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

POLARCAT

A Proposed International Polar Year Activity

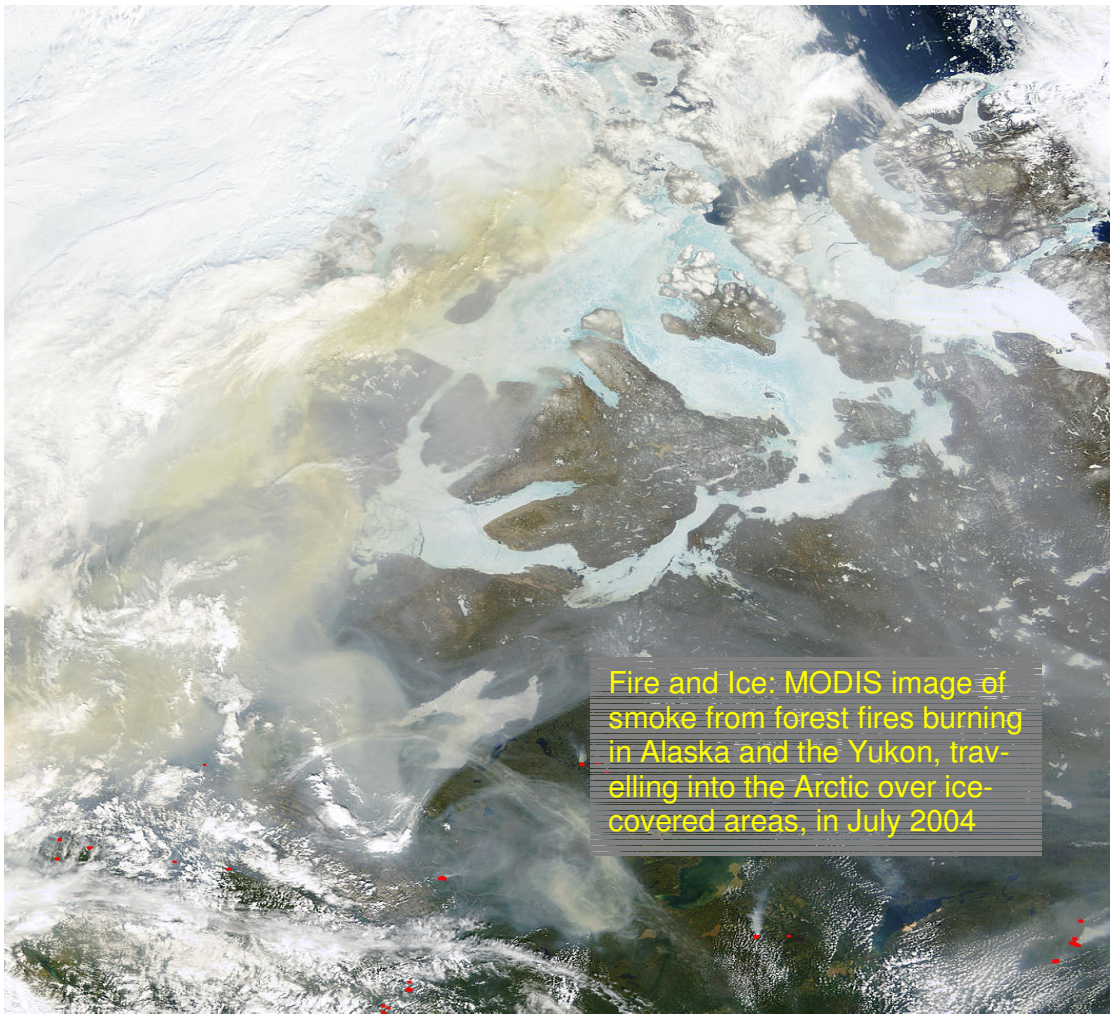

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Table of Contents

POLARCAT website (in development):
http://zardoz.nilu.no/~andreas/POLARCAT/

This document:
http://zardoz.nilu.no/~andreas/POLARCAT/polarcat_white_paper.pdf

1. 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. This 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 (see title page) 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). 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.

After specifying POLARCAT's overall objectives, the scientific background for POLARCAT and open questions shall briefly be reviewed. Thereafter, the POLARCAT scientific activities shall be described, as they are currently planned.

2. Overall Goals

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***. Particular objectives are to

- better characterize the transport pathways of pollution into the Arctic free troposphere and boundary layer, including quantification of the residence times of polluted air masses in the polar dome.
- determine the vertical layering of Arctic pollution from different sources and the associated optical properties of Arctic aerosol particles.
- study the seasonal and interannual variability of transport pathways and removal processes for aerosols and pollutants, and to investigate likely changes in a future climate.
- characterize the direct radiative effects (solar and terrestrial) within pollution layers in the Arctic.
- investigate the interactions of aerosols with clouds (e.g., their role as ice nuclei), and their impact on radiative forcing.
- quantify the albedo changes of snow and ice surfaces and resulting radiative effects due to the deposition of black carbon from anthropogenic and biomass burning sources.
- study the impact of boreal forest fire emissions on the chemical composition of the troposphere and on the deposition of soot in the Arctic.
- determine the fate and effects of aerosols and chemical compounds injected into the stratosphere by pyro-convection, including their role for ozone formation and ozone depletion in the polar stratosphere.
- improve our understanding of the $O_3/NO_x/HO_x$ chemistry in the Arctic troposphere.
- investigation the role of halogen atom chemistry in the Arctic boundary layer and free troposphere.
- study the impact of snow pack emissions of NO_x, OVOC, and halogens on the Arctic troposphere.
- validate aerosol, trace gas, and cloud products of space observations from polar orbital satellites.

3. Transport Processes

Seasonal aspects of pollution transport. Field studies 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).

Since black carbon (BC) emissions in south Asia are increasing, Koch and Hansen (2005) suggested that nowadays 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; the NAO is

strongly correlated to the so-called Arctic Oscillation). The NAO is identified by variations in the NAO index, which is typically defined by the difference in surface pressure between Iceland and the Azores/Lisbon (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). For instance, there is a positive correlation between carbon monoxide concentrations at Arctic measurement stations and the NAO index (Table 1). Transport of emissions from Europe is particularly enhanced under positive NAO conditions. GOME tropospheric NO_2 columns, for instance, show much stronger transport of NO_x from European sources towards the Arctic (and reduced transport towards the Atlantic and south-central Asia) for high-NAO conditions (Figure 1).

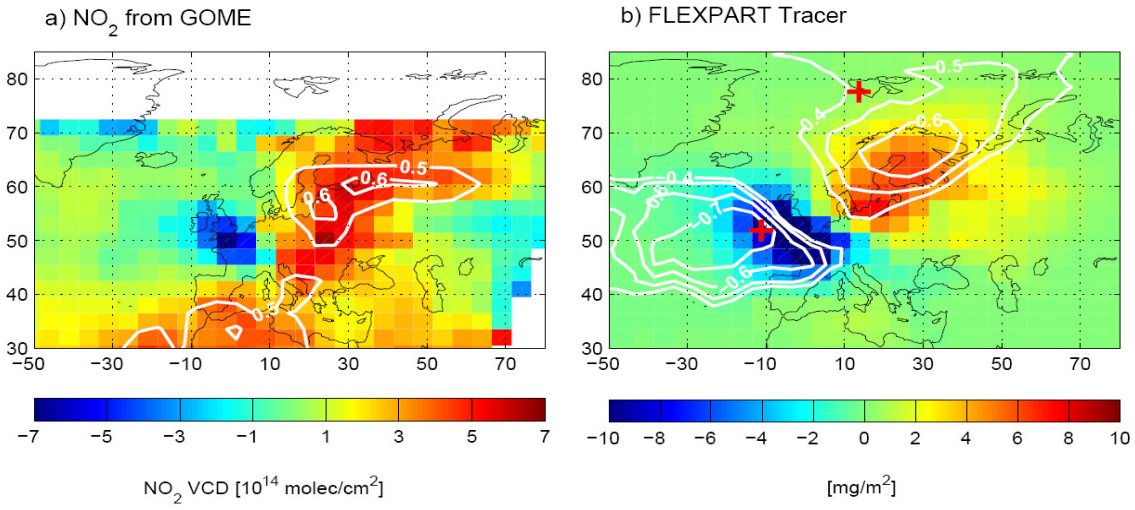

Figure 1. Comparison of observed and simulated NAO signal in pollution transport from Europe (from Eckhardt et al., 2003). Map of residual NO_2 vertical columns [10^{14} molecules cm−2] retrieved from GOME satellite observations for NAO+ minus NAO− composites during seven (1996–2002) winters (a). Same 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.

Table 1. Table (after Eckhardt et al., 2003) shows coefficients obtained from a multiple linear regression with carbon monoxide as dependent, and time and the NAO index as independent variables: r^2 explained variance, a_1 intercept (unit ppbv), a_2 trend (ppbv/year), a_3 slope with the NAO index (ppbv). The analysis was done for the years 1994-2001. Correlations are significant at the 1% level for Spitsbergen and Alert, and at the 0.1% level for Barrow. The major explanatory factor is the NAO index.

Location	r^2	a_1	a_2	a_3
Spitsbergen	0.41	170	-0.9	3.3
Barrow	0.44	186	-2.4	4.0
Alert	0.28	187	-2.7	2.8

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 (Chapter 9) states, “a few studies have shown increasingly positive trends in the indices of the NAO/AO

in simulations with increased greenhouse gases, although this is not true in all models, and the magnitude and character of the changes varies across models." 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. They showed that the two centers of action of the first empirical orthogonal function of mean sea level pressure (an alternative definition of the NAO) shifted downstream in the scenario run, such that the poleward center of action moved from the east coast of Greenland into the Norwegian Sea. The lack of trend in NAO index was attributed to the weak projection of the increase in European storm activity on the spatially-fixed NAO pattern derived from the recent climate. However the variability is defined, the implication 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, the increase in O_3 in spring is likely caused by photochemical production (Browell et al., 2003). Model studies suggest photochemical production of O_3 becomes the major source in late spring and into summer (e.g., Mauzerall et al., 1996; Wang et al., 2003). A year-long study at Alert used the radionuclide tracers ^{7}Be and ^{10}Be to investigate the STT in the high Arctic (Dibb et al., 1994). This investigation confirmed that STT was significant even at the surface throughout the year. 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.
- 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 2). 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.

