

ARCTIC AIR POLLUTION

New Insights from POLARCAT-IPY

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POLARCAT provided a wealth of data on the concentrations and sources of short-lived climate pollutants (aerosols, ozone) and their precursors in the Arctic.

In the face of rapid climate change in the Arctic (increasing temperatures, earlier onset of spring snowmelt, sea ice loss, etc.), it is important to improve our knowledge about processes driving these changes. In general, climate models are able to reproduce enhanced warming in the Arctic, the so-called Arctic amplification. However, discrepancies are apparent between observations and global climate model predictions of, for example, Arctic summer sea ice, resulting in significant differences between model-based and extrapolated estimates for the complete disappearance of the summer ice (Overland and Wang 2013). While increases in carbon dioxide (CO_2) and associated atmosphere-ice-ocean feedbacks are major contributing factors, short-lived (with respect to chemical lifetime) climate forcers, such as absorbing (heating) aerosols like black carbon (BC), and trace gases ozone (O_3) and methane, are also likely to be playing an important role (Quinn et al. 2008). Short-lived climate pollutants (SLCPs; e.g., BC, O_3) can impact Arctic warming as a result of ►

NASA P-3B over Yukon River, Alaska in spring 2008 with pollution haze layer visible in the background. (Credit J. Cozic NOAA/CIRES.)

(i) direct warming either locally in the Arctic or remotely via heat transport resulting from SLCP-induced warming at lower latitudes (e.g., Shindell 2007), (ii) decreased surface albedo and subsequent surface warming due to deposition of light-absorbing aerosols (e.g., BC) on snow/ice surfaces (e.g., Hansen and Nazarenko 2004; Flanner et al. 2007), or (iii) indirect aerosol–cloud feedbacks such as increases in infrared emissivity to the surface in winter/spring from anthropogenic aerosols (Garrett and Zhao 2006; Lubin and Vogelmann 2006).

It is also important to consider that aerosols contain a significant fraction of scattering aerosols such as sulfate (or organics), which cool the atmosphere, and that they also have natural sources, making the diagnosis and attribution of direct and indirect radiative effects a complex task. In fact, decreasing trends in midlatitude emissions of sulfur dioxide, resulting in declining sulfate concentrations in the Arctic (Quinn et al. 2009), may have led to warming in the Arctic. Increasing trends of BC, as well as O_3 precursor emissions, over Asia are predicted to have contributed as much as two-thirds of observed Arctic warming (e.g., Quinn et al. 2008; Shindell and Faluvegi 2009). This is despite recent negative trends in observed BC at surface sites in the Arctic due to emission reductions in Europe and the former Soviet Union (FSU) and points to the potentially important role of BC transport from Asia in the upper troposphere (Sharma et al. 2013), as well as midlatitude warming resulting in heat transport to the Arctic. At the same time, significant discrepancies between simulated seasonal cycles of trace gases and aerosols and surface observations in the Arctic point to gaps in our knowledge about pollution origins and processing during long-range transport to the Arctic (e.g., Shindell et al. 2008). Deficiencies in climate model treatments of aerosols and clouds, as well as in the Arctic radiation budget, have been recently highlighted as a possible reason for

poor climate model performance in the Arctic (e.g., Overland and Wang 2013).

The study of Arctic air pollution is not a new topic. As discussed by Garrett and Verzella (2008), the presence of some kind of visibility-reducing haze in the Arctic was noted by early explorers in the late nineteenth century and discussed, in particular, by Nordenskiöld (1883). It was also observed later by pilots in the 1950s (Mitchell 1957). It was not until the 1970s that it was suggested that this pollution or “Arctic haze” originated from anthropogenic emissions in northern midlatitudes (Rahn et al. 1977) and Eurasia (defined in this paper as Europe and the FSU, including Russia) in particular (Barrie et al. 1981). Long-term records of surface observations in the Arctic clearly show an increase in aerosol abundance in winter and early spring every year (e.g., Shaw 1975; Sharma et al. 2004). This springtime peak in total aerosol mass concentration is caused primarily by increases in submicrometer particles (or fine mode aerosols) composed of non-sea-salt (nss) sulfate and sea salt with smaller amounts of ammonium, nitrate, dust, trace elements of combustion, and a residual likely composed of particulate organic matter (e.g., Quinn et al. 2002). Arctic haze also contains elevated levels of gas-phase O_3 precursors such as volatile organic compounds (VOCs) and carbon monoxide (CO), as well as nitrogen oxides ($NO_x = NO + NO_2$) and the nitrogen reservoir peroxy-acetyl nitrate (PAN), in particular (e.g., Hov et al. 1989; Beine et al. 1996).

Previous aircraft campaigns in the 1980s (Schnell 1984; Radke et al. 1984; Leaitch et al. 1989; Brock et al. 1990) and the Tropospheric Ozone Production about the Spring Equinox (TOPSE) experiment in 2000 (Atlas et al. 2003) provided some of the first comprehensive information about the vertical structure of haze layers, finding them to be highly variable both vertically (tens of meters to 1 km thick) and horizontally (20–200 km). These measurements

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suggested that the seasonal maximum in aerosols in the free troposphere occurs later than at the surface (Scheuer et al. 2003). Despite these observations, the extent to which aerosol haze layers aloft are associated with the seasonal Arctic haze phenomenon at the surface remains an open question. In the summer, surface observations show a clear minimum in aerosol concentrations due to more efficient washout and less efficient transport to the Arctic at this time of year (e.g., Sharma et al. 2004; Garrett et al. 2010), although certain natural aerosols, such as those produced from oceanic emissions, peak in the summer months (Ferek et al. 1995; Quinn et al. 2002).

Observations of CO at sea level sites also show a summer minimum and winter–early spring maximum due to more active summertime photochemical destruction of CO (e.g., Shindell et al. 2008). Tropospheric O₃ is influenced by depletion events in the Arctic; the complex halogen chemistry found over sea ice can lead to very low or even near-zero O₃ concentrations in the spring months in the surface layer (e.g., Barrie et al. 1988). Re-emission of deposited nitrate as NO_x may contribute to increasing or sustaining O₃ at the surface (Honrath et al. 1999; Dibb et al. 2002). Summertime pollution layers, originating from boreal forest fires, have also been observed in the free troposphere as part of the airborne Arctic Boundary Layer Expeditions (ABLE-3) in July–August 1988 and 1990 (Wofsy et al. 1992). Ozone production in these summertime forest fire plumes was estimated to be negligible (Mauzerall et al. 1996) but the contribution from this source relative to anthropogenic emissions remains uncertain. During the TOPSE winter–spring campaign, photochemical O₃ production was diagnosed to dominate the O₃ budget in the Arctic troposphere with only a rather small contribution (20%) from the stratosphere (Emmons et al. 2003; Browell et al. 2003).

POLARCAT-IPY: OBJECTIVES AND MAIN ACTIVITIES. Although airborne campaigns prior to 2008 provided very useful first data about the composition and possible origin of pollution in the Arctic,

many scientific questions remained (Law and Stohl 2007), and major advances in airborne measurement techniques and modeling capabilities had occurred. These factors, coupled with the arrival of several key satellite instruments capable of making aerosol and trace gas measurements in the Arctic [e.g., the *Atmospheric Infrared Sounder* (AIRS), *Infrared Atmospheric Sounding Interferometer* (IASI), and *Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observations* (CALIPSO)] and the announcement of an International Polar Year (IPY) in 2007/08, motivated the atmospheric chemistry community to design an international project called the Polar Study using Aircraft, Remote Sensing, Surface Measurements and Models, Climate, Chemistry, Aerosols and Transport (POLARCAT), which is aimed at further improving

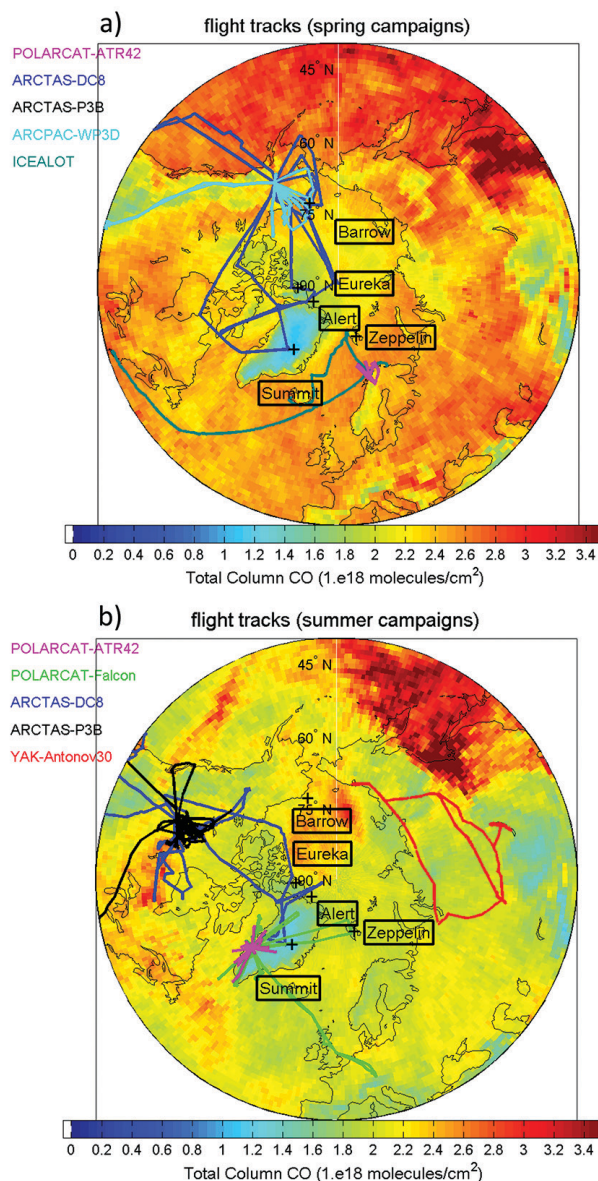


FIG. 1. Overview of all flights of the DC-8 (blue), P-3B (black), ATR-42 (magenta), WP-3D (cyan), DLR-Falcon (green), Antonov-30 (red) aircraft campaigns, and the ICEALOT ship cruise (green), during the (a) spring and (b) summer 2008 campaigns superimposed on IASI CO total column monthly mean maps (daytime). The CO data are averaged over 1° × 1°. The spring map is represented by the April mean and the summer by the July mean. Locations of surface sites referred to in this paper are also indicated. Adapted from Pommier et al. (2010).

our understanding of Arctic pollution and impacts on climate.

