

The chemical composition of the polar atmosphere—the IPY contribution

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The pristine polar atmosphere?

The polar atmosphere is remote from pollution sources of chemical trace species. Its composition has traditionally been seen as a clean background for more polluted air over the continents and their adjacent marine areas. This view of the polar regions as both clean and simple has gradually changed over the last decades, starting with polar route aircraft pilots observing haze and reduced visibility. Meanwhile, polar stratospheric ozone depletion, particularly strong in the Antarctic, was discovered more than two decades ago.

In the mid-1980s also, tropospheric ozone-depletion events of a surprising magnitude were discovered within the Arctic boundary layer. Later, these events were found to coincide with the depletion of gaseous elementary mercury. Equally unexpected has been the finding that the snowpack is a source of several photochemically important chemicals, e.g. bromine, oxides of nitrogen, nitrous acid and formaldehyde, and that, in some places, they dominate the chemistry of the lowest parts of the atmosphere.

Both tropospheric ozone depletion events and snow photochemical emissions are seen around both poles, and are indicative of unexpected chemistry that is not directly related to pollution, but whose effects might be strongly altered by the changing climate. The climate is indeed changing in the Arctic and parts of the Antarctic with a variability and trend that are significantly larger than the average for the globe. The area of sea ice in the Arctic has been in decline for more than two decades. The chemistry within sunlit snowpacks is surface mediated and a changing polar surface is likely to change that chemistry.

International Polar Year provides the opportunity to organize, in an Earth system perspective, an international study of the chemical composition of the polar atmosphere in both hemispheres. Important are the exchange mechanisms and drivers of exchange of chemical trace species between the atmosphere, land surfaces, the oceans and snow and ice surfaces, which depend on the physical, chemical and biological state of the Earth system components. Attention is required to the coupled local, regional and global spatial scales and the coupling mechanisms

between climate, ozone depletion in the stratosphere and the boundary layer and the environmental changes linked to the long-range transport of pollutants (in particular, aerosols, including persistent organic pollutants). We need to understand the system well enough that the effects of future changes in climate and in the relative proportions of snow, land and ocean surfaces on the atmosphere, with associated feedbacks, can be quantified.

How remote is the polar troposphere?

The Arctic and Antarctic present rather different geographical and dynamical situations. The Arctic region consists of an ocean, often covered in sea ice, and surrounded by land masses at relatively high latitude. During winter, the Arctic can have rather high concentrations of long-range transported anthropogenic pollutants, since many of the removal mechanisms are dormant. The Antarctic on the other hand is a polar continent surrounded by ocean (again covered in sea ice), but with no inhabited land masses until South America at 55°S and other continents much further north. The large distance to major pollution sources, the outflowing katabatic winds at low level and the isolating nature of the atmospheric circulation all combine to keep the Antarctic

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relatively remote and clean. Ice-core measurements show, for example, that, in contrast to the Arctic, there is not yet any significant increase in the concentration in Antarctic snow (and therefore air) of sulphate or nitrate, although increased levels of metals including lead have been observed and, of course, Antarctic ice cores have been a rich source of information about the global increase in trace gases such as carbon dioxide, methane and nitric oxide (MacFarling Meure *et al.*, 2006).

Surface temperatures in the Arctic can become very low, in particular over snow and ice surfaces during winter, leading to stable stratification with frequent and persistent surface inversions that reduce the exchange of air between the surface and the troposphere above. Under such conditions, the removal at the ground of atmospheric pollutants is inhibited, extending the lifetimes of species that are removed via dry deposition. This applies, for example, to atmospheric aerosols, ozone, sulphur dioxide, nitrogen dioxide and nitric acid.

The weak exchange with air above also partly explains why processes on the ground, such as ozone depletion events and photochemical production in snow, have such a strong effect on the boundary layer chemistry. Slow vertical mixing tends to enhance the rate of ozone depletion in the near surface layer, since it is a function of $[BrO]^2$; the latter decreases rapidly with increased turbulence. Slow mixing also slows mixing of ozone rich air from aloft down to the depleted surface layer.

