Science

∥AAAS

Arctic Air Pollution: Origins and Impacts
Kathy S. Law, *et al.*
Science **315**, 1537 (2007);
DOI: 10.1126/science.1137695

REVIEW

Arctic Air Pollution: Origins and Impacts

Kathy S. Law[1] and Andreas Stohl[2]

Notable warming trends have been observed in the Arctic. Although increased human-induced emissions of long-lived greenhouse gases are certainly the main driving factor, air pollutants, such as aerosols and ozone, are also important. Air pollutants are transported to the Arctic, primarily from Eurasia, leading to high concentrations in winter and spring (Arctic haze). Local ship emissions and summertime boreal forest fires may also be important pollution sources. Aerosols and ozone could be perturbing the radiative budget of the Arctic through processes specific to the region: Absorption of solar radiation by aerosols is enhanced by highly reflective snow and ice surfaces; deposition of light-absorbing aerosols on snow or ice can decrease surface albedo; and tropospheric ozone forcing may also be contributing to warming in this region. Future increases in pollutant emissions locally or in mid-latitudes could further accelerate global warming in the Arctic.

Even though early Arctic explorers had noticed atmospheric haze and dirty deposits on the snow (1), the remote Arctic atmosphere was long believed to be extremely clean. However, pilots flying over the North American Arctic in the 1950s observed widespread haze (2) that could be seen every winter and early spring. It took until the 1970s for scientists to realize that the haze was air pollution transported from the middle latitudes (3). Arctic haze continues to be an air quality problem, and the acidic compounds (mainly sulfate) associated with it can be washed out with precipitation or deposited at the surface, leading to increased acidity in natural ecosystems (4). Long-range transport of pollution to the Arctic also carries toxic substances, such as mercury or persistent organic pollutants, that can have adverse effects on ecosystems and human health.

Over the past 20 years there has been much research on the climatic consequences of this pollution, which is also present in summer, albeit at lower concentrations. Climate change is proceeding fastest at the high latitudes of the Arctic. Surface air temperatures have increased more than the global average over the past few decades and are predicted to warm by about 5°C over a large part of the Arctic by the end of the 21st century, the most rapid of any region on Earth (5). Models also predict that summer sea ice may completely disappear by 2040 (6). These changes are caused by global increases in long-lived greenhouse gases (GHGs), whose effects are enhanced in the Arctic through feedback mechanisms such as the sea-ice albedo feedback. However, air pollution also affects Arctic climate, particularly through changes in surface radiative forcing.

Arctic Haze

Arctic haze is a mixture of sulfate and particulate organic matter and, to a lesser extent, ammonium, nitrate, black carbon (BC) (7), and dust aerosols (8). It also contains relatively high levels of ozone precursors such as nitrogen oxides (NO_x) and volatile organic compounds (VOCs) (9). Aerosol haze particles are well aged, very efficient at scattering solar radiation, and also weakly absorbing. The haze has a distinct seasonal cycle with a maximum in late winter and early spring (3) when the removal processes in the dry and stable Arctic atmosphere are very slow. For example, Fig. 1 shows the seasonal cycle in BC measured at Alert [62.3°W, 82.5°N, 210 m above sea level (ASL)] (10). Near the surface, the haze starts disappearing in April, but layers at higher altitudes may persist into May. Trends in trace constituents and aerosols are complex in the Arctic region. Although sulfate, aerosol light scattering, and absorption exhibit significant downward trends at most Arctic stations (8) because of emission reductions in the haze's source regions, nitrate concentrations have been increasing over the past two decades (4).

