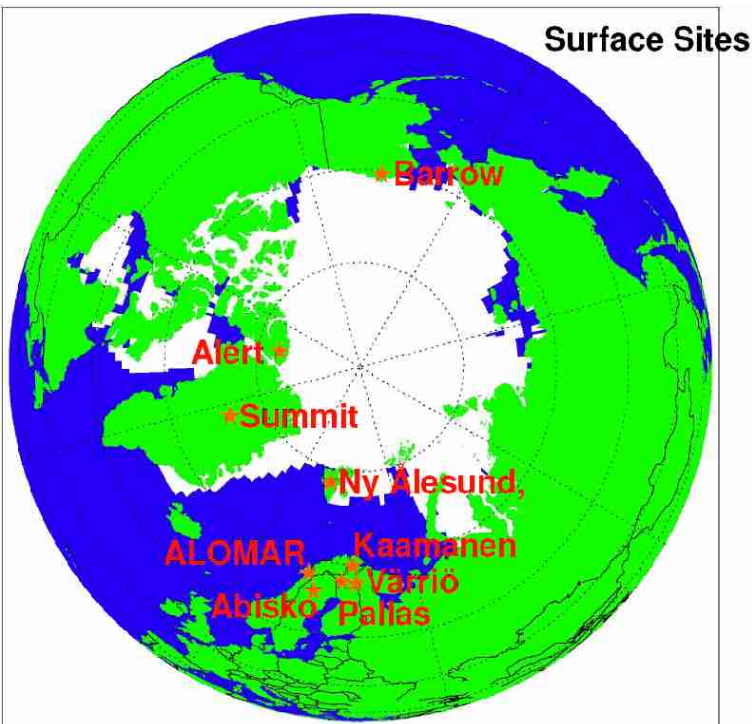

Figure 10. Location of major POLARCAT surface measurement sites.

POLARCAT will bring together intensive aircraft experiments, research ship cruises, monitoring activities at surface stations, ground-based remote sensing, balloon releases, satellite measurements, and a range of different models. In order to achieve its overall goals, POLARCAT will closely co-ordinate these different activities. For instance, the aircraft and shipboard experiments will be supported by forecasts from meteorological and chemical models, satellite observations, surface networks and enhanced ozone sonde releases. In some cases, pathfinder aircraft carrying remote sensing instrumentation will be used to guide other aircraft carrying in-situ instrumentation into pollution layers.

Aircraft measurements will also be closely co-ordinated with each other and with the releases of Lagrangian balloons, in order to sample the same polluted air masses repeatedly. Such a Lagrangian approach will allow constraining the overall chemical budget in an air parcel between individual observations.

The airborne measurements will be coordinated with satellite overpasses, especially of Aura, Aqua, Terra, Envisat, and Calipso. Validation of the satellite observations of tropospheric composition and aerosol parameters will receive a high priority. Vertical aircraft profiling will also be done above surface stations and the ship, in order to characterize the vertical (and horizontal) extent of phenomena observed at the stations. POLARCAT will also work together closely with other IPY core activities (i.e. OASIS, AICI-IPY). For instance, it is planned that aircraft perform over-flights over the icebreakers used in other programs.
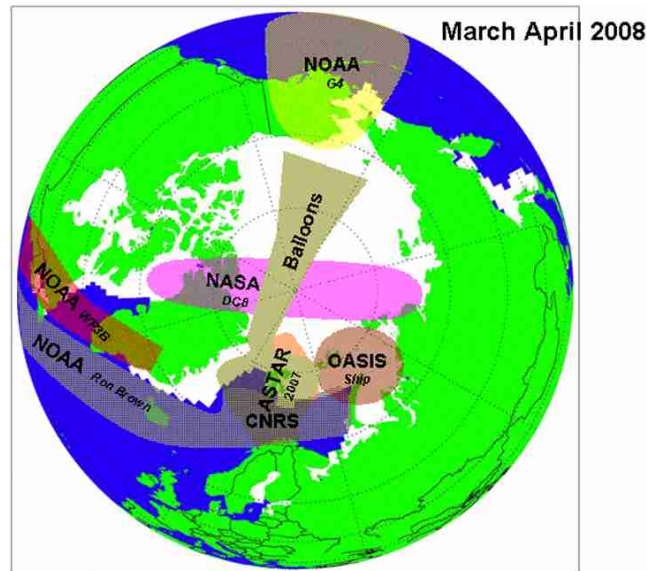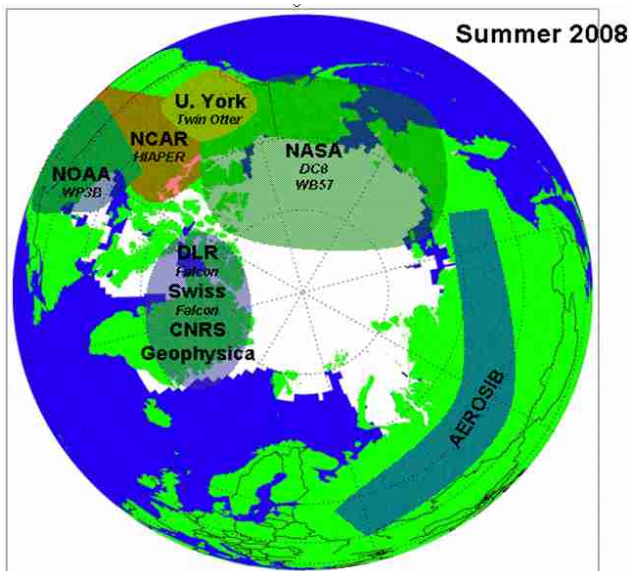POLARCAT is a bottom-up project that will remain open for others to join.