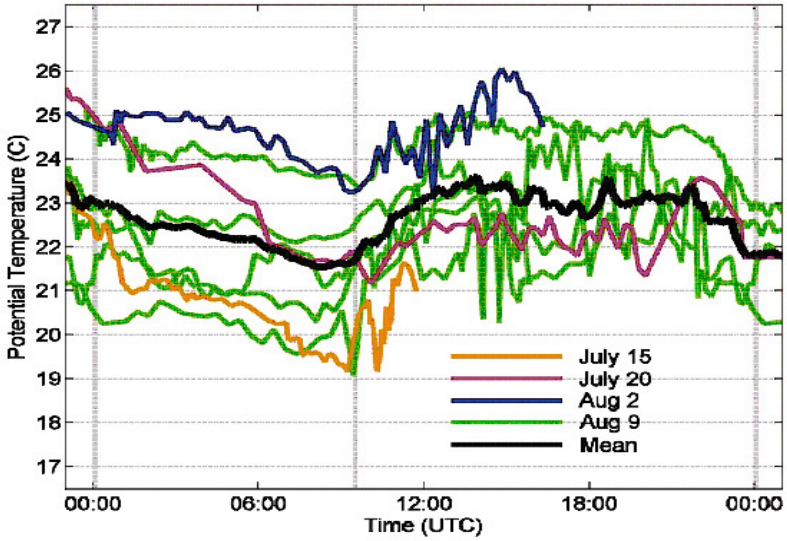

Figure 2. Air temperature observed over eight days of constant-altitude Lagrangian balloon flights in the New York City plume during the ICARTT experiment in summer 2004. The magnitude, direction, and regularity of the daily cycle as well as the stability of the marine atmosphere where the balloons were 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.

4. 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. This particle size range is very efficient at scattering solar radiation since the peak in the particle surface-area size distribution is near the maximum efficiency for Mie scattering. The haze also is weakly absorbing due to the presence of black carbon. The result of the strong scattering and weaker absorption 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. 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. Figure 3 shows ground-based measurements which depict the pronounced increase in light scattering during March and April. 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). 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 (at the 95% confidence level) 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 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. Leck and 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 pre-

vailing 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).

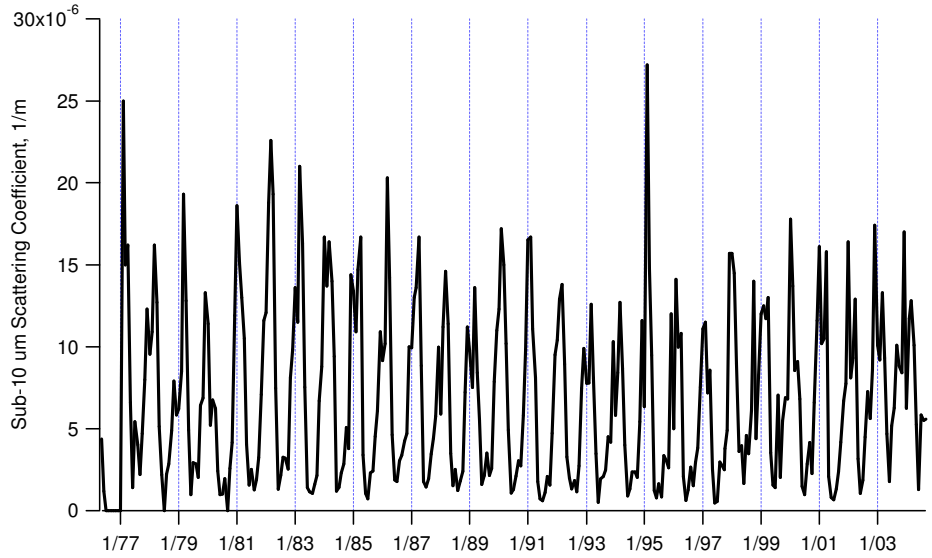

Figure 3. Monthly averaged light scattering at 550 nm by sub-10 micron aerosol at Barrow, Alaska. Data made available by NOAA CMDL.

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 increase solar cloud albedo due to increasing the number concentration and decreasing the average size of cloud droplets provided the liquid water content in the clouds remains constant (Twomey, 1977). An increase in the number concentration of pollution aerosol particles that act as cloud condensation nuclei (CCN) will affect Arctic stratus and stratocumulus by increasing the cloud droplet number concentration which results in more radiation being reflected back to space (Albrecht, 1989; Twomey, 1991). The relatively 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) showed 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. In addition, Arctic stratus appears to be more sensitive to pollutant particles than clouds outside of the Arctic. 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.)

5. Boreal Forest Fires and their Effects on the Arctic

Climate change and boreal forest fires. Climate change is an accepted reality, and observed and forecast impacts 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., 1999; 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 in the Arctic free troposphere (e.g., during the ABLE 3A and 3B campaigns; Harriss et al., 1994, Shipham et al., 1992) and at the surface (e.g., Ily-Tuomi et al., 2003) exist. 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. An exciting new development with respect to climate change is the recent discovery of transport of biomass burning emissions into the lower stratosphere through an explosive combination of intense forest fires and extreme convection (see Figure 4). 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 (An-

dreae 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 17 km (remote sensing observations exist even at higher altitudes), several kilometers above the tropopause and at potential temperatures greater than 380 K (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 AURA, CALIPSO and METOP 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.

Figure 4. 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.

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 can remain 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.

6. Composition and Chemistry of the Arctic Troposphere

Background. The Arctic 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, it is impacted by emissions from many of the world's largest industrial regions (e.g., Rahn et al., 1977; Barrie, 1986). Initial research into Arctic Haze found that emissions from northern Eurasia were most significant, though recent evidence suggests that contributions from rapidly developing economies in eastern Asia are growing in importance (Koch and Hansen, 2005). In summer, the arctic free troposphere frequently receives "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. The discovery of Arctic Haze in the late 1950s led to the development of a network of surface observatories. Between 1983 and 1991 four AGASP airborne campaigns complemented the surface network by documenting the vertical and horizontal distribution and composition of haze in the lower troposphere over the western sector of the Arctic in late winter and spring. Sampling at Alert and Barrow in support of haze investigations discovered severe ozone depletion events (ODEs) in the boundary layer over the Arctic Ocean at the time of polar sunrise (Oltmans, 1981; Barrie et al., 1988). This lead to a series of increasingly intensive ground based campaigns in which the importance of halogen chemistry and the link to mercury depletion events were discovered (e.g., Schroeder et al., 1998; Bottenheim et al., 2002). The TOPSE airborne sampling campaign in 2000 helped to establish the vertical and geographic extent of ODEs in the Canadian Arctic, though the primary focus of this mission was to establish the cause of the springtime maximum in ozone in the Arctic mid troposphere (Atlas et al., 2003). The discovery of the stratospheric ozone hole over Antarctica led to a series of airborne campaigns probing also the Arctic stratosphere between 1989 and 2005. Unfortunately these missions devoted very few flight hours to sampling in the Arctic troposphere. The ABLE 3 summer campaigns in 1988 and 1990 did sample the North American free troposphere, but platform limitations and a focus on constraining surface fluxes resulted in much of the sampling at low levels (Harriss et al, 1992, 1994).

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 con-

cert with ozone/HO_x/NO_x raising the specter of a potential major contamination of the Arctic biosphere (Schroeder et al., 1998). Figure 5 and Figure 6 show examples of such coincident depletions and their vertical extent.

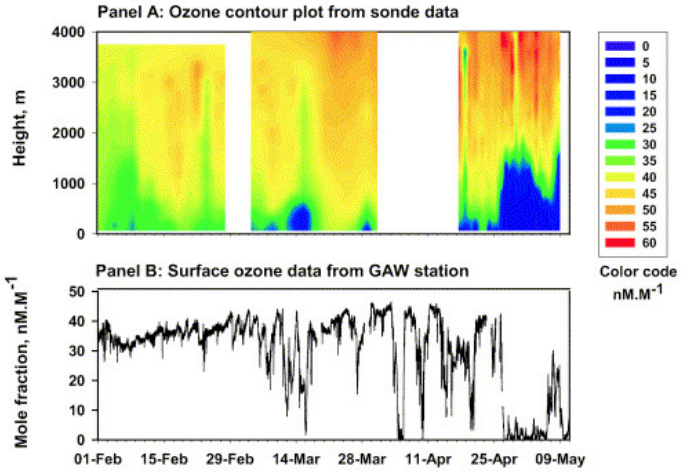

Figure 5. Surface ozone depletion at Alert (80ºN) and its vertical extent (from Bottenheim et al., 2002).

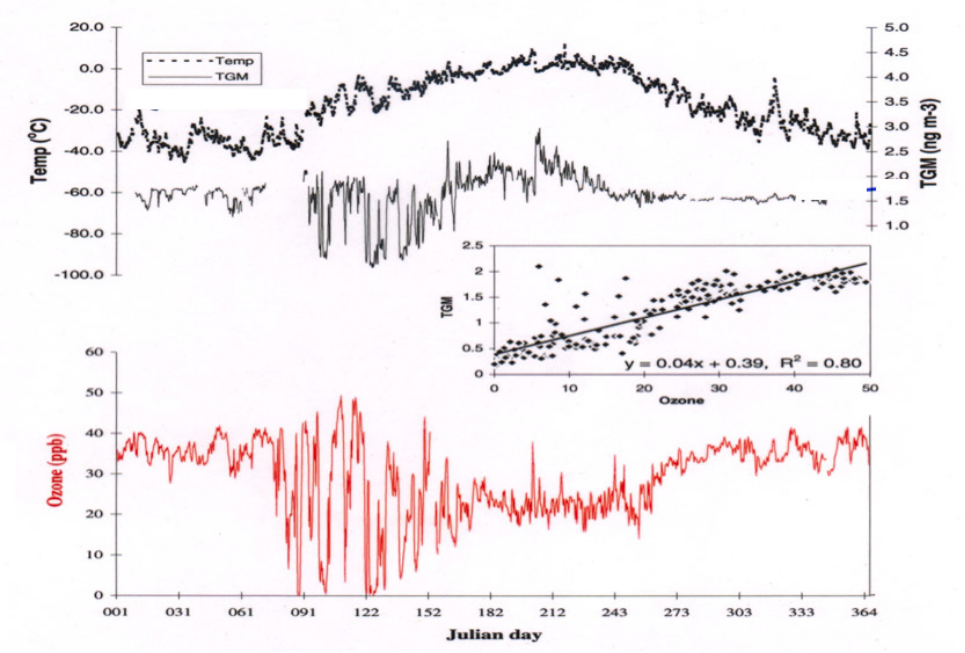

Figure 6. Simultaneous mercury and ozone depletion at Alert (82ºN) (Schroeder et al. 1998).