Air Pollution Transport into the Arctic

Practically all pollution in the high Arctic originates from more southerly latitudes. Local pollution sources are currently small and limited to near the Arctic Circle. These include vol-

canic emissions in Alaska and Kamchatka; anthropogenic emissions from conurbations like Murmansk; industrial emissions, most notably in the northern parts of Russia; and emissions from the oil industry and shipping (4). Surfaces of constant potential temperature (11) form a dome above the cold Arctic lower troposphere, forcing air parcels traveling northward to ascend (12, 13). This isolates the Arctic lower troposphere from the rest of the atmosphere by a transport barrier, the Arctic front. On time scales of a few days to weeks, the Arctic lower troposphere is accessible only to pollution originating from very cold source regions (14, 15). The polar dome is not zonally symmetric and can extend to about 40°N over Eurasia in January, thus making northern Eurasia the major source region for the Arctic haze. Air masses leaving densely populated areas on the east coasts of Asia and North America are too warm and moist to directly penetrate the polar dome, but they can ascend to the Arctic middle or upper troposphere. However, Greenland, because of its high topography, is exposed to pollution from southeast Asia and North America more strongly than is the rest of the Arctic (16).

The polar dome also makes it difficult for stratospheric air masses to reach the Arctic lower troposphere. A recent model study suggested a strong vertical gradient in the influence of stratospheric air masses (16). For a transport time scale
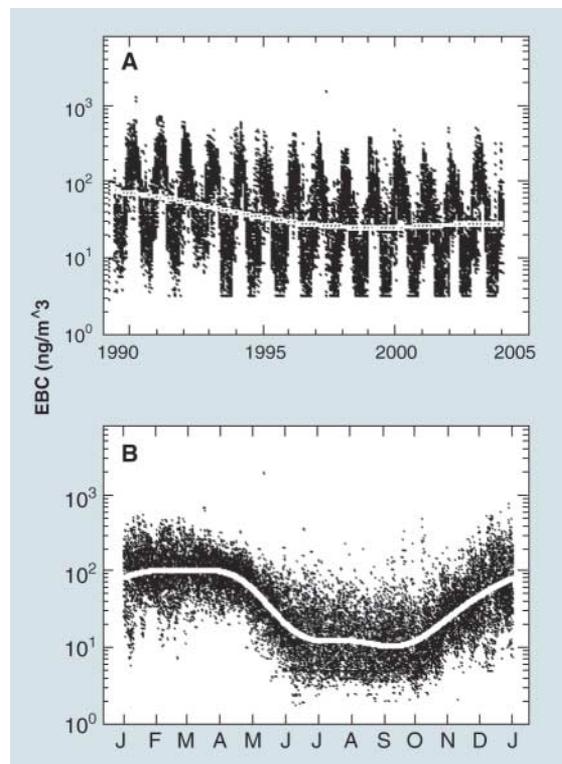

Fig. 1. Long-term trends (**A**) and seasonal variation (**B**) of 6-hourly equivalent BC concentrations at Alert. [Reproduced/modified from (10) by permission of the American Geophysical Union. Copyright 2006 American Geophysical Union.]

[1]Service d' Aéronomie, CNRS, IPSL/Université Pierre et Marie Curie, Boîte 102, 4 Place Jussieu, Paris Cedex 05, 75252 France. E-mail: kathy.law@aero.jussieu.fr. [2]Norwegian Institute for Air Research (NILU), Instituttveien 18, 2027 Kjeller, Norway. E-mail: ast@nilu.no

Polar Science

of 10 days, a 10% contribution from stratospheric air masses was found between 3 and 5 km, but near sea level these contributions had decreased to only 1% and 0.3% in winter and summer, respectively, much less than in the mid-latitudes.

The mean circulation in winter is characterized by low-level transport from northern Eurasia across the Arctic toward North America. In January, boundary layer air in the North American part of the Arctic has, on average, experienced about 2 weeks of complete darkness en route (16). In summer, the overall pathway is directed from the North Atlantic Ocean across the high Arctic toward the North Pacific Ocean, and the transport is only half as fast as in winter. Because of the slower transport and more-efficient removal processes in summer, pollution concentrations are lower and high-latitude sources of air pollution become even more important, relative to more southerly sources, than in winter.