Precipitation amounts in the Arctic are very low (annual precipitation on Svalbard is 150-300 mm). This means that wet scavenging is not efficient and the lifetime for soluble species like aerosols is longer than on the continents to the south. Gas-phase chemical removal of trace compounds all but stops in the Arctic atmosphere during the polar night. In the absence of sunshine

the production rate of the hydroxyl radical, which is the main gas-phase scavenger, is low. Also, in the sunlit part of the year, the chemical lifetime of trace compounds is relatively long in the polar atmosphere (except in the near-surface layer that is impacted by snowpack photochemical processes), due to the strong attenuation of short wave visible sunlight as the solar elevation is low and the low specific humidity. All these factors indicate that most gases and aerosols have a longer lifetime in the polar atmosphere than at lower latitudes. There are, however, exceptions for species that specifically react with halogen atoms/radicals, such as gaseous elementary mercury.

The removal by chemistry, precipitation or dry deposition competes with the efficiency of the exchange of air between the polar atmosphere and air at more southerly latitudes. In the absence of diabatic heating or the release of latent heat, transport processes are adiabatic and follow isentropic surfaces. Such surfaces form closed domes over the Arctic with minimum values in the Arctic boundary layer, as explained by Iversen (1984) and Stohl (2006). Most pollution source regions are too warm to allow isentropic pollution transport along the surface into the Arctic.

Northern Eurasia during winter is the most likely source region for Arctic pollution (e.g. sulphur dioxide and sulphate aerosol) and, in this case also, diabatic cooling can take place during transport over snow-covered surfaces. Transport from northern Eurasia into the Arctic takes place in blocking situations where the prevailing flow direction from the west towards the east is replaced by meridional transport with a strong north-south component. Such transport is highly episodic.

North American pollution is often heated diabatically through frontal advection ("warm conveyor belts") and brought into the Arctic free troposphere well above the

boundary layer. South-East Asian sources are found at even higher potential temperature and episodic pollution events, involving man-made emissions or desert dust, can be found as elevated polar atmosphere haze layers. A climatological study (Stohl, 2006) shows that air masses north of 80°N and near the surface reside there on the average about one week in winter and two weeks in summer, corresponding to a higher probability of meridional transport in winter/spring than in the summer. The average residence time decreases rapidly with height to about three days on average in the upper troposphere. In the most isolated parts of the Arctic, air is exposed to continuous darkness for an average 10-14 days in December.

Long range transport of air pollution into the Arctic

The discovery and start of intensive scientific investigation of polar atmospheric pollution coincided with the decade (the 1980s) of maximum anthropogenic emissions of sulphur dioxide in Europe, North America and the former USSR. The field studies of the Arctic atmospheric composition a quarter of a century ago reflected this. A major surface- and aircraft-based investigation of the pollution load in the Norwegian Arctic atmosphere was carried out in the first half of the 1980s (Ottar *et al.*, 1986). It followed the spirit of the report of the Organization for Economic Cooperation and Development (1977), which documented the contribution of continental European emissions to acid deposition in Scandinavia. In the Arctic study, it was found that "The wintertime Arctic haze, with concentration levels of man-made pollutants which are comparable to average concentrations over the industrialized continents, is due to pollutants emitted from sources within the Arctic air mass."