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

$Br + O_3 \rightarrow BrO + O_2$ (x2)
$BrO + BrO \rightarrow Br_2 + O_2$
$Br_2 + h\nu \rightarrow 2\ Br$

$2O_3 \rightarrow 3O_2$ (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 particles are likely implicated. Observational data are critically needed to analyze the coupled evolution of $BrO_x/ClO_x/IO$-NO_x-HO_x-O_3 chemistry during O_3 and Hg depletion events in the Arctic spring.

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 3-5 x 10^{13} mol cm^{-2} (Figure 7) 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.

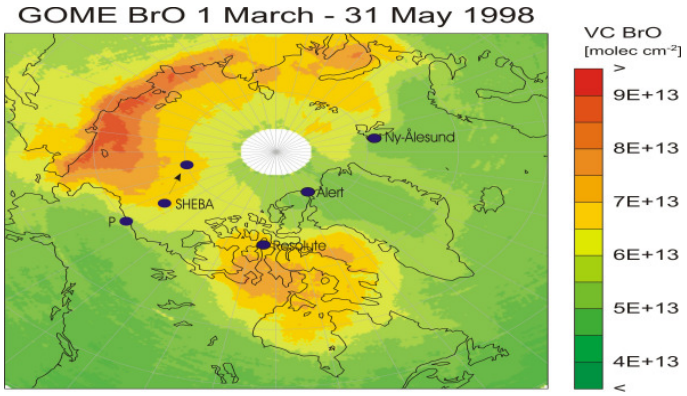

Figure 7. Tropospheric BrO column derived from the GOME satellite.

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 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 O_3 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 8 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 (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.

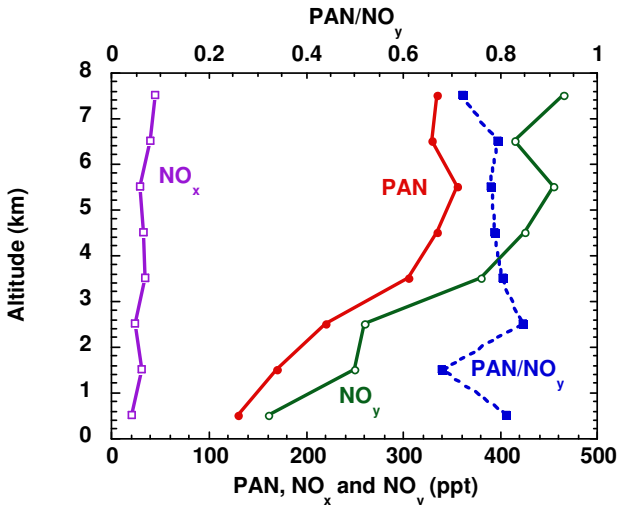

Figure 8. PAN, NO_x and total reactive nitrogen (NO_y) in the Arctic troposphere (50-80˚N) in May. Median mixing ratios as measured during TOPSE (see Atlas et al, 2003).

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.

7. Use of Satellite Data during POLARCAT

The Arctic is an ideal 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 (see Figure 9, for an example), 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, but also makes detection of light scattering by aerosols more difficult.

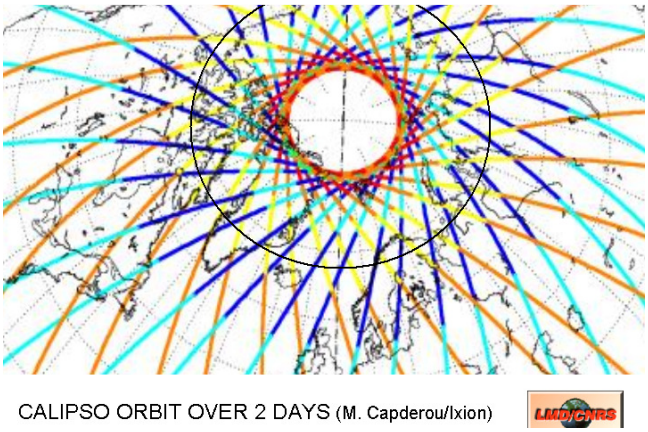

Figure 9. Coverage of CALIPSO measurements during ascending (orange/yellow/red) and descending (blue) orbits for two consecutive days. The polar circle is shown in black.

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 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.

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. Table 2 has been compiled from information given by people involved in the POLARCAT consortium.

Table 2. Satellite observations of atmospheric constituents and aerosols.

Instrument platform, launch year	*Total column*	*Tropospheric products*	*Stratospheric products*	*Other products*
GOME-1 ERS-2, 1995 GOME-2 Metop, 2005	O_3, BrO, NO_2, O_2, O_4 IO (potentially)	SO_2, HCHO, H_2O (column) O_3 (profiles) Clouds: fraction, top altitude, optical thickness & albedo	BrO, NO_2, OClO, O_3 (profiles) OClO (column) Aerosol optical thickness	
MOPITT Terra, 1999		CO (column/4 km)		
MISR Terra, 1999		Aerosol optical depth (column)		
MODIS Terra, 1999		Aerosol optical depth (column)		
SMR/OSIRIS Odin, 2001		Cirrus cloud occurrence	O_3, ClO, N_2O, HNO_3, H_2O, CO, NO, NO_2, isotopes of H_2O & O_3 (profiles) PSC occurence	NLCs
AATSR Envisat, 2002		aerosol optical depth & particle size		SST
MIPAS Envisat, 2002		H_2O, N_2O, CH_4, O_3, HNO_3, CFC-11/12, HCFC-22 (upper troposphere) Cirrus: occurrence, optical thickness	H_2O, N_2O, CH_4, O_3, HNO_3, CFC-11/12, HCFC-22, $ClONO_2$, N_2O_5, NO_2, NO, CO, HNO_4, CCl_4, CF_4 (profiles) PSCs volcanic aerosols	
SCIAMACHY Envisat, 2002	O_3, BrO, NO_2, O_2, O_4 IO (planned)	SO_2, HCHO, H_2O, CO, CH_4 (column) Clouds: fraction, top altitude, droplet/grain size, optical thickness, albedo, liquid/ice water path, thermodynamic state	BrO, NO_2, OClO, O_3 (profiles ~10-40 km) OClO (column) Aerosol optical thickness PSCs	NLCs
AIRS Aqua, 2002		CO (column/4 km)		
AMSR-E Aqua, 2002				Sea ice, snow
MODIS Aqua, 2002		Aerosol optical depth (column)		
ACE-FTS SCISAT-1, 2003		H_2O, O_3, N_2O, CO, CH_4, NO, NO_2, HNO_3, HF, HCl, N_2O_5, $ClONO_2$, CFC-11/12, COF_2, HCFC-	H_2O, O_3, N_2O, CO, CH_4, NO, NO_2, HNO_3, HF, HCl, N_2O_5, $ClONO_2$, CFC-11/12, COF_2, HCFC-22, HDO, SF_6, CF_4	

Instrument platform, launch year	*Total column*	*Tropospheric products*	*Stratospheric products*	*Other products*
		22, HDO, SF_6, HCN, CH_3Cl, CF_4, C_2H_2, C_2H_6 (profiles)	(profiles)	
TES Aura, 2004		CO, CH_4, O_3, HNO_3, NO_2 (column/4 km)		
OMI Aura, 2004		O_3, NO_2, SO_2, HCHO, BrO (column)		
MLS Aura, 2004		H_2O, HCN (upper troposphere)	H_2O, HCN (profiles)	
IASI Metop, 2006	H_2O, CO, O_3, CH_4, HNO_3	O_3 (profiles), CO, H_2O (profiles)		
CALIOP Calipso		Aerosol distribution (profiles)		
CPR CloudSat		Cloud liquid and ice water profiles, precipitation		
POLDER Parasol		Cloud and fine mode optical depths and particle size		
AVHRR on NOAA 15-18 MODIS on Terra & Aqua Geostat. imagers on GMS, GOES 10 & 12, Meteosat				composite Arctic visible, infrared and water vapor images

8. 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 using the measurements made, 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.

In the 20th century, the anthropogenic emissions resulted in a general increase of the aerosol load (sulfate as well as carbonaceous) in North America, Europe, and East Asia. In the first part of the 21st century, according to the IPCC SRES scenario A1B, the most polluted regions are found at lower latitudes (Brazil, Africa, the Arabian Peninsula, India and China) whilst sulphate and carbonaceous aerosols have both decreased in North America and Europe. To 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.

9. Overview of the Planned Activities

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. For instance, it is planned that aircraft perform overflights over the icebreakers used in other programs (e.g., OASIS).

POLARCAT is a bottom-up project that will remain open for others to join. Therefore, the following description of activities reflects the current stage of planning. Additional activities can be suggested by others and added at any time. 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. In the following, short descriptions of all planned activities (aircraft experiments, ship cruises, balloon flights, surface stations, remote sensing, models) will be given. Table 3 gives an overview of the time schedule for the various activities.

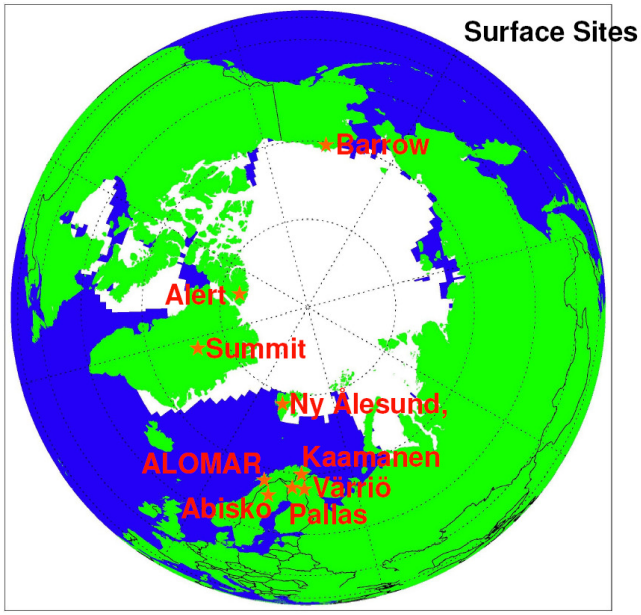

Figure 10. Location of major POLARCAT surface measurement sites.