POLARCAT was recognized as an IPY activity, cosponsored by the International Geosphere Biosphere Programme core projects International Global Atmospheric Chemistry (IGAC) and Integrated Land Ecosystem–Atmosphere Processes Study (iLEAPS) and the World Climate Research Programme (WCRP) core project Stratosphere–Troposphere Processes and Their Role in Climate (SPARC). The main scientific objectives of POLARCAT, which continues as a task within IGAC, are described in the white paper available online at www.igacproject.org/POLARCAT. These objectives are shown in Table 1, grouped according to the main topics discussed in this review: namely, 1) pollution transport and origins, 2) aerosol composition and processes, and 3) trace gas chemical

composition and processes. To investigate these issues, POLARCAT organized several airborne campaigns at high northern latitudes during the spring and summer of 2008, focusing primarily on the Arctic free troposphere (up to 9 km). Six instrumented aircraft from the United States, Germany, France, and Russia participated, flying from bases in Alaska, Canada, Greenland, Sweden, and Russia and sampling across the entire Arctic. Figure 1 shows the flight tracks of the main aircraft campaigns and a ship cruise that took place, as well as the locations of ground-based sites referred to in this paper. Table 2 provides further details about the different POLARCAT projects; the platforms, including altitude ranges covered by the aircraft measurements; and references providing detailed information about the aircraft and ship deployments and payloads. Information about complementary

TABLE 1. POLARCAT-IPY scientific objectives.

| Theme | Scientific objectives |
|--|---|
| Pollution transport and origins | <ul style="list-style-type: none"> • Better characterize pollution transport pathways into the Arctic • Improve knowledge about sources of Arctic aerosols and trace gases • Quantify the residence times of pollution in the Arctic and their seasonal/interannual variability • Determine the vertical layering of pollution from different source regions including its seasonal variation • Quantify the transport pathways for boreal fire plumes into the Arctic |
| Aerosol composition and processes | <ul style="list-style-type: none"> • Improve knowledge about the sources, evolution, and removal of aerosols in Arctic haze • Characterize vertical distributions of chemical, physical, and optical properties and their impacts on direct radiative effects • Make detailed in situ observations of microphysical and optical properties of Arctic clouds including ice/mixed phase and examine aerosol–cloud interactions • Determine the impacts of soot deposition from forest fire and anthropogenic emissions on surface albedo of snow/ice surfaces and atmospheric radiative effects • Validate satellite observations of aerosols and clouds |
| Trace gas chemical composition and processes | <ul style="list-style-type: none"> • Improved understanding about O₃ photochemistry in the Arctic troposphere • Better quantify sources of Arctic O₃ during spring and summer • Study the impact of boreal fire emissions on chemical composition of the Arctic troposphere • Investigate the role of local photochemistry (halogens, snow emissions) on chemical composition in the Arctic boundary layer and free troposphere • Validate satellite observations of tropospheric trace gases |

campaigns and measurement programs is also given, as well as details of all project acronyms. We also note other significant research programs, not directly affiliated with POLARCAT: the Indirect and Semi-Direct Aerosol Campaign (ISDAC), investigating properties of Arctic stratus clouds and aerosol–cloud interactions in spring 2008 (McFarquhar et al. 2011), and the Arctic Summer Cloud Ocean Study (ASCOS;

Tjernström et al. 2014), studying low-level Arctic clouds and aerosol–cloud–ocean interactions. In this review, it is not possible to cover all issues listed in Table 2, such as aerosol–cloud interactions, to which POLARCAT also contributed.

Briefly, gas-phase measurements on board the POLARCAT aircraft included O₃ and its precursors (NO_x, CO, VOCs); intermediate VOC oxidation

TABLE 2. POLARCAT-IPY and related field campaigns. Asterisk (*) indicates complementary activities to POLARCAT.

| | | | | |
|-------------------------------------|---------------------------|---|---|--|
| ASTAR ^a | Mar–Apr 2007 | Dornier 228–101 (Polar-2), Deutsches Zentrum für Luft- und Raumfahrt (DLR) Falcon-20 | Arctic cloud and aerosol properties | Gayet et al. (2009) |
| ARCPAC ^b | Mar–Apr 2008 | National Oceanic and Atmospheric Administration (NOAA) WP-3D (0–7 km) | Aerosols plus radiation budget | Brock et al. (2011) |
| ARCTAS-A + ARCTAS-B ^c | Apr 2008/ Jul 2008 | National Aeronautics and Space Administration (NASA) DC-8 (0–12 km), NASA P-3B (0–7 km) | Aerosols, O ₃ photochemistry, boreal fires, satellite validation | Jacob et al. (2010), Fuelberg et al. (2010) |
| POLARCAT- France | Mar/Apr + Jun/Jul 2008 | French ATR-42 (0–7km) | Pollution transport (aerosols + O ₃) | de Villiers et al. (2010), Pommier et al. (2010) |
| POLARCAT- GRACE ^d | Jun–Jul 2008 | DLR Falcon-20 (0–11 km) | Pollution transport (O ₃) | Roiger et al. (2011b) |
| YAK- AEROSIB ^e | July 2008 | Russian Antonov-30 (0–6 km) | Siberian fires | Paris et al. (2009) |
| ICEALOT ^f | Mar–Apr 2008 | NOAA RV-Knorr ship | Orgins of aerosols over Arctic Ocean | Frossard et al. (2011) |
| ISDAC ^{*g} | Spring 2008 | National Research Council (NRC) Convair-580 (0–6 km) | Aerosol-cloud properties | McFarquhar et al. (2011) |
| POLAR- AOD ^{*h} | 2007–09 | Surface measurements | Aerosol optical and radiative properties | Tomasi et al. (2012) |
| IASOA ^{*i} | Continuous | Arctic surface sites | Air mass origins and trends of aerosols, O ₃ , CO | e.g., Quinn et al. (2009), Hirdman et al. (2010b) |
| ARC-IONS ^j | Periodic 2008 | Ozonesonde (multiple sites) | Vertical O ₃ distribution + origins | Tarasick et al. (2010) |
| GSHOX ^{*k} | Summer 2008 | Summit, Greenland | O ₃ /nitrogen photochemistry over snow | Thomas et al. (2012) |

^a Arctic Study of Tropospheric Aerosol, Clouds and Radiation.

^b Aerosol, Radiation, and Cloud Processes affecting Arctic Climate.

^c Arctic Research of the Composition of the Troposphere from Aircraft and Satellites.

^d POLARCAT-Greenland Aerosol and Chemistry Experiment.

^e YAK-Airborne Extensive Regional Observations in Siberia.

^f International Chemistry Experiment in the Arctic Lower Troposphere.

^g Indirect and Semi-direct Aerosol Campaign.

^h Polar Aerosol Optical Depth.

ⁱ International Arctic Systems for Observing the Atmosphere.

^j ARCTic–Intensive Ozonesonde Network Study.

^k Greenland Summit Halogen-HO_x Experiment.

products (e.g., carbonyls, peroxides); tracers of combustion (CO), industrial emissions (sulfur dioxide, SO₂), and biomass burning (acetonitrile, CH₃CN); and greenhouse gases. Detailed data were also collected on aerosol chemical composition, and their physical and optical properties. Additional data were provided by observations from satellites and ozonesondes (e.g., Pommier et al. 2010, 2012a; Tarasick et al. 2010) as well as by enhanced observations at long-term surface measurement sites (see Table 2). Studies using regional and global models were also a key part of POLARCAT ranging from forecasts for flight planning to post-campaign data analysis. Overall, analyses of POLARCAT data have so far resulted in more than 80 published papers, many of which appear in a special issue of *Atmospheric Chemistry and Physics* (www.atmos-chem-phys.net/special_issue182.html). In this review, we highlight some of the key results from POLARCAT. The discussion is focused around transport and origins of Arctic air pollution, Arctic aerosols, and Arctic gas-phase chemical composition (see the following three sections). Conclusions and future perspectives are discussed in the final section.

ARCTIC AIR POLLUTION: TRANSPORT AND ORIGINS. At the onset of the POLARCAT campaigns, while established concepts held that Arctic haze originated from long-range transport of Eurasian pollution coupled to inefficient pollutant removal, there was increasing evidence to support

some paradigm shifts in pollutant sources and transport processes impacting the Arctic troposphere. Most significantly, the potential for boreal forest fires (Stohl 2006; Stohl et al. 2006) and South Asian emissions (Koch and Hansen 2005) to be efficiently transported to the Arctic troposphere was highlighted, although controversy remained regarding the scale of these contributions. The main transport pathways are illustrated in Fig. 2.

The POLARCAT campaigns provided an unprecedented “snapshot” of the state of Arctic composition in spring and summer 2008. In terms of meteorological conditions, Fuelberg et al. (2010) noted that mid-latitude cyclones were more frequent and followed a more northerly course than usual, over eastern Asia and the northern Pacific, but were less common over the North Atlantic during spring. Frequent cyclone activity also occurred over the Pacific during summer 2008. At the same time, the North Atlantic Oscillation (NAO) transitioned toward a negative state in spring and remained so for the summer campaigns. Such a negative NAO state is associated with reduced pollution transport toward the Arctic (Burkhart et al. 2006), especially from Europe, compared to the mean (Eckhardt et al. 2003). Examination of AIRS satellite CO anomalies over the Arctic also suggested that transport of pollution to the Arctic was hindered in spring 2008 because of negative El Niño–Southern Oscillation (ENSO) conditions (Fisher et al. 2010).

