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1 Arctic Air Pollution: New Insights From POLARCAT-IPY

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Capsule

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

Abstract

1
2 Given the rapid nature of climate change occurring in the Arctic and the difficulty for climate
3 models to quantitatively reproduce observed changes such as sea ice loss, it is important to
4 improve understanding of the processes leading to climate change in this region, including the
5 role of short-lived climate pollutants such as aerosols and ozone. It has long been known that
6 pollution produced from emissions at mid-latitudes can be transported to the Arctic resulting
7 in a winter/spring aerosol maximum known as Arctic Haze. However, many uncertainties
8 remain about the composition and origin of Arctic pollution throughout the troposphere; for
9 example, many climate-chemistry models fail to reproduce the strong seasonality of aerosol
10 abundance observed at Arctic surface sites, the origin and deposition mechanisms of black
11 carbon (soot) particles that darken the snow and ice surface in the Arctic is poorly understood,
12 and chemical processes controlling the abundance of tropospheric ozone are not well
13 quantified. The International Polar Year (IPY) core project POLARCAT (Polar Study using
14 Aircraft, Remote Sensing, Surface Measurements and Models, Climate, Chemistry, Aerosols
15 and Transport) had the goal to improve understanding of the origins of pollutants transported
16 to the Arctic, to detail the chemical composition, optical properties, and climate forcing
17 potential of Arctic aerosols, to evaluate the processes governing tropospheric ozone, and
18 quantify the role of boreal forest fires. This article provides a review of the many results now
19 available based on analysis of data collected during the POLARCAT aircraft, ship and
20 ground-based field campaigns in spring and summer 2008. We highlight major findings and
21 discuss areas requiring further investigation.

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1 **1. Introduction**

2 In the face of rapid climate change in the Arctic (increasing temperatures, earlier onset of
3 spring snow melt, sea ice loss, etc.), it is important to improve our knowledge about processes
4 driving these changes. In general, climate models are able to reproduce enhanced warming in
5 the Arctic, the so-called Arctic amplification. However, discrepancies are apparent between
6 observations and global climate model predictions of, for example, Arctic summer sea-ice,
7 resulting in significant differences between model-based and extrapolated estimates for the
8 complete disappearance of the summer ice (Wang and Overland, 2013). Whilst increases in
9 carbon dioxide (CO₂), and associated atmosphere-ice-ocean feedbacks, are major contributing
10 factors, short-lived (with respect to chemical lifetime) climate forcers, such as absorbing
11 (heating) aerosols like black carbon (BC), and trace gases ozone (O₃) and methane, are also
12 likely to be playing an important role (Quinn et al., 2008). Short-lived climate pollutants
13 (SLCPs) (BC, O₃) can impact Arctic warming as a result of (i) direct warming either locally
14 in the Arctic or remotely via heat transport resulting from SLCP-induced warming at lower
15 latitudes (e.g. Shindell, 2007), (ii) decreased surface albedo and subsequent surface warming
16 due to deposition of light-absorbing aerosols (e.g. BC) on snow/ice surfaces (e.g. Hansen and
17 Nazarenko, 2004; Flanner et al., 2007), or (iii) through indirect aerosol-cloud feedbacks such
18 as increases in infra-red emissivity to the surface in winter/spring from anthropogenic
19 aerosols (Garrett and Zhao, 2006; Lubin and Vogelmann, 2006).

20
21 It is also important to consider that aerosols contain a significant fraction of scattering
22 aerosols like sulfate (or organics), which cool the atmosphere, and that also have natural
23 sources, making the diagnosis and attribution of direct and indirect radiative effects a complex
24 task. In fact, decreasing trends in mid-latitude emissions of sulphur dioxide, resulting in
25 declining sulfate concentrations in the Arctic (Quinn et al., 2009), may have led to warming in
26 the Arctic. Increasing trends of BC, as well as O₃ precursor emissions, over Asia are predicted

1 to have contributed as much as two-thirds of observed Arctic warming (e.g. Quinn et al.,
2 2008; Shindell and Faluvegi, 2009). This is despite recent negative trends in observed BC at
3 surface sites in the Arctic due to emission reductions in Europe and the Former Soviet Union
4 (FSU) and points to the potentially important role of BC transport from Asia in the upper
5 troposphere (Sharma et al., 2013) as well as mid-latitude warming resulting in heat transport
6 to the Arctic. At the same time, significant discrepancies between simulated seasonal cycles
7 of trace gases and aerosols and surface observations in the Arctic point to gaps in our
8 knowledge about pollution origins and processing during long-range transport to the Arctic
9 (e.g. Shindell et al. (2008)). Deficiencies in climate model treatments of aerosols and clouds,
10 as well as in the Arctic radiation budget, have been recently highlighted as a possible reason
11 for poor climate model performance in the Arctic (e.g. Wang and Overland (2013)).

12
13 The study of Arctic air pollution is not a new topic. As discussed by Garrett and
14 Verzella (2008), the presence of some kind of visibility-reducing haze in the Arctic was noted
15 by early explorers in the late 19th century, and discussed, in particular, by Nordenskiöld
16 (1883). It was also observed later by pilots in the 1950s (Mitchell, 1957). It was not until the
17 1970s that it was suggested that this pollution or “Arctic Haze” originated from anthropogenic
18 emissions in northern mid-latitudes (Rahn et al., 1977), and Eurasia (defined in this paper as
19 Europe and FSU including Russia) in particular (Barrie et al., 1981). Long-term records of
20 surface observations in the Arctic clearly show an increase in aerosol abundance in winter and
21 early spring every year (e.g. Shaw, 1975; Sharma et al., 2004). This springtime peak in total
22 aerosol mass concentration is caused primarily by increases in submicrometer particles (or
23 fine mode aerosols) composed of non sea-salt (nss) sulfate and sea salt with smaller amounts
24 of ammonium, nitrate, dust, trace elements of combustion, and a residual likely composed of
25 particulate organic matter (e.g. Quinn et al. (2002)). Arctic Haze also contains elevated levels
26 of gas-phase O₃ precursors such as volatile organic compounds (VOCs) and carbon monoxide

1 (CO), as well as nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) and the nitrogen reservoir peroxy-acetyl
2 nitrate (PAN), in particular (e.g. Hov et al., 1989; Beine et al., 1996).

3
4 Previous aircraft campaigns in the 1980s (Schnell, 1984; Radke et al., 1984; Leitch et
5 al., 1989; Brock et al., 1990) and the Tropospheric Ozone Production about the Spring
6 Equinox (TOPSE) experiment in 2000 (Atlas et al., 2003) provided some of the first
7 comprehensive information about the vertical structure of haze layers, finding them to be
8 highly variable both vertically (tens of meters to 1 km thick) and horizontally (20 – 200 km).
9 These measurements suggested that the seasonal maximum in aerosols in the free troposphere
10 occurs later than at the surface (Scheuer et al., 2003). Despite these observations, the extent to
11 which aerosol haze layers aloft are associated with the seasonal Arctic Haze phenomenon at
12 the surface remains an open question. In the summer, surface observations show a clear
13 minimum in aerosol concentrations due to more efficient washout and less efficient transport
14 to the Arctic at this time of year (e.g. Sharma et al., 2004; Garrett et al., 2010) although
15 certain natural aerosols, such as those produced from oceanic emissions, peak in the summer
16 months (Ferek et al., 1995; Quinn et al., 2002).

17
18 Observations of CO at sea-level sites also show a summer minimum and winter-early
19 spring maximum due to more active summertime photochemical destruction of CO (e.g.
20 Shindell et al. (2008)). Tropospheric O_3 is influenced by depletion events in the Arctic; the
21 complex halogen chemistry found over sea-ice can lead to very low or even near-zero O_3
22 concentrations in the spring months in the surface layer (e.g. Barrie et al. (1988)). Re-
23 emission of deposited nitrate as NO_x may contribute to increasing or sustaining O_3 at the
24 surface (Honrath et al., 1999; Dibb et al., 2002). Summertime pollution layers, originating
25 from boreal forest fires, have also been observed in the free troposphere as part of the airborne
26 Arctic Boundary Layer Expeditions (ABLE-3) in July-August 1988 and 1990 (Wofsy et al.,

1 1992). Ozone production in these summertime forest fire plumes was estimated to be
2 negligible (Mauzerall et al., 1996) but the contribution from this source relative to
3 anthropogenic emissions remains uncertain. During the TOPSE winter-spring campaign,
4 photochemical O₃ production was diagnosed to dominate the O₃ budget in the Arctic
5 troposphere with only a rather small contribution (20%) from the stratosphere (Emmons et al.,
6 2003; Browell et al., 2003).

7

8 **2. POLARCAT-IPY: objectives and main activities**

9 Although airborne campaigns prior to 2008 provided very useful first data about the
10 composition, and possible origin of pollution in the Arctic, many scientific questions
11 remained (Law and Stohl, 2007), and major advances in airborne measurement techniques
12 and modeling capabilities had occurred. These factors, coupled with the arrival of several key
13 satellite instruments capable of making aerosol and trace gas measurements in the Arctic (e.g.
14 Aqua Atmospheric Infrared Sounder (AIRS), Infrared Atmospheric Sounding Interferometer
15 (IASI), Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO)), and
16 the announcement of an International Polar Year (IPY) in 2007/08, motivated the atmospheric
17 chemistry community to design an international project, POLARCAT, aimed at further
18 improving our understanding of Arctic pollution and impacts on climate.

19

20 POLARCAT was recognised as an IPY activity, co-sponsored by the International
21 Geosphere Biosphere Programme core projects International Global Atmospheric Chemistry
22 (IGAC) and Integrated Land Ecosystem – Atmosphere Processes Study (iLEAPS) and the
23 World Climate Research Programme (WCRP) core project Stratosphere-
24 troposphere Processes And their Role in Climate (SPARC). The main scientific objectives of
25 POLARCAT, which continues as a task within IGAC, are described in the White Paper
26 available at <http://www.igacproject.org/CurrentActivities>. These objectives are shown in

1 Table 1, grouped according to the main topics discussed in this review, namely 1) pollution
2 transport and origins, 2) aerosol composition and processes, and 3) trace gas chemical
3 composition and processes. To investigate these issues, POLARCAT organised several
4 airborne campaigns at high northern latitudes during the spring and summer of 2008, focusing
5 primarily on the Arctic free troposphere (up to 9km). Six instrumented aircraft from the
6 United States, Germany, France, and Russia participated, flying from bases in Alaska,
7 Canada, Greenland, Sweden, and Russia, and sampling across the entire Arctic. Figure 1
8 shows the flight tracks of the main aircraft campaigns, and a ship cruise which took place, as
9 well as the locations of ground based sites referred to in this paper. Table 2 provides further
10 details about the different POLARCAT projects, the platforms, including altitude ranges
11 covered by the aircraft measurements, and references providing detailed information about the
12 aircraft and ship deployments and payloads. Information about complementary campaigns
13 and measurement programs is also given as well as details of all project acronyms. We also
14 note other significant research programs, not directly affiliated to POLARCAT, the Indirect
15 and Semi-Direct Aerosol Campaign (ISDAC) investigating properties of Arctic stratus clouds
16 and aerosol-cloud interactions in spring 2008 (McFarquhar et al., 2011), and Arctic Summer
17 Cloud Ocean Study (ASCOS) (Tjernström et al., 2014) studying low-level Arctic clouds and
18 aerosol-cloud-ocean interactions. In this review, it is not possible to cover all issues listed in
19 Table 2, such as aerosol-cloud interactions, and to which POLARCAT also contributed.