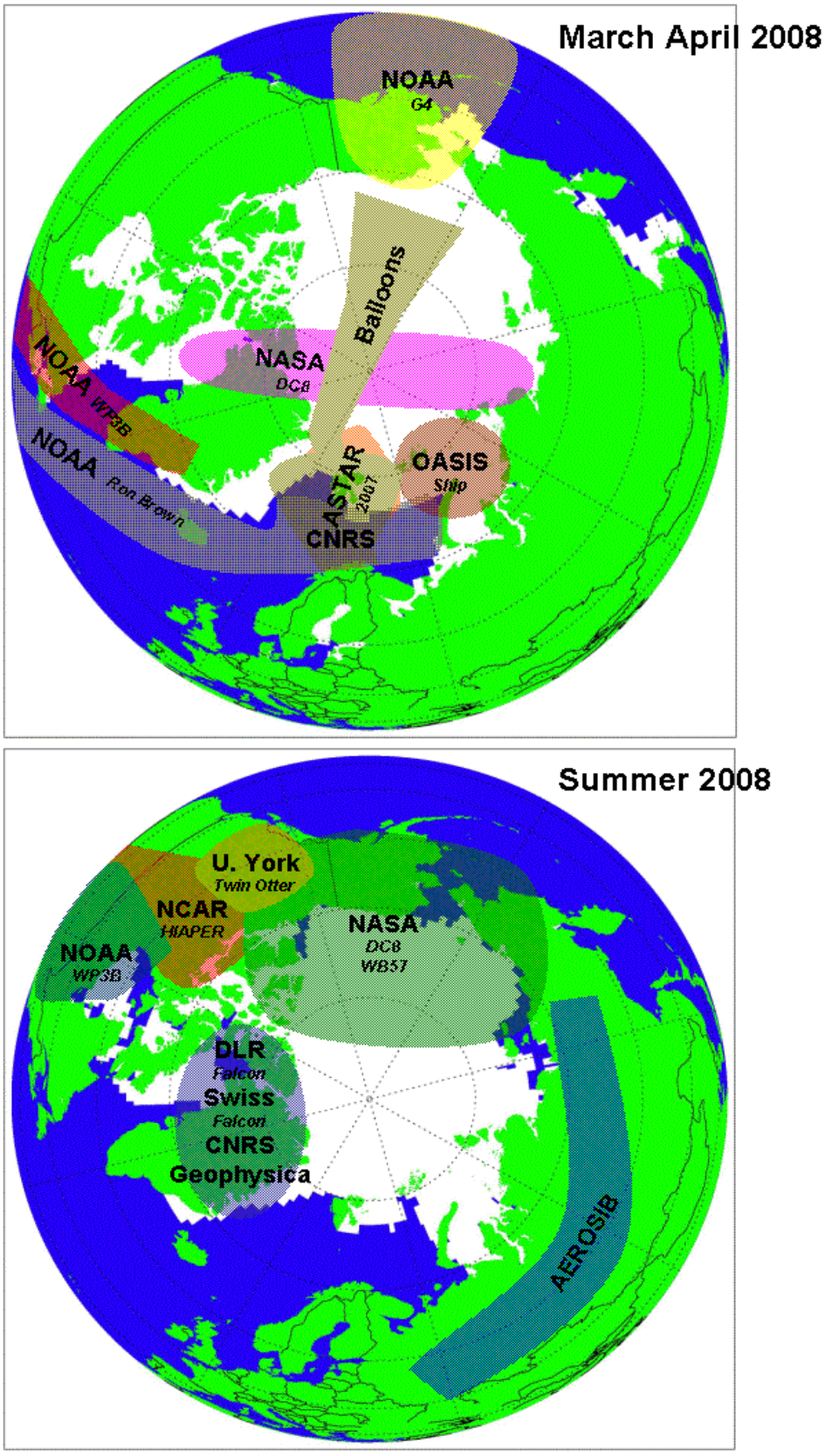

Figure 11. Summary of major field activities in spring and summer 2008. White areas show typical sea ice extent in the respective season.

Table 3. Time schedule for the major POLARCAT activities. T indicates tentative but with firm plans to submit a proposal, P indicates that a proposal has already been submitted, S indicates that this is an activity for which funding is already secured. Planned aircraft bases are indicated.

Planned platform availability	***2007***												***2008***												***2009***		
	M	A	M	J	J	A	S	O	N	D	J	F	M	A	M	J	J	A	S	O	N	D	J	F	M
Aircraft																										
Twin Otter, Northern Canada																T	T									
French Falcon, Northern Europe												T		T		T	T									
DLR Falcon, Svalbard		P																								
DLR Falcon, Kiruna/Greenl.																T	T									
AWI Polar 2, Kiruna or Greenl.																T	T									
Geophysica, Kiruna or Greenl.																T	T									
AWI Do-228, Svalbard		S																								
Bae-146, Norway								T				T		T												
DC-8, Thule, GL; Barrow, Alaska														T			T									
WB-57, Barrow Alaska																	T									
NOAA Twin Otter, N. America																P	P									
NOAA P3, N. Am., to Greenl.												P	P			P	P									
NOAA GIV, Alaska													P	P												
NCAR HIAPER, CO – Alaska																P	P									
York Univ. Twin Otter, Canada																	T	T								
Swiss Learjet, Svalbard					T											T										
YAK-SIB			T	T	T	T								T	T	T	T									
Ships																										
NOAA R.H. Brown												P	P	P												
Amundsen Icebreaker, Hg																	P	P	P	P	P					
Balloons																										
Lagrangian Balloons												T		T												
Surface stations																										
Lidar, AOD, etc., measurements at Andøya	s	o	m	e		a	c	t	i	v	i	t	i	e	s		f	u	n	d	e	d			
Surface measurements at Summit	s	o	m	e		a	c	t	i	v	i	t	i	e	s		f	u	n	d	e	d			
Surface chemical measurements at Zeppelin mountain, Svalbard	s	o	m	e		a	c	t	i	v	i	t	i	e	s		f	u	n	d	e	d			
Aerosol lidar, spectrophotometers, surface chemistry, etc., at Ny-Ålesund, Svalbard	re	gu	lar		m	e	a	s	u	r	e	m	e	n	t	s		f	u	n	d	e	d		
Precipitation chemistry, aerosol spectrophotometer, meteorological station at Hornsund, Svalbard	re	gu	lar		m	e	a	s	u	r	e	m	e	n	t	s		f	u	n	d	e	d		
Aerosol, chemistry, trace gas and ecological measurements at SMEAR I-station, Värriö	s	o	m	e		a	c	t	i	v	i	t	i	e	s		f	u	n	d	e	d			
Aerosol, chemistry and trace gas measurements at GAW Pallas	s	o	m	e		a	c	t	i	v	i	t	i	e	s		f	u	n	d	e	d			

Gas, gas exchange and ecological measurements at Kaamanen	s	o	m	e		a	c	t	i	v	i	t	i	e	s		f	u	n	d	e	d			
Meteorological and ecological measurements at Abisko	s	o	m	e		a	c	t	i	v	i	t	i	e	s		f	u	n	d	e	d			
CO_2 and O_2 continuous monitoring at Cape Farewell	s	o	m	e		a	c	t	i	v	i	t	i	e	s		f	u	n	d	e	d			

Aircraft experiments

- **ASTAR 2007 Experiment** - The ASTAR (Arctic Study on Tropospheric Aerosol, Clouds and Radiation) field experiment in April 2007 involving two research aircraft as well as ground measurements based in Longyearbyen and Ny-Ålesund, Svalbard (78ºN). The two aircraft (the AWI Polar 2 aircraft, a Dornier 228 turboprop type, and, to be confirmed, the DLR Falcon 20) will carry an extensive payload to measure: complete aerosol size distribution, aerosol volatility, aerosol chemical composition, aerosol extinction (vertical profiles), air mass tracers, SO_2, spectral radiance and as well as cloud physical properties (particle morphology and size, in-cloud partitioning of ice/water content) with the aim to study the aerosol direct and indirect effects. An additional focus is CALIPSO validation.
- **NOAA 2008 IPY Proposed Climate Study based on the NOAA WP-3D aircraft**. Thirty hours of flight time have been requested to be concurrent with the *RV Ronald H. Brown* cruise in February/March 2008. This limited flight time will allow several flights into the far northern Atlantic. In addition, 60 flight hours have been requested for a summertime study (concurrent with the NOAA Twin Otter aircraft) for a climate research study on cloud-aerosol interaction. The study will be conducted somewhere in North America, and the arctic region may be selected.
- **NOAA 2008 IPY Proposed Climate Study onboard the NOAA Twin Otter aircraft.** Fifty flight hours have been requested for a summertime study (concurrent with the NOAA WP-3D aircraft) for a climate research study on cloud-aerosol interaction. The study will be conducted somewhere in North America, and the arctic region may be selected.
- **NOAA 2008 IPY Proposed Climate Study onboard the NOAA G-IV aircraft.** In March and April, 2008 150 flight hours have been requested to investigate transport across the Pacific and the impact on climate forcing. It is expected that at least a fraction of these flights will be flown into the Arctic from a base in Anchorage, Alaska or the Aleutian Islands.
- **NASA Proposed IPY Experiment in spring (April) 2008**. NASA DC-8 will be the primary airborne platform for this activity. Its long range and high altitude capability (0-12 km) makes it ideal to cover most of the arctic troposphere and coordinate activity with IPY partners. Table 4 shows the expected DC-8 instrument payload with the capability to measure nearly all important gaseous and aerosol constituents in the arctic troposphere. The platform is also ideal for validation of satellite data. It is envisioned that approximately 100 DC-8 flight hours will be available for this phase of the experiment. Thule, Greenland would be a suitable base of operation.
- **NASA Proposed IPY Experiment in summer (July) 2008.** The summer experiment would employ both the DC-8 and the WB-57 aircrafts. The DC-8 payload would be the same as described in Table 4. The evolution of arctic tropospheric chemistry from spring to summer and the influences of boreal fires would be the principal focus of this summer

study. The WB-57, with its ability to penetrate the polar stratosphere, would be especially useful to investigate stratospheric pollution resulting from deep pyro-convection events. The WB-57 payload (Table 4) will be tailored to assess this important phenomenon in the UTLS region including satellite validation. Approximately 100 DC-8 and 60 WB-57 flight hours would be requested for the 2008 summer experiment. Tentatively both of these aircraft could be based in Barrow Alaska making it easier for them to coordinate activities.

Table 4. Anticipated NASA DC-8 and WB-57 payload.