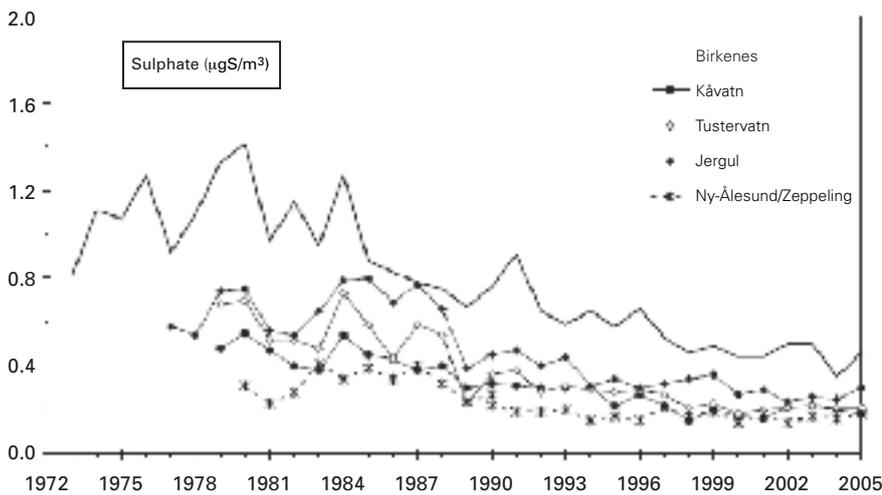


Figure 1 — Annual mean non-sea-salt sulphate aerosol concentrations for the last 2-3 decades for rural measuring sites in a north-south transect of Norway and Spitsbergen, starting at Birkenes (58°N), Kårvatn (62°N), Tustervatn (65°N), Jergul (69°N) and Ny Ålesund, Svalbard (78°N) (Aas et al., 2006).

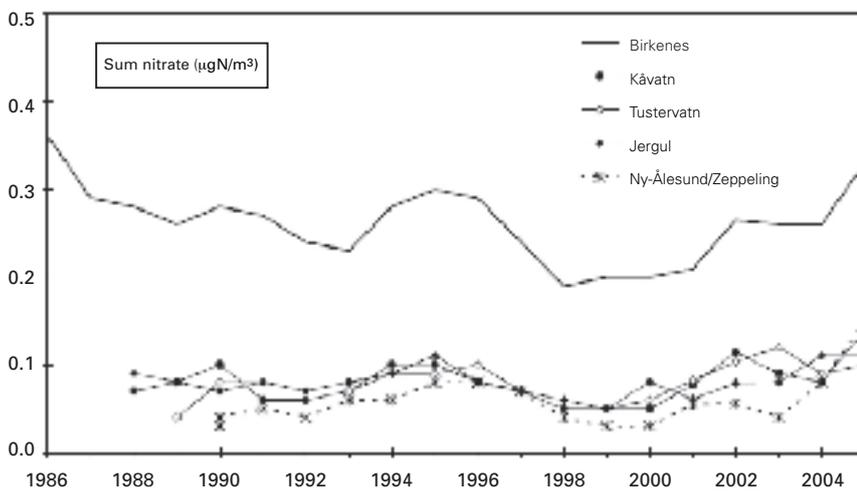


Figure 2 — Annual mean aerosol concentrations of the sum of nitrate and nitric acid for the last 2-3 decades for rural measuring sites in a north-south transect of Norway and Spitsbergen, starting at Birkenes (58°N), Kårvatn (62°N), Tustervatn (65°N), Jergul (69°N) and Ny Ålesund, Svalbard (78°N) (Aas et al., 2006).

In Figure 1 (Aas et al., 2006) is shown the annual mean non-sea-salt sulphate aerosol concentrations for the last 2-3 decades for rural measuring sites in a north-south transect across Norway and Spitsbergen, starting at Birkenes (58°N), Kårvatn (62°N), Tustervatn (65°N), Jergul (69°N) and Ny Ålesund, Svalbard (78°N). There is a significant decline in the concentrations with a factor of four drop in the annual average concentration in Ny Ålesund since the early 1980s, reflecting the decline in European and Eurasian sulphur