20

21 Briefly, gas-phase measurements on board the POLARCAT aircraft included O₃ and its
22 precursors (NO_x, CO, VOCs), intermediate VOC oxidation products (e.g. carbonyls,
23 peroxides), tracers of combustion (CO), industrial emissions (sulfur dioxide, SO₂), and
24 biomass burning (acetonitrile, CH₃CN), and greenhouse gases. Detailed data were also
25 collected on aerosol chemical composition, and their physical and optical properties.
26 Additional data were provided by observations from satellites and ozonesondes (e.g. Pommier

1 et al., 2010, 2012; Tarasick et al., 2010) as well as by enhanced observations at long-term
2 surface measurement sites (see Table 2). Studies using regional and global models were also a
3 key part of POLARCAT ranging from forecasts for flight planning to post-campaign data
4 analysis. Overall, analysis of POLARCAT data has so far resulted in more than 80 published
5 papers, many of which appear in a journal special issue ([http://www.atmos-chem-](http://www.atmos-chem-phys.net/special_issue182.html)
6 [phys.net/special_issue182.html](http://www.atmos-chem-phys.net/special_issue182.html)). In this review, we highlight some of the key results from
7 POLARCAT. The discussion is focused around transport and origins of Arctic air pollution
8 (Section 3), Arctic aerosols (Section 4), and Arctic gas-phase chemical composition (Section
9 5). Conclusions and future perspectives are discussed in Section 6.

10

11 **3. Arctic air pollution: transport and origins**

12 At the onset of the POLARCAT campaigns, while established concepts held that Arctic
13 Haze originated from long-range transport of Eurasian pollution coupled to inefficient
14 pollutant removal, there was increasing evidence to support some paradigm shifts in pollutant
15 sources and transport processes impacting the Arctic troposphere. Most significantly, the
16 potential for boreal forest fires (Stohl, 2006; Stohl et al., 2006) and south Asian emissions
17 (Koch and Hansen, 2005) to be efficiently transported to the Arctic troposphere was
18 highlighted, although controversy remained regarding the scale of these contributions. The
19 main transport pathways are illustrated in Figure 2.

20

21 The POLARCAT campaigns provided an unprecedented ‘snap shot’ of the state of
22 Arctic composition in spring and summer 2008. In terms of meteorological conditions,
23 Fuelberg et al. (2010) noted that mid-latitude cyclones were more frequent, and followed a
24 more northerly course than usual, over eastern Asia and the northern Pacific but were less
25 common over the North Atlantic during spring. Frequent cyclone activity also occurred over
26 the Pacific during summer 2008. At the same time, the North Atlantic Oscillation (NAO)

1 transitioned toward a negative state in spring and remained so for the summer campaigns.
2 Such a negative NAO state is associated with reduced pollution transport toward the Arctic
3 (Burkhart et al., 2006), especially from Europe, compared to the mean (Eckhardt et al., 2003).
4 Examination of AIRS satellite CO anomalies over the Arctic also suggested that transport of
5 pollution to the Arctic was hindered in spring 2008 because of negative ENSO (El Nino
6 Southern Oscillation) conditions (Fisher et al., 2010).

7
8 Nevertheless, despite large-scale meteorological patterns that did not favor transport
9 from mid-latitudes, a surprising finding was the strong influence of Eurasian fire emissions, in
10 particular from agricultural fires, during the spring campaigns (Warneke et al., 2009, 2010;
11 Brock et al., 2011; McNaughton et al., 2011). In 2008, negative precipitation anomalies
12 contributing to particularly large fires over Siberia were not driven by ENSO, even though in
13 other years ENSO has been shown to be strongly linked to boreal fire activity and emissions
14 of trace gases like CO (Monks et al., 2012). In addition, these emissions occurred further east
15 where the NAO has a weaker influence compared to high-latitude anthropogenic emissions in
16 Europe. In addition, emissions of total carbon from agricultural fires in western Russia and
17 Kazakhstan were twice as high (29 TgC) in 2008 as the 1997-2011 average while boreal
18 wildfire emissions were 21% higher than the mean (214 TgC in 2008 compared to 180 ± 111
19 TgC yr⁻¹ over 1997-2011) (van der Werf et al., 2010). Plumes originating from anthropogenic
20 sources in eastern Asia and Europe were also sampled over the Arctic in spring 2008 (Singh
21 et al., 2010; Adam de Villiers, 2010). Transport by warm conveyor belts associated with
22 frontal systems was an important mechanism for the transport of smoke and Asian pollution
23 into the free troposphere whereas transport of pollution at low altitudes was primarily from
24 Eurasia. During spring 2008, this resulted in higher aerosol loadings in the free troposphere as
25 shown in Figure 3 by satellite aerosol data from CALIPSO. This springtime feature was more
26 pronounced in 2008 compared to other years (2006-2012) (Di Pierro et al., 2013). Aerosol

1 loadings in the lower troposphere were lower in spring having declined from the winter-early
2 spring Arctic Haze maximum observed every year. However, we note that CALIPSO data,
3 based on validation against POLARCAT data, may underestimate aerosol loadings by 20-
4 30% below 4 km due to the presence of clouds, and overestimate contributions from smoke
5 and pollution/dust sources (Di Pierro et al., 2013; Ancellet et al., 2013; Burton et al., 2013).
6 During summer 2008, the central Arctic was very clean (as shown in Fig. 3) due to more
7 efficient washout, and due to the Arctic front retreating to high latitudes. Nevertheless, small
8 enhancements in aerosols were observed in polluted air masses transported to the Arctic in the
9 free troposphere from Asian/North American anthropogenic or boreal fire emission regions
10 (Schmale et al., 2011).

11
12 Source attribution studies often use CO as a tracer of pollutant origins due to its rather
13 long chemical lifetime of 1-3 months. Total CO columns observed by the IASI satellite
14 instrument provided an Arctic-wide view of CO distributions in spring and summer 2008 as
15 illustrated in Figure 1. Higher CO was prevalent throughout the Arctic in spring compared to
16 summer, especially downwind of Asia and boreal fire regions. In summer, whilst lower CO
17 was observed due to enhanced photochemical destruction at this time of year, there is a
18 suggestion of higher CO over the western Arctic. Global model studies, run with tagged CO
19 for different sources, suggest that Asian anthropogenic emissions dominated the CO budget
20 (for CO concentrations above the baseline due to methane oxidation) throughout the depth of
21 the troposphere in spring and summer 2008 (Fisher et al., 2010; Bian et al., 2013). An
22 example is shown from Bian et al. (2013) in Figure 4. Anthropogenic CO emissions from
23 Europe were also important in the lower troposphere but results differ regarding this
24 contribution. Also, in contrast to a multi-model study, for the year 2001, (Shindell et al.,
25 2008), Bian et al. (2013) did not find a significant contribution from North American
26 emissions in 2008. Bian et al. (2013) also found that boreal fires were a significant source of

1 CO. This illustrates that CO source attribution depends on inter-annual variations in emissions
2 and transport pathways. It is also important to note that, in general, global models
3 underestimate CO in the Arctic (and Northern Hemisphere) (Shindell et al., 2008). Such
4 discrepancies between observed and modeled CO concentrations, as well as other species like
5 O₃ and PAN, motivated the POLARCAT Model Inter-comparison Project (POLMIP) (Monks
6 et al., 2014; Emmons et al., 2014; Arnold et al., 2014) where several models were run with
7 the same 2008 emissions. Analysis of results show that, in this case, variability between
8 modeled Arctic CO is driven mainly by differences in oxidative (OH) chemistry (Monks et
9 al., 2014). Differences in simulated vertical export efficiency over mid-latitude emission
10 regions may also play an important role in determining large-scale pollutant transport to the
11 Arctic.

12
13 The transport of smoke from wildfires in Siberia and pollution from eastern Asian
14 anthropogenic sources was particularly well documented during the POLARCAT summer
15 campaigns. This transport may occur frequently enough to affect average pollutant
16 distributions in the summer (Hirdman et al., 2010a), as illustrated by the enhanced IASI CO
17 over the western Arctic mentioned earlier (see Fig. 1). Pollution was lifted into the Arctic by
18 frontal systems and transported directly across the North Pole in the mid- and upper
19 troposphere (Harrigan et al., 2011). In one particularly notable case, pollution was transported
20 by this mechanism from eastern Asia into the lowermost stratosphere where it was observed
21 over Greenland (Roiger et al., 2011a). When the aircraft sampled the plume ~1 km above the
22 tropopause, the polluted air from East Asia was mixed with stratospheric air (see Fig. 5).
23 Detailed comparison of Eulerian and Lagrangian model simulations with aircraft data and CO
24 satellite retrievals demonstrated that the models were able to simulate this complex cross-
25 polar transport episode rather accurately (Sodemann et al., 2011). Whilst transport of air from
26 the troposphere to the stratosphere has been studied previously (e.g. Sprenger et al. (2003);

1 Stohl et al. (2003)), the POLARCAT campaigns provided some of the first detailed in-situ
2 observations of cross-polar pollution transport into the lowermost stratosphere. These findings
3 suggest that the Arctic lower stratosphere (and upper troposphere) may hold substantial
4 amounts of pollution, primarily from low-latitude Asia, which could eventually descend into
5 the Arctic and mid-latitude troposphere. In fact, Trickl et al. (2014) show a downward trend
6 (since 1990) in observed CO concentrations at Zugspitze mountain (11.0E, 47.4N), Germany,
7 with the exception of air masses originating from the high-latitude lower stratosphere.
8 Increasing CO concentrations in these air masses can best be explained by increasing CO
9 emissions in East Asia and transport events similar to those observed by Roiger et al. (2011a).