Figure 11. Summary of major field activities in spring and summer 2008. White areas show typical sea ice extent in the respective season.

Figure 10 shows the location of the most important POLARCAT surface sites, and Figure 11 shows an overview of the major field activities using mobile platforms planned for spring and summer 2008. Note that activities are also planned during other periods, but these will be the periods with most concentrated efforts.

Use of satellite data during the POLARCAT campaign

The Arctic is an advantageous region for analyzing satellite measurements. Most of the instruments suitable for the characterization of aerosols and trace gases in the troposphere are carried on polar orbiting satellites and their tracks are most densely packed at the poles. Therefore, overpasses over a fixed location are much more frequent close to the poles than at lower latitudes (Figure 13), providing excellent opportunities for comparisons with ground-based or aircraft measurements in the Arctic. The Arctic is also special because of the high albedo of ice and snow surfaces, which can enhance signals in DOAS-type retrievals of trace gas columns, though this also makes detection of light scattering by aerosols more difficult.

Meteorological satellite data.

Detection of cloud, water vapor and surface features at high temporal resolution (< 1 hour) in the mid-latitudes at all longitudes is made possible by six geostationary satellites positioned around the equator. However, north of the Arctic Circle the geostationary images are distorted by the curvature of the Earth's surface. Therefore, at high latitudes polar orbiting weather satellites must be relied upon to provide images of clouds and surface features at high temporal resolution. While each of the four NOAA Polar-orbiting Operational Environmental Satellites (POES) passes over the Arctic region once every 102 minutes, their inclined orbit, coupled with the rotation of the Earth means the portion of the Arctic viewed by a particular satellite changes with each
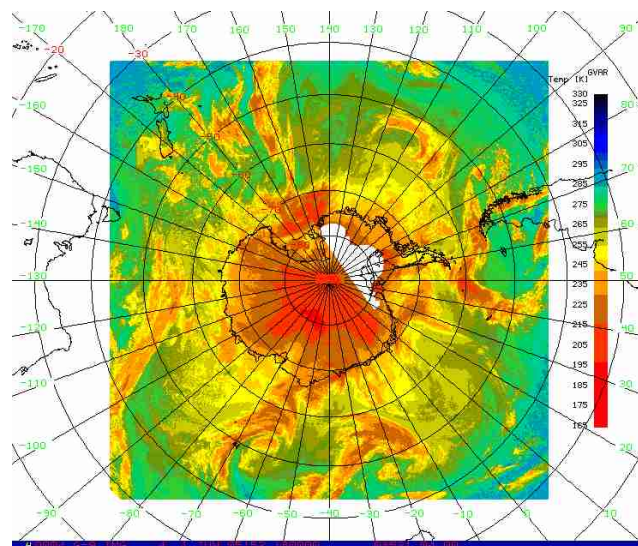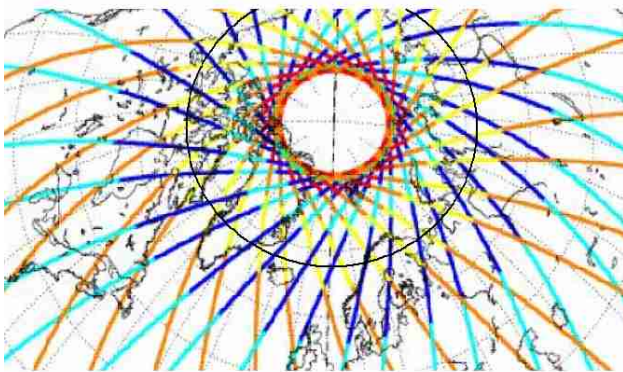

Figure 12. Infrared composite image above Antarctica for 18 UTC, June 1, 2005. This is a routinely produced product from the Antarctic Meteorological Research Center. Similar images for the Arctic will be provided.
al., 2004).

orbit. The inconsistency of the viewable region of the Arctic makes it difficult to track meteorological features associated with air pollution transport. For POLARCAT, merged visible, IR, and water vapor products using images from the Terra and Aqua satellites, all four NOAA/POES satellites and geostationary satellites will be created (Figure 12).

