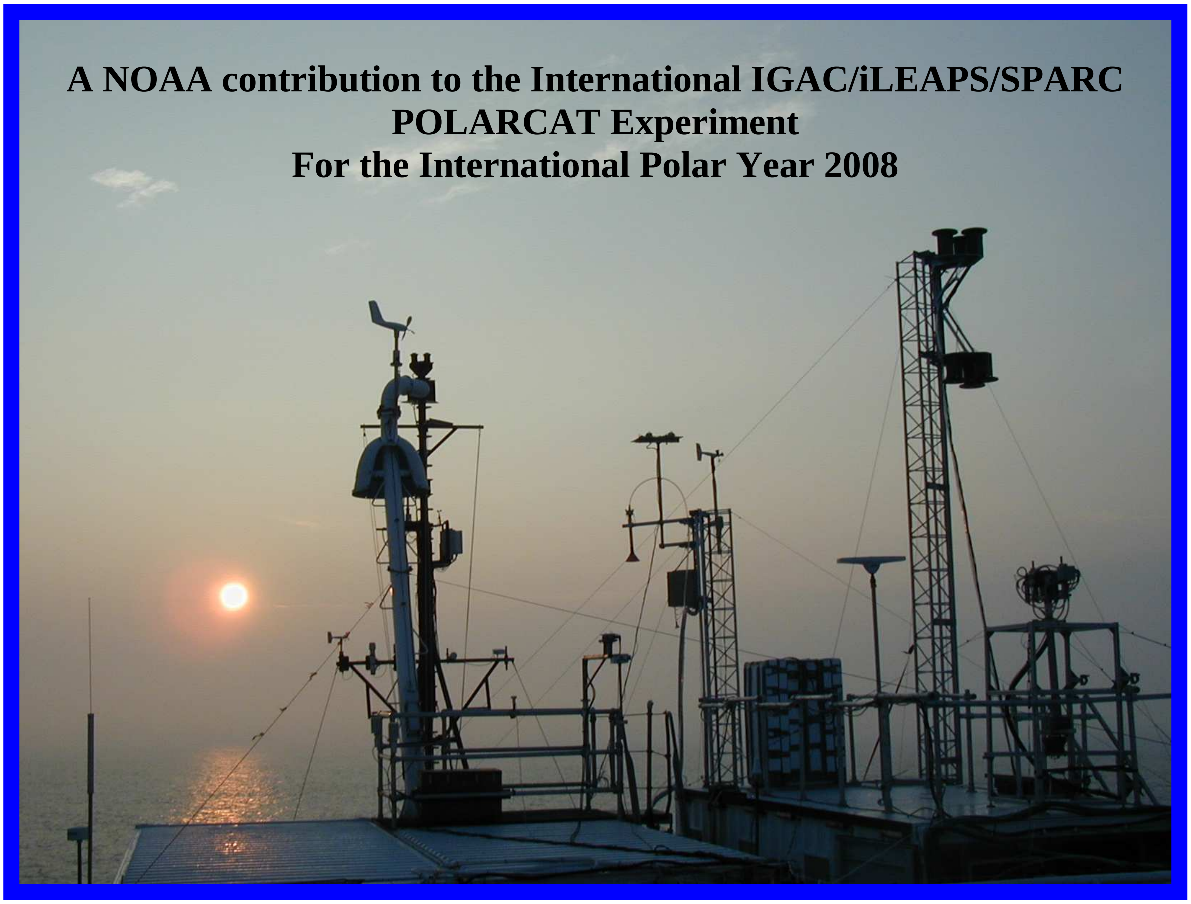
ICEALOT
International Chemistry Experiment in the Arctic LOwer Troposphere

A Springtime Study of Aerosol Properties and Atmospheric Chemistry over an Ice-Free Region of the Arctic

Scientific Objectives
September 2007



A NOAA contribution to the International IGAC/iLEAPS/SPARC
POLARCAT Experiment
For the International Polar Year 2008

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Overview

As part of POLARCAT, NOAA will undertake a research cruise in an ice-free region of the Arctic during March and April of 2008. The study area will include the Greenland, Norwegian, and Barents Seas. Scientific issues to be addressed include springtime sources and transport of pollutants to the Arctic, evolution of aerosols and gases into and within the Arctic, and climate impacts of haze and ozone in the Arctic.

Platform:

WHOI R/V *Knorr*
http://www.whoi.edu/page.do?pid=8157

Schedule:

Leg 1 – Woods Hole to Tromso, March 17 – April 11, 2008
Leg 2 – Tromso to Reykjavik, April 15 – 28, 2008

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Background

In the late 1950s, pilots flying over the Canadian and Alaskan Artic observed a haze of unknown origin that significantly decreased visibility. This “Arctic Haze” is a phenomenon that recurs every winter and spring and is now understood to be due to long range transport of anthropogenic aerosols primarily from Europe and Western Asia. The haze is composed of a varying mixture of sulfate, nitrate, particulate organic matter (POM), dust, and black carbon. Long-term measurements at ground sites within the Arctic (Barrow and Alert), reveal a decreasing trend in concentrations of aerosol black carbon during March and April throughout the 1990s. Since the beginning of the 21st century, however, concentrations have increased not only during the Arctic Haze months but also during the summer. Aerosol light scattering follows a similar trend with levels decreasing through the 1990s and increasing since 2000. In addition, concentrations of nitrate have increased at Alert from the early 1980s to present. In contrast, levels of sulfate have decreased from the 1990s to present. The lack of long term measurements of POM in the Arctic makes it difficult to assess trends in POM. Reasons for the changing trends, especially the decoupling of sulfate from nitrate and black carbon, are uncertain as are the impacts on the climate of the region.