10

11 Sources of pollutants near the surface are very different from those sampled by the
12 aircraft in the free troposphere. Statistical analysis of long-term monitoring data from the
13 stations Alert (82.5N, 62.3W), Barrow (71.3N, 156.6W) and Zeppelin (78.9N, 11.9E) (Fig. 1)
14 confirmed earlier results (e.g. Sirios and Barrie, 1999; Stohl, 2006) that the main source of
15 BC aerosols near the surface in the Arctic is northern Eurasia in winter/spring, and that
16 smelting activities in Norilsk, northern Russia, are a significant source of sulfate in the eastern
17 Arctic (Hirdman et al., 2010a). Only during summer is there clear evidence of influence by
18 boreal forest fires at surface sites. In addition, a recent study (Stohl et al., 2013) suggests that
19 emissions from flaring of natural gas by the oil industry in northern Russia may be an
20 important missing winter-spring source of Arctic BC. Trend analysis by Hirdman et al.
21 (2010b) also confirmed earlier studies showing decreases in observed equivalent BC and
22 sulfate at surface locations since the late 1980s (e.g. Sharma et al., 2004; Quinn et al., 2009).
23 Hirdman et al. (2010b) showed that decreases in northern Eurasian emissions appear to be the
24 main cause, and that changes in atmospheric transport to the Arctic were not important. This
25 finding is supported by the global model study of Sharma et al. (2013) although they point out
26 that while BC concentrations may be decreasing at the surface, it is possible that they may be

1 increasing aloft due to increasing Asian emissions. We note that POLARCAT aircraft
2 measurements only showed pollution layers present above the sea-ice inversion layer and not
3 reaching the (sea-ice) surface during spring 2008 (Brock et al., 2011).

4

5 **4. Arctic aerosols**

6 Advances in instrumentation made the characterization of the vertical distribution of
7 aerosol chemical composition and optical properties a particular focus during POLARCAT.
8 This is illustrated in Figure 6 which shows an example of the vertical distribution of aerosols
9 collected during the ARCPAC spring campaign north of Alaska on 18 April 2008 (Brock et
10 al., 2011). Of particular note are enhanced concentrations of organic aerosols (OA) in the mid
11 and upper troposphere attributed primarily to boreal fires over Eurasia based on enhancements
12 in tracers like CH_3CN (e.g. Warneke et al., 2009). Enhanced sulfate (non sea-salt component)
13 was also observed and attributed to anthropogenic origins. Fisher et al. (2011) diagnosed that,
14 in free troposphere, sulfate from East Asian sources exceeded that from European sources, in
15 contrast to previous studies suggesting that Europe was the main source (Shindell et al.,
16 2008). Overall, OA and sulfate dominated the aerosol mass in the free troposphere during
17 spring 2008. Figure 6 also shows enhanced BC in the free troposphere together with enhanced
18 sulfate and OA, a feature also observed during ARCTAS (Wang et al., 2011). Enhanced BC
19 was attributed to Eurasian fires (Warneke et al., 2010) and Asian anthropogenic emissions
20 (Wang et al., 2011) in spring 2008 with differences likely due to different campaign sampling
21 strategies. Since sulfate enhancements were primarily of anthropogenic origin, positive
22 correlations with OA enhancements suggest mixing of Asian anthropogenic and Siberian
23 boreal fire plumes during transport to the Arctic (Wang et al., 2011). There is also evidence
24 for plumes of mixed anthropogenic (Asian) and Russian fire origin containing soot-like
25 particles in air masses transported from Asia to the north of Scandinavia (Adam de Villiers et
26 al. 2010; Quennehen et al., 2012).

1
2 During POLARCAT, BC mass concentrations were directly measured for the first time using
3 airborne single particle soot photometer (SP2), with an estimated uncertainty of +/- 40%, in
4 the Arctic free troposphere with the highest concentrations found aloft,(Jacob et al., 2010;
5 Warneke et al. 2009). Overall, BC concentrations observed during POLARCAT in spring
6 2008 were higher than direct measurements reported during PAM-ARCMIP (Pan-Arctic
7 Measurements and Arctic Regional Climate Model Inter-comparison Project) in spring 2009
8 when cleaner air masses were sampled (Stone et al., 2010). During summer 2008, aerosol
9 concentrations in the free troposphere were generally 2-3 times lower than in spring and BC
10 concentrations were ten times lower (based on all available data) (Matsui et al., 2011).
11 Organics, and to a lesser extent sulfate, were rather pervasive during the summer
12 POLARCAT-France campaign over Greenland, albeit at low concentrations, and the organic
13 fraction of the submicrometer aerosol increased during strong gas-phase pollution episodes
14 observed between 4-6 km (Schmale et al., 2011). Sampled plumes in July 2008 originated
15 primarily from Canadian fires and contained higher organic fractions. Mixed Asian fossil fuel
16 and Siberian fire plumes were also sampled exhibiting higher sulfate fractions (Schmale et al.,
17 2011).

18
19 New information was also collected about aerosol composition at or near to the surface,
20 particularly in spring 2008. Aerosol mass was dominated by sulfate at several Arctic sites at
21 this time of year and as noted earlier, attributed to Eurasian emissions, in agreement with
22 previous studies (Fisher et al., 2011; Hirdman et al., 2010a). This was also shown to be the
23 case during the spring 2008 ICEALOT ship cruise in the eastern Arctic where Frossard et al.
24 (2011) attributed air masses with elevated organosulfates and sulfate to coal burning in
25 eastern Europe. Results from this study are illustrated in Figure 7 which shows aerosol
26 composition in air masses with Arctic and European origins. Whilst sulfate clearly dominated

1 the eastern Arctic during spring 2008, Arctic air masses had a larger fraction (on average
2 approximately 40%) of marine aerosols compared to European air masses which had a higher
3 fraction (70%) from combustion sources. In both air mass types, the proportion of submicron
4 organic mass was lower compared to 2 years of spring data from Barrow, Alaska (also shown
5 in Fig. 7). Shaw et al. (2010) examined one year of measurements at Barrow (March 2008 –
6 March 2009) and found higher particulate organic matter (POM) concentrations in
7 winter/spring compared to summer. In winter, analysis of the organic component indicated a
8 higher oceanic contribution (i.e. hydroxyl groups and POM correlated with inorganic
9 seawater components), while in spring, anthropogenic sources dominated with alkane and
10 carboxylic acid functional groups correlated with elemental tracers of Asian, shipping and
11 boreal fire emissions. Overall, organic fractions made up a significant fraction of the total
12 aerosol mass, for example, reaching up to 35% at Barrow in spring (Frossard et al. 2011;
13 Shaw et al., 2010). These results highlight the complex nature of organic aerosols which can
14 sometimes make up a substantial fraction of the total aerosol mass, and which are still poorly
15 characterized in models (e.g. Lapina et al. (2011)).

16
17 The findings discussed so far suggest that our picture of Arctic aerosol sources,
18 distribution, and composition is more complex than previously thought. Brock et al. (2011)
19 used long-term ground-based measurements, and the short-term aircraft observations provided
20 by ARCPAC (in the western Arctic), to conclude that Arctic aerosol pollution has both
21 chronic and episodic components. Aerosol light scattering, submicrometer sulfate mass
22 concentration, and particle number concentrations measured at Barrow during winter and
23 early spring have limited temporal variability compared to episodic dense aerosol layers
24 observed aloft. Brock et al. (2011) noted that this chronic aerosol haze, as measured at the
25 surface, originates primarily from Eurasian sources and increases in the winter-spring
26 boundary layer in the Arctic due to continued emissions into the polar dome, limited vertical

1 mixing, and inefficient removal (Fig. 2). The haze dissipates in summer with the poleward
2 retreat of the arctic front and increased Arctic precipitation (Garrett et al., 2010). Although
3 there have been occasional observations of dense haze layers at the surface (e.g. Stohl et al.,
4 2006), these do not dominate the seasonal cycle of average aerosol values (Brock et al.,
5 2011).

6
7 Aloft in the free troposphere, long-range transport of pollution from mid-latitude
8 sources, often associated with lifting in mid-latitude warm conveyor belts, can lead to
9 episodically enhanced aerosol loadings. Brock et al. (2011) suggested that since such
10 intercontinental long-range transport is not unique to the Arctic, it can be considered a
11 separate phenomenon from the chronic pollution reported at surface sites in the Arctic and
12 commonly referred to as Arctic Haze. Indeed, the aerosol composition measured in
13 background air outside of discrete aerosol layers (Fig. 6) was very similar to the average
14 composition of non-refractory components reported in springtime at the surface at Barrow
15 (Brock et al., 2011); namely, anthropogenic sulfate made up the largest mass fraction with a
16 smaller contribution of organics resulting from biomass or biofuel combustion. Jacob et al.
17 (2010) also reported anthropogenic aerosol dominating below 2 km. In contrast, in the free
18 troposphere, fire emissions were an important contributor to aerosol layers between 2 and 7
19 km in spring 2008, particularly for organic and BC aerosols, and were superimposed on or
20 mixed with, aerosols of anthropogenic origin (Wang et al., 2011; Adam de Villiers et al.,
21 2010). Taken together, these observations are consistent with a background of a largely
22 anthropogenic aerosol that results from the emission of pollution into the polar dome (Fig. 2),
23 and that has been identified as ‘Arctic Haze’ by surface observations. Superimposed on the
24 background Arctic Haze are pollution and biomass burning plumes from midlatitude sources
25 due to episodic transport events to the Arctic free troposphere during both spring and summer.
26 This picture of a background pollution aerosol perturbed by periodic transport events aloft,

1 which was particularly evident in spring 2008, is consistent with the analysis of sources and
2 transport described in Section 3, and with Stohl (2006) who demonstrated that seasonal
3 differences in transport processes could potentially explain much of the seasonality and
4 vertical distribution of air pollution in the Arctic.