Aerosol and trace gas satellite measurements.

A wide range of satellite data on the composition of both the troposphere and the stratosphere will be available for POLARCAT. The specific satellite instruments that will be

Figure 13. Coverage of CALIPSO measurements during ascending (orange/yellow/red) and descending (blue) orbits for two consecutive days. The polar circle is shown in black.

utilized are still being finalized as the funding and other (e.g. the CALIPSO satellite launch) situations evolve. We refer you to the POLARCAT web page(*http://zerdoz.nilu.no/~andreas/POLARCAT*), which should have specifics available at the end of this year.

Use of Models during POLARCAT

Numerical models of atmospheric composition will play an important role in POLARCAT, both in addressing the key scientific issues of the project, and in operational support of the field campaign periods. A hierarchy of models will be used, ranging from process-based photochemical box models capable of detailed representation of reactive radical chemistry and aerosol microphysics to 3-D Chemical Transport Models (CTMs) and General Circulation Models (GCMs) able to resolve the interaction of aerosol, chemical and dynamical processes and to determine the impacts on regional or global climate.

The Arctic presents a unique photochemical environment characterized by low UV intensities, cold temperatures, halogen radical chemistry, and ice-covered surfaces, and strongly stratified conditions lead to aging times of several weeks. The dynamical and chemical conditions here are challenging to model, and the current generation of global CTMs show widely divergent behaviour over polar regions, as seen in recent studies of surface ozone (Stevenson et al., 2005). POLARCAT observations will provide a valuable test of the ability of CTMs to simulate the chemical and microphysical evolution of air masses in the region, and in combination with more detailed process-based box model analysis will contribute to improved treatments of stratification, slow and/or novel chemistry and surface processes. Reducing uncertainties in modelling the Arctic region is an important goal of the project and will contribute to an improved understanding of the impacts on regional chemistry and climate.

Analysis of POLARCAT observations will make use of box models, trajectory models and CTMs. Photochemical box (0-D) models including detailed representation of

chemical processes (Crawford et al., 1999; Evans et al., 2003) will be used to interpret aircraft observations in terms of radical chemistry in the Arctic with particular attention to processes involving halogen radicals and heterogeneous reactions. Another class of box models including detailed representation of aerosol microphysics will be needed to describe the unique Arctic environment for nucleation and growth of particles. 3-D particle dispersion models will be used to derive flow climatologies for the Arctic region, and to determine source-receptor relationships to aid in interpretation of aircraft observations (Stohl et al., 2003). Photochemical box models following air mass trajectories will be used to trace the chemical evolution of air masses entering and leaving the region (e.g., Methven et al., 2003). Finally, 3-D chemical transport models (CTMs) will integrate the information from the surface, aircraft, and satellite platforms in terms of the constraints that they provide on source regions affecting the Arctic atmosphere, transport between mid-latitudes and the Arctic, large-scale vertical motions, and the chemical and aerosol evolution coupled with these dynamical processes. The CTMs will need to be at least hemispheric in scale to describe the range of motions affecting Arctic atmospheric composition.

Beyond their value for post-mission data analysis, the CTMs will be of critical importance for the planning and execution phases of the POLARCAT field missions. Model simulations conducted before the mission using hindcast meteorological fields, and evaluated with pre-existing surface and satellite observations in the Arctic, will provide critical input for selecting optimal mission time windows, bases of operations, and flight regions. The hindcast simulations will be used to develop a menu of flights to guide mission execution. During the execution phase of the missions, the same CTMs driven by meteorological forecasts will provide chemical forecasts to guide the aircraft on a day-to-day basis. These forecasting activities will involve a number of CTMs to provide different perspectives and to address the broad range of mission objectives. This hindcast-forecast methodology has been applied very successfully in a number of recent aircraft measurement campaigns including TRACE-P (Jacob et al., 2003; Kiley et al., 2003), ITCT-2K2 (Parrish et al., 2004; Forster et al., 2004) and the recent ICARTT/INTEX campaign.

Finally, study scenarios of possible future climate conditions and emission distributions, fully coupled aerosol-chemistry-climate models are needed. Transient as well as time-slice simulations using these models will be performed to study how Arctic Haze will develop in the future. Furthermore, these simulations will be used to identify possibly important feedback processes in the climate system involving aerosol and pollution transport at high latitudes.

Activity coordinators:

Andreas Stohl, Norway
Kathy Law, France
Steering committee:
Hajime Akimoto, Japan
Boris Belan, Russia
Jack Dibb, U.S.A.

Mike Fromm, U.S.A.
Ewa Krawczyk, Poland
Lauri Laakso, Finland
Kathy Law, France (co-chair)
John Methven, U.K.
David Parrish, U.S.A.
Laurier Poissant, Canada
Phil Rasch, U.S.A.
Hans Schlager, Germany
Hanwant Singh, U.S.A.
Andreas Stohl, Norway (chair)

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