Tropospheric Ozone

Observed ozone trends in the Arctic are complex, with Canadian sites showing increases in the 1990s, particularly in the winter and spring, whereas decreases were observed in the 1980s (17). Although changes in anthropogenic emissions at mid-latitudes are likely to have played a role, changes in natural emissions or transport patterns also could have been important. Recent climate model simulations suggest that increases in tropospheric ozone, caused by increases in anthropogenic emissions, could have contributed 0.4° to 0.5°C during winter and spring to 20th-century surface temperature trends (roughly 30% of observed trends) in the Arctic (18). This is likely due to longer ozone lifetimes and enhanced atmospheric infrared absorption in the dry Arctic winter and subsequent feedbacks on snow-ice albedo. Note that these calculations did not include boreal forest fire emissions or possible changes in the stratospheric ozone flux. The latter

has been put forward to explain ozone trends in the Canadian Arctic upper troposphere (19).

To date, the main focus has been on understanding the annual springtime increase in ozone. Ozone lidar data collected from February to May 2000 between Colorado (United States) and northern Canada and Greenland as part of the Tropospheric Ozone Production about the Spring Equinox (TOPSE) airborne campaign clearly show this transition throughout the free troposphere (Fig. 2) (20). Analysis of odd nitrogen (NO_y) and its constituents confirmed previous ground-based studies showing that, as temperatures warm, thermal decomposition of peroxy acetyl nitrate (PAN) (produced from oxidation of VOCs) is the primary driving factor, leading to enhanced photochemical ozone production from released NO_x (21, 22). This springtime breakdown of the Arctic haze's pollution reservoir and its subsequent southward transport may also contribute to spring ozone maxima observed at remote mountain sites in Northern mid-latitudes (17, 23). However, inconsistencies were found between observations and model calculations of oxygenated organic and nitrogen-containing species {e.g., nitrous acid (HONO), pernitric acid (HNO_4), and PAN [e.g., (21)]}, and also, more recently in hydroxyl radical (OH) data collected at the high-altitude Summit site in Greenland (38.4°W, 72.6°N, 3208 m ASL) (24), suggesting that our knowledge of Arctic photochemistry and processes, such as wet scavenging, is incomplete. New measurements of other important nitrogen species such as the nitrate radical (NO_3) and dinitrogen pentoxide (N_2O_5), which play a key role in nighttime chemistry, may also reveal new insights, as has recently been shown at mid-latitudes (25).

An analysis of TOPSE ozone lidar data also suggested a rather low contribution from the stratospheric flux of ozone during the winter-spring (less than 15%) (20), confirmed by more

recent transport analyses (16). Few global model estimates exist and include more southerly latitudes, where net exchange is stronger, leading to higher estimates for the fraction of ozone originating from the stratosphere [e.g., 30 to 50% in spring, 30° to 90°N (22)] than what would have been obtained for the Arctic alone. Stratospheric air masses could also be a source of NO_y in the troposphere and therefore nitrate in snow.

The springtime ozone budget is also complicated by the occurrence of dramatic ozone depletion events in the Arctic boundary layer, first observed in the 1980s (26) and evident in surface ozone records (17) (see also Fig. 2). These events are strongly correlated to deposition of gaseous mercury onto snow surfaces [e.g., (27)]. They are related to the rapid release of bromine radicals (so-called bromine explosions), seen by satellite as clouds of bromine oxide (BrO) over regions of sea ice, which then destroy ozone through a series of autocatalytic reactions. The bromine originates from seawater, but the exact mechanism by which it is transformed to reactive bromine in the atmosphere is unclear. Mechanisms involving formation of sea-salt aerosols from frost flowers on sea ice (28), or sea-salt–contaminated snow on newly formed sea ice (29), have been proposed. The wider impact of these halogens on the Arctic ozone budget (and indeed the oxidizing capacity of the troposphere) is also uncertain. Recent calculations constrained by BrO satellite measurements suggest that ozone levels in the Arctic boundary layer could be reduced by more than 50% because of halogen chemistry over the high northern latitudes in spring (30).