5
6 Another research focus during POLARCAT was improving understanding about the
7 impacts of BC-containing aerosols on the radiative budget in the Arctic including the
8 deposition of BC to snow and ice and its impact on surface albedo. Wang et al. (2011) used a
9 global model to estimate BC deposition in the Arctic during the period January to May 2008
10 and diagnosed that wet deposition accounted for 85-91% of total BC deposition during this
11 period. This is in agreement with certain previous studies (e.g. Bourgeois and Bey, 2011) but
12 contrasts with other studies estimating that dry deposition dominates in the Arctic winter (e.g.
13 Sharma et al., 2013). While dry depositional fluxes calculated by Wang et al. (2011) were
14 only 15% and 9% of total deposition in winter and spring, respectively, they were similar in
15 magnitude to those derived from independent estimates based on analysis of springtime
16 POLARCAT aircraft data in the range of $1500 \text{ ng m}^{-2} \text{ d}^{-1}$ (Spackman et al., 2010). It had been
17 suggested previously that dry deposition may occur preferentially over regions of open leads
18 (linear cracks of open water within the sea-ice) as a result of convectively driven turbulence
19 due to temperature differences between open leads and surface-layer air (Strunin et al., 1997).
20 During several low altitude ARCPAC flights, a positive vertical gradient in BC mass was
21 indeed observed in the vicinity of open leads providing evidence for dry deposition of BC
22 from low altitude, well-aged air masses in the Arctic (Spackman et al., 2010). Wang et al.
23 (2011) estimated that anthropogenic emissions over Eurasia made the largest contribution to
24 total BC deposition in the Arctic winter (January to March) whereas Russian fires were the
25 main source in spring 2008 (April-May) (see Figure 8). They also calculated a 0.4% winter

1 and 0.8% spring decrease in snow albedo averaged over the Arctic corresponding to a surface
2 radiative forcing from deposited BC of 0.1 W m^{-2} in winter and 1.7 W m^{-2} in spring.

3
4 Evaluation of global models using POLARCAT airborne observations has led to some
5 improvements in modeled vertical aerosol distributions using adjusted wet and dry deposition
6 schemes (Wang et al., 2011; Fisher et al., 2011). However, many global aerosol models,
7 including those used to analyze POLARCAT data, still struggle with reproducing observed
8 BC concentrations in both snow and air. This may be partly due to deficiencies in emissions
9 (Stohl et al., 2013) but treatment of both wet and dry deposition in models is a challenge.
10 Even if, for example, improved parameterizations of wet scavenging leads to better agreement
11 with data in some cases (e.g. Browse et al. (2011)), differences in modeled estimates of the
12 relative contributions of wet versus dry deposition requires further attention (Bourgeois and
13 Bey, 2011; Liu et al., 2011; Wang et al., 2011; Sharma et al., 2013, Lee et al., 2013). Analysis
14 of POLARCAT summertime data suggest large variability in the efficiency of aerosol wet
15 scavenging, with an average aerosol lifetime of 7-11 days (Schmale et al., 2011). Additional
16 observational constraints are needed to improve our knowledge about wet and dry deposition
17 mechanisms and rates, and their treatment in models.

18
19 BC-containing particles can also directly perturb the energy balance in the Arctic by
20 absorbing solar radiation and heating the surrounding air. Using aircraft observations of light
21 scattering and extinction, and a radiative transfer model, Brock et al., (2011) calculated the
22 radiative forcing for measurements made during ARCPAC in the springtime Alaskan Arctic.
23 Instantaneous, maximum noontime heating rates were found to be higher in the dense smoke
24 layers aloft (up to $\sim 0.6 \text{ K day}^{-1}$) than in more diffuse background haze layers ($\sim 0.1 \text{ K day}^{-1}$).
25 The presence of both background diffuse and dense haze layers resulted in a net cooling at the
26 surface and a heating of the atmospheric column. Dense smoke from springtime biomass

1 burning is a potentially large, but highly variable, source of aerosol to the Arctic during the
2 snowmelt season. The net impact of smoke, in terms of cooling the surface through absorption
3 in the atmosphere or warming the surface through deposition of BC on snow and ice and
4 downward transport of heat from the atmosphere, has yet to be fully resolved (Bond et al.,
5 2013) and represents a critical area for future research.

6

7 **5. Arctic gas-phase chemical composition**

8 POLARCAT airborne campaigns provided new information about the vertical
9 distribution of Arctic chemical composition in the troposphere and lowermost stratosphere
10 during spring and summer 2008. Again, advances in measurement techniques allowed
11 improved assessment of vertical distributions of many trace species including reactive
12 nitrogen (NO_y) components (NO_x , PAN, HNO_3 , organic nitrates), oxidized volatile organic
13 compounds (OVOCs) (e.g. acetone, formaldehyde (HCHO)), free radicals (e.g. hydroxyl
14 (OH)), and tracers of pollution sources (e.g. acetonitrile, CO) (Singh et al., 2010; Neuman et
15 al., 2010; Olson et al., 2012; Wespes et al., 2012; Thomas et al., 2013). A number of these
16 species had not been measured in the Arctic free troposphere prior to the POLARCAT
17 missions, especially during the summer.

18

19 Figure 9 shows profiles of selected trace gases (CO, O_3 , PAN and NO) in the spring and
20 summer from various airborne campaigns. Profiles of CO reflect seasonal variations in
21 photochemical destruction, as noted earlier, as well as influences from different sources.
22 During spring 2008, ARCPAC sampled enhanced concentrations of CO and PAN in aged
23 Siberian fire plumes (Fig. 9 top panels). In summer 2008, ARCTAS-B made flights close to
24 boreal fires in Canada and measured significant enhancements in CO, NO and PAN (as well
25 as other trace species and aerosols) in the lower troposphere (Fig. 9 bottom panels).
26 POLARCAT-GRACE and POLARCAT-France sampled some of these fire plumes

1 downwind over Greenland as well as anthropogenic plumes from North America (Thomas et
2 al., 2013) and pollution from Asia/Siberia resulting in elevated CO and PAN concentrations
3 between 4-9 km (Roiger et al., 2011b). Enhanced PAN downwind of source regions in spring
4 (Fig. 9d) and summer (Fig 9h) can be explained by fast conversion of NO_x to PAN near
5 emission regions, followed by rapid lofting into the colder free troposphere, where PAN is
6 thermally stable (Roiger et al., 2011b). Boreal fires sampled over Canada in summer 2008
7 were a particularly important source of PAN (Alvarado et al., 2010). NO_x released from PAN
8 decomposition, when air masses descend to lower warmer altitudes, can produce O₃. Overall,
9 PAN was the dominant nitrogen species accounting for 50% of NO_y in the spring and 70% in
10 summer (Liang et al., 2011). The measured NO_y budget was largely balanced with PANs,
11 alkyl nitrates, NO_x and HNO₃ accounting for more than 90% of total measured NO_y (Singh et
12 al., 2010). Although little NO was found in the mid-troposphere, mixing ratios were higher
13 above 8 km due to stratospheric and lightning influences (in summer) (Liang et al., 2011).
14 Observed O₃ mixing ratios show a compact vertical distribution, particularly in summer.
15 Higher O₃ concentrations in the upper troposphere are attributable to greater stratospheric
16 influence, particularly in the spring.

17

18 Active local fires over Canada and Siberia were sampled in summer 2008, providing new
19 information about fire emissions from these important sources (Singh et al., 2010; Paris et al.,
20 2009). The fires over Canada were not only sources of PAN and other nitrogen species (e.g.
21 NO, NO₂, CH₃CN), but were also an important source of CO, VOCs (e.g. alkanes, alkenes,
22 aromatics), and OVOCs (e.g. HCHO, acetone, methanol) (Simpson et al., 2011).
23 Monoterpenes, usually considered to originate from live biogenic emissions, were reported,
24 for the first time, to have large enhancements in plumes from boreal fires – an interesting
25 finding because monoterpenes are important precursors for secondary organic aerosols.
26 Surprisingly, many emission ratios measured in smoke from boreal and mid-latitude

1 Californian forest fires were largely indistinguishable between the two climate zones (Singh
2 et al., 2010).

3
4 The large body of data collected on Arctic chemical composition are being used to
5 evaluate a range of models. This includes measurements of OH, which is the main
6 tropospheric oxidant for CO, VOCs and methane. For example, vertical distributions of HO_x
7 (OH plus hydroperoxyl (HO₂)) radicals, together with profiles of OVOCs, which can act as
8 radical sources or sinks, were used to constrain a photochemical box model, and showed that
9 springtime HO₂ is overpredicted despite modeled OH agreeing reasonably well with the data
10 (Olson et al., 2012). During the summer, Olson et al. (2012) found opposite discrepancies
11 between modeled and observed OH (underpredicted) and HO₂ (overpredicted), pointing to
12 significant difficulties in reproducing observed concentrations of radical sources and sinks
13 (e.g. HCHO, hydrogen peroxide). A global model analysis of these data suggested a new
14 chemical mechanism, involving conversion of HO₂ on aerosols, to reconcile differences
15 between modeled and observed HO_x partitioning (Mao et al., 2010). This mechanism may
16 also explain the underestimation of modeled CO in northern mid and high latitudes (Mao et
17 al., 2013), although further research is needed to investigate these processes.

18
19 The impact of boreal fires on the production of tropospheric O₃ was a focus of
20 POLARCAT, particularly in the summer when photochemistry is more active. As noted in a
21 review of fire impacts on O₃ (Jaffe and Wigder, 2012), O₃ is generally formed in aged air
22 masses downwind of boreal fire emission regions due to PAN decomposition releasing NO_x
23 (e.g. Real et al. (2007)) although there can be some O₃ formation even close to the fires.
24 However, Alvarado et al. (2010) found little or no enhancement in O₃ relative to CO in fresh
25 plumes over Canada due to rapid PAN formation limiting NO_x availability and O₃ formation
26 potential close to the fires. In another study, Singh et al. (2010) noted that aged fire plumes

1 transported at higher altitudes over North America were generally mixed with some urban
2 emissions and did have enhanced O₃. However, these fire plumes may have also been
3 influenced by NO_x produced by lightning or by air masses of stratospheric origin, both having
4 higher NO_x mixing ratios (Liang et al., 2011). Plumes measured over Siberia during summer,
5 in the lower troposphere, exhibited depleted O₃ relative to CO, probably due to widespread
6 dry deposition of O₃ onto vegetation, thus limiting the potential for O₃ enhancements in aged
7 fire plumes at low altitudes (Engvall Stjernberg et al., 2011; Paris et al., 2010).

8
9 Wespes et al. (2012) carried out a detailed evaluation of Arctic O₃ sources in spring and
10 summer 2008 based on tagged tracers in the global MOZART-4 model (see Figure 10).
11 Diagnosing O₃ sources is more complex than for CO (discussed in Section 2) due to
12 photochemical production/destruction, natural sources (stratosphere, lightning NO_x) and loss
13 by dry deposition. During spring, Arctic O₃ was influenced primarily by anthropogenic
14 emissions from Europe in the lower troposphere and from Asia/North America in the mid-
15 troposphere. Wespes et al. (2012) found that the lower stratosphere was an important source
16 of upper tropospheric O₃ in spring, but much less so during the summer campaign period
17 when O₃ production from Asian emissions and lightning NO_x were found to be important
18 sources of mid and upper tropospheric O₃. Their findings, together with other studies based on
19 POLARCAT data (e.g. Roiger et al., 2011a), suggest that summertime O₃ in the Arctic upper
20 troposphere, which shows a similar vertical distribution over Greenland (GRACE data in Fig.
21 9f) to the spring (Fig. 9b), is considerably influenced by tropospheric sources including
22 anthropogenic emissions. In the summertime lower and mid-troposphere Wespes et al. (2012)
23 also diagnosed that European emissions made an important contribution to O₃ over the eastern
24 Arctic and that O₃ produced downwind of North America was also an important source. This
25 is in contrast to the multi-model analysis of Shindell et al. (2008) which found North America
26 to be the main O₃ source at all altitudes in the Arctic. Wespes et al. (2012) estimated that

1 boreal fire emissions made a significant contribution only in the summer months. However,
2 the authors note that O₃ production may be underestimated in the mid-troposphere in their
3 model since simulated O₃ is underestimated in this region. This was also the case in Alvarado
4 et al. (2010) who diagnosed low contributions from boreal fires to Arctic O₃ using the GEOS-
5 Chem model.