A. DC-8
Gas Phase/in-situ:
Nitrogen - NO, NO_2, PANs, HNO_3, HNO_4, $RONO_2$, NO_y, NH_3
Carbon - CO_2, CO, CH_4, C_2-C_{10} NMHCs
Oxidants - O_3, $OH/HO_2/RO_2$, H_2O_2, ROOH
Oxidized organics - CH_2O, CH_3CHO, $(CH_3)_2CO$, CH_3OH, others
Sulfur - SO_2, DMS
Halogens – ClO/BrO/IO, Cl_2/Br_2 (?)
Others- H_2O, VOCs, Halocarbon tracers, organic acids, mercury, POPs
Aerosol/in-situ:
Fine/ultra-fine CN : CN- volatile (>3 nm), CN- nonvolatile (>10 nm)
Size distribution (3-20,000 nm), surface area etc.
Black carbon
Aerosol bulk composition (SO_4, NO_3, organic)
Scattering, absorption, extinction
Remote: O_3 lidar, aerosol lidar, optical depth, T
Physical: Spectral irradiances, MMS (T, P, u, v, w), DP, RH, albedo
B. WB-57
Gas Phase/in-situ: Carbon- CO, CO_2, CH_4 Nitrogen- $NO/HNO_3/NO_y$ Other- , H_2O (total and vapor), O_3, PAN, CFCs
Aerosol/in-situ: Black carbon, aerosol size and shape, aerosol bulk and single particle composition
Remote Sensing: Cloud Lidar, Temperature Profile

- **DLR POLARCAT Summer 2008 Experiment** - Flights with the DLR Falcon are proposed to study the transport of forest fire pollutant plumes into the Arctic troposphere and lower stratosphere. A special focus will be on the role of pyro-convection in injecting pollutants deep into the lower stratosphere. The DLR Falcon will be based down-wind of the major North American forest fire regions, most probably in northern Greenland or northern Sweden. The DLR Falcon will be instrumented to measure in situ chemical species and aerosol properties. It is proposed to combine the DLR Falcon campaign with the deployment of an additional path-finding aircraft using on-board lidar measurements to guide the DLR Falcon into pollution layers (e.g., the French ATR).
- **Geophysica Summer 2008 Arctic Experiment.** It is proposed to employ the high-altitude aircraft M55 Geophysica during the POLARCAT summer campaign in 2008. The principle focus would be the impact of boral fires, especially pollution from pyro-convection, on the chemistry and aerosol properties in the lower stratosphere. It is foreseen to base the Geophysica together with the DLR Falcon and a lidar pathfinding aircraft in Northern Greenland or Northern Sweden. The Geophysica will be instrumented with in situ tracer, chemistry, and aerosol instruments, as well as lidars.

- **DLR's TROPOLEX airborne ozone/aerosol lidar**, currently installed on a Cessna-Caravan, is proposed to study Arctic haze outbreaks during the breakdown of the polluted polar dome in late winter. In coordination with other aircraft and balloons, the distribution (layers, gradients) of polluted air masses during persisting isolation will be characterized. Then, quasi-Lagrangian flights along forecasted transport pathways will follow the chemical and dynamical transformations along the flow in order to validate and improve 3-D CTM's. The lidar measures vertical profiles of particle backscatter ratio @ 286-296 nm and 355nm and ozone (down to 5 ppb) from the ground up to ≈ 3-4 km with horizontal and vertical resolutions of $\Delta_{x,aer}$≈100m/$\Delta_{x,O3}$≈1km and $\Delta_{z,aer}$≈30m/$\Delta_{z,O3}$≈300m, respectively. The preferred aircraft operation base will be Northern Scandinavia or the west coast of Greenland. A deployment onboard a DO-228 may also be considered, eventually in connection with IPY activities of the AWI.
- **AWI Summer 2008 Experiment** - Part of the DLR POLARCAT activities. Different aerosol systems will be installed at the DLR Falcon to investigate the aerosol distribution and their transport pathways from anthropogenic aerosol in the Arctic. The Polar 2 will operate also from Kiruna or Greenland and will be equipped with different systems for measurements of the cloud micro-physical parameters, with the aim to investigation of cloud micro-physical and optical properties in the Arctic as a function of different tropospheric aerosol load and the regional extent of aerosol and cloud structures (indirect climate effect). The Airborne Mobile Aerosol Lidar (AMALi) will measure the horizontal aerosol distribution and will be used to identify polluted layers.
- **CNRS/INSU POLARCAT Experiment** - Flights are proposed using a French aircraft (ATR or F20) equipped with airborne ozone and aerosol lidars. The ATR (max. altitude 7km) or the F20 (max. altitude 12km) can be flown with lidars in upward or downward looking mode. It is proposed to make flights in the late winter/spring timeframe either in combination with ASTAR in 2007 or possibly with other aircraft making in-situ measurements in spring 2008, ideally from a base in northern Scandinavia. The focus will be on European outflow/inflow and quantification of the relative contributions of photochemistry and STE. The aircraft will act as a pathfinder for in-situ measurements of trace gases and aerosol plumes and also to map out the vertical structure and transport of such plumes in the Arctic region. Flights would also be made, if within range, near the OASIS ship and over Ny Ålesund to link with surface experiments looking at ODEs and mercury cycling by providing information about the structure of the lower troposphere and entrainment of trace gases, such as O3 into the PBL. It is also proposed to make flights in the summer 2008, possibly with the DLR Falcon, with the focus on transport of forest fire pollutant plumes. In this case, it is proposed to base the aircraft in southern Greenland at Kangerlussuaq. Here, the ozone lidar can be used to provide information about the structure of the tropopause or the lower/mid troposphere whilst the aerosol lidar, in conjunction with CALIPSO aerosol lidar data can be used to map out the transport of forest fire layers into the PBL and in the lower stratosphere. In the case of flights mapping out the structure in the lower troposphere/PBL, overpasses of Summit are envisaged to examine, if possible, deposition of soot on snow surfaces and the impact on surface albedo. Flights will be also be linked, possibly in a Lagrangian sense with other aircraft making measurements at the same time (e.g. Canadian Twin Otter, NASA DC8, AEROSIB).
- **York University POLARCAT Experiment** – Flights with a Twin Otter targeting pollution from boreal forest fires are proposed for the summer 2008. The flights would be made within 100 km of the pyro-convection in order to determine the height of the material injected into the upper troposphere and lower stratosphere. The Twin Otter will carry two

lidars: one for measuring vertical profiles of ozone, the other for detecting clouds and aerosol layers.

- **AEROSIB (Airborne Extensive Regional Observations in Siberia)** – As part of an on-going CNRS/INTAS initiative, a series of regular flights are planned (depending on positive outcome of current funding request) over Siberia during summer periods starting in 2006 (see Figure 12). During each period, from June to September, 3 to 4 missions are proposed making transects from Tomsk across to Yakutsk in the lower troposphere below 4 km using a Russian Antonov-30 (OPTIK-E) aircraft, operated by Inst. Atmos. Optics (IAO-SB-RAS), Tomsk, equipped to measure CO_2, O_3 and CO continuously. Air samples will also be collected for further trace gas and isotope analysis. The main objective is to use the aircraft data, in conjunction with surface site measurements of CO_2 fluxes, to improve our knowledge about the role of the Russian forest and tundra/permafrost ecosystems in the carbon balance. The other main objective is to collect data on O_3 and CO in order to characterise the transport of O_3 and its precursors from industrial areas west of the Urals and from boreal forest fires in Siberia which have been shown to have a significant impact on free tropospheric and lower stratospheric composition downwind (Nedelec et al., 2005). Inverse modelling will be used to analyse results.

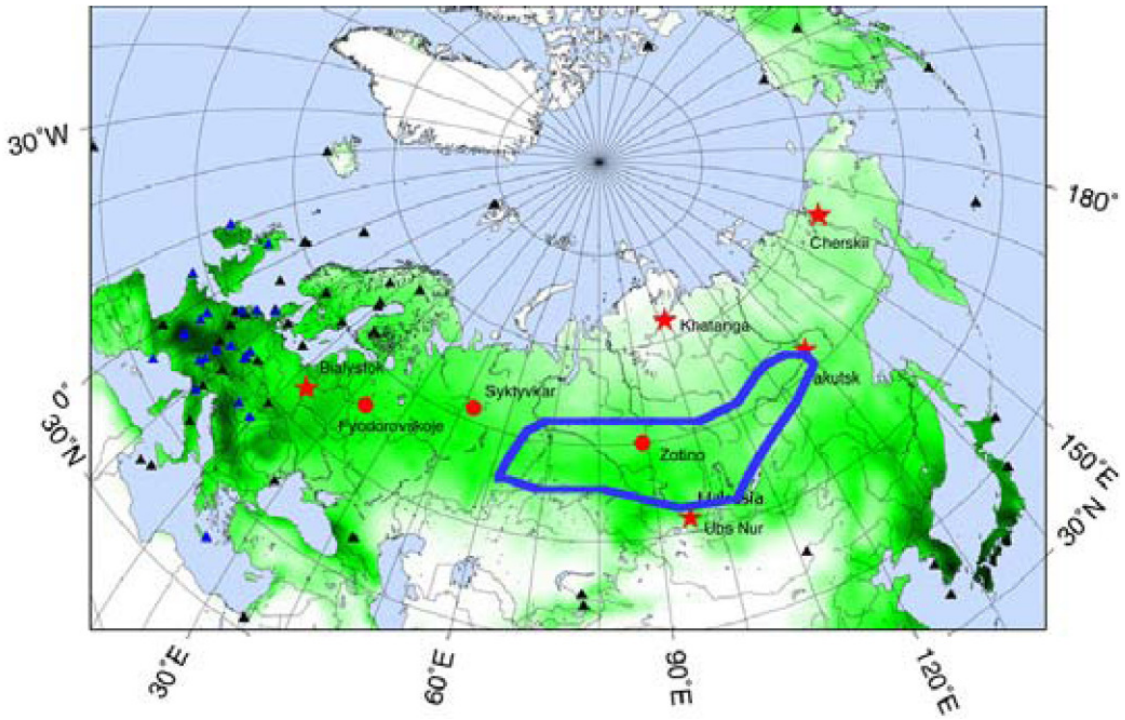

Figure 12. AEROSIB flights (courtesy – Ciais et al.) - The blue line indicates the proposed flight route. Monitoring stations over northern Eurasia region. The transcontinental flights will complete a set of ground-based and infrequent (1 per month) pointwise airborne observations (Red Symbols) carried out by European researchers (EU funded project TCOS-Siberia 2002-2004). Each station is co-located with continuous long-term eddy covariance surface CO_2 flux measurements. Triangles show the denser network of surface CO_2 monitoring sites and eddy covariance flux measurements existing in Western Europe.

- **Swiss Learjet POLARCAT experiment.** The Institute of Applied Physics (IAP) at the University of Bern, Switzerland, operates the airborne microwave radiometer AMSOS. This instrument is capable of retrieving water vapour profiles from approximately 12-60 km above the aircraft along the flight track. The instrument has been operating from a Learjet of the Swiss Air Force in yearly campaigns since 1998.