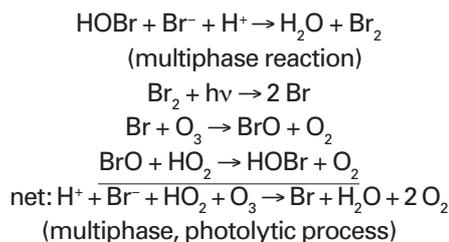
dioxide emissions (including volcanic and natural marine emissions) in Europe and the European part of the Russian Federation and ship traffic decreased from almost 50 Mt in 1990 to 21 Mt in 2004). In Figure 2, trend curves are shown for the annual average concentration of the sum of nitrate aerosol and nitric acid. The emissions of nitrogen oxides changed from 28 Mt (as nitrogen dioxide) in 1990 to 22 Mt in 2004. At Ny Ålesund, there is no obvious trend in the annual average airborne nitrate concentrations and 2005 had

the highest annual concentration on record. The concentration trends reflect the substantial decline in sulphur dioxide emissions (resulting from abatement in power plant emissions), while, for the emissions of nitrogen oxides, the benefit of technology to reduce emissions to a large extent has been compensated by a growth in car traffic in particular.

Rapid ozone and mercury depletion events

In the mid-1980s rapid ozone depletion events in the cold and stable polar boundary layer were discovered at Barrow in Alaska, at Alert in northern Canada and in Ny Ålesund on Svalbard (see Simpson et al., 2007 for references). Ozone levels drop from typical levels of more than 30 parts per billion (ppb) to very low levels and even below detection limits in a matter of hours in events during the spring. Such depletion events have also been observed at coastal sites in the Antarctic. Halogens have been found to be involved in the depletion process with a bromine radical-catalyzed cycle involving bromine (Br) and bromine monoxide (BrO) as the most important one for the depletion of ozone. Br+BrO-concentration levels of the order of 40 parts per trillion (ppt) ($1 \text{ ppt} = 1 \times 10^{-12}$) can be sufficient to reduce ozone from 30 ppb or more to virtually zero over a time period of the order of a few hours ($1 \text{ ppb} = 1 \times 10^{-9}$) in the catalytic destruction process, although the rapidity of many events can also be ascribed to transport of ozone-depleted air masses from over the sea-ice zone to the measurement site. A time series of air temperature, total gaseous mercury and ozone concentrations at Alert, Canada, in 1995, is shown in Figure 3.

It is believed that the main source for reactive bromine (Br and BrO) is bromide from sea salt that is released via photochemical reactions known as the bromine explosion reaction sequence:



This chemistry ties to both chlorine chemistry and snowpack emissions of formaldehyde and nitrous acid, as these are all important sources of HO_x which is needed to propagate the bromine explosion, via hypobromous acid (HOBr) production. Models have been developed to explain the role of bromine and iodine chemistry in the mercury-depletion events. Gaseous elementary mercury is converted to a water soluble form of mercury with a much shorter lifetime than that of mercury, increasing the deposition of mercury to polar ecosystems very significantly. Elemental mercury is re-emitted from the surface after reduction of the deposited products, and this may limit the magnitude of the impact.

Sunlit snow and ice play an important role in processing atmospheric species. Photochemical production of chemicals occur in snow/ice and the photochemically generated species can subsequently be released to the atmosphere. A review is given

by Grannas *et al.* (2007). Carbonyl compound and nitrogen oxide fluxes have been measured in a number of snow-covered environments and, in some cases, the emissions significantly impact the overlying boundary layer. For example, photochemical ozone production of 3–4 ppbv/day has been observed at South Pole, due to high hydroxyl and nitric oxide (even up to 1 parts per billion by volume (ppbv)) levels present in a relatively shallow boundary layer. Field and laboratory experiments have determined that the origin of the observed oxides of nitrogen flux is the photochemistry of nitrate within the snowpack. Low molecular weight organic compounds are also emitted from sunlit snowpacks, the source of which can be photo-oxidation of natural organic materials present in the snow. The fundamental chemistry occurring in snow packs remains poorly understood, especially for organic compounds. The role of biological processes, that impact the iodine chemistry, for example, is also ill-defined at this time.

Polar atmospheric chemical change in the long term

IPY is an intensive study period that will help improve the understanding of the Earth system processes that

control the exchanges of trace chemical species inside the polar atmosphere and their interaction with the global Earth system and the human community. The chemical composition of the polar atmosphere also strongly reflects the variability and long term changes induced by climate change and the underlying changes in the biosphere, cryosphere and oceans.