Nevertheless, despite large-scale meteorological

patterns that did not favor transport from the midlatitudes, a surprising finding was the strong influence of Eurasian fire emissions, in particular from agricultural fires, during the spring campaigns (Warneke et al. 2009, 2010; Brock et al. 2011; McNaughton et al. 2011). In 2008, negative precipitation anomalies contributing to particularly large fires over Siberia were not driven by ENSO, even though in other years ENSO had been shown to be strongly linked to boreal fire activity and emissions of trace gases like CO (Monks et al. 2012). These emissions also occurred

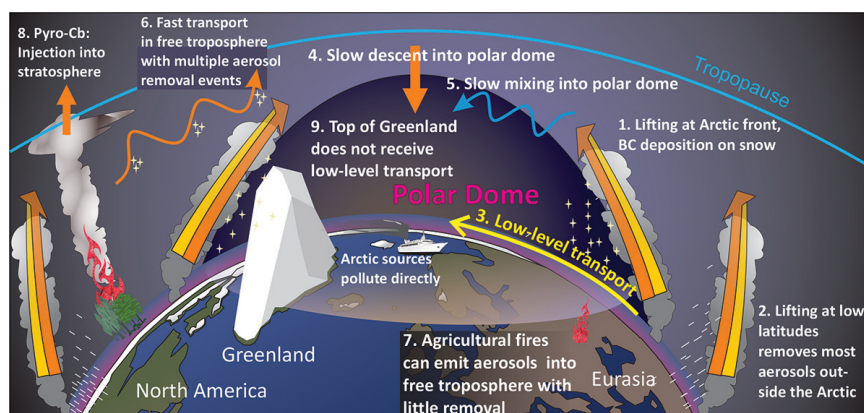


FIG. 2. Schematic showing pathways for the transport of air pollution into the Arctic. Following Stohl (2006), three main routes are evident: 1) low-level transport from midlatitude emission regions followed by uplift at the Arctic front; 2) lifting of pollutants at lower latitudes followed by upper tropospheric transport and eventual slow descent (due to radiational cooling) or mixing into the polar dome—a frequent transport route from North America and Asia but prone to significant wet scavenging; and 3) wintertime low-level transport of already cold air into the polar dome mainly from northern Eurasia. Emissions from strong boreal fires could be lofted by pyroconvection (Fromm et al. 2005) and later entrained into the polar dome.

farther east where the NAO has a weaker influence compared to high-latitude anthropogenic emissions in Europe. In addition, emissions of total carbon from agricultural fires in western Russia and Kazakhstan were twice as high (29 TgC) in 2008 as the 1997–2011 average while boreal wildfire emissions were 21% higher than the mean (214 TgC in 2008 compared to 180 ± 111 TgC yr⁻¹ over 1997–2011) (van der Werf et al. 2010).

Plumes originating from anthropogenic sources in eastern Asia and Europe were also sampled over the Arctic in spring 2008 (Singh et al. 2010; de Villiers et al. 2010). Transport by warm conveyor belts associated with frontal systems was an important mechanism for the transport of smoke and Asian pollution into the free troposphere whereas transport of pollution at low altitudes was primarily from Eurasia. During spring 2008, this resulted in higher aerosol loadings in the free troposphere as shown in Fig. 3 by satellite aerosol data from CALIPSO. This springtime feature was more pronounced in 2008 compared to other years (2006–12) (Di Pierro et al. 2013). Aerosol loadings in the lower troposphere were lower in spring having declined from the winter–early spring Arctic haze maximum observed every year. However, we note that CALIPSO data, based on validation against POLARCAT data, may underestimate aerosol loadings by 20%–30% below 4 km due to the presence of clouds and overestimate contributions from smoke and pollution/dust sources (Di Pierro et al. 2013; Ancellet et al. 2014; Burton et al. 2012). During summer 2008, the central Arctic was very clean (as shown in Fig. 3) because of more efficient washout, and the Arctic front retreating to high latitudes. Nevertheless, small enhancements in aerosols were observed in polluted air masses transported to the Arctic in the free troposphere from Asian/North American anthropogenic or boreal fire emission regions (Schmale et al. 2011; Quennehen et al., 2011).

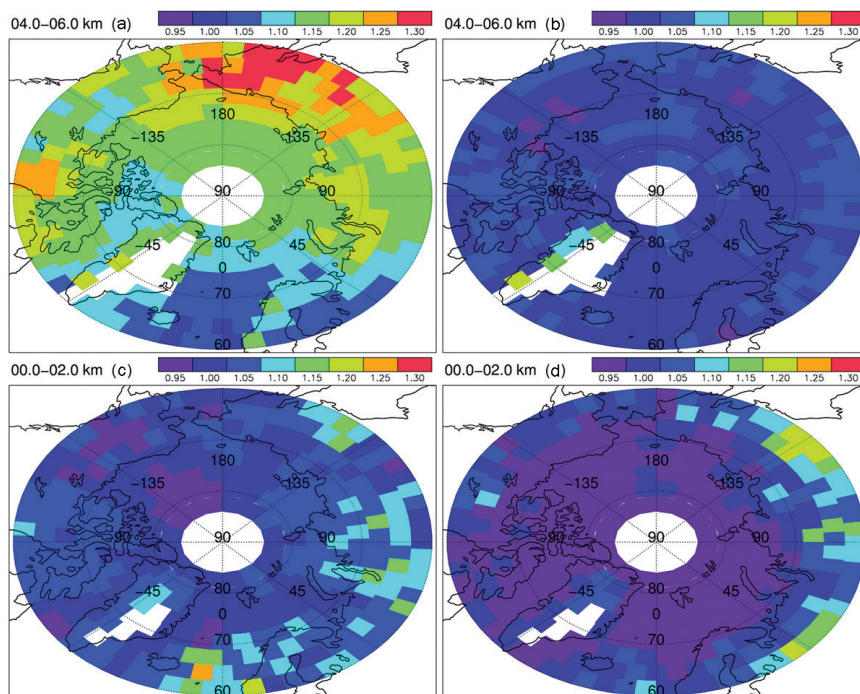


FIG. 3. Seasonal mean of the CALIPSO scattering ratio at 532 nm (I is for Rayleigh scattering) providing the spatial distribution of aerosol loadings over the Arctic for (a),(c) March–May and (b),(d) June–August 2008 and for two altitude ranges: (bottom) 0–2 km and (top) 4–6 km. A cloud mask was applied to keep only the aerosol contribution. Data averaged over 300 km² grid cells. Based on methods described in Ancellet et al. (2014).

Source attribution studies often use CO as a tracer of pollutant origins due to its rather long chemical lifetime of 1–3 months. Total CO columns observed by the IASI satellite instrument provided an Arctic-wide view of CO distributions in spring and summer 2008 as illustrated in Fig. 1. Higher CO was prevalent throughout the Arctic in spring compared to summer, especially downwind of Asia and boreal fire regions. In summer, while lower CO was observed due to enhanced photochemical destruction at this time of year, there is a suggestion of higher CO over the western Arctic. Global model studies, run with tagged CO for different sources, suggest that Asian anthropogenic emissions dominated the CO budget (for CO concentrations above the baseline due to methane oxidation) throughout the free troposphere in spring and summer 2008 (Fisher et al. 2010; Bian et al. 2013). An example is shown from Bian et al. (2013) in Fig. 4. Anthropogenic CO emissions from Europe were also important in the lower troposphere but models differ regarding the contribution from different source regions. Also, in contrast to a multimodel study, for the year 2001 (Shindell et al. 2008), Bian et al. (2013) did not find a significant contribution from North American emissions in 2008. Bian et al. (2013) also

found that boreal fires were a significant source of CO. This illustrates that CO source attribution depends on interannual variations in emissions and transport pathways. It is also important to note that, in general, global models underestimate CO in the Arctic (and Northern Hemisphere) (Shindell et al. 2008). Such discrepancies between observed and modeled CO concentrations, as well as other species such as O₃ and PAN, motivated the POLARCAT Model Intercomparison Project (POLMIP; Monks et al. 2014; Arnold et al. 2014; Emmons et al. 2014, manuscript submitted to *Atmos. Chem. Phys. Discuss.*), where several models were run with the same 2008 emissions. Analysis of results shows that, in this case, variability between modeled Arctic CO is driven mainly by differences in oxidative (OH) chemistry (Monks et al. (2014). Differences in simulated vertical export efficiency over midlatitude emission regions may also play an important role in determining large-scale pollutant transport to the Arctic.

The transport of smoke from wildfires in Siberia and pollution from eastern Asian anthropogenic sources was particularly well documented during the POLARCAT summer campaigns. This transport may occur frequently enough to affect average pollutant distributions in the summer (Hirdman et al. 2010a), as illustrated by the enhanced IASI CO over the western

Arctic mentioned earlier (see Fig. 1). Pollution was lifted into the Arctic by frontal systems and transported directly across the North Pole in the mid- and upper troposphere (Harrigan et al. 2011). In one particularly notable case, pollution was transported by this mechanism from eastern Asia into the lowermost stratosphere where it was observed over Greenland (Roiger et al. 2011a). When the aircraft sampled the plume ~1 km above the tropopause, the polluted air from East Asia was mixed with stratospheric air (see Fig. 5). Detailed comparison of Eulerian and Lagrangian model simulations with aircraft data and CO satellite retrievals demonstrated that the models were able to simulate this complex cross-polar transport episode rather accurately (Sodemann et al. 2011). While transport of air from the troposphere to the stratosphere has been studied previously (e.g., Sprenger et al. 2003; Stohl et al. 2003), the POLARCAT campaigns provided some of the first detailed in situ observations of cross-polar pollution transport into the lowermost stratosphere. These findings suggest that the Arctic lower stratosphere (and upper troposphere) may hold substantial amounts of pollution, primarily from low-latitude Asia, which could eventually descend into the Arctic and midlatitude troposphere. In fact, Trickl et al. (2014) show a downward trend (since 1990) in observed CO