The summertime ozone budget has yet to be quantified in the Arctic. In particular, the impact of boreal forest fire emissions, which have already been shown at mid-latitudes to produce large amounts of ozone because of their large loading of
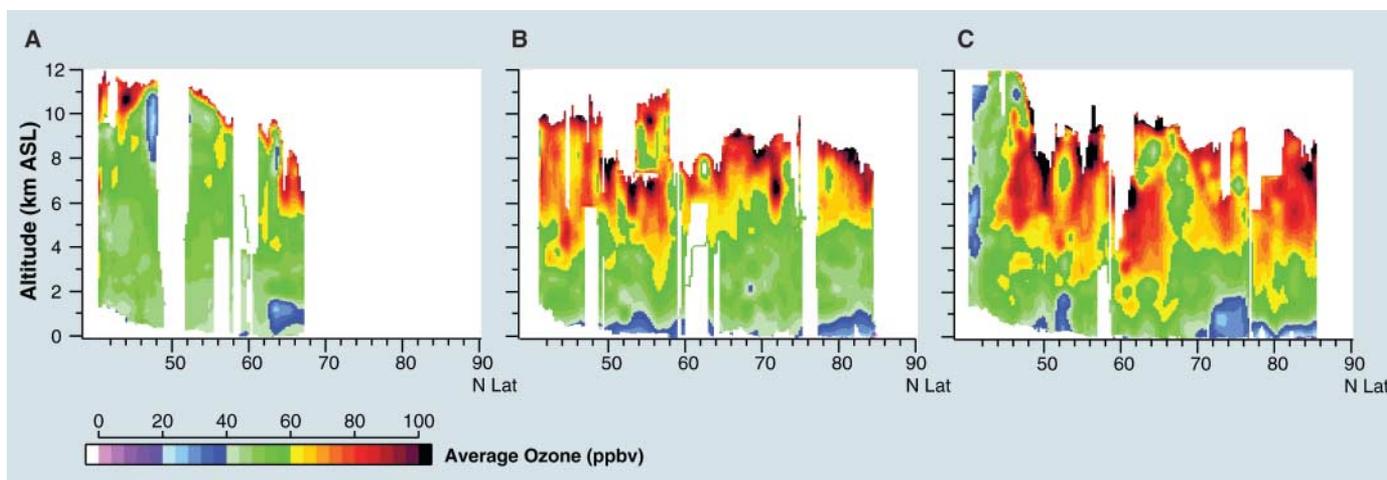

Fig. 2. Average latitudinal distribution of vertical ozone concentrations in parts per billion (ppb), measured by airborne lidar, during deployments (**A**) 4–9 February 2000, (**B**) 19–26 March 2000, and (**C**) 15–23 May 2000. Data was collected during each deployment on flights between Broomfield, Colorado (United States) (40°N, 105°W), and northern Canada and northern Greenland as part of the TOPSE experiment. [Reproduced/modified from (20) by permission of the American Geophysical Union. Copyright 2003 American Geophysical Union]

nitrogen-containing constituents, most notably PAN (*31*), is poorly known. Another process, identified relatively recently, is the production of NO_x (and HONO) from photolysis of nitrate in the snowpack in the presence of sunlight (*32*). Although very high levels of NO_x [>600 parts per trillion] have been observed at South Pole (elevation of 2840 m) because of the existence of prolonged periods with a very stable shallow boundary layer (*33*), much lower enhancements have so far been reported in the Arctic (*34*), making it unlikely that these emissions are important on regional scales at northern high latitudes.

Climatic Effects of Light-Absorbing Aerosols

Measurements at Barrow (156.6°W, 71.3°N, 11 m ASL) have shown that the single scattering albedo of haze aerosols in the Arctic can be as low as 0.9 in winter (*35*), indicating that these aerosols contain large amounts of light-absorbing material. In the Arctic, the efficiency of sunlight absorption in aerosol layers is greater than the efficiency at lower latitudes because of the high albedo of snow and ice and multiple reflection and scattering of light between the surface and the aerosol layers. BC, which is responsible for most of the aerosol light absorption, is a minor but important component of the Arctic haze (*10*) and causes heating in the haze layers (*8*). In addi-

tion, deposition of BC onto snow and ice results in a reduction of the surface albedo (*36*, *37*). It has been suggested that the climate forcing due to this albedo effect is relevant when compared with the effect of GHGs (*38*). Its efficacy, measured as the effectivity in increasing the surface air temperature per unit of forcing, is twice as large as that of carbon dioxide, and it may be even more effective in melting snow and ice.