The term “tipping point” is used for some of the greenhouse effects in the Arctic (News feature in *Nature*, 15 June 2006, Vol. 441, 802-805), where tipping point is taken to mean the moment at which internal dynamics start to promote a change previously driven by external forces. One such tipping point is related to the shrinking of Arctic summertime sea ice at an average rate of 8 per cent a decade over the past 30 years (Stroeve *et al.*, 2007) and with a thinning of about one metre in the period 1987-1997.

Open water reflects much less sunlight than does ice and, as the ice cover shrinks, more of the incoming solar energy is absorbed in the Arctic Ocean in the summer; there is a greater heat flux from the ocean to the atmosphere, resulting in a positive climate feedback; and a reduction in the likelihood of stable surface inversions for the future. Changes are induced in processes that are of first-order importance for the polar atmospheric physical structure, and chemical and biological composition: ice-atmosphere interaction including ice-based photosynthetic plankton; the stability of air near the ground; and the presence of sea ice as a source of halogens.

The decrease in ice area also seems to warm the atmosphere with a rise in Arctic surface temperatures in particular in the springtime. In 2006, Svalbard experienced an unprecedented heat wave, when January was warmer than ever before and April was more than 12°C warmer than the long-term average. In April-May 2006 there was

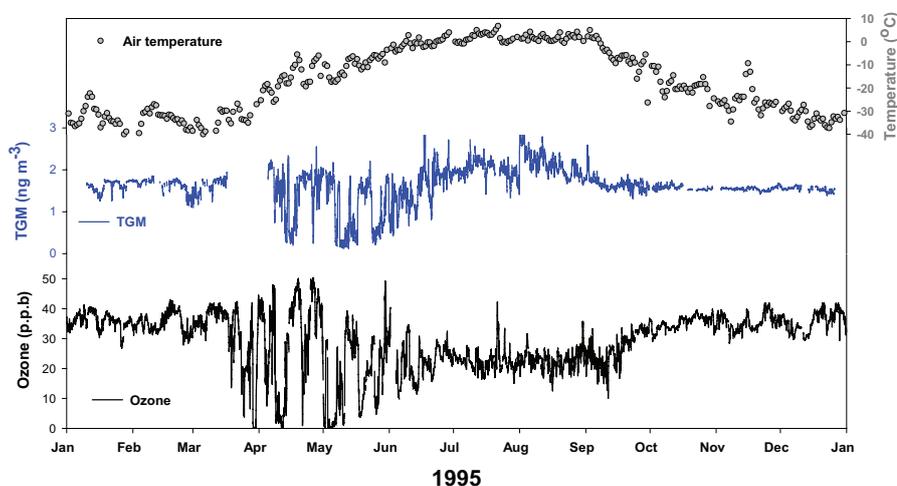


Figure 3 — Time series of air temperature, total gaseous mercury (TGM), and ozone concentrations at Alert, Canada, in 1995. Adapted by A. Steffen from Schroeder *et al.* (1998) with permission from Macmillan Publishers Ltd., *Nature*, 394, 331-332, copyright 1998.

an extraordinary pollution event on Svalbard with record high pollution levels measured on the Zeppelin mountain station of Ny Ålesund on the west coast of the Archipelago (78°N latitude) (Stohl *et al.*, 2007). The hourly ozone concentration reached 83 ppb, up from 61 ppb as the highest concentration until then in the 1989-2006 period.

The aerosol number density showed an increase in accumulation mode particles and a complete absence of Aitken particles, while, under normal circumstances, the aerosol number density distribution would have new particle formation within the Aitken mode. The particulate matter mass reached 29 µg/m³ on a 24-h basis, more than one order of magnitude higher than just before or after the episode and with a dominating organic matter fraction (60 per cent by mass compared to 4-9 per cent in the preceding weeks), reflecting the dominating contribution to the pollution event by smoke from agricultural burning in eastern Europe.