6
7 In contrast to large-scale model simulations, a high-resolution regional study, using the
8 WRF-Chem model, examined the transport of anthropogenic and boreal fire plumes from
9 North America to Greenland in July 2008, and diagnosed significant O₃ production in
10 individual plumes (Thomas et al., 2013). Figure 11 shows the average impact on O₃
11 concentrations from anthropogenic and fire plumes transported to latitudes > 55N (as function
12 of altitude) during the study period within the regional model domain. O₃ increased by up to
13 18% from anthropogenic emissions and about 5% from boreal fire emissions (with emissions
14 from fires representing 21% of total CO emitted during the model run). Other non-
15 POLARCAT related studies have also noted significant contributions from boreal fires to
16 Arctic O₃ (e.g. Parrington et al. (2012)), and that the PAN contained in such plumes can lead
17 to significant O₃ production during warmer summer months (Walker et al., 2012). Reasons for
18 differences in model responses may be due to a number of factors such as the injection height
19 of fire emissions (e.g. Sessions et al., 2011). As part of POLMIP, Arnold et al. (2014)
20 demonstrate that global models, run using the same fire emissions for 2008 (Wiedinmyer et
21 al., 2011), all produce O₃ in fire-dominated air masses, with increasing O₃ production as the
22 fire emissions age. Ozone production efficiency in the Arctic is strongly sensitive to model
23 VOC chemistry because PAN, which originates from VOC oxidation, dominates summertime
24 Arctic NO_y. Differences in the efficiency of PAN formation, linked to differences in vertical
25 export efficiency from emission regions, also contribute to model diversity.

26

1 Improved understanding regarding the source of reactive bromine responsible for near
2 surface O₃ depletion, and its vertical extent in the Arctic troposphere, was also a focus of the
3 POLARCAT campaigns. Significant inconsistencies exist in tropospheric column retrievals of
4 bromine oxide (BrO) from satellite sensors (e.g. SCIAMACHY). Because these satellite
5 measurements provide some of the few observations available to understand free tropospheric
6 halogen chemistry in the Arctic, validation of satellite BrO was an important objective (Jacob
7 et al., 2010; Brock et al., 2011). The ARCTAS and ARCPAC campaigns were the first
8 airborne missions to use chemical ionization mass spectrometry (CIMS) to speciate inorganic
9 bromine (Neuman et al., 2010). In-situ measurements of active bromine (sum of bromine
10 (Br₂) and hypobromous acid, HOBr) as well as of BrO were reported in the Arctic free
11 troposphere (Neuman et al., 2010), for the first time, and following extensive laboratory
12 investigations (Liao et al, 2012).

13
14 POLARCAT clearly established that BrO hotspots in the Arctic observed by satellite
15 cannot routinely be interpreted as boundary layer enhancements. Salawitch et al. (2010)
16 showed satellite observations of BrO hotspots where no evidence of bromine chemistry or O₃
17 depletion was found *in-situ*. The aircraft also occasionally sampled air with nearly complete
18 O₃ depletion together with enhanced active bromine and soluble bromide that was not in a
19 satellite-detected BrO hotspot. This study established that spatial variations in the
20 stratospheric contribution to the total column measurement of BrO from space must be taken
21 into account in order to estimate the tropospheric residual, and indicated that a significant
22 fraction of the tropospheric BrO column might sometimes be in the free troposphere. Choi et
23 al. (2012) confirmed that BrO in the free troposphere often contributed substantially to the
24 tropospheric column. They noted that true boundary layer BrO maxima observed from space
25 reflect active bromine chemistry and O₃ destruction, while air masses with severe O₃
26 depletion cannot be identified by the satellite sensors because BrO quickly converts to other

1 undetectable inorganic bromine compounds in the absence of O₃. Koo et al. (2012) combined
2 the aircraft and satellite observations with observations of O₃ and bromine compounds at
3 several surface stations and ozone sondes launched around the Arctic during POLARCAT to
4 examine O₃ depletion events across the region. Their study provided additional evidence that
5 the impact of active bromine chemistry is not restricted to the shallow Arctic boundary layer
6 but extends into the Arctic free troposphere. The sources of Arctic free tropospheric BrO
7 enhancements may not be completely dominated by halogens emitted in the Arctic boundary
8 layer or of stratospheric origins, as pointed out by Pommier et al. (2012). Further evidence for
9 halogen chemistry was found during the ICEALOT cruise in the northern North Atlantic
10 where air masses exposed to sea ice in the high Arctic showed depleted O₃ and perturbed
11 ratios of selected hydrocarbons (Gilman et al., 2010). We note that many global models do
12 not take tropospheric halogen chemistry into account and its impact on tropospheric O₃
13 requires further investigation.

14

15 **6. Conclusions and future perspectives**

16 POLARCAT-IPY provided a wealth of new data on Arctic air pollution. In particular,
17 data were collected on the vertical distributions of aerosol chemical composition and trace gas
18 species throughout the free troposphere and lowermost stratosphere during spring and
19 summer 2008. These data have been used, and are continuing to be used, to evaluate and
20 improve chemistry-climate models, for example, as part of POLMIP, and by the Arctic
21 Council AMAP (Arctic Monitoring and Assessment Program) expert group on Black Carbon
22 and Ozone, which is examining the impact of different sources of short-lived pollutants on
23 climate.

24

25 Analysis of POLARCAT data highlighted important contributions from agricultural
26 and boreal fire emissions, as well as anthropogenic emissions from east Asia, to aerosols and

1 trace gases observed over the Arctic in spring and summer 2008. In the free troposphere, fires
2 were the main source of organic aerosols during spring whereas Asian pollution was the main
3 source of sulfate with BC originating from both these sources. While sulfate dominated
4 aerosol mass at Arctic surface sites, organic matter also contributed substantially; the origins
5 of this organic mass warrant further investigation. Although aerosol concentrations were
6 lower in summer in the Arctic free troposphere, enhancements were observed in polluted
7 plumes.

8
9 During spring 2008, transport of pollution into the Arctic free troposphere produced
10 high aerosol concentrations in layers above the surface, in particular due to active fires in
11 boreal regions. Multi-year analysis of CALIPSO aerosol lidar satellite data, which was
12 extensively validated using POLARCAT aircraft and ground-based data, was used to show
13 that transport of pollution into the Arctic free troposphere was more pronounced in 2008.
14 Surface observations, in contrast, agreed with the long-term seasonal cycle of the chronic
15 Arctic Haze aerosol. These observations are consistent with the conceptual model that
16 northern midlatitude and Arctic sources within the cold polar dome contribute most to near-
17 surface Arctic pollution, and that a variety of mid- and high-latitude emissions may be
18 advected to the Arctic aloft. Further, these findings imply that Arctic surface measurements
19 are often not representative of the atmosphere aloft, and suggest that Arctic composition in
20 the free troposphere is particularly sensitive to yearly and seasonal variations in transport
21 patterns and fire emissions.

22
23 The first direct measurements of Arctic black carbon vertical distributions were
24 collected during POLARCAT and used to estimate dry deposition rates as well as local
25 heating rates in aerosol layers observed in the free troposphere. The impact of these lofted
26 pollution layers, which often contain absorbing and non-absorbing aerosols from more than

1 one source, on the Arctic radiation budget and their effects on cloud properties, as well as wet
2 and dry depositional fluxes to the Arctic surface, and effects of deposited absorbing aerosol
3 on surface albedo, are still areas of active research. While global aerosol models continue to
4 be improved, it is clear that further attention needs to be paid to improving treatments of wet
5 and dry deposition. In addition, emissions of light-absorbing particles near and within the
6 Arctic, such as BC from shipping and oil/gas extraction, require better quantification since
7 they are likely to increase in the future.

8
9 Asian, and to a lesser extent European and North American anthropogenic pollution as
10 well as boreal fires, were identified as important sources of O₃ precursors in spring/summer
11 2008 in the lower and mid-troposphere. Episodes of cross-Arctic pollution transport were
12 documented in the summer resulting in elevated concentrations of trace gases, such as CO and
13 PAN, in the free troposphere and lower stratosphere. Together with lightning NO_x emissions,
14 these tropospheric sources of O₃ appear to be making a significant contribution to Arctic
15 summertime O₃ in the free troposphere. Direct trans-polar transport of East Asian pollution
16 toward Europe may lead to downwind O₃ production but the contribution of this pathway has
17 yet to be fully assessed. Anthropogenic pollution appears to make a larger contribution to O₃
18 over the Arctic compared to boreal fires. However, evidence exists for O₃ production
19 downwind of fires in the Arctic and further work is required to evaluate the role of PAN as a
20 source of NO_x, particularly during summer.

21
22 Airborne in-situ measurements of reactive bromine during POLARCAT established that
23 regions of enhanced BrO observed from satellites are not always evidence of bromine
24 activation and loss of O₃ in the boundary layer. Spatial and temporal variations, in both the
25 stratospheric and free-tropospheric BrO columns, can erroneously be interpreted as boundary
26 layer enhancements in BrO. Because satellite sensors have provided the primary means of

1 determining the spatial and temporal distribution of active bromine in the Arctic boundary
2 layer, many uncertainties remain in understanding the scale and magnitude of bromine-
3 catalyzed O₃ loss in the Arctic lower troposphere.

4
5 In the future, global warming and economic factors will likely lead to increased
6 exploitation of Arctic resources and increased emissions of SLCPs within the Arctic.
7 Pollutant emissions in the developed and developing world will also change rapidly, with
8 expected continuing declines in North America and Europe and increases in eastern and
9 southern Asia. The POLARCAT experiments have provided improved understanding of the
10 sources, transport, transformation, and sinks for key SLCPs in the Arctic. The observations,
11 and their analysis, provide valuable constraints for global models, which remain the primary
12 tool for developing policies to limit future climate forcing.