Ship cruises

- **NOAA 2008 IPY Proposed Climate Study onboard the *RV Ronald H. Brown.*** A cruise is proposed for February and March 2008 starting with two weeks in the Gulf of Maine (New England Air Quality Study) followed by a 9 day transit to Iceland, and 3 weeks in the Greenland, Norwegian, and Barents Seas. Measured parameters will include gas phase species (O_3, CO, CO_2, SO_2, NO, NO_2, NOy, PANS, $RONO_2$, HNO_3, VOCs), particulate phase species (inorganic ions, OC, EC), aerosol light scattering, absorption, extinction, and optical depth, aerosol size distributions, aerosol vertical profiles, and meteorological parameters.
- **Mercury measurements will be made on board the Amundsen icebreaker** on a cruise through Hudson Bay and the Canadian Maritimes.
- **The OASIS icebreaker m/s Antarctica** will be frozen in initially at approximately 85°N, 100°E and, via co-operation with OASIS, is also available as a field site for POLARCAT. Furthermore, POLARCAT overflights of the m/s Antarctica are planned.

Balloon flights

- **POLARCAT proposed Lagrangian balloon study.** University of Massachusetts and Smith College proposed deployment of controlled meteorological balloons in March and April 2008. Release of balloons from sites in North America and northern Europe to explore Lagrangian transport into the Arctic, quantify in-situ heating/cooling rates, and to characterize the fine-scale thermal stratification of the lower troposphere in the remote Arctic. Co-ordination with the aircraft will allow quantifying net chemical production/loss and aerosol development in a Lagrangian framework.

Surface stations, remote sensing

- **Ny-Ålesund** (78°54'N, 11°53'E). The Atmospheric Observatory at Koldewey station is continuously monitoring meteorological and radiation parameters, aerosol optical depth, aerosol profiles, water vapor profiles, and various trace gas column densities using lidars, photometers, spectrometers and standard meteorological instruments. Such long term data is analyzed for interannual variability and sets the scene for intensified measurements during the IPY period. Campaigns will address Arctic Haze features during spring periods of 2007-2009. Provided parameters will include: AOD, vertical structure of aerosols, temporal development of aerosol and trace gases, and their radiative effects. A particular nvesttive of the remote sensing investigations is the validation of new satellite-born sensors (e.g., CALIPSO).
- **Ny-Ålesund, TOPAS** (lead-U. Bremen). Ground-based remote sensing measurements of atmospheric trace gases using the infrared (FTIR) and microwave (MW) spectrometry with the sun (or moon during the polar night) as light source or in emission. The FTIR-observations yield the total column concentrations of up to 20 trace gases. Those determined by tropospheric contribution: CO_2, N_2O, CFC-12, CFC-22, CH_4, C_2H_2, C_2H_6, CH_2O, CO, SF_6, HCN, H_2O, OCS. Those determined by stratospheric contribution: HCl, HNO_3, NO, NO_2, $ClONO_2$, O_3. The MW observations yield the stratospheric trace gas profiles of O_3, ClO, and H_2O.

Surface stations, in-situ

- **Summit, Greenland** (72°34'N, 38°28'W). Continuous baseline measurements that will be maintained throughout the IPY period include: standard meteorological parameters, radiation and micrometeorology to constrain energy balance at the surface, elemental com-

position of aerosols, elemental carbon in aerosols, aerosol-associated radionuclide tracers, in-situ ozone, and flask sampling as part of the NOAA CMDL carbon cycle and halocarbon networks. A MAX DOAS should be installed in summer, 2006, and will be operated as long as the station remains open. Ozone sondes are released routinely during winter and spring to study stratospheric ozone depletion, launches will be extended to overlap the POLARCAT campaigns. In addition, intensive sampling campaigns studying active photochemistry within and above the snow, and fluxes of reactive compounds into the overlying free troposphere have been proposed for summer during the IPY time frame.

- **Alert, Nunavut** (82.5°N, 62.3°W). The extensive routine measurement program which is part of the Global Atmospheric Watch (GAW) program will be continuing. In addition to meteorological records going back to 1950, Alert features an aerosol chemistry program since 1980 (elemental carbon since 1987), a very extensive trace gas program since 1987 (both in-situ and flask sampling, including extensive carbon cycle monitoring, ozone, PAN, VOCs, Hg, and POPs, weekly ozone soundings). Recent additions to the program include continuous MAX-DOAS measurements (since 2003) and a renewed aerosol physics program. Alert is also part of the SEARCH network.
- **HgCanEurasia** (lead Met. Service, Canada) – Speciated Hg measurements in Kuujjuarapik (Quebec) and French Alps and proposed in Jilin Province (China) to study human impacts on mercury in the Arctic and the contribution of different source regions.
- **GRAMM** (lead – LSCE-CNRS/CEA) – New proposal for CO_2, O_2 measurements at Cape Farewell, southern coast of Greenland (as part of CARBO-Ocean) to improve knowledge about carbon fluxes and role of long-range transport from anthropogenic source regions.
- **Zeppelin mountain** (78°54'N, 11°53'E)**.** The Zeppelin mountain station in close vicinity to Ny-Ålesund, but at 475 m asl provides a number of aerosol properties in situ, including aerosol size distributions, total aerosol number density, absorption coefficients, etc. Further data is sampled in the village, including precipitation of snow and rain, aerosol number density, scattering coefficients, and aerosol chemical composition.
- **Hornsund, Svalbard** (77°00'N 15°33'E). Recording of changes in components of Earth's magnetic and electric fields, measurements of structure of the ionosphere. Aerosol measurements within AERONET. Standard meteorological data. Sampling of rain and snow, measurements of pH and SpC, chemical analyses. Sampling of air for organic compounds (for ATMOPOL). Chemical laboratory with two ion chromatographs.
- **POLAP (lead Institute of Geophysics PAS and U. Silesia).** Collecting of precipitation samples at several surface sites (Kinnvika, Hornsund, Longyearbyen, Barentsburg, Ny Ålesund (Svalbard); Kola Peninsula, Kotelny Is., Tiksi, Wrangel Is. (Russia) and Tromso, Tarfala, Rovaniemi, Andoya, Bjørnoya, Hopen, Jan Mayen – northern Scandinavia, see also Figure 13) with aim to quantify man-made impacts and wet deposition of soluble species in the Arctic. Each precipitation event (rainfall and snowfall) will be sampled with the aim to combine pH and ion concentrations with meteorological data and synoptic maps, in order to determine the pollution origin. All inorganic ions (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, NH_4^{+}, SO_4^{2-}, Cl^{-}, NO_3^{-}) will be analysed and chemical composition of precipitation will be divided into marine and anthropogenic components. Isotopes of sulphur will be determined to confirm the anthropogenic sulphate source areas. Large volume precipitation events will be sampled for organic pollutants (PAHs, POPs).

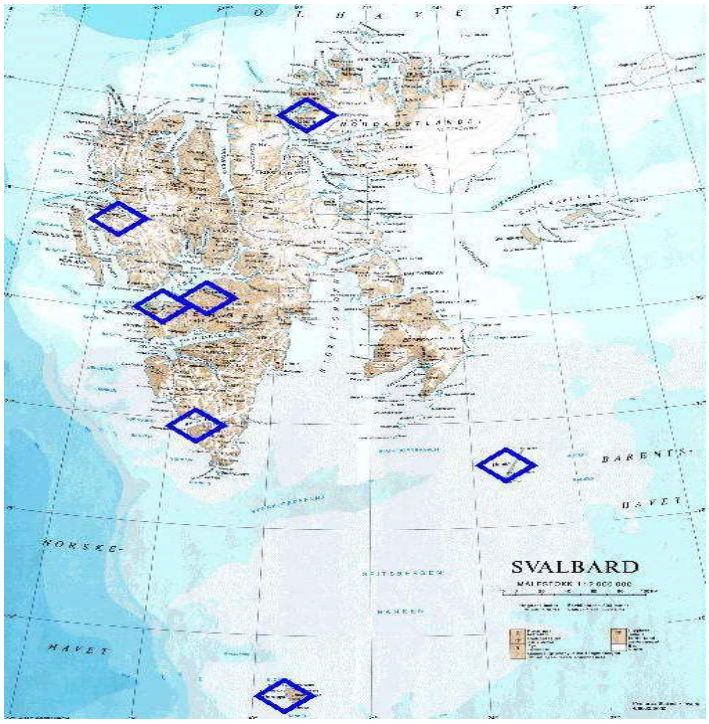

Figure 13. Location of precipitation sampling sites on Svalbard, Bjørnøya, and Hopen.

- **SMEAR I-station in Värriö, northern Finland** (69°46'N, 29°35'E). SO_2, O_3, NO_x (trace gases), aerosol particle number size distribution, photosynthesis, gas fluxes and meteorological data.
- **Pallas GAW-station in northern Finland** (67°58'N, 24°07'E, 565 m a.s.l.) has five separate stations within about 12 km of each other. Measured gaseous components are CO_2, CO, CH_4, N_2O, SF6, O_3, SO_2, NO_x, and radon. Aerosol properties measured are number concentration, scattering coefficient, size distribution, black carbon, and aerosol mass concentration..
- **Abisko station in northern Sweden** (68°21'N,18°49'E, 385 m a.s.l.). Instrumentation include eddy correlation flux tower for CO_2 and H_2O exchange, chambers for flux measurements of CO_2, CH_4 and total hydrocarbons, and meteorological measurements. It has also instrumentation for measuring emissions of volatile organic carbon (BVOC) compounds, a mass spectrometer, a scanning mobility particle system and an air ion spectrometer and a gas chromatograph.
- **ALOMAR, Andoya** (69.3ºN, 16.0ºE, 380 m a.s.l)**.** The core instrumentation consists of a steerable Rayleigh-Mie-Raman double lidar system, a (stratospheric) ozone lidar, a resonance lidar, and a tropospheric aerosol lidar. Furthermore, a MST radar covering the lowermost 15 km of the atmosphere and the upper mesosphere, an MF radar for mesospheric studies, a water vapour microwave spectrometer, Brewer, Bentham, OH spectrometers, several DOAS instruments and UV-Vis filter instruments, covering a wide range of atmospheric trace gases and UV irradiance. Currently, Aerosol Optical Density (AOD) photometers and ground spectral imaging spectrometers are being built up. ALOMAR receives EU support for international Access to Research Infrastructure.
- **Kaamanen CARBOEUROPE supersite, northern Finland** (69°08'N, 27°17'E, 155 m a.s.l.). Continuous micrometeorological CO_2 flux measurements.