Two other tipping points are closely related to the Arctic: the possible loss of the Greenland ice sheet and its associated change in sea-level, and changes in thermohaline circulation, which could be related partly to changes in freshwater in the Arctic. Both these points would have global effects, including atmospheric chemical feedbacks in the Arctic.

More frequent high-latitude forest fires in the boreal forest zone with large carbon stocks may impact the polar atmospheric composition and climate in the long term. Fire frequency and intensity are strongly sensitive to climate change and variability and to land-use practices. Over the last century, trends in burned area have been largely driven by land-use practices, through fire suppression policies in mid-latitude temperate regions and increased use of fire to clear forest in tropical regions (for references, see IPCC Fourth

Assessment Report, 2007, page 527 Denman *et al.*, 2007). The Fourth Assessment Report also discusses evidence that climate change has contributed to an increase in fire frequency in Canada. The decrease in fire frequency in regions like the USA and Europe has contributed to the land carbon sink there, while increased fire frequency in regions like Amazonia, South-East Asia and Canada has contributed to the carbon source. At high latitudes, the role of fire appears to have increased in recent decades: fire disturbance in boreal forests was higher in the 1980s than in any previous decade on record. In the future, the carbon dioxide source from fire can increase (Fourth Assessment Report, IPCC, 2007, Chapter 7). High-latitude biomass burning is of particular significance for the polar atmosphere as the smoke has a high black carbon content. A thin layer of light absorbing dark aerosols can heat the Earth-atmosphere system, in particular above surfaces of high solar albedo such as ice and snow. In winter, haze layers are important for the radiation budget through their direct and indirect control of the infrared part of the spectrum. Black carbon deposition to ice and snow surfaces has the same effect. Forest-fire emissions thus not only can change the polar atmospheric composition in episodes, but can also trigger more long-lasting changes in the radiation balance and stratification of the lower polar atmosphere, as well as impact the surface albedo. In addition to these major "tipping points", there are also chronic changes in the environment that can have feedbacks to the atmosphere. Increasing carbon dioxide with associated increase in ocean bicarbonate and lower pH has significant impacts on marine biota. As an example, changes in carbon dioxide have been shown to significantly impact coccolithophore development (Riebesell *et al.*, 2000). How such impacts in turn affect biogenic emissions, e.g. of dimethyl sulphide, is as yet quite unclear. But, as sea-ice extent continues to decline, biogenic emissions in the Arctic are

likely to become more important to atmospheric composition and chemistry.

Summarizing, the polar atmospheric chemistry responds significantly to major climate-change-controlling processes: Arctic sea-ice extent; the stability of the Greenland ice cap; the freshwater run off to the Arctic ocean and other changes in ocean properties; and the prevalence of biomass burning in particular at high latitudes.

Polar atmospheric chemistry studies during IPY

IPY provides the opportunity to organize an international study of the chemical composition of the polar atmosphere in both hemispheres, in an Earth system perspective. The project "Polar study using aircraft, remote sensing, surface measurements and models, of climate, chemistry, aerosols and transport (POLARCAT)" will carry out a series of aircraft experiments at different times of the year to follow pollution plumes of different origin as they are transported into the Arctic and to 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 take advantage of the long residence times of pollutants in the stably stratified Arctic atmosphere to study aging 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. See <http://www.polarcat.no/polarcat> for more details.

The Ocean-Atmosphere-Sea Ice-Snow-pack (OASIS) mission, see <http://www.oasishome.net/Docs/Science%20Plan%20Version%202.2.pdf> aims to determine the importance of chemical, physical

and biological exchange processes in the ocean-atmosphere-sea ice-snowpack system on tropospheric chemistry, the cryosphere, and the marine environment and their feedback mechanisms in the context of a changing climate. The overarching questions are:

- What is the nature of feedback loops between atmosphere-sea ice-snowpack exchange processes and global climate change?
- What are the fundamental physical, chemical and biologically-mediated mechanisms of atmosphere-sea ice-snowpack exchange processes involving halogens, dimethyl sulphide, oxides of nitrogen, ozone, volatile organic compounds, persistent organic pollutants, mercury, sulphur constituents, particulate matter and carbon dioxide in the polar regions?
- What are the impacts of atmosphere-sea ice-snowpack on exchange processes on the marine cryosphere (ice/snow) and the underlying polar ocean?
- What is the relationship of - atmosphere-sea ice-snowpack exchange processes with the chemistry, physics and biology of airborne gases, airborne particles and cloud/snow formation?
- Environmental pollution: what is the impact on, and by, atmosphere-sea ice-snowpack exchange and the role of long term changes?

Figure 4 illustrates how OASIS is directed to the study of the processes that modify biogeochemical fluxes between the atmosphere, the ice and ocean surfaces.

Air-Ice Chemical Interactions (AICI) (<http://classic.ipy.org/development/eoi/proposal-details.php?id=20>) is set up to determine how the snowpack over the polar regions controls the

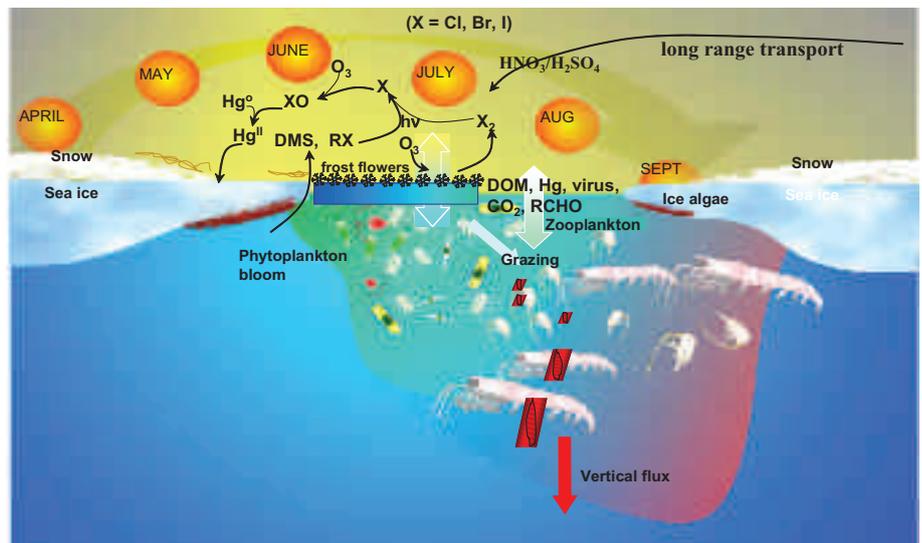


Figure 4 — Illustration of how OASIS is directed to the study of the processes that modify biogeochemical fluxes between the atmosphere, the ice and ocean surfaces (<http://www.oasishome.net/Docs/Science%20Plan%20version%202.2.pdf>).

chemistry of the lower atmosphere: Halogen chemistry over the sea ice zone depletes boundary layer ozone, and causes mercury deposition. Snow photochemical production alters the nitrogen oxide chemistry and, in some cases, the oxidation state of large parts of the polar boundary layer. Persistent organic compounds undergo a distillation which leads to their deposition in polar regions. Biochemical processes in open leads play a major role in the formation of cloud condensation and ice-forming nuclei. Through cloud formation, this process may play a vital role in ice-albedo climate feedbacks. AICI will contribute to the assessment of how these processes may change with a warming climate and shrinking cryosphere. In this context, IPY offers the opportunity to determine the spatio-temporal pattern of chemistry and processes from the ice surface through the boundary layer, including cloud formation, by linking various field activities in both polar regions carried out in the same year. AICI-IPY will provide an overall framework, arrange supporting laboratory and modelling studies and integration of remote-sensing data and organize synthesis meetings. AICI-IPY will also coordinate the publication of major reviews of polar air-surface exchange and chemistry processes, and

demonstrate how IPY has improved our understanding of connections between a changing climate, a changing surface and atmosphere.

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