13
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24 348 (SCHL1857/3-1).

25

1 *List of figures*

2 FIG. 1. Overview of all flights of the DC-8 (blue), P-3B (black), ATR-42 (magenta), WP-3D
3 (cyan), Falcon (green), Antonov-30 (red) aircraft campaigns, and the ICEALOT ship cruise
4 (green), during the a) spring and b) summer 2008 campaigns superimposed on IASI CO total
5 column monthly mean map (daytime). The CO data are averaged 1x1 degrees. The spring
6 map is represented by the April mean and the summer by the July mean. Locations of surface
7 sites referred to this paper are also indicated. Adapted from Pommier et al. (2010).

8

9 FIG. 2. Schematic showing pathways for the transport of air pollution into the Arctic.

10 Following Stohl (2006), three main routes are evident: (1) low-level transport from mid-
11 latitude emission regions followed by uplift at the Arctic front, (2) lifting of pollutants at
12 lower latitudes followed by upper tropospheric transport and eventual slow descent (due to
13 radiational cooling) or mixing into the polar dome – a frequent transport route from North
14 America and Asia, but prone to significant wet scavenging, (3) wintertime low-level transport
15 of already cold air into the polar dome mainly from northern Eurasia. Emissions from strong
16 boreal fires could be lofted by pyro-convection (Fromm et al., 2005) and later entrained into
17 the polar dome.

18

19 FIG. 3. Seasonal mean of the CALIPSO scattering ratio at 532 nm (1 is for Rayleigh
20 scattering) providing the spatial distribution of aerosol loadings over the Arctic for
21 March/April/May (A, C) and June/July/August (B, D) 2008 and for two altitude ranges: 0-2
22 km(C, D) and 4-6 km (A, B). A cloud mask was applied to keep only the aerosol contribution.
23 Data averaged over 300 km² grid cells. Based on methods described in Ancellet et al. (2014).

24

25 FIG. 4. Vertical distribution of CO mixing ratios (ppbv) from the ARCTAS DC-8
26 measurements (solid thick red line plus standard deviation shown as horizontal bars) and from

1 the GEOS-5 global model simulations sampled along the flights for a) April and b) July
2 (bottom). Model results are shown for total CO (thick black line) and for tagged components
3 from Asian (AS), European (EU) and North American (NA) fossil fuel (FF) and boreal
4 biomass burning (BOBB) and non-boreal biomass burning emissions (NBBB). “Other CO”
5 refers to global CO other than the five tagged components and is attributed mainly to CO
6 from methane oxidation. From Bian et al. (2013).

7

8 FIG. 5. (a) Time series of in-situ CO (red), in-situ O₃ (blue), and pressure at flight altitude
9 (black) for POLARCAT-GRACE flight on 10 July 2008, between 15:30 and 17:00 UTC, (b)
10 Simulated CO tracers from the FLEXPART parcel dispersion model; the red pattern
11 represents the FLEXPART excess-CO originating from Asian anthropogenic pollution; the
12 blue line gives the stratospheric fraction along the flight path. (c) CO source contribution plot
13 for 16:11UTC (maximum CO concentration at 11.3 km altitude) with color scaling on the
14 right. From Roiger et al. (2011a).

15

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

24

25 FIG. 7. Track of the R/V Knorr ship during the ICEALOT cruise (19 March to 24 April 2008)
26 colored by regions of air mass origin including Long Island (purple), North America (pink),

1 North Atlantic (dark blue), Europe (bronze), and Arctic (teal). Gray represents air masses of
2 with no distinct origin or periods without sampling. The pies represent the submicron particle
3 composition (left) and organic functional group composition (right) for the European and
4 Arctic source regions during ICEALOT and Barrow, Alaska (see Fig. 1) (dark purple). The
5 submicron particle composition excludes the elemental carbon (estimated on average less than
6 3% during ICEALOT and at Barrow). OM is submicron organic mass. IOM is inorganic
7 oxidized material (dust and fly ash). Note that the Barrow data is an average of 2 years (2008
8 and 2009). From Frossard et al. (2011).

9
10 FIG. 8. Contributions of boreal and agricultural fire emissions and anthropogenic (fuel
11 combustion) emissions to total (wet plus dry) deposition fluxes of black carbon from the
12 GEOS-Chem global model simulations for winter (January to March) and spring (April/May)
13 2008. From Wang et al. (2011).

14
15 FIG. 9. Mean vertical distribution of trace gas mixing ratios (CO/ppbv, O₃/ppbv, NO/pptv and
16 PAN/pptv) measured during different POLARCAT aircraft deployments (see Table 2). Top
17 row shows data from spring campaigns covering the period 18 March to 23 April 2008,
18 bottom row shows measurements from summer deployments covering the period 29 June to
19 29 July 2008. The horizontal bars at each altitude bin indicate the extent of the inter-quartiles
20 (25th and 75th percentile).

21
22 FIG. 10. Principal contributions to O₃ columns (molecules cm⁻²) from the ground to 300 hPa
23 during spring (1 to 19 April 2008) and summer (18 June to 13 July 2008) ARCTAS
24 campaigns as simulated by the global MOZART-4 model. The flight tracks corresponding to
25 these periods are superimposed in black on the stratospheric contribution plots. From Wespes
26 et al. (2012).

1
2 FIG. 11. The average increase in ozone (ΔO_3) in ppbv from WRF-Chem model simulations
3 north of 55N from anthropogenic emissions over North America (left panel) and boreal fire
4 emissions over Canada (right panel). Excess ozone (ΔO_3) was calculated as the average
5 increase in O_3 north of 55N based on model runs with and without anthropogenic and fire
6 emissions. From Thomas et al. (2013).

7

1 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/ inter-annual 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 of 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

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TABLE 2. POLARCAT-IPY and Related Field Campaigns

ASTAR ^{a,*}	Mar-Apr 2007	Dornier 228-101 (Polar-2), DLR Falcon-20	Arctic cloud and aerosol properties	Gayet et al. (2009)
ARCPAC ^b	Mar-Apr 2008	NOAA WP-3D (0-7km)	Aerosols plus radiation budget	Brock et al. (2011)
ARCTAS ^c -A + ARCTAS ^c -B	Apr. 2008/ July 2008	NASA DC-8 (0- 12km), NASA P-3B (0-7km)	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)	Adam de Villiers et al. (2010), Pommier et al. (2010)
POLARCAT- GRACE ^d	Jun-Jul 2008	DLR Falcon-20 (0-11km)	Pollution transport (O ₃)	Roiger et al. (2011b)
YAK- AEROSIB ^e	July 2008	Russian Antonov- 30 (0-6km)	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	NRC Convair-580 (0-6km)	Aerosol-cloud properties	McFarquhar (et al. (2011)
POLAR- AOD ^{h,*}	2007-2009	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. (2007), Hirdman et al. (2010)
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)

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

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

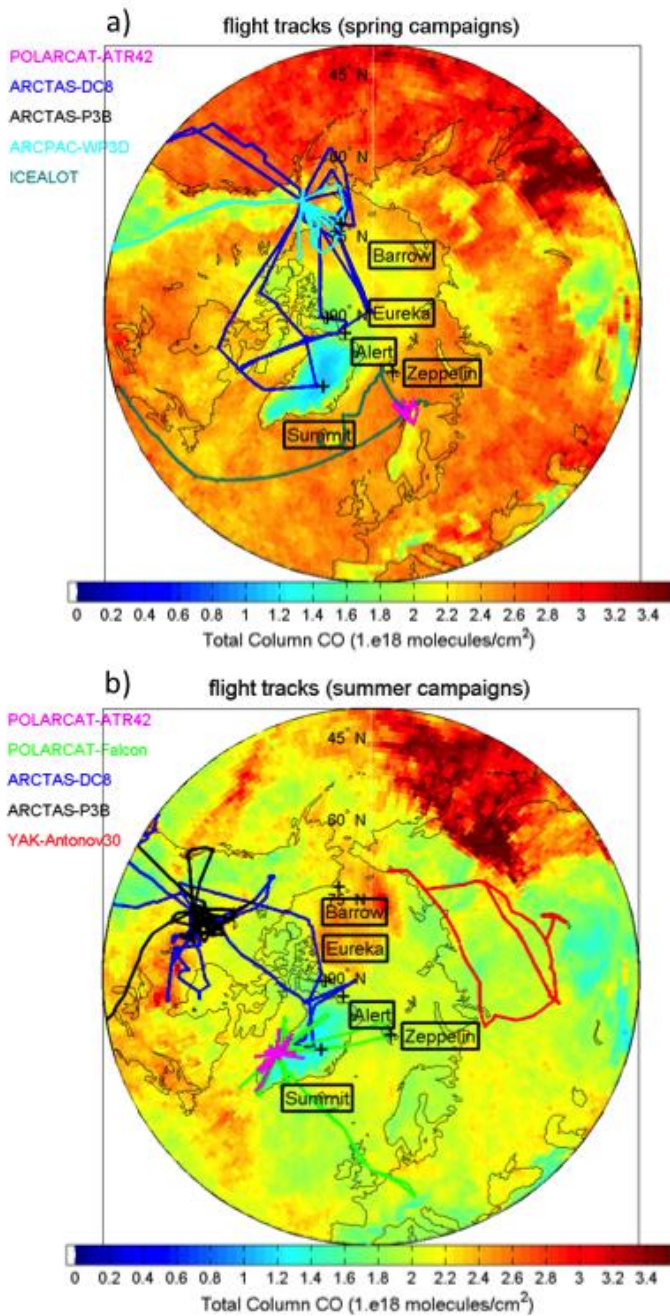
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^c Arctic Research of the Composition of the Troposphere from Aircraft and Satellites.

8

^d POLARCAT-Greenland Aerosol and Chemistry Experiment.