BC concentrations are highest during the Arctic haze season and lowest in summer (*10*). As a result of emission reductions, BC concentrations have declined by 54% at Alert and 27% at Barrow from 1989 to 2003, but with some indication of a recent trend reversal (Fig. 1). In winter, BC originates mostly from anthropogenic activities, but the regional distribution of sources is debated. In a climate model study, it was argued that, after recent strong emission increases in southeast Asia and decreases elsewhere, southeast Asia is now the largest BC source for the Arctic (*39*). However, this result also has been questioned (*16*), because the large temperature difference between southeast Asia and the Arctic lower troposphere does not allow for direct transport between the two regions. Observations linked with trajectory calculations suggested Russian sources have the strongest influence on BC levels at Alert and Barrow (*10*). More BC measurements in the Arctic, especially at

higher altitudes, are required to clarify the relative importance of different BC sources.

During summer, atmospheric BC concentrations are much lower than in late winter and early spring (*10*) but still are important for the Arctic radiation budget because of the abundance of solar radiation. A recent model study suggests that, in summer, boreal forest fires are the dominant source for BC in the Arctic because many of the fires burn at high latitudes (*16*). Chemical signatures of biomass burning emissions have been preserved in Arctic snow and ice records (*40*), and biomass burning plumes have been observed in the Arctic (*41*, *42*). For example, large pan-Arctic enhancements of atmospheric BC concentrations occurred as a result of strong burning in the boreal forests of North America in summer 2004 (Fig. 3A), which also lead to a decrease in the snow albedo at Summit (Greenland) during one episode (*43*). In spring 2006, smoke from agricultural fires in eastern Europe was transported into the European Arctic and led to the highest concentrations of many pollutants ever measured at the Zeppelin station (11.9°E, 78.9°N, 478 m ASL) on Svalbard, Norway, as well as a dramatic reduction in visibility (Fig. 3, B and C) (*44*). Atmospheric BC concentrations reached record levels and also led to a visible discoloration of drifting snow on a glacier. All this points toward a strong influence of biomass burning on Arctic BC levels, snow-ice albedo, and radiation transmission in the Arctic atmosphere.

Pyro-Convection

It has been known for some time that forest fires can inject emissions into the upper troposphere, but it was discovered only recently that injections deep into the stratosphere also occur and are in fact quite common (*45–47*). The highest altitude where smoke from boreal forest fires was observed in situ is 17 km, several kilometers above the tropopause, and at potential temperatures greater than 380 K (*46*). Remote sensing observations indicate that even deeper injections into the stratospheric overworld are possible (*47*). The lifetime of aerosols (and also many trace gases) at these altitudes can be months, thus prolonging their possible radiative effects. It has been suggested that a cold bias in the high-latitude lower stratosphere that exists in many climate models could be removed by including high-altitude BC injections from boreal fires (*48*). However, nothing is known about the impact of pyro-convection on stratospheric chemistry.
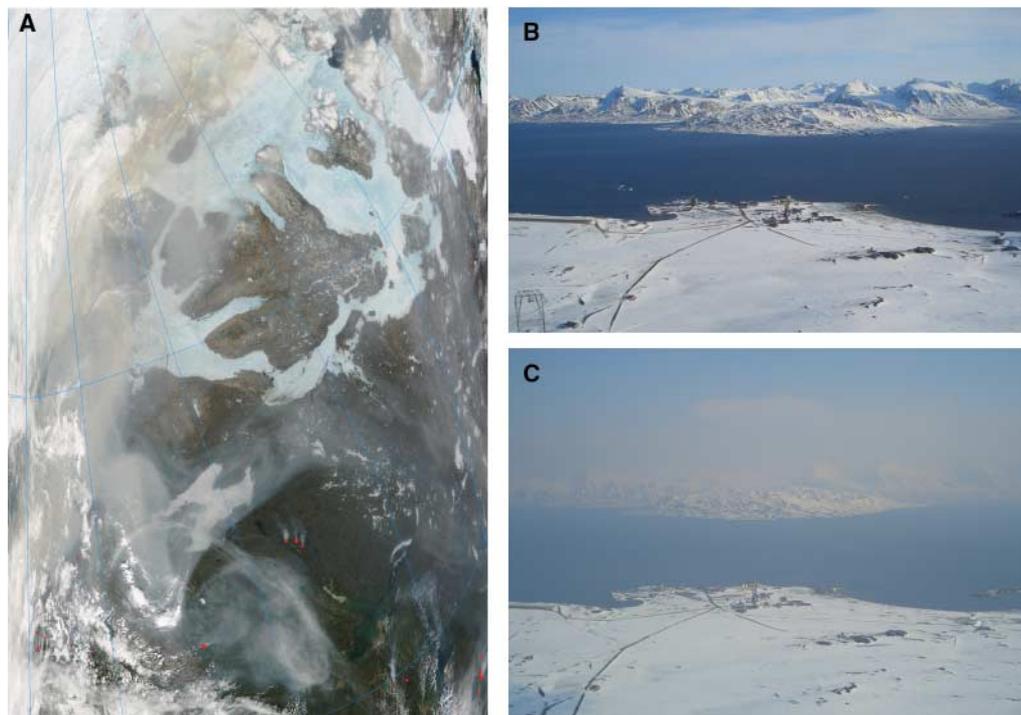