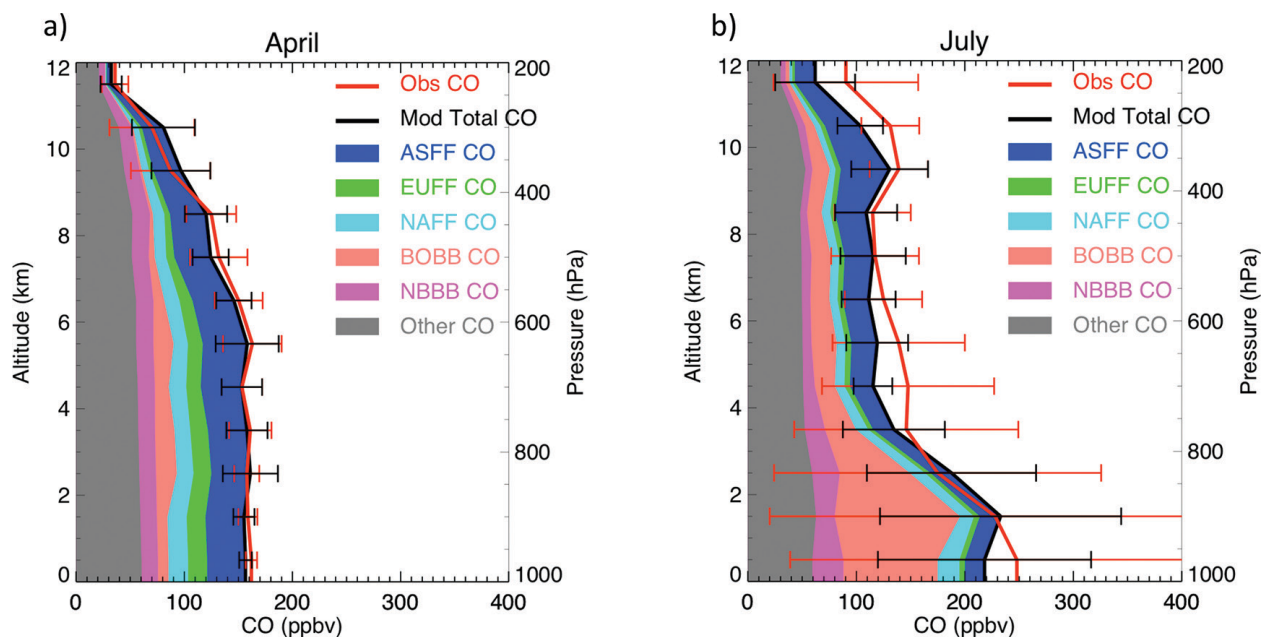


FIG. 4. Vertical distribution of CO mixing ratios (ppbv) from the ARCTAS DC-8 measurements (solid thick red line plus standard deviation shown as horizontal bars) and from the Goddard Earth Observing System Model, version 5 (GEOS-5) global model simulations sampled along the flights for (a) April and (b) July. Model results are shown for total CO (thick black line) and for tagged components from Asian (AS), European (EU), North American (NA), fossil fuel (FF), boreal biomass burning (BOBB) and nonboreal biomass burning emissions (NBBB). “Other CO” refers to global CO other than the five tagged components and is attributed mainly to CO from methane oxidation. From Bian et al. (2013).

concentrations at Zugspitze mountain (47.4°N, 11.0°E), Germany, with the exception of air masses originating from the high-latitude lower stratosphere. Increasing CO concentrations in these air masses can best be explained by increasing CO emissions in East Asia and transport events similar to those observed by Roiger et al. (2011a).

Sources of pollutants near the surface are very different from those sampled by the aircraft in the free troposphere. Statistical analysis of long-term monitoring data from the stations Alert (82.5°N, 62.3°W), Barrow (71.3°N, 156.6°W), and Zeppelin (78.9°N, 11.9°E) (Fig. 1) confirmed earlier results (e.g., Sirois and Barrie 1999; Stohl 2006) that the main source of BC aerosols near the surface in the Arctic is northern Eurasia in winter/spring and that smelting activities in Norilsk, northern Russia, are a significant source of sulfate in the eastern Arctic (Hirdman et al. 2010a). Only during summer is there clear evidence of influence by boreal forest fires

at surface sites. In addition, a recent study (Stohl et al. 2013) suggests that emissions from flaring of natural gas by the oil industry in northern Russia may be an important missing winter–spring source of Arctic BC. Trend analysis by Hirdman et al. (2010b) also confirmed earlier studies showing decreases in observed equivalent BC and sulfate at surface locations since the late 1980s (e.g., Sharma et al. 2004; Quinn et al. 2009). Hirdman et al. (2010b) showed that decreases in northern Eurasian emissions appear to be the main cause, and that changes in atmospheric transport to the Arctic were not important. This finding is supported by the global model study of Sharma et al. (2013), although they point out that, while BC concentrations may be

decreasing at the surface, it is possible that they may be increasing aloft due to increasing Asian emissions. We note that POLARCAT aircraft measurements only showed pollution layers present above the sea ice inversion layer and not reaching the (sea ice) surface during spring 2008 (Brock et al. 2011).

ARCTIC AEROSOLS. Advances in instrumentation made the characterization of the vertical distribution of aerosol chemical composition and optical properties a particular focus during POLARCAT.

This is illustrated in Fig. 6, which shows an example of the vertical distribution of aerosols

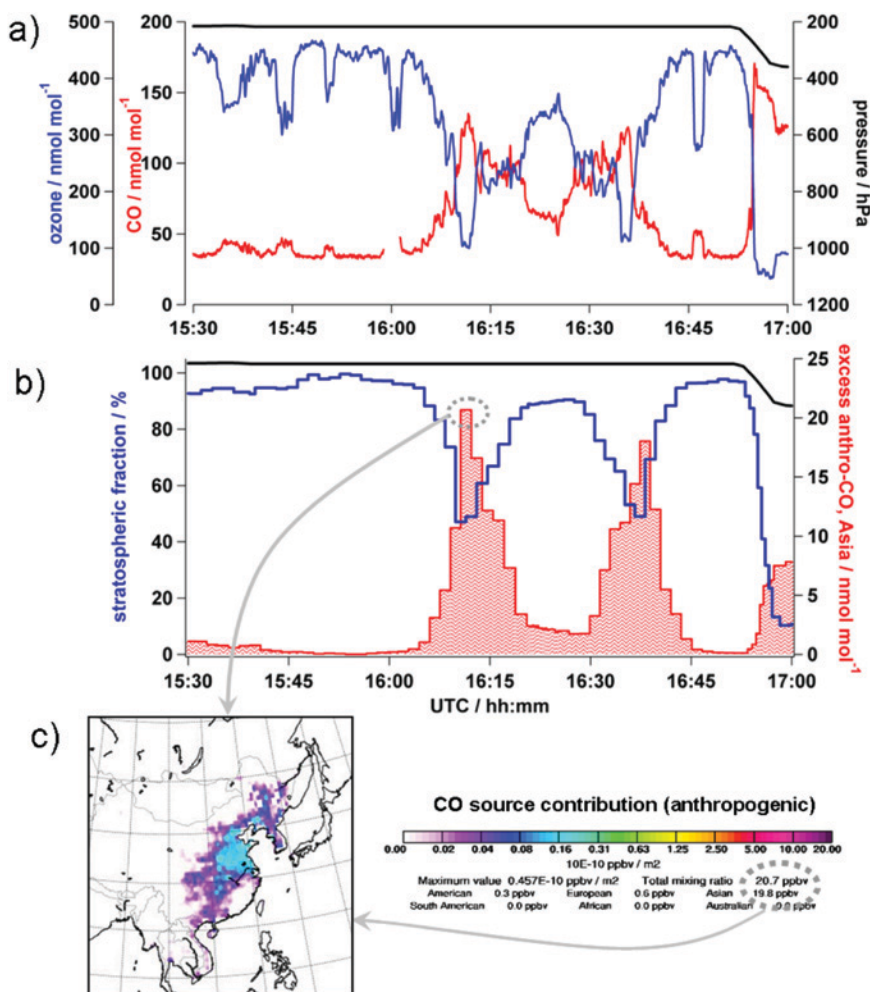


FIG. 5. (a) Time series of in situ CO (red), in situ O₃ (blue), and pressure at flight altitude (black) for the POLARCAT-GRACE flight on 10 Jul 2008, between 1530 and 1700 UTC. (b) Simulated CO tracers from the FLEXible PARTICle Dispersion Model (FLEXPART); the red pattern represents the FLEXPART excess CO originating from Asian anthropogenic pollution; the blue line gives the stratospheric fraction along the flight path. (c) CO source contribution plot for 1611 UTC (maximum CO concentration at 11.3-km altitude) with color scaling on the right. From Roiger et al. (2011a).

collected during the Aerosol, Radiation, and Cloud Processes Affecting Arctic Climate (ARCPAC) spring campaign north of Alaska on 18 April 2008 (Brock et al. 2011). Of particular note are enhanced concentrations of organic aerosols (OA) in the mid and upper troposphere attributed primarily to boreal fires over Eurasia based on enhancements in tracers like CH_3CN (e.g., Warneke et al. 2009). Enhanced sulfate (non-sea-salt component) was also observed and attributed to anthropogenic origins. Fisher et al. (2011) diagnosed that, in the free troposphere, sulfate from East Asian sources exceeded that from

European sources, in contrast to previous studies suggesting that Europe was the main source (Shindell et al. 2008). Overall, OA and sulfate dominated the aerosol mass in the free troposphere during spring 2008. Figure 6 also shows enhanced BC in the free troposphere together with enhanced sulfate and OA, a feature also observed during Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS; Wang et al. 2011). Enhanced BC was attributed to Eurasian fires (Warneke et al. 2010) and Asian anthropogenic emissions (Wang et al. 2011) in spring 2008 with differences likely due to different campaign sampling strategies. Since sulfate enhancements were primarily of anthropogenic origin, positive correlations with OA enhancements suggest mixing of Asian anthropogenic and Siberian boreal fire plumes during transport to the Arctic (Wang et al. 2011). There is also evidence for plumes of mixed anthropogenic (Asian) and Russian fire origin containing sootlike particles in air masses transported from Asia to the north of Scandinavia (de Villiers et al. 2010; Quennehen et al. 2012).

During POLARCAT, BC mass concentrations were directly measured for the first time using airborne single particle soot photometry (SP2), with an estimated uncertainty of $\pm 40\%$, in the Arctic free troposphere with the highest concentrations found aloft (Jacob et al. 2010; Warneke et al. 2009). Overall, BC concentrations observed during POLARCAT in spring 2008 were higher than direct measurements reported during Pan-Arctic Measurements and Arctic Regional Climate Model Intercomparison Project (PAM-ARCMIP) in spring 2009 when cleaner air masses were sampled (Stone et al. 2010). During summer 2008, aerosol concentrations in the free troposphere were generally 2–3 times lower than in spring and BC concentrations were 10 times lower (based on all available data) (Matsui et al. 2011). Organics, and to a lesser extent sulfate, were rather pervasive during the summer POLARCAT-France campaign over Greenland, albeit at low concentrations, and the organic fraction of the submicrometer aerosol increased during strong gas-phase pollution episodes observed between 4 and 6 km (Schmale et al. 2011). Sampled plumes in July 2008 originated primarily from Canadian fires and contained higher organic fractions. Mixed Asian fossil fuel and Siberian fire plumes were also sampled, exhibiting higher sulfate fractions (Schmale et al. 2011).