Just as anthropogenic aerosol is transported to the Arctic during the spring, so are gas phase compounds that impact the oxidative capacity of the atmosphere and Arctic climate. The peak in average surface level Arctic ozone concentrations occurs coincidentally with the Arctic haze during springtime due to the presence of reactive nitrogen and other ozone precursors. There are uncertainties surrounding the partitioning of reactive nitrogen as it is transported into the Arctic and the mechanism for the conversion and cycling between NO_x (=NO + NO_2) and NO_y (= the sum of all reactive nitrogen). The uncertainty in reactive nitrogen chemistry leads to uncertainty in the rate of photochemical ozone production in relation to processes such as long range transport and stratosphere-troposphere exchange during the arctic spring ozone maximum. For example, photochemical HO_x production, a key component of ozone photochemistry, has a potentially large but still uncertain contribution from long wavelength photolysis of HO_x and NO_x reservoir compounds at the high solar zenith angles that occur during the spring in the Arctic. Furthermore, the production and photochemical cycling of halogen species has a profound effect on the local O_3 in the lower arctic troposphere, leading to intense ozone destruction events (ODEs). There are considerable uncertainties in this chemistry, including the processes that are responsible for its initiation, the magnitude and extent of halogen radical processing in this environment, the interplay between chlorine and bromine, and the broader implications of this chemistry, especially with respect to hydrocarbon processing.

Changes in surface air temperature and ice extent over the past decade suggest that anthropogenically-induced climate change is occurring in the Arctic. Arctic warming is primarily a manifestation of global warming such that reducing global-average warming will reduce Arctic warming and the rate of melting. Reductions in the atmospheric burden of CO_2 are the backbone of any meaningful effort to mitigate climate forcing. But, even if swift and deep reductions were made, given the long lifetime of CO_2, the reductions may not be achieved in time to delay a rapid melting of the Arctic. Hence, the goal of constraining the length of the melt season and, in particular, delaying the onset of spring melt, may best be achieved by targeting shorter-lived climate forcing agents which also impact Arctic climate. Addressing these species has the advantage that emission reductions will be felt immediately. A better understanding of the climatic effects of the short lived pollutants is required to guide mitigation strategies and, in particular, to determine to what extent reducing concentrations of aerosols and tropospheric ozone in the source regions will reduce the rate of warming in the Arctic.

NOAA will undertake a research cruise in the eastern Arctic in March and April of 2008 to address scientific questions related to the sources, transport, and climatic impacts of anthropogenic aerosol and gas phase species. This experiment, which will be part of POLARCAT (an IPY activity), will take place in the Greenland, Norwegian, and Barents Seas (Figure 1). One unique aspect of the project is the focus on the ice free region of the Arctic at a time when the fraction of Arctic ice coverage is decreasing. In addition, measurements made of aerosol and gas phase species associated with ship emissions will serve as a “baseline” before the possibility of an increase in ship traffic as a result of the decrease in ice coverage is realized along the Northern Sea Route and Northwest Passage. Specific scientific questions to be addressed are listed below. Instrumentation that will be onboard is listed in Table 1.

Scientific Questions

Q1. Springtime sources and transport of pollutants to the Arctic

Measurements of aerosol properties coupled with chemical transport models are required to understand the apparently changing trends in certain components of Arctic Haze. Measurements of aerosol composition will be made and these data will be used in conjunction with chemical transport models to determine:

- What is the composition of the aerosol during March and April over the ice-free regions of the Arctic?
- What are the sources of the aerosol to this region during March and April?
    - How significant is local production of aerosols (e.g., oceanic emissions of particles and trace gases, emissions from ships, local point sources in the Arctic)? What are the dominant oxidation pathways in the production of aerosols from these sources?
    - How significant is the North Atlantic as a marine boundary layer transport pathway for mid-latitude pollutants into this region of the Arctic?
    - How significant is the exchange of aerosols between the MBL and free troposphere?

Q2. Evolution of aerosols and gases into and within the Arctic

The impact of aerosols on climate is determined by the size and composition of the particles which, in turn, is affected by processing during transport and the spring progression. Measurements will be made to determine:

- How do aerosol precursor gases and the chemical, physical, optical and cloud nucleating properties of the aerosol evolve along the North Atlantic transport route?
- How do aerosol precursor gases and the chemical, physical, optical and cloud nucleating properties of the aerosol evolve as the spring progresses?