- 1 ^e YAK-Airborne Extensive Regional Observations in SIBeria.
- 2 ^f International Chemistry Experiment in the Arctic Lower Troposphere.
- 3 ^g Indirect and Semi-direct Aerosol Campaign
- 4 ^h Polar Aerosol Optical Depth
- 5 ⁱ International Arctic Systems for Observing the Atmosphere.
- 6 ^j ARCTic - Intensive Ozonesonde Network Study.
- 7 ^k Greenland Summit Halogen-HO_x Experiment
- 8 * Complementary activities to POLARCAT
- 9



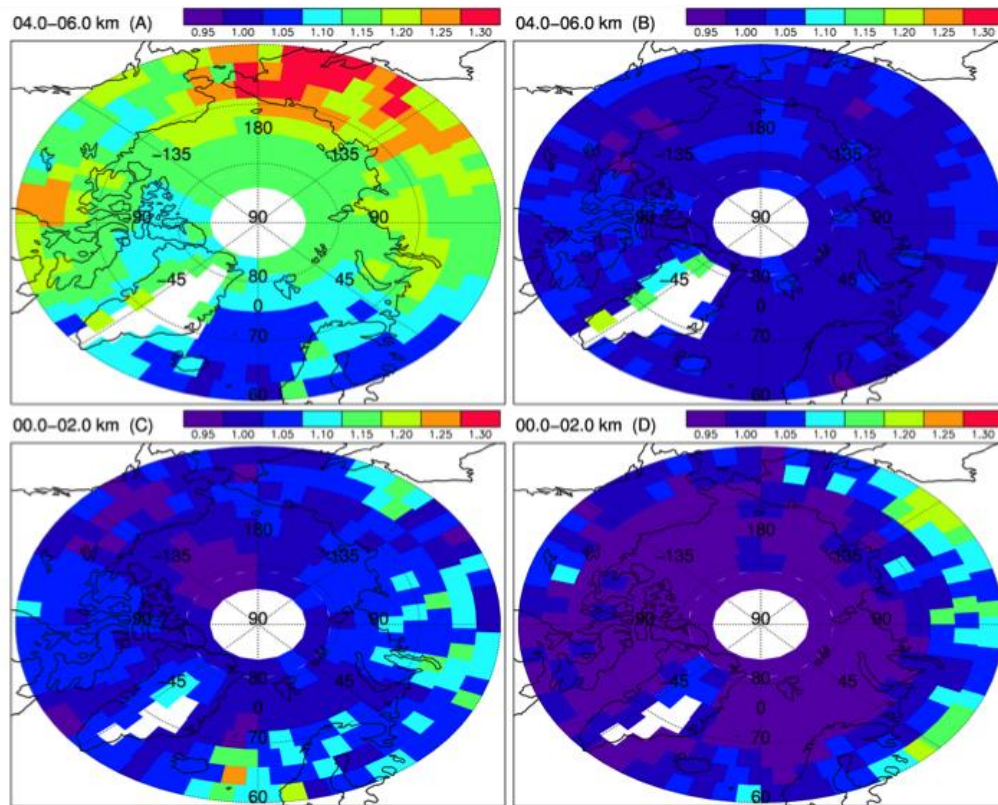
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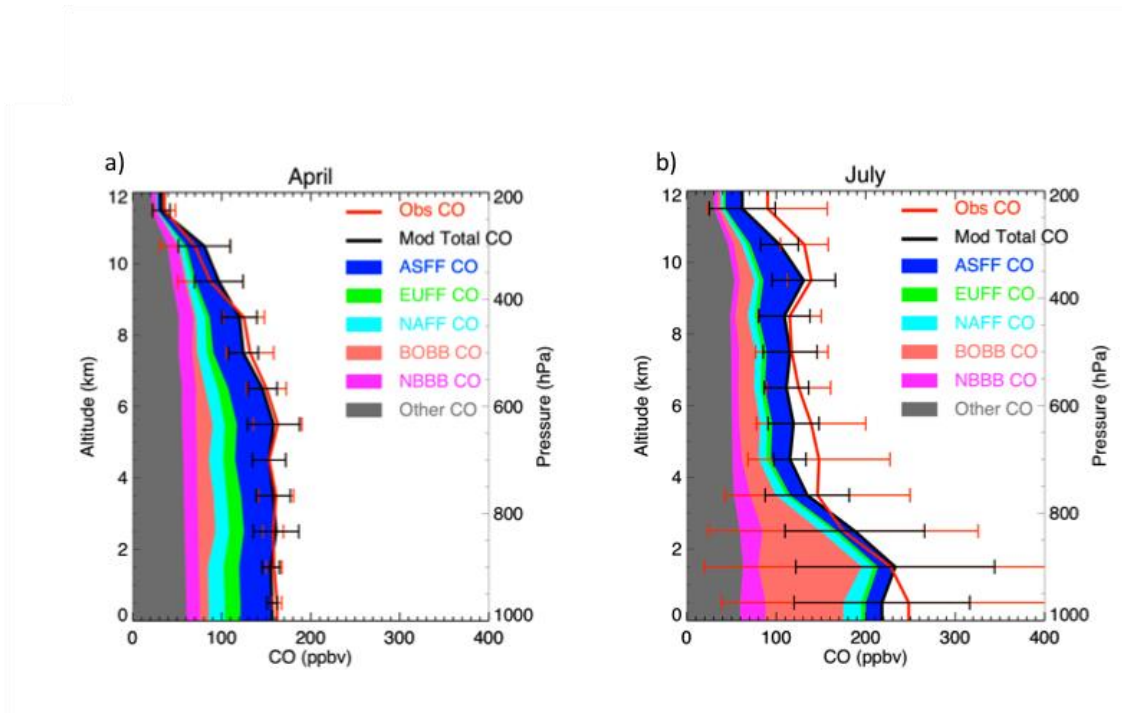
1 **Figure 1:** Overview of all flights of the DC-8 (blue), P-3B (black), ATR-42 (magenta), WP-
2 3D (cyan), Falcon (green), Antonov-30 (red) aircraft campaigns, and the ICEALOT ship
3 cruise (green), during the a) spring and b) summer 2008 campaigns superimposed on IASI
4 CO total column monthly mean map (daytime). The CO data are averaged 1×1 degrees. The
5 spring map is represented by the April mean and the summer by the July mean. Locations of
6 surface sites referred to this paper are also indicated. Adapted from Pommier et al. (2010).



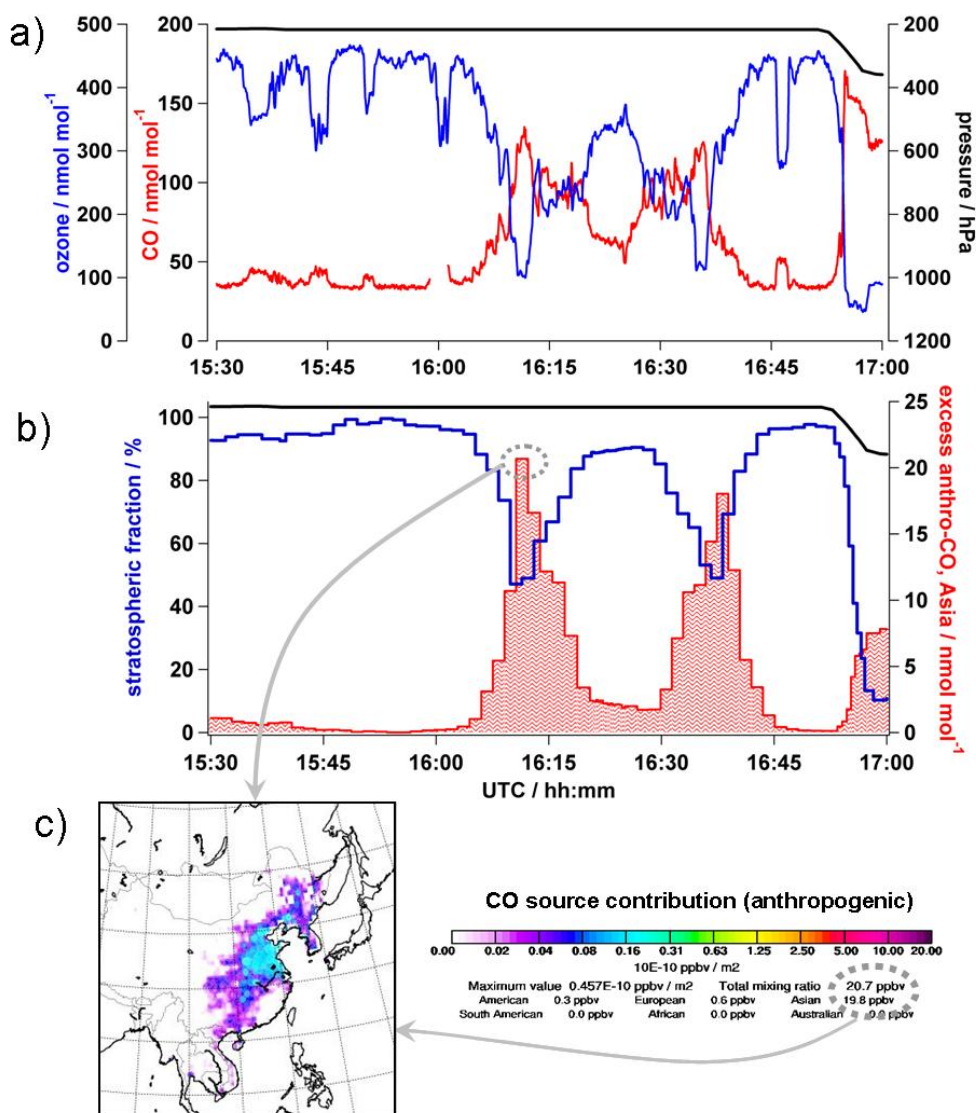
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 2 **Figure 2:** Schematic showing pathways for the transport of air pollution into the Arctic.
 3 Following Stohl (2006), three main routes are evident: (1) low-level transport from mid-
 4 latitude emission regions followed by uplift at the Arctic front, (2) lifting of pollutants at
 5 lower latitudes followed by upper tropospheric transport and eventual slow descent (due to
 6 radiational cooling) or mixing into the polar dome – a frequent transport route from North
 7 America and Asia, but prone to significant wet scavenging, (3) wintertime low-level transport
 8 of already cold air into the polar dome mainly from northern Eurasia. Emissions from strong
 9 boreal fires could be lofted by pyro-convection (Fromm et al., 2005) and later entrained into
 10 the polar dome.
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 2 **Figure. 3** Seasonal mean of the CALIPSO scattering ratio at 532 nm (1 is for Rayleigh
 3 scattering) providing the spatial distribution of aerosol loadings over the Arctic for
 4 March/April/May (A, C) and June/July/August (B, D) 2008 and for two altitude ranges: 0-2
 5 km(C, D) and 4-6 km (A, B). A cloud mask was applied to keep only the aerosol contribution.
 6 Data averaged over 300 km² grid cells. Based on methods described in Ancellet et al. (2014).