- **Siberian Sites**. Continuous surface measurements of ozone and CO will be made at three locations in Siberia, at Mondy, Yakutsk and Tiksi, by Frontier Research Institute. These will focus on monitoring Arctic outflow and on quantifying high and mid-latitude transport (e.g., from European sources) affecting the Arctic region. It is hoped to add aerosol instrumentation at these sites.
- **Pico-NARE**. An integrated set of measurements of ozone and ozone precursors (NMHCs, nitrogen oxides, CO) and of aerosol size distribution, CCN, and aerosol absorption will be made at this free tropospheric station in the Azores islands. This station will provide information on the impact of arctic outflow, including boreal fire emissions, on the composition of the midlatitude free troposphere.

Use of satellite data

- **Merged Arctic meteorological satellite images**. University of Colorado/NOAA Aeronomy Laboratory and University of Wisconsin propose to combine the images from all four NOAA/POES satellites, Terra and Aqua satellites and the geostationary satellites, to produce high resolution, composite images of the Arctic every 2 hours in near real-time. These images will be generated using the same methodology developed by the Antarctic Meteorological Research Center that currently provides operational composite images of the Antarctic (Figure 14). Acquisition of data from other POES satellites and channels may also be possible and is being investigated.

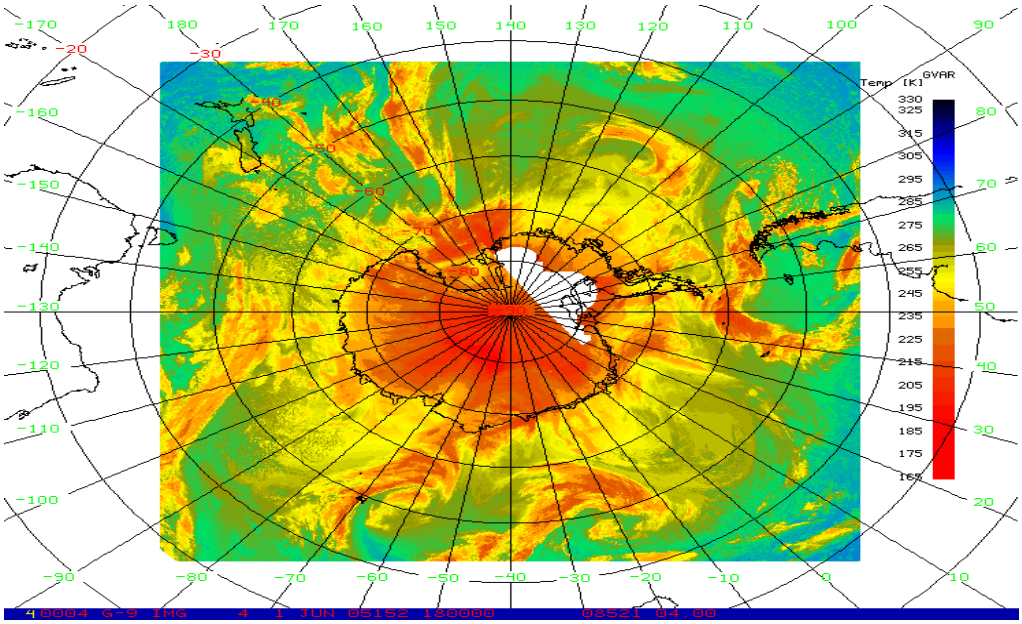

Figure 14. Infrared composite image above Antarctica for 18 UTC, June 1, 2005. This is a routinely produced product from the Antarctic Meteorological Research Center. Similar images for the Arctic will be provided.

- **Proposed satellite snow albedo study.** University of California proposes to study the influence of temperature and aerosol on the polar surface snow and icepack using satellite data, especially from MODIS and MISR. The objectives are estimating temperature, temperature gradient, and aging effects on snowpack specific surface area and reflectance, detecting dirty snow's radiative signature, and estimating the soot-albedo feedback effect.
- **Proposed ACE study.** ACE (Atmospheric Chemistry Experiment), also known as SCI-SAT-1 – lead Bernath (U. Waterloo, Canada) to provide columns and some vertical profile data in Arctic region with good spatial/temporal coverage (e.g. N_2O, CH_4, O_3, CO,

NO in polar stratosphere; C_2H_2 and C_2H_6 in upper troposphere). Measurements started in 2004.

- **Proposed CALIPSO data use.** Of special relevance for POLARCAT is the CALIPSO mission, which is developed as a joint effort between NASA in the USA, and CNES and CNRS in France. The satellite is to be launched end of summer 2005 to join the AQUA-Train on a polar orbit (98° inclination). CALIPSO instruments will provide an unprecedented set of measurements in the polar regions (see Figure 9). The CALIOP lidar offers the capability of aerosol profiling and identification using its two wavelength and depolarisation measurement capabilities. It also allows detecting cloud properties and haze profiling. For cloud property analysis, and to some extent for aerosol identification, it is complemented by the Infrared Imaging Radiometer (IIR) operating in three spectral bands in the thermal infrared window (8-13 µm). Launched at the end of the summer 2005, and with a lifetime of 3 years, it should be fully operational during the IPY. CALIOP and the IIR will help analysing polar haze properties during the winter season and bring new information on seasonal variations of high latitude fires and transport of biomass burning and pollution aerosols.

- **CO retrievals from IASI and ACE.** Service d'Aéronomie/CNRS and University of Brussels propose to use the combined CO distributions as retrieved from the IASI/Metop and ACE missions to study the transport of pollution plumes over the Artic area. IASI will provide total columns (Turquety et al., 2004) with an excellent horizontal coverage in that region and ACE is delivering profiles ranging from the mid-troposphere to the thermosphere (Clerbaux et al., 2005) but is limited by the solar occultation geometry of the measurement. A joint analysis will allow studying the CO transport over the pole due to boreal forest fires and due to anthropogenic pollution. Previous analysis has shown the usefulness of infrared sounders to follow pollution plumes over long distances (Figure 15). Other species such as ozone, HNO_3 and C_2H_6 will also be investigated.

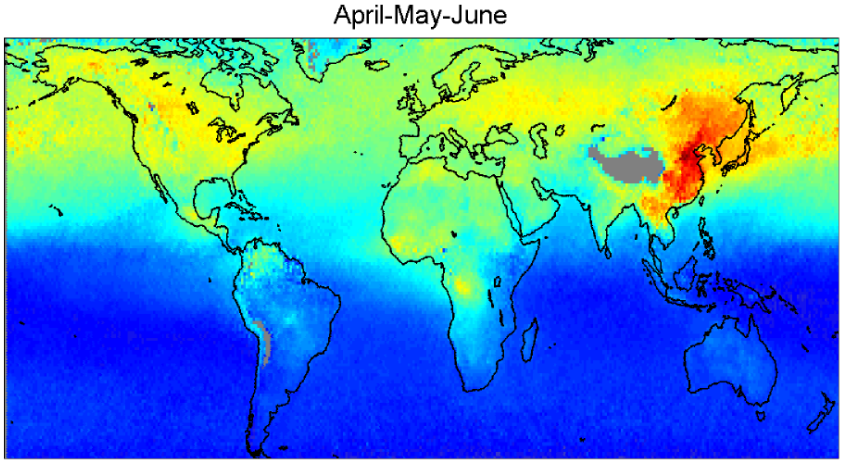

Figure 15. This composite image depicts the MOPITT CO measurements at an altitude of 850 hPa. The seasonal plot shows the observations averaged over 4 years for April-May-June. In the Northern hemisphere, most of the pollution is associated with urban activity and fires, such as those frequently observed over Alaska and Siberia (Clerbaux et al., 2004).

- **Use of combined data from GOME, SCIAMACHY, OMI, and GOME-2.** The Belgian Institute for Space Aeronomy at Brussels, the University of Bremen and the University of Heidelberg propose a joint effort to provide trace gas measurements (NO_2, BrO, SO_2, HCHO, OclO) from the GOME, SCIAMACHY, OMI and possibly GOME-2 instruments during POLARCAT. One of the two main scientific issues would be the quantification of

halogens (especially BrO) in the free troposphere and investigation of the respective roles of local production and transport processes. The other would be the impact of boreal fires and long-range transport on the content of pollutants (NO_2, HCHO) in the polar troposphere. These activities could be supported by ground-based measurements from Harestua, Norway, at 60°N.

Modeling

Here, the types of modeling activities planned for POLARCAT are listed. The activities planned by individual groups are not described, as too many of them will be involved in POLARCAT. Model products will include:

- Flow climatologies based on re-analysis data.
- Trajectory models, both in forecast and hindcast mode.
- Lagrangian particle dispersion models, both in forecast and hindcast mode.
- Chemistry transport models, both in forecast and hindcast mode.
- Fully coupled chemistry-aerosol-climate models will be used to simulate the effects resulting from scenarios for expected changes in emissions.

10. Project Management

Project structure. The POLARCAT activity forms part of the sub-cluster 4.1 on Clouds, aerosol and chemical composition. It has been agreed that within this sub-cluster there will be meetings of the activity leads in order to foster collaboration between the different activities. It is also proposed to have joint workshops and after the main field phase joint publications in journal special sections involving several activities. The cluster lead by the OASIS project will also form part of this grouping as their activities are closely linked to the 4.1 sub-cluster activities and in particular POLARCAT and AICI.

Within this overarching IPY umbrella, POLARCAT has defined its own management structure (see Figure 16). A Steering Group has been formed led by the activity lead (chair) and co-chair. It is made up of representatives from the original ITCT-Arctic proposal and other EoIs joining this activity within POLARCAT (e.g. POLAP, STEP, HgCanEurasia). The Steering Group will report directly to the Steering Committees of IGAC and SPARC who are in the process of endorsing this activity. This will ensure that the project is also overseen by independent experts within an international framework. It will also aide the synergistic co-ordination of POLARCAT with other major international atmospheric science activities. Furthermore, SPARC and IGAC could help provide support for workshops and other overarching activities. Note also that the POLARCAT SG includes current members of the IGAC SC (Law, Parrish) who are also IGAC representatives on the SPARC SC.