Previous aircraft and surface measurements in the Arctic have provided evidence for reactive nitrogen transport into the Arctic during spring. Although most of the measured NO_y is in the form of PAN, modeling studies suggest that N_2O_5 hydrolysis is responsible for much of the conversion of NO_x to NO_y during transport. Measurements will be made to determine:

- What is the partitioning of reactive nitrogen in the springtime Arctic?
- What is the rate of N_2O_5 hydrolysis in the Arctic, and how does it impact NO_y?
- What are the lifetimes of and loss processes for NO_3 in the Arctic?

The lower tropospheric ozone maximum that occurs in the Arctic spring has contributions from long range transport, stratosphere-troposphere exchange, and *in-situ* production. Modeling of aircraft data from 2000 showed an unexpectedly large contribution from the latter. Additional uncertainties surrounding the Arctic springtime ozone budget include the importance of HO_x in the photochemical production of ozone and ozone depletion events linked to halogen activation in the Arctic spring. Measurements will be made to determine:

- What are the *in-situ* ozone production rates during the spring in the Arctic?
- What is the role of HO_x chemistry at high solar zenith angles?
- Are ozone depletion events due to halogen activation significant in the ice-free regions of the Arctic?
- What mechanism activates halogens to initiate arctic ozone depletion events?
- What is the role of reactive nitrogen uptake by sea salt?

Q3. Climate Impacts of aerosols and ozone in the Arctic

The contribution of aerosols to anthropogenically-induced climate change in the Arctic is uncertain yet may be significant through direct interaction with solar and longwave radiation, aerosol – cloud interactions, and feedback processes. Measurements will be made to determine:

- What is the impact of anthropogenic aerosol on the clear-sky radiation balance of the ice-free regions of the Arctic during March and April?
- How do anthropogenic aerosols affect the radiative properties of clouds in this region?
    - What are the cloud nucleating properties of the aerosol?
    - What is the impact of anthropogenic aerosol on cloud drop effective radius and reflectivity?
    - What is the impact of anthropogenic aerosol on longwave downwelling radiation, atmospheric heating rates, and surface warming?

During the winter and early spring, tropospheric ozone is sufficiently long-lived to be transported from lower latitude source regions to the Arctic. Since ozone absorbs both infrared and shortwave radiation, it can induce large warming over highly reflective surfaces which may, in turn, contribute to snow/ice melting.

- Given the observed surface concentrations and vertical profiles of tropospheric ozone, what is the radiative impact in the springtime western Arctic?
- How does radiative forcing by tropospheric ozone vary as a function of ozone production and depletion in the ice-free western Arctic?

Table 1. Parameters to be measured and required instrumentation for the March and April 2008 IPY research cruise.

Parameter	**Method**
Size-resolved aerosol composition and gravimetric mass	Impactors (IC, XRF, and thermal-optical OC/EC)
Water soluble organic carbon	PILS-WSOC
Ionic Aerosol Composition	Particle In Liquid Sampler (PILS)-IC
Aerosol Size and Composition	Aerosol Mass Spectrometer
Organic function groups	FTIR
Single particle size and composition	ATOFS
Aerosol number	CNC
Aerosol size distribution	Twin DMAs and an APS
Cloud condensation nuclei concentration	CCN Counter
Aerosol scattering (400, 550, 700 nm)	TSI Model 3563 Nephelometer
Aerosol absorption (400, 550, 700 nm)	Radiance Research PSAP
Aerosol absorption	Photoacoustic
Aerosol light scattering hygroscopic growth f(RH)	Twin TSI 3563 nephelometers
Aerosol light extinction hygroscopic growth f(RH)	Cavity ring-down spectrometer
Total and sub-micron aerosol extinction	Cavity ring-down spectrometer
Vertical profiles of aerosol backscatter	Micropulse lidar
Aerosol optical depth	MicroTOPS
Temperature/relative humidity profiles	Radiosondes
Cloud Liquid Water Path	Microwave radiometer
Cloud droplet effective radius	Cloud radar
Radiative fluxes (near UV to near IR)	Spectral radiometers
Surface energy balance (fluxes)	Eddy covariance (bow mounted)
High resolution BL turbulence structure	Doppler mini-Sodar
VOC Speciation	GC/MS
Persistent Organic Pollutants (POPS)	GCMS
Radon (Rn)	Radon gas decay
Seawater DMS	GC chemiluminescence
Ozone (O3)	UV absorbance
Ozone	NO chemiluminescence
Carbon monoxide (CO)	Nondispersive IR
Sulfur dioxide (SO2)	Pulsed UV fluorescence
Nitric oxide (NO)	Chemiluminescence
Nitrogen dioxide (NO2)	Photolysis/chemiluminescence
Total reactive nitrogen oxides (NOy)	Au tube/chemiluminescence
Peroxyacyl nitric anhydrides (PANs)	PAN-CIMS
Alkyl nitrates (RONO2)	GC/MS
Acyl peroxy nitrates, ClNO2	CIMS
Nitrate radical (NO3); Dinitrogen pentoxide (N2O5)	Cavity ring-down spectroscopy
BrO	MAX DOAS
Water vapor (H2O)	Capacitance probe
Photolysis rates (j-values)	Spectral radiometer
Carbon dioxide (CO2)	Nondispersive IR

Figure 1. Working area for the March and April 2008 IPY research cruise.