New information was also collected about aerosol composition at or near the surface, particularly in spring 2008. Aerosol mass was dominated by sulfate at several Arctic sites at this time of year and, as noted earlier, attributed to Eurasian emissions, in

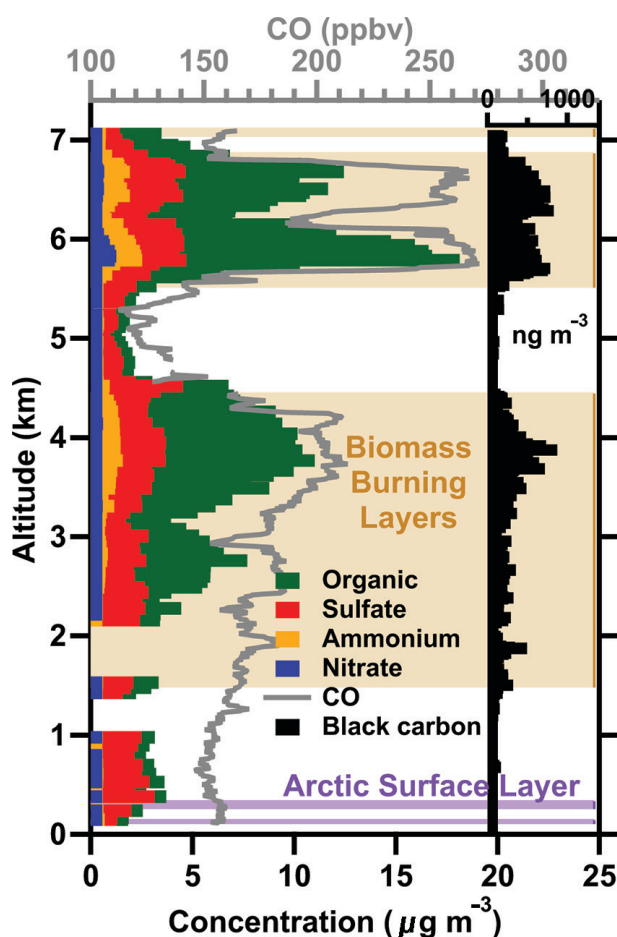


FIG. 6. Vertical profile from the ARCPAC spring campaign measured during a slantwise ascent of the WP-3D aircraft over sea ice north of Alaska on 18 Apr 2008. Submicron particle composition measured by aerosol mass spectrometry providing concentrations of sulfate (red), organic aerosols (green), ammonium (yellow), and nitrate (blue) aerosols (in $\mu\text{g m}^{-3}$). Measured CO mixing ratios in ppbv (gray) and BC concentrations in ng m^{-3} (black) are also shown. Tan and purple shading show regions identified as being directly influenced by biomass burning, and being within the boundary layer, respectively. Regions with no shading are in the “background” category. Adapted from Brock et al. (2011).

agreement with previous studies (Fisher et al. 2011; Hirdman et al. 2010a). This was also shown to be the case during the spring 2008 International Chemistry Experiment in the Arctic Lower Troposphere (ICEALOT) ship cruise in the eastern Arctic where Frossard et al. (2011) attributed air masses with elevated organosulfates and sulfate to coal burning in eastern Europe. Results from this study are illustrated in Fig. 7, which shows aerosol composition in air masses with Arctic and European origins. While sulfate clearly dominated the eastern Arctic during spring 2008, Arctic air masses had a larger fraction (on average approximately 40%) of marine aerosols compared to European air masses, which had a higher fraction (70%) from combustion sources. In both air mass types, the proportion of submicron organic mass was lower compared to 2 years of spring data from Barrow, Alaska (also shown in Fig. 7). Shaw et al. (2010) examined one year of measurements at Barrow (March 2008–March 2009) and found higher particulate organic matter (POM) concentrations in winter/spring compared to summer. In winter, analysis of the organic component indicated a higher oceanic contribution (i.e., hydroxyl groups and POM correlated with inorganic seawater components), whereas in spring anthropogenic sources dominated with alkane and carboxylic acid functional groups correlated with elemental tracers of Asian, shipping, and boreal fire emissions. Overall, organic fractions made up a significant fraction of the total aerosol mass, for example, reaching up to 35% at Barrow in spring (Frossard et al. 2011; Shaw et al. 2010). These results highlight the complex nature of organic aerosols, which can sometimes make up a substantial fraction of the total aerosol mass and are still poorly characterized in models (e.g., Lapina et al. 2011).

The findings discussed so far suggest that our picture of Arctic aerosol sources, distribution, and composition is more complex than previously thought. Brock et al. (2011) used long-term ground-based measurements and the short-term aircraft observations provided by ARCPAC (in the western Arctic) to conclude that Arctic aerosol pollution has both chronic and episodic components. Aerosol light scattering, submicrometer sulfate mass concentration, and particle number concentrations measured at Barrow during winter and early spring have limited temporal variability compared to episodic dense aerosol layers observed aloft. Brock et al. (2011) noted that this chronic aerosol haze, as measured at the surface, originates primarily from Eurasian sources and increases in the winter–spring boundary layer in the Arctic due to continued emissions into the

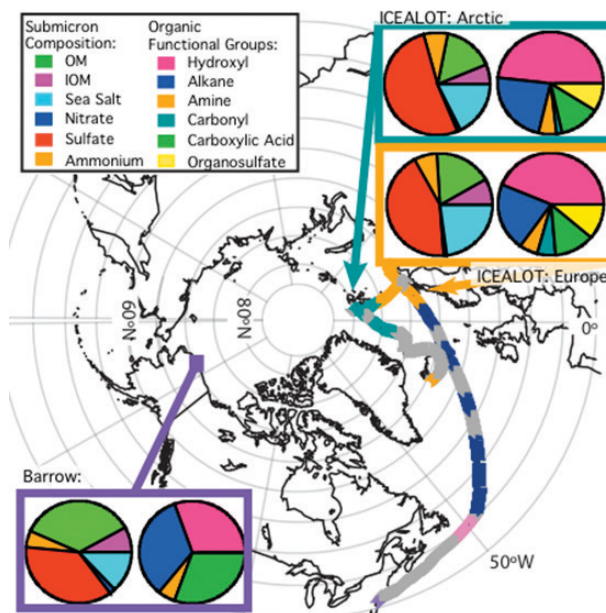


FIG. 7. Track of the R/V Knorr ship during the ICEALOT cruise (19 Mar–24 Apr 2008) colored by regions of air mass origin including northeast United States (purple), North America (pink), North Atlantic (dark blue), Europe (bronze), and Arctic (teal). Gray represents air masses of with no distinct origin or periods without sampling. The pies represent the (left) submicron particle composition and (right) organic functional group composition for the European and Arctic source regions during ICEALOT and Barrow, Alaska (dark purple). The submicron particle composition excludes the elemental carbon (estimated on average less than 3% during ICEALOT and at Barrow). OM is submicron organic mass. IOM is inorganic oxidized material (dust and fly ash). Note that the Barrow data are an average of 2 years (2008 and 2009). From Frossard et al. (2011). Copyright 2011 by the American Geophysical Union.

polar dome, limited vertical mixing, and inefficient removal (Fig. 2). The haze dissipates in summer with the poleward retreat of the Arctic front and increased Arctic precipitation (Garrett et al. 2010). Although there have been occasional observations of dense haze layers at the surface (e.g., Stohl et al. 2006), these do not dominate the seasonal cycle of average aerosol values (Brock et al. 2011).

Aloft in the free troposphere, long-range transport of pollution from midlatitude sources, often associated with lifting in midlatitude warm conveyor belts, can lead to episodically enhanced aerosol loadings. Brock et al. (2011) suggested that since such intercontinental long-range transport is not unique to the Arctic, it can be considered a separate phenomenon from the chronic pollution reported at surface sites in the Arctic and commonly referred to as Arctic haze. Indeed, the aerosol composition measured in background air outside

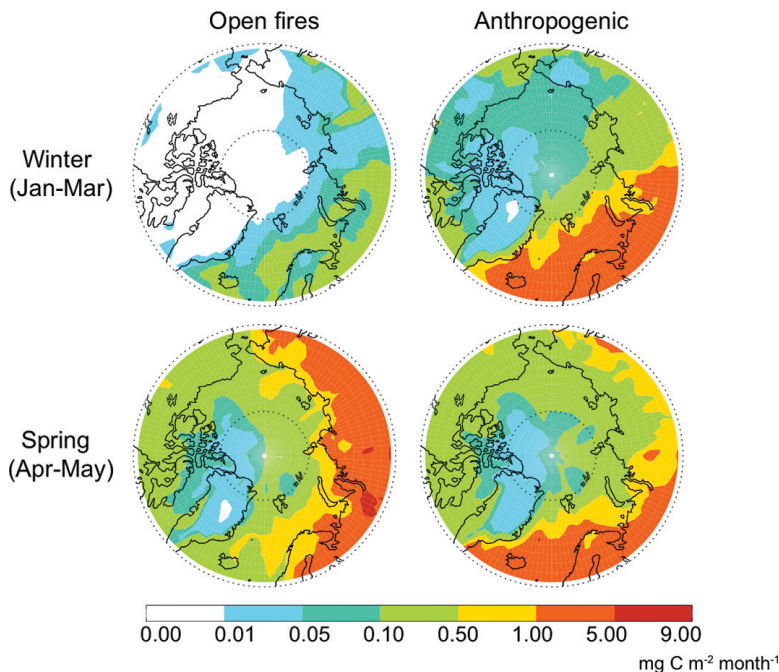


FIG. 8. Contributions of boreal and agricultural fire emissions and anthropogenic (fuel combustion) emissions to total (wet plus dry) deposition fluxes of black carbon from GEOS-Chem global model simulations for winter (January–March) and spring (April–May) 2008. From Wang et al. (2011).

of discrete aerosol layers (Fig. 6) was very similar to the average composition of nonrefractory components reported in springtime at the surface at Barrow (Brock et al. 2011); that is, anthropogenic sulfate made up the largest mass fraction with a smaller contribution of organics resulting from biomass or biofuel combustion. Jacob et al. (2010) also reported anthropogenic aerosol dominating below 2 km. In contrast, in the free troposphere, fire emissions were an important contributor to aerosol layers between 2 and 7 km in spring 2008, particularly for organic and BC aerosols, and were superimposed on, or mixed with, aerosols of anthropogenic origin (Wang et al. 2011; de Villiers et al. 2010). Taken together, these observations are consistent with a background of a largely anthropogenic aerosol that results from the emission of pollution into the polar dome (Fig. 2) and that has been identified as Arctic haze by surface observations. Superimposed on the background Arctic haze are pollution and biomass burning plumes from midlatitude sources due to episodic transport events to the Arctic free troposphere during both spring and summer. This picture of a background pollution aerosol perturbed by periodic transport events aloft, which was particularly evident in spring 2008, is consistent with the analysis of sources and transport described in the previous section, and with the work of Stohl (2006), who demonstrated that

seasonal differences in transport processes could potentially explain much of the seasonality and vertical distribution of air pollution in the Arctic.