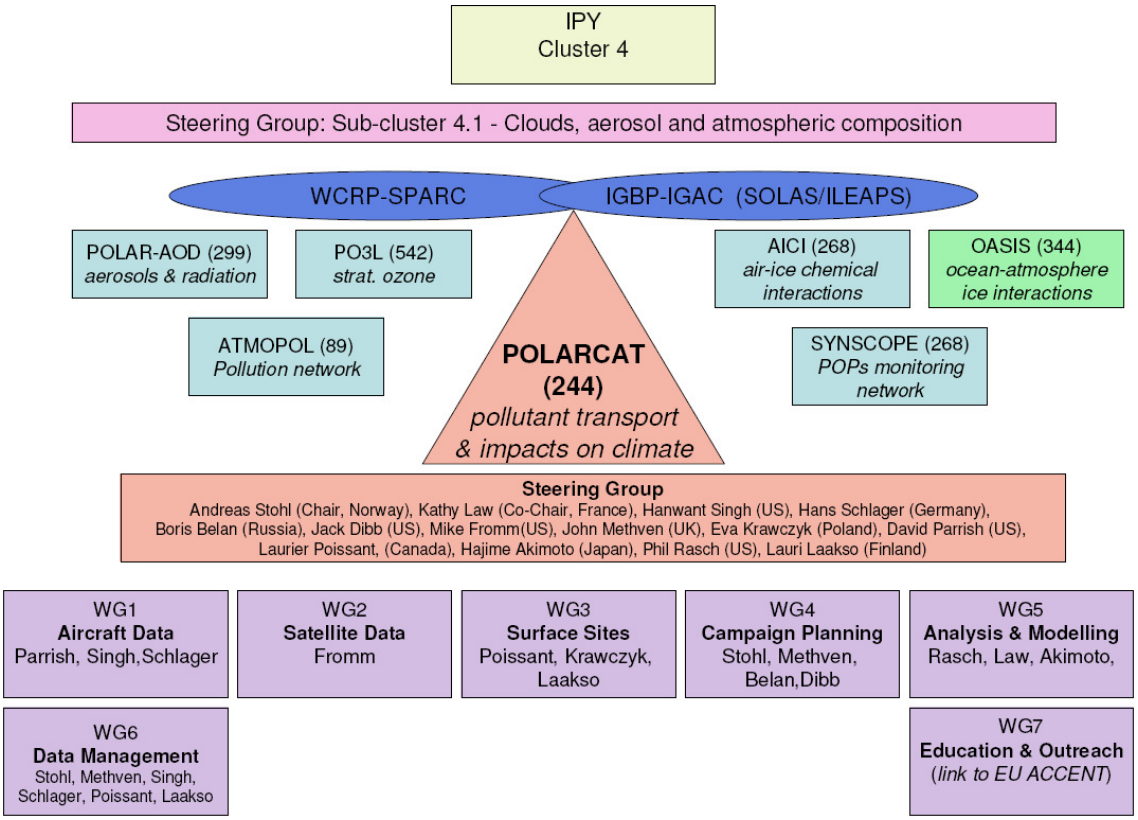

Figure 16. Position of POLARCAT in the cluster "Clouds, aerosol and atmospheric composition", and POLARCAT management structures.

POLARCAT has also defined several Working Groups which will be coordinated by task teams (SG members) responsible for coordinating activities within and between WGs. Towards the end of 2005, an implementation plan will be written describing in detail the activities that will take place as part of POLARCAT and links with other sub-cluster 4.1 and related (e.g. OASIS) activities. SG meetings and meetings/discussions with other sub-cluster leads will facilitate this process. Workshops will be held to plan and coordinate field intensives in 2007/8.

Quality Assurance/Quality Control. Each institution will be responsible for QA/QC following accepted guidelines. In addition to this, POLARCAT's Working Group 4 ("Campaign Planning") will organize a range of intercomparison activities, such as data intercomparison flights between different aircraft, overflights of ships and ground stations, satellite validation flights, etc. This will allow identifying possible problems with individual instruments.

Data management. POLARCAT will produce a wealth of data, which will be stored in internationally recognized data centers (e.g., NASA, British Atmospheric Data Centre, NILU database), which will all be linked from the POLARCAT website. All aircraft and ship data will be made available in a common format that was used already during the ICARTT field campaign. Common data formats for ground stations will be discussed at a common meeting of Working Groups 6 ("Data Management") and 3 ("Surface Sites").

POLARCAT will follow the IPY Data Policy, which will guarantee free data access after a period needed for validation and quality control of the data. Working Group 6 ("Data Management"), during its first meeting, will produce a viable plan for implementing the IPY Data Policy.

Education, Capacity Building, and Outreach. POLARCAT will provide PhD positions to train young researchers. Some of the participants will also offer courses for students at their universities. Outreach activities will also offer material for school teachers in order to attract young people into polar and atmospheric research. Courses will also be held at field stations (e.g. in STEP: Nordic graduate school CBACCI – Biosphere-Carbon-Aerosol-Cloud-Climate Interactions). Participants have a good record in communicating results to the public and making new results quickly available to the media. The project will make use of facilities being developed related to e-learning, particularly for undergraduates, in the framework of the EU ACCENT network. Outreach activities targeted towards school children will include "Teacher at Sea" (which already in the past has taken teachers onto the research vessel R.H. Brown during research cruises) or "Teachers and researchers exploring and collaborating" (TREC). A POLARCAT web site will be created that will serve the communication among researchers, the communication between researchers and journalists, and will also communicate information on the program to the public. Press conferences will be held during the field campaigns. Communication of polar atmospheric research to other atmospheric researchers will be carried out via reviewed publications, established newsletters (e.g., IGAC and SPARC) and presentations at international conferences.

11. List of Researchers and Institutes Involved in the IPY Expression of Intent Submission

POLARCAT is and will remain an open project. The following list gives the principal nvesttigator names and institutions involved in the submission of POLARCAT to the IPY International Programme Office. Most of these persons (and some not named here) have contributed to this document.

Name	Organisation	Country
Michel Carleer	U. Libre de Bruxelles	Belgium
Pierre Coheur	U. Libre de Bruxelles	Belgium
Jim Whiteway	York University	Canada
Laurier Poissant	Environment Canada	Canada
Brian Stocks	Canadian Forest Service	Canada
Peter Bernath	University of Waterloo	Canada
Jan Bottenheim	Environment Canada	Canada
Xinbin Feng	State Key Lab. Env. Geochem.	China
Bastidas/Rodriguez	U. Cauca	Columbia
Urmas Hõrrak	U. Tarto	Estonia
Lauri Laakso/ Markku Kulmala	U. Helsinki	Finland
John Moore	U. Lapland	Finland
Jussi Paatero / Risto Hillamo	Finnish Met. Inst.	Finland
Jacques Pelon	Service d'Aéronomie, CNRS, Paris	France
Gerard Ancellet	Service d'Aéronomie, CNRS, Paris	France
Cathy Clerbaux	Service d'Aéronomie, CNRS, Paris	France
J.-P. Cammas/P. Nedelec	Laboratoire d'Aérologie, Toulouse	France
Philippe Ricaud	Laboratoire d'Aérologie, Toulouse	France
Philippe Ciais/ M. Ramonet	LSCE/CEA-CNRS, Paris	France
Marc Delmotte	LSCE/CEA-CNRS, Paris	France
Christophe Ferreira	LGGE/CNRS, Grenoble	France
Jean-Francois Gayet	LaMP, U. Blaise Pascale, Clermont Ferrand	France
Hans Schlager	DLR	Germany
Andreas Minikin	DLR	Germany
Heidi Huntrieser	DLR	Germany
Andreas Fix	DLR	Germany
Fred Stroh	FZK, Julich	Germany
John Burrows, A. Kokhanovsky	U. Bremen	Germany
Thomas Wagner/ Ulrich Platt	U. Heidelberg	Germany
Roland Neuber	Alfred Wegener Institute, Potsdam	Germany
Andreas Herber	Alfred Wegener Institute, Bremerhaven	Germany
Justus Notholt	U. Bremen	Germany
Joerg Trentmann	U. Mainz	Germany
Gunnar Luderer	MPIC, Mainz	Germany
Thomas von Clarmann	IMK-FZ, Karlsruhe	Germany
Hans Feichter, Stefan Kinne	MPI Meteorology, Hamburg	Germany
Ugo Cortesi	IFAC/CNR	Italy
Francesco Cairo	CNR	Italy
Leopoldo Steffanuti	CNR/EEIG	Italy
Hajime Akimoto	Frontiers Research	Japan
Oliver Wild	Frontiers Research	Japan
Oystein Hov	Meteorological Office	Norway
Jon Egill Kristjansson	U. Oslo	Norway
Kjetil Torseth, Jozef Pacyna	NILU	Norway
Michael Gausa	Andoya Rocket Range	Norway
Jon Borre Oerbaek	Norsk Polarinstitutte, Tromso	Norway

Wieslawa Ewa Krawczyk	U. Silesia	Poland
Piotr Glowacki	Institute of Geophysics, PAS	Poland
Sandra Mogo	U. Beira Interior	Portugal
Boris Belan	Institute Atmospheric Optics, Tomsk	Russia
Nikolai Elansky	Institute Atmospheric Physics, Moscow	Russia
Sergey Pyramikov	AARIR, Saint Petersburg	Russia
Angel Frutos Baraja	U. Valladolid	Spain
Per Holmlund	Stockhom Univ.	Sweden
HC Hansson	U. Stockholm	Sweden
Jo Urban/ Donal Murtagh	Chalmers U.	Sweden
Michael Tjernstrom/ C. Leck	U. Stockholm	Sweden
Almut Arneth	U. Lund	Sweden
Dietrich Feist	U. Bern	Switzerland
John Methven	U. Reading	UK
Mathew Evans	U. Leeds	UK
Hugh Coe/ Tom Choularton	U. Manchester	UK
Claire Reeves	U. East Anglia	UK
Paul Monks/ John Remedios	U. Leicester	UK
Alastair Lewis	U. York	UK
Rod Jones/ John Pyle	U. Cambridge	UK
Tim Heaton	British Geological Survey	UK
Andy Hodson	U. Sheffield	UK
Eric Wolff	British Antarctic Survey	UK
Brian Kerridge	Rutherford Appleton Laboratory	UK
Hanwant Singh	NASA Ames	USA
Hans-Jurg Jost	NASA	USA
David Parrish/Michael Trainer	NOAA Aeronomy Lab	USA
Mike Fromm	Naval Research Lab	USA
Jack Dibb	U. New Hampshire	USA
Daniel Jacob	U. Harvard	USA
Paul Voss	U. Massachusetts	USA
Philip Russell	NASA	USA
John Burkhart	U. California, Merced	USA
Tim Bates / Patricia Quinn	NOAA	USA
Phil Rasch	NCAR	USA
Laura Pan	NCAR	USA
Steve Massie	NCAR	USA
AR Ravishankara	NOAA Aeronomy Lab	USA
Richard Honrath	Michigan Technological University	USA
Mark Hermanson	U. Pennsylvania	USA
Charlie Zender/ Mark Flanner	U. California-Irvine	USA
Dong Wu	JPL/Caltech	USA
Owen Cooper	CIRES/U. Colorado/NOAA	USA
Matthew Lazzara	Antarctic Met. Research Centre, U. Wisconsin	USA

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