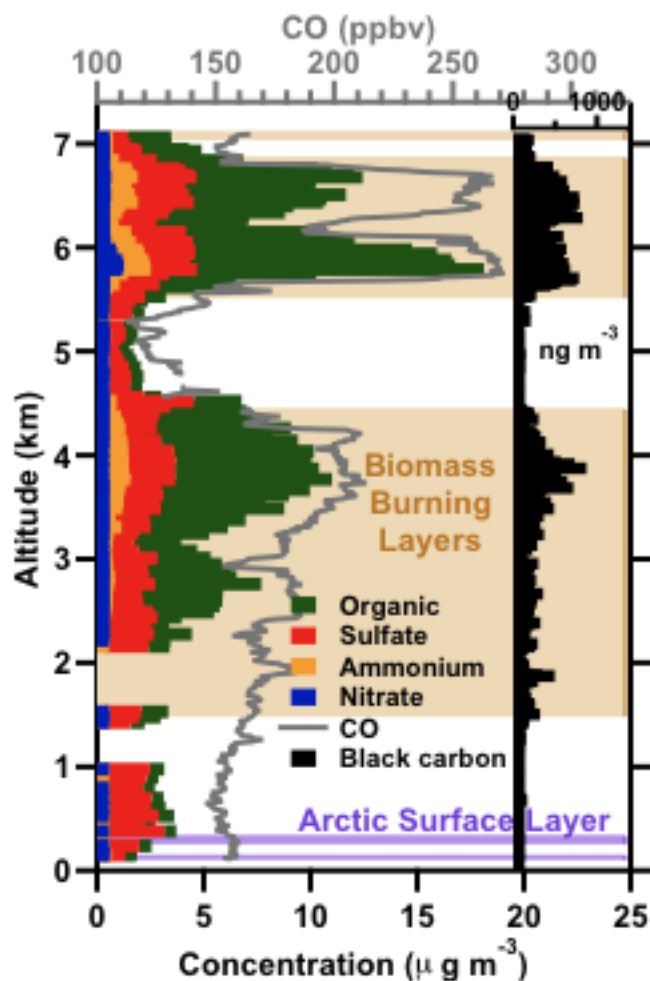


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 2 **Figure 4:** Vertical distribution of CO mixing ratios (ppbv) from the ARCTAS DC-8
 3 measurements (solid thick red line plus standard deviation shown as horizontal bars) and from
 4 the GEOS-5 global model simulations sampled along the flights for a) April and b) July.
 5 Model results are shown for total CO (thick black line) and for tagged components from
 6 Asian (AS), European (EU) and North American (NA) fossil fuel (FF) and boreal biomass
 7 burning (BOBB) and non-boreal biomass burning emissions (NBBB). “Other CO” refers to
 8 global CO other than the five tagged components and is attributed mainly to CO from
 9 methane oxidation. From Bian et al. (2013).
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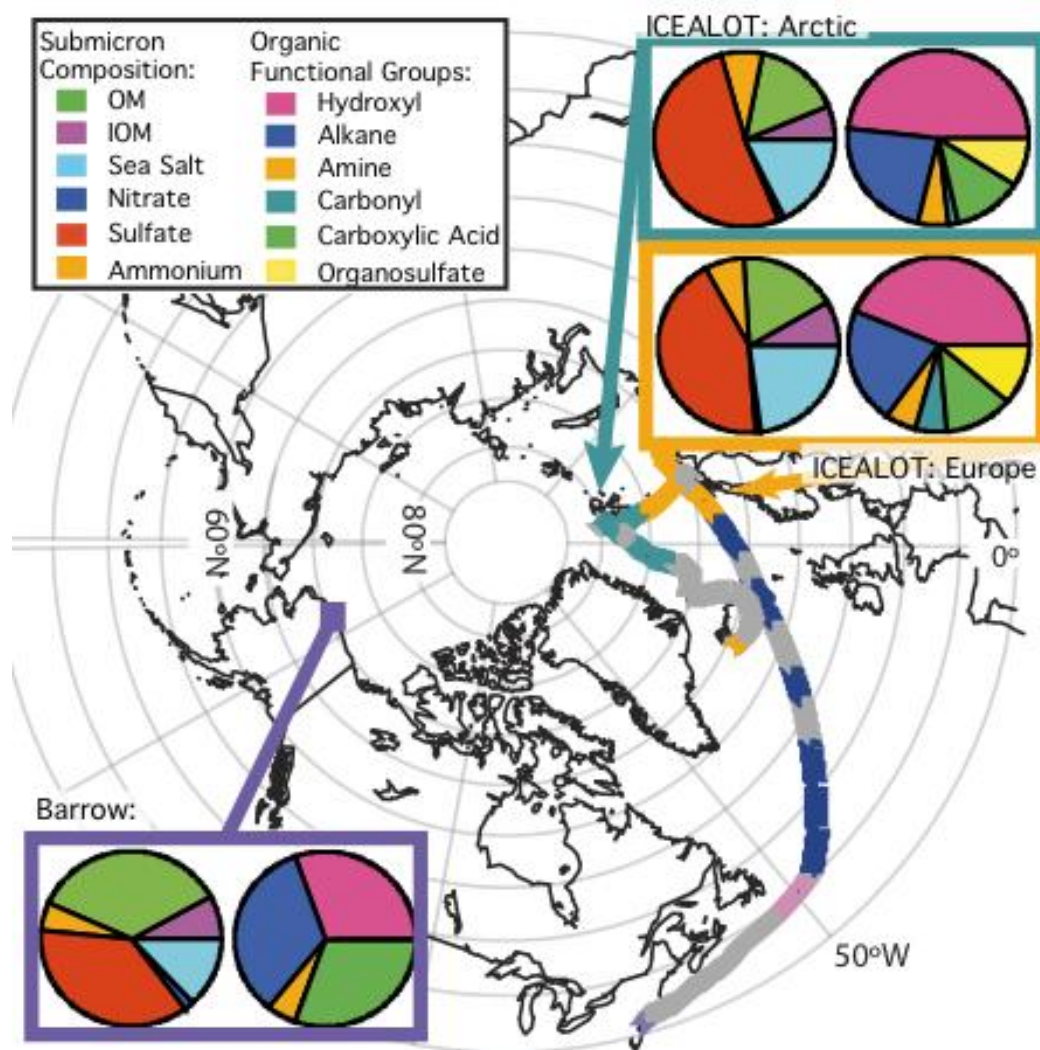
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 2 **Figure 5.** (a) Time series of in-situ CO (red), in-situ O₃ (blue), and pressure at flight altitude
 3 (black) for POLARCAT-GRACE flight on 10 July 2008, between 15:30 and 17:00 UTC, (b)
 4 Simulated CO tracers from the FLEXPART parcel dispersion model; the red pattern
 5 represents the FLEXPART excess-CO originating from Asian anthropogenic pollution; the
 6 blue line gives the stratospheric fraction along the flight path. (c) CO source contribution plot
 7 for 16:11UTC (maximum CO concentration at 11.3 km altitude) with color scaling on the
 8 right. From Roiger et al. (2011a).

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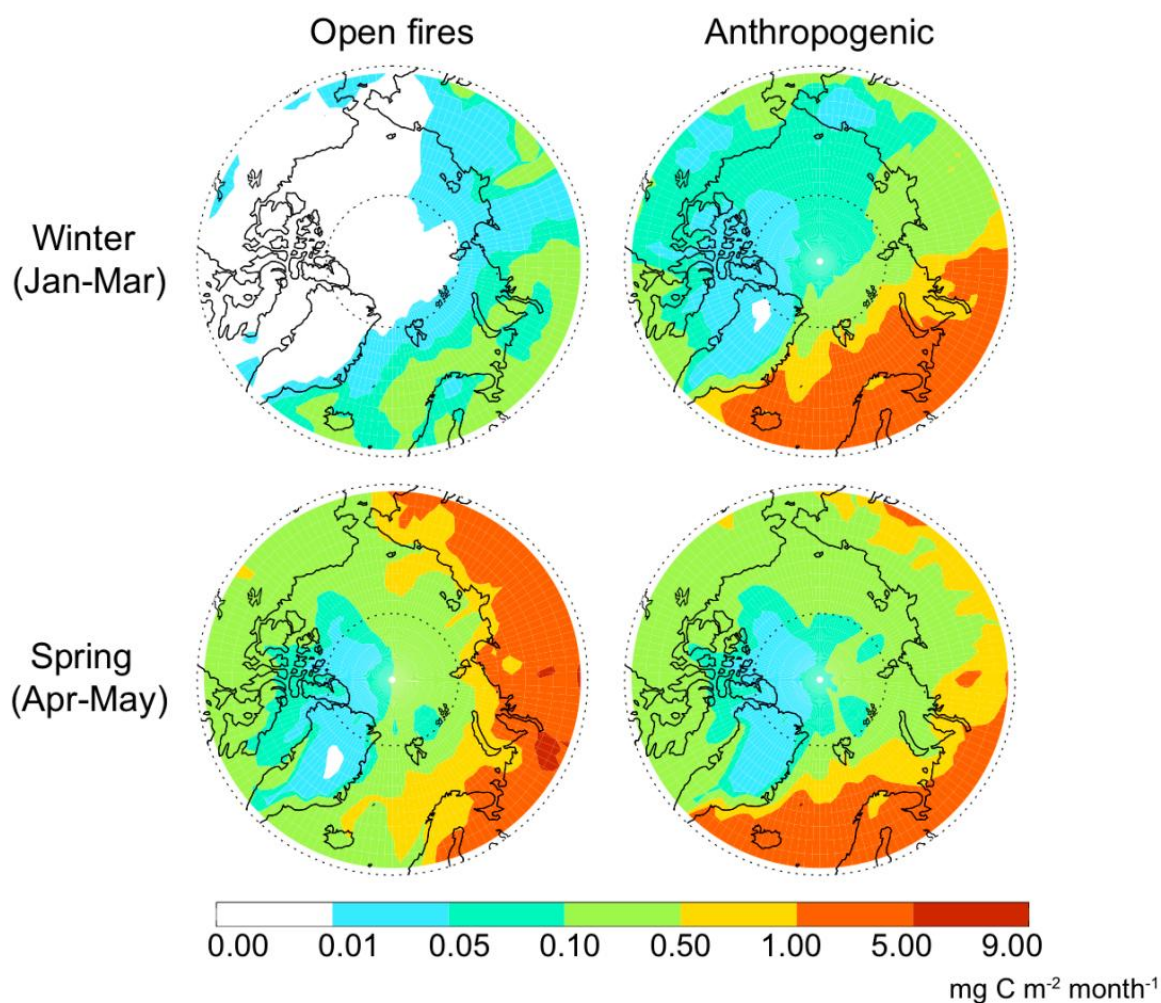
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 2 **Figure 6.** Vertical profile from the ARCPAC spring campaign measured during a slantwise
 3 ascent of the WP-3D aircraft over sea-ice north of Alaska on 18 April 2008. Submicron
 4 particle composition measured by the aerosol mass spectrometry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

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