Another research focus during POLARCAT was improving understanding about the impacts of BC-containing aerosols on the radiative budget in the Arctic including the deposition of BC to snow and ice and its impact on surface albedo. Wang et al. (2011) used a global model to estimate BC deposition in the Arctic during the period January to May 2008 and diagnosed that wet deposition accounted for 85%–91% of total BC deposition during this period. This is in agreement with certain previous studies (e.g., Bourgeois and Bey 2011) but contrasts with other studies estimating that dry deposition dominates in the Arctic winter (e.g., Sharma et al. 2013). While dry depositional fluxes calculated by Wang et al. (2011) were only 15%

and 9% of total deposition in winter and spring, respectively, they were similar in magnitude to those derived from independent estimates based on analysis of springtime POLARCAT aircraft data in the range of $1500 \text{ ng m}^{-2} \text{ day}^{-1}$ (Spackman et al. 2010). It had been suggested previously that dry deposition may occur preferentially over regions of open leads (linear cracks of open water within the sea ice) as a result of convectively driven turbulence due to temperature differences between open leads and surface-layer air (Strunin et al. 1997). During several low-altitude ARCPAC flights, a positive vertical gradient in BC mass was indeed observed in the vicinity of open leads, providing evidence for dry deposition of BC from low-altitude, well-aged air masses in the Arctic (Spackman et al. 2010). Wang et al. (2011) estimated that anthropogenic emissions over Eurasia made the largest contribution to total BC deposition in the Arctic winter (January–March) whereas Russian fires were the main source in spring 2008 (April–May) (see Fig. 8). They also calculated a 0.4% winter and 0.8% spring decrease in snow albedo averaged over the Arctic corresponding to a surface radiative forcing from deposited BC of 0.1 W m^{-2} in winter and 1.7 W m^{-2} in spring.

Evaluation of global models using POLARCAT airborne observations has led to some improvements in modeled vertical aerosol distributions using

adjusted wet and dry deposition schemes (Wang et al. 2011; Fisher et al. 2011). However, many global aerosol models, including those used to analyze POLARCAT data, still struggle with reproducing observed BC concentrations in both snow and air. This may be partly due to deficiencies in emissions (Stohl et al. 2013) but treatment of both wet and dry deposition in models is a challenge. Even if, for example, improved parameterizations of wet scavenging leads to better agreement with data in some cases (e.g., Browse et al. 2012), differences in modeled estimates of the relative contributions of wet versus dry deposition requires further attention (Bourgeois and Bey 2011; Liu et al. 2011; Wang et al. 2011; Sharma et al. 2013; Lee et al. 2013). Analysis of POLARCAT summertime data suggests large variability in the efficiency of aerosol wet scavenging, with an average aerosol lifetime of 7–11 days (Schmale et al. 2011). Additional observational constraints are needed to improve our knowledge about wet and dry deposition mechanisms and rates and their treatment in models.

BC-containing particles can also directly perturb the energy balance in the Arctic by absorbing solar radiation and heating the surrounding air. Using aircraft observations of light scattering and extinction and a radiative transfer model, Brock et al. (2011) calculated the radiative forcing from measurements made during ARCPAC in the springtime Alaskan Arctic. Instantaneous, maximum noontime heating rates were found to be higher in the dense smoke layers aloft (up to $\sim 0.6 \text{ K day}^{-1}$) than in more diffuse background haze layers ($\sim 0.1 \text{ K day}^{-1}$). The presence of both background diffuse and dense haze layers resulted in a net cooling at the surface and a heating of the atmospheric column. Dense smoke from springtime biomass burning is a potentially large, but highly variable, source of aerosol to the Arctic during the snowmelt season. The net impact of smoke, in terms of cooling the surface through absorption in the atmosphere or warming the surface through deposition of BC on snow and ice and downward transport of heat from the atmosphere, has yet to be fully resolved (Bond et al. 2013) and represents a critical area for future research.

ARCTIC GAS-PHASE CHEMICAL COMPOSITION. POLARCAT airborne campaigns provided new information about the vertical distribution of Arctic chemical composition in the troposphere and lowermost stratosphere during spring and summer 2008. Again, advances in measurement techniques allowed improved assessment of vertical distributions of many trace species including total odd nitrogen

(NO_y) components (NO_x , PAN, HNO_3 , organic nitrates), oxidized volatile organic compounds [OVOCs; e.g., acetone, formaldehyde (HCHO)], free radicals [e.g., hydroxyl (OH)], and tracers of pollution sources (e.g., acetonitrile, CO) (Singh et al. 2010; Neuman et al. 2010; Olson et al. 2012; Wespes et al. 2012; Thomas et al. 2013). A number of these species had not been measured in the Arctic free troposphere prior to the POLARCAT missions, especially during the summer.

Figure 9 shows profiles of selected trace gases (CO , O_3 , PAN, and NO) in the spring and summer from various airborne campaigns. Profiles of CO reflect seasonal variations in photochemical destruction, as noted earlier, as well as influences from different sources. During spring 2008, ARCPAC sampled enhanced concentrations of CO and PAN in aged Siberian fire plumes (Fig. 9, top panels). In summer 2008, ARCTAS-B made flights close to boreal fires in Canada and measured significant enhancements in CO , NO , and PAN (as well as other trace species and aerosols) in the lower troposphere (Fig. 9, bottom panels). POLARCAT Greenland Aerosol and Chemistry Experiment (POLARCAT-GRACE) and POLARCAT-France sampled some of these fire plumes downwind over Greenland as well as anthropogenic plumes from North America (Thomas et al. 2013) and pollution from Asia/Siberia resulting in elevated CO and PAN concentrations between 4 and 9 km (Roiger et al. 2011b). Enhanced PAN downwind of source regions in spring (Fig. 9d) and summer (Fig. 9h) can be explained by fast conversion of NO_x to PAN near emission regions, followed by rapid lofting into the colder free troposphere, where PAN is thermally stable (Roiger et al. 2011b). Boreal fires sampled over Canada in summer 2008 were a particularly important source of PAN (Alvarado et al. 2010). NO_x released from PAN decomposition, when air masses descend to lower warmer altitudes, can produce O_3 . Overall, PAN was the dominant nitrogen species accounting for 50% of NO_y in the spring and 70% in summer (Liang et al. 2011). The measured NO_y budget was largely balanced with PANs, alkyl nitrates, NO_x , and HNO_3 accounting for more than 90% of total measured NO_y (Singh et al. 2010). Although little NO was found in the midtroposphere, mixing ratios were higher above 8 km due to stratospheric and lightning influences (in summer) (Liang et al. 2011). Observed O_3 mixing ratios show a compact vertical distribution, particularly in summer. Higher O_3 concentrations in the upper troposphere are attributable to greater stratospheric influence, particularly in the spring.

Active local fires over Canada and Siberia were sampled in summer 2008, providing new information

about fire emissions from these important sources (Singh et al. 2010; Paris et al. 2009). The fires over Canada were not only sources of PAN and other nitrogen species (e.g., NO, NO₂, CH₃CN) but also an important source of CO, VOCs (e.g., alkanes, alkenes, aromatics), and OVOCs (e.g., HCHO, acetone, methanol) (Simpson et al. 2011). Monoterpenes, usually considered to originate from live biogenic emissions, were reported, for the first time, to have large enhancements in plumes from boreal fires—an interesting finding because monoterpenes are important precursors for secondary organic aerosols. Surprisingly, many emission ratios measured in smoke from boreal and midlatitude Californian forest fires were largely indistinguishable between the two climate zones (Singh et al. 2010).

The large body of data collected on Arctic chemical composition is being used to evaluate a range of models. This includes measurements of OH, which is the main tropospheric oxidant for CO, VOCs, and

methane. For example, vertical distributions of HO_x [OH plus hydroperoxyl (HO₂)] radicals, together with profiles of OVOCs, which can act as radical sources or sinks, were used to constrain a photochemical box model, and showed that springtime HO₂ is overpredicted despite modeled OH agreeing reasonably well with the data (Olson et al. 2012). During the summer, Olson et al. (2012) found opposite discrepancies between modeled and observed OH (underpredicted) and HO₂ (overpredicted), pointing to significant difficulties in reproducing observed concentrations of radical sources and sinks (e.g., HCHO, hydrogen peroxide). A global model analysis of these data suggested a new chemical mechanism, involving conversion of HO₂ on aerosols, to reconcile differences between modeled and observed HO_x partitioning (Mao et al. 2010). This mechanism may also explain the underestimation of modeled CO in northern mid and high latitudes (Mao et al. 2013), although further research is needed to investigate these processes.