Fig. 3. (**A**) Moderate Resolution Imaging Spectroradiometer (MODIS) satellite image from 5 July 2004, showing the intrusion of thick smoke from boreal forest fires (red dots) into the Canadian Maritime Arctic. Image courtesy of MODIS Rapid Response Project at NASA Goddard Space Flight Center. View from the Zeppelin station near Ny Ålesund on Svalbard, Norway, under clear conditions (**B**) on 26 April 2006 and (**C**) on 2 May 2006, when smoke from agricultural fires burning in Eastern Europe was transported to the station (*43*). [Image courtesy of A.-C. Engvall, Stockholm University]

Indirect Aerosol Effects

Aerosols also influence irradiances in the Arctic indirectly via changes in the

microphysical properties of clouds. Enhanced particle concentrations increase the number concentration and decrease the size of cloud droplets (*49*), which increases the cloud albedo. They can also reduce rain formation and increase cloud lifetime. The Arctic is particularly susceptible to aerosol indirect effects, because the low aerosol number concentrations result in a large fraction of particles being activated during cloud formation (*8*). It has also been suggested that aerosols can increase the longwave emissivity of Arctic liquid-phase clouds (*50*). Most liquid-phase clouds contain enough water to be considered blackbodies with unit emissivity at thermal wavelengths; in which case, aerosol effects can be ignored. However, Arctic clouds are often so thin that their emissivity increases with increasing cloud droplet number concentrations. This enhances the downwelling thermal radiation fluxes, an effect opposed to the indirect effects on the solar radiation. The effect is most important in winter and early spring, when Arctic haze aerosols are abundant, thin clouds exist, and the radiation balance is tied toward thermal fluxes because of the absence or small magnitude of solar radiation. It may trigger more rapid warming of the Arctic in spring and, thus, an earlier snowmelt. However, a quantitative understanding of aerosol indirect effects, including those involving mixed-phase and ice clouds, remains elusive.

Future Changes

The disappearance of summertime sea ice could have a huge impact on trace gas and aerosol distributions in the Arctic. For example, increased areas of open ocean could lead to increases in natural dimethyl sulfide emissions and production of sulfate aerosols (*51*), whereas emissions of halogens and NO_x from the ice and snow could be reduced. There is evidence that ship traffic is already affecting the summertime Arctic atmosphere, with strong signatures seen in marine aerosols (*52*). Increased deposition of soot from increased shipping after the reduction of summertime sea ice could further accelerate sea-ice melting. Increases in surface ozone by a factor of 2 to 3, to levels currently observed at Northern Hemisphere mid-latitudes as a result of increasing ship NO_x emissions, have been predicted (*53*). Similar effects might also be expected from an increase in Arctic oil drilling.