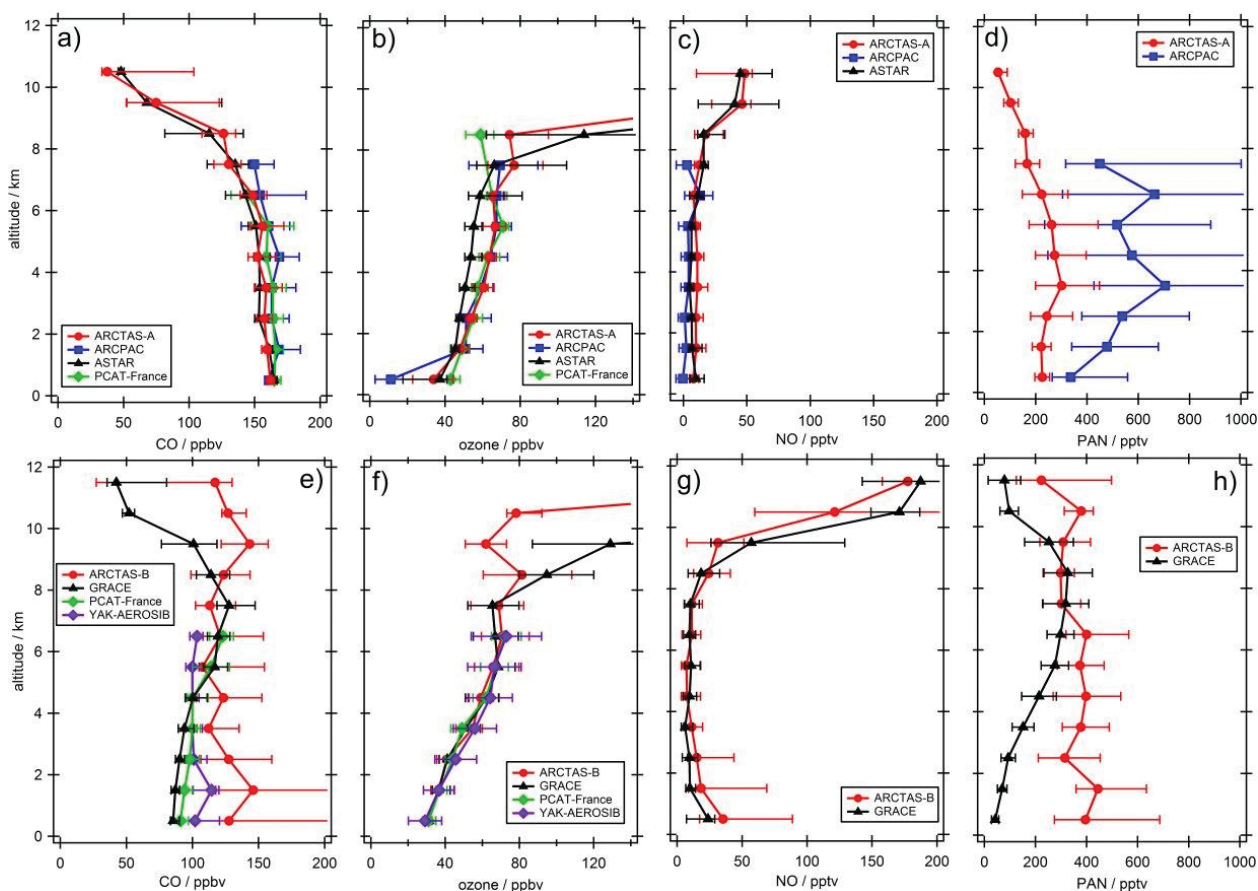


FIG. 9. Mean vertical distribution of trace gas mixing ratios (CO/ppbv, O₃/ppbv, NO/pptv, and PAN/pptv) measured during different POLARCAT aircraft deployments (see Table 2). (top) Data from spring campaigns covering the period 18 Mar–23 Apr 2008 except for ASTAR/DLR (26 Mar–17 Apr 2007); (bottom) Measurements from summer deployments covering the period 29 Jun–29 Jul 2008. The horizontal bars at each altitude bin indicate the extent of the interquartiles (25th and 75th percentile).

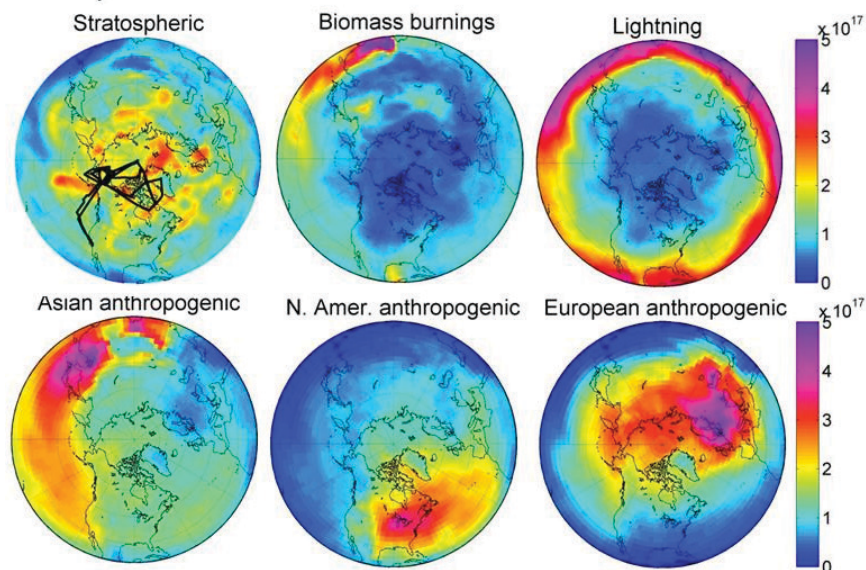
The impact of boreal fires on the production of tropospheric O_3 was a focus of POLARCAT, particularly in the summer when photochemistry is more active. As noted in a review of fire impacts on O_3 (Jaffe and Wigder 2012), O_3 is generally formed in aged air masses downwind of boreal fire emission regions due to PAN decomposition releasing NO_x (e.g., Real et al. 2007), although there can be some O_3 formation even close to the fires. However, Alvarado et al. (2010) found little or no enhancement in O_3 relative to CO in fresh plumes over Canada due to rapid PAN formation limiting NO_x availability and O_3 formation potential close to the fires. In another study, Singh et al. (2010) noted that aged fire plumes transported at higher altitudes over North America were generally mixed with some urban emissions and did have enhanced O_3 . However, these fire plumes may have also been influenced by NO_x produced by lightning or by air masses of stratospheric origin, both having higher NO_x mixing ratios (Liang et al. 2011). Plumes measured over Siberia during summer, in the lower troposphere, exhibited depleted O_3 relative to CO, probably due to widespread dry deposition of O_3 onto vegetation, thus limiting the potential for O_3 enhancements in aged fire plumes at low altitudes (Engvall Stjernberg et al. 2011; Paris et al. 2010).

Wespes et al. (2012) carried out a detailed evaluation of Arctic O_3 sources in spring and summer 2008 based on tagged tracers in the global Model for Ozone and Related Chemical Tracers, version 4 (MOZART-4; see Fig. 10). Diagnosing O_3 sources is

more complex than for CO (discussed previously in “Arctic air pollution: transport and origins” section) due to photochemical production/destruction, natural sources (stratosphere, lightning NO_x), and loss by dry deposition. During spring, Arctic O_3 was influenced primarily by anthropogenic emissions from Europe in the lower troposphere and from Asia/North America in the midtroposphere. Wespes et al. (2012) found that the lower stratosphere was an important source of up-

O_3 contributions

1-19 April:



18 June-13 July:

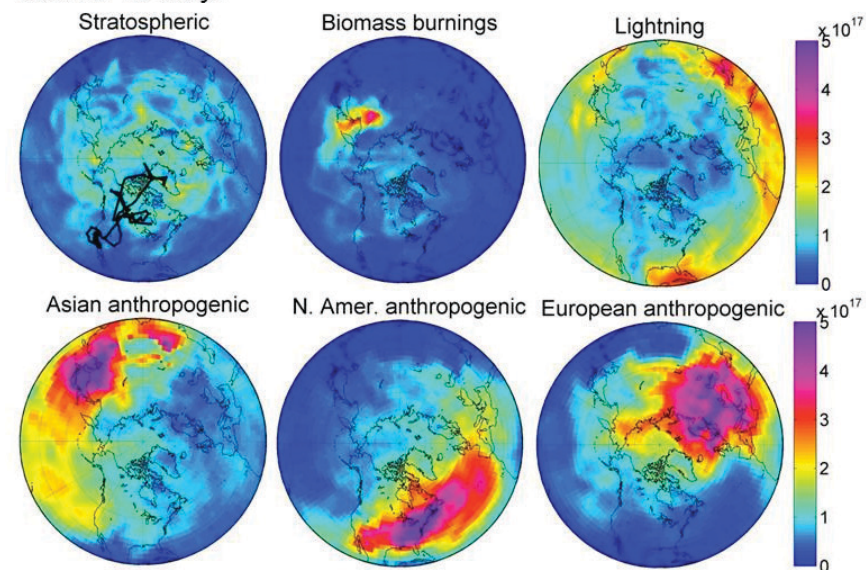


FIG. 10. Principal contributions to O_3 columns (molecules cm^{-2}) from the ground to 300 hPa during spring (1–19 Apr 2008) and summer (18 Jun–13 Jul 2008) ARCTAS campaigns as simulated by the global MOZART-4 model. The flight tracks corresponding to these periods are superimposed in black on the stratospheric contribution plots. From Wespes et al. (2012).

per tropospheric O_3 in spring but much less so during the summer campaign period when O_3 production from European/Asian emissions and lightning NO_x were found to be important sources of mid and upper tropospheric O_3 . Their findings, together with other studies based on POLARCAT data (e.g., Roiger et al. 2011a), suggest that summertime O_3 in the Arctic upper troposphere, which shows a similar vertical distribution over Greenland (GRACE data in Fig. 9f) to the spring (Fig. 9b), is considerably influenced by tropospheric sources including anthropogenic emissions. In the summertime lower and midtroposphere, Wespes et al. (2012) also diagnosed that European emissions made an important contribution to O_3 over the eastern Arctic and that O_3 produced downwind of North America was also an important source. This is in contrast to the multimodel analysis of Shindell et al. (2008), which found North America to be the main O_3 source at all altitudes in the Arctic. Wespes et al. (2012) estimated that boreal fire emissions made a significant contribution only in the summer months. However, the authors note that O_3 production may be underestimated in the midtroposphere in their model since simulated O_3 is underestimated in this region. This was also the case in Alvarado et al. (2010), who diagnosed low contributions from boreal fires to Arctic O_3 using the Goddard Earth Observing System (GEOS)-Chem model.

In contrast to large-scale model simulations, a high-resolution regional study, using the Weather Research and Forecasting (WRF)-Chem model,

examined the transport of anthropogenic and boreal fire plumes from North America to Greenland in July 2008 and diagnosed significant O_3 production in individual plumes (Thomas et al. 2013). Figure 11 shows the average impact on O_3 concentrations from anthropogenic and fire plumes transported to latitudes higher than $55^\circ N$ (as function of altitude) during the study period within the regional model domain. O_3 increased by up to 18% from anthropogenic emissions and about 5% from boreal fire emissions (with emissions from fires representing 21% of total CO emitted during the model run). Other non-POLARCAT-related studies have also noted significant contributions from boreal fires to Arctic O_3 (e.g., Parrington et al. 2012) and that the PAN contained in such plumes can lead to significant O_3 production during warmer summer months (Walker et al. 2012). Reasons for differences in model responses may be due to a number of factors such as the injection height of fire emissions (e.g., Sessions et al. 2011). As part of POLMIP, Arnold et al. (2014) demonstrate that global models, run using the same fire emissions for 2008 (Wiedinmyer et al. 2011), all produce O_3 in fire-dominated air masses, with increasing O_3 production as the fire emissions age. Ozone production efficiency in the Arctic is strongly sensitive to model VOC chemistry because PAN, which originates from VOC oxidation, dominates summertime Arctic NO_y . Differences in the efficiency of PAN formation, linked to differences in vertical export efficiency from emission regions, also contribute to model diversity.

Improved understanding regarding the source of reactive bromine responsible for near-surface O_3 depletion, as well as its vertical extent in the Arctic troposphere, was also a focus of the POLARCAT campaigns. Significant inconsistencies exist in tropospheric column retrievals of bromine oxide (BrO) from satellite sensors [e.g., the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY)]. Because these satellite measurements provide some of the few observations available to understand

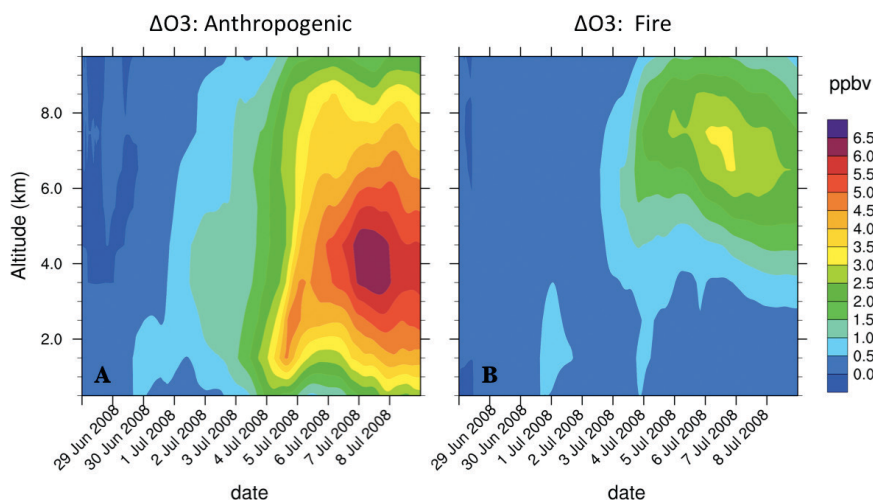


FIG. 11. The average increase in ozone (ΔO_3) in ppbv from WRF-Chem model simulations north of $55^\circ N$ from (left) anthropogenic emissions over North America and (right) boreal fire emissions over Canada. Excess ozone (ΔO_3) was calculated as the average increase in O_3 north of $55^\circ N$ based on model runs with and without anthropogenic and fire emissions. From Thomas et al. (2013).

free tropospheric halogen chemistry in the Arctic, validation of satellite BrO was an important objective (Jacob et al. 2010; Brock et al. 2011). The ARCTAS and ARCPAC campaigns were the first airborne missions to use chemical ionization mass spectrometry (CIMS) to speciate inorganic bromine (Neuman et al. 2010). In situ measurements of active bromine [sum of bromine (Br_2) and hypobromous acid (HOBr)] as well as of BrO were reported in the Arctic free troposphere (Neuman et al. 2010), for the first time, following extensive laboratory investigations (Liao et al. 2012).

POLARCAT clearly established that BrO hotspots in the Arctic observed by satellite cannot routinely be interpreted as boundary layer enhancements. Salawitch et al. (2010) showed satellite observations of BrO hotspots where no evidence of bromine chemistry or O_3 depletion was found in situ. The aircraft also occasionally sampled air with nearly complete O_3 depletion together with enhanced active bromine and soluble bromide that was not in a satellite-detected BrO hotspot. This study established that spatial variations in the stratospheric contribution to the total column measurement of BrO from space must be taken into account in order to estimate the tropospheric residual and indicated that a significant fraction of the tropospheric BrO column might sometimes be in the free troposphere. Choi et al. (2012) confirmed that BrO in the free troposphere often contributed substantially to the tropospheric column. They noted that true boundary layer BrO maxima observed from space reflect active bromine chemistry and O_3 destruction, while air masses with severe O_3 depletion cannot be identified by the satellite sensors because BrO quickly converts to other undetectable inorganic bromine compounds in the absence of O_3 . Koo et al. (2012) combined the aircraft and satellite observations with observations of O_3 and bromine compounds at several surface stations and ozonesondes launched around the Arctic during POLARCAT to examine O_3 depletion events across the region. Their study provided additional evidence that the impact of active bromine chemistry is not restricted to the shallow Arctic boundary layer but extends into the Arctic free troposphere. The sources of Arctic free tropospheric BrO enhancements may not be completely dominated by halogens emitted in the Arctic boundary layer or be of stratospheric origin, as pointed out by Pommier et al. (2012b). Further evidence for halogen chemistry was found during the ICEALOT cruise in the northern North Atlantic, where air masses exposed to sea ice in the high Arctic showed depleted O_3 and perturbed ratios of selected hydrocarbons (Gilman et al. 2010). We

note that many global models do not take tropospheric halogen chemistry into account and its impact on tropospheric O_3 requires further investigation.

CONCLUSIONS AND FUTURE PERSPECTIVES. POLARCAT-IPY provided a wealth of new data on Arctic air pollution. In particular, data were collected on the vertical distributions of aerosol chemical composition and trace gas species throughout the free troposphere and lowermost stratosphere during spring and summer 2008. These data have been used, and are continuing to be used, to evaluate and improve chemistry–climate models, for example, as part of POLMIP and by the Arctic Council Arctic Monitoring and Assessment Program (AMAP) expert group on black carbon and ozone, which is examining the impact of different sources of short-lived pollutants on Arctic climate.

Analysis of POLARCAT data highlighted important contributions from agricultural and boreal fire emissions, as well as anthropogenic emissions from East Asia, to aerosols and trace gases observed over the Arctic in spring and summer 2008. In the free troposphere, fires were the main source of organic aerosols during spring whereas Asian pollution was the main source of sulfate with BC originating from both these sources. While sulfate dominated aerosol mass at Arctic surface sites, organic matter also contributed substantially; the origins of this organic mass warrant further investigation. Although aerosol concentrations were lower in summer in the Arctic free troposphere, enhancements were observed in polluted plumes.

During spring 2008, transport of pollution into the Arctic free troposphere produced high aerosol concentrations in layers above the surface, in particular due to active fires in boreal regions. Analysis of CALIPSO aerosol lidar satellite data, which was extensively validated using POLARCAT aircraft and ground-based data, was used to show that transport of pollution into the Arctic free troposphere was more pronounced in 2008. Surface observations, in contrast, agreed with the long-term seasonal cycle of the chronic Arctic haze aerosol. These observations are consistent with the conceptual model that northern midlatitude and Arctic sources within the cold polar dome contribute most to near-surface Arctic pollution and that a variety of mid- and high-latitude emissions may be advected to the Arctic aloft. Further, these findings imply that Arctic surface measurements are often not representative of the atmosphere aloft and suggest that Arctic composition in the free troposphere is particularly sensitive

to yearly and seasonal variations in transport patterns and fire emissions.

The first direct measurements of Arctic black carbon vertical distributions were collected during POLARCAT and used to estimate dry deposition rates as well as local heating rates in aerosol layers observed in the free troposphere. The impact of these lofted pollution layers, which often contain absorbing and nonabsorbing aerosols from more than one source, on the Arctic radiation budget and their effects on cloud properties, as well as wet and dry depositional fluxes to the Arctic surface, and effects of deposited absorbing aerosol on surface albedo, are still areas of active research. While global aerosol models continue to be improved, it is clear that further attention needs to be paid to improving treatments of wet and dry deposition. In addition, emissions of light-absorbing particles near and within the Arctic, such as BC from shipping and oil/gas extraction, require better quantification since they are likely to increase in the future.

Asian, European, and to a lesser extent North American, anthropogenic pollution and boreal fires were identified as important sources of O_3 precursors in spring/summer 2008 in the lower and midtroposphere. Episodes of cross-Arctic pollution transport were documented in the summer resulting in elevated concentrations of trace gases, such as CO and PAN, in the free troposphere and lower stratosphere. Together with lightning NO_x emissions, these tropospheric sources of O_3 appear to be making a significant contribution to Arctic summertime O_3 in the free troposphere. Direct transpolar transport of East Asian pollution toward Europe may lead to downwind O_3 production but the contribution of this pathway has yet to be fully assessed. Anthropogenic pollution appears to make a larger contribution to O_3 over the Arctic compared to boreal fires. However, evidence exists for O_3 production downwind of fires in the Arctic and further work is required to evaluate the role of PAN as a source of NO_x , particularly during summer.

Airborne in situ measurements of reactive bromine during POLARCAT established that regions of enhanced BrO observed from satellites are not always evidence of bromine activation and loss of O_3 in the boundary layer. Spatial and temporal variations, in both the stratospheric and free-tropospheric BrO columns, can erroneously be interpreted as boundary layer enhancements in BrO. Because satellite sensors have provided the primary means of determining the spatial and temporal distribution of active bromine in the Arctic boundary layer, many uncertainties remain in understanding the scale and magnitude of bromine-catalyzed O_3 loss in the Arctic lower troposphere.

In the future, global warming and economic factors will likely lead to increased exploitation of Arctic resources and increased emissions of SLCPs within the Arctic. Pollutant emissions in the developed and developing world will also change rapidly, with expected continuing declines in North America and Europe and increases in eastern and southern Asia. The POLARCAT experiments have provided improved understanding of the sources, transport, transformation, and sinks for key SLCPs in the Arctic. The observations and their analysis provide valuable constraints for models, which are the primary tools for developing policies to limit future pollution impacts on climate, ecosystems and air quality.

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