As northern high latitudes warm because of climate change, boreal forest fires are becoming more frequent (*54*), thus increasing pollution transport into the Arctic. This may also trigger a feedback cycle, where forest fire emissions lead to earlier melting of Arctic snow and ice and thus further warming. Furthermore, the polar dome, which currently presents a barrier to pollution transport into the Arctic, may weaken in the future as the Arctic continues to warm relatively faster than the lower latitudes, thus allowing more efficient pollution import into the Arctic. This could be facilitated by, for instance, an upward trend in the North Atlantic Oscillation, which correlates with the transport of pollutants into the Arctic (*55, 56*).

Future Directions

Clearly many uncertainties still exist in our knowledge of processes governing the buildup of air pollution in the Arctic and its role in climate change. Within the framework of the International Polar Year (IPY), the scientific community is mobilizing to tackle these issues through a series of coordinated field measurement programs and analysis using climate models that also include trace gases and aerosols.

Most of the ongoing and predicted rapid changes in the Arctic climate are a direct consequence of the increasing levels of long-lived greenhouse gases and positive feedbacks specific to the Arctic (*5*). In order to combat these changes, reductions in the emissions of long-lived greenhouse gases, particularly carbon dioxide, are urgently needed. However, the Arctic may also benefit more than other regions from reductions in the emissions of short-lived climate agents. In particular, reducing BC emissions could slow atmospheric warming and the melting of snow and ice, and reducing tropospheric ozone concentrations could slow the increase in Arctic surface air temperatures. Increases in emissions of BC and ozone precursors in the Arctic itself should be strictly avoided.

References and Notes

1. A. E. Nordenskiöld, *Science* ns-**2**, 732 (1883).
2. J. M. Mitchell, *J. Atmos. Terr. Phys.* (special suppl.), 195 (1957).
3. L. A. Barrie, *Atmos. Environ.* **20**, 643 (1986).
4. "Acidifying pollutants, Arctic haze, and acidification in the Arctic," Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway (2006).
5. Intergovernmental Panel on Climate Change (IPCC), *Climate Change 2001: The Scientific Basis*, J. T. Houghton, Ed. (Cambridge Univ. Press, New York, 2001).
6. M. M. Holland, C. M. Blitz, B. Trembley, *Geophys. Res. Lett.* **33**, 10.1029/2006GL028024 (2006).
7. We use the term black carbon (BC) here, although it is poorly defined. Often, light absorption is measured and converted to "equivalent" BC.
8. P. K. Quinn *et al.*, *Tellus* **59B**, 99 (2007).
9. S. Solberg, C. Dye, N. Schmidbauer, *J. Atmos. Chem.* **25**, 33 (1996).
10. S. Sharma, E. Andrews, L. A. Barrie, J. A. Ogren, D. Lavoué, *J. Geophys. Res.* **111**, 10.1029/2005JD006581 (2006).
11. Potential temperature (Θ) is almost a conserved quantity in the atmosphere. Radiational cooling decreases Θ by about 1 K/day in the free troposphere, whereas condensation of water vapor increases Θ.
12. A. Klonecki *et al.*, *J. Geophys. Res.* **108**, 10.1029/2002JD002199 (2003).
13. The ascent of moist air parcels is even stronger because of the extra heat released by the condensation of water vapor.
14. T. N. Carlson, *Atmos. Environ.* **15**, 1473 (1981).
15. T. Iversen, *Geophys. Res. Lett.* **11**, 457 (1984).
16. A. Stohl, *J. Geophys. Res.* **111**, 10.1029/2005JD006888 (2006).
17. S. J. Oltmans *et al.*, *Atmos. Environ.* **40**, 10.1016/j.atmosenv.2006.01.029 (2006).
18. D. Shindell *et al.*, *J. Geophys. Res.* **111**, 10.1029/2005JD006348 (2006).
19. D. W. Tarasick, V. E. Fioletov, D. I. Wardle, J. B. Kerr, J. Davies, *J. Geophys. Res.* **110**, 10.1029/2004JD004643 (2005).
20. E. V. Browell *et al.*, *J. Geophys. Res.* **108**, 10.1029/2001JD001390 (2003).
21. C. Stroud *et al.*, *Atmos. Environ.* **37**, 10.1016/S1352-2310(03)00353-4 (2003).
22. L. K. Emmons *et al.*, *J. Geophys. Res.* **108**, 10.1029/2002JD002665 (2003).
23. S. A. Penkett, K. A. Brice, *Nature* **319**, 655 (1986).
24. S. J. Sjostedt *et al.*, *Atmos. Environ.* **41**, 10.1016/j.atmosenv.2006.08.058 (2007).
25. S. S. Brown *et al.*, *Science* **311**, 67 (2006).
26. L. A. Barrie, J. W. Bottenheim, J. W. Schnell, P. J. Crutzen, R. A. Rasmussen, *Nature* **334**, 138 (1988).
27. P. A. Ariya *et al.*, *Tellus* **B56**, 397 (2004).
28. A. M. Rankin, E. W. Wolff, S. Martin, *J. Geophys. Res.* **107**, 4683 (2002).
29. W. R. Simpson *et al.*, *Atmos. Chem. Phys. Discuss.* **6**, 11051 (2006).
30. T. Zeng *et al.*, *J. Geophys. Res.* **111**, 10.1029/2005JD006706 (2006).
31. E. Real *et al.*, *J. Geophys. Res.*, 10.1029/2006JD007576, in press (2007).
32. F. Dominé, F. P. B. Shepson, *Science* **297**, 1506 (2002).
33. D. Davis *et al.*, *Atmos. Environ.* **38**, 5375 (2004).
34. R. E. Honrath *et al.*, *Atmos. Environ.* **36**, 2629 (2002).
35. P. K. Quinn *et al.*, *J. Geophys. Res.* **107**, 10.1029/2001JD001248 (2002).
36. S. G. Warren, W. J. Wiscombe, *J. Atmos. Sci.* **37**, 2734 (1980).
37. A. D. Clarke, K. J. Noone, *Atmos. Environ.* **19**, 2045 (1985).
38. J. Hansen, L. Nazarenko, *Proc. Natl. Acad. Sci. U.S.A.* **101**, 423 (2004).
39. D. Koch, J. Hansen, *J. Geophys. Res.* **110**, 10.1029/2004JD005296 (2005).
40. S. Whitlow, P. Mayewski, J. Dibb, G. Holdsworth, M. Twickler, *Tellus* **46B**, 234 (1994).
41. N. C. Hsu *et al.*, *Geophys. Res. Lett.* **26**, 1165 (1999).
42. M. G. Iziomon, U. Lohmann, P. K. Quinn, *J. Geophys. Res.* **111**, 10.1029/2005JD006223 (2006).
43. A. Stohl *et al.*, *J. Geophys. Res.* **111**, 10.1029/2006JD007216 (2006).
44. A. Stohl *et al.*, *Atmos. Chem. Phys.* **7**, 511 (2007).
45. M. Fromm *et al.*, *Geophys. Res. Lett.* **27**, 1407 (2000).
46. H.-J. Jost *et al.*, *Geophys. Res. Lett.* **31**, 10.1029/2003GL019253 (2004).
47. M. Fromm *et al.*, *J. Geophys. Res.* **110**, 10.1029/2004JD005350 (2005).
48. P. J. Rasch *et al.*, *Eos* **86** (suppl.), abstr. A22B–01 (2005).
49. S. Twomey, *J. Atmos. Sci.* **34**, 1149 (1977).
50. T. J. Garrett, C. F. Zhao, *Nature* **440**, 787 (2006).
51. A. J. Gabric, B. Qu, P. Matrai, A. C. Hirst, *Tellus* **57B**, 391 (2005).
52. Z. Xie, L. Sun, J. D. Blum, Y. Huang, W. He, *J. Geophys. Res.* **111**, 10.1029/2005JD006253 (2006).
53. C. Granier *et al.*, *Geophys. Res. Lett.* **33**, 10.1029/2006GL026180 (2006).
54. B. J. Stocks *et al.*, *Clim. Change* **38**, 1 (1998).
55. S. Eckhardt *et al.*, *Atmos. Chem. Phys* **3**, 1769 (2003).
56. J. F. Burkhart, R. C. Bales, J. R. McConnell, M. A. Hutterli, A. Manuel, *J. Geophys. Res.* **111**, 10.1029/2005JD006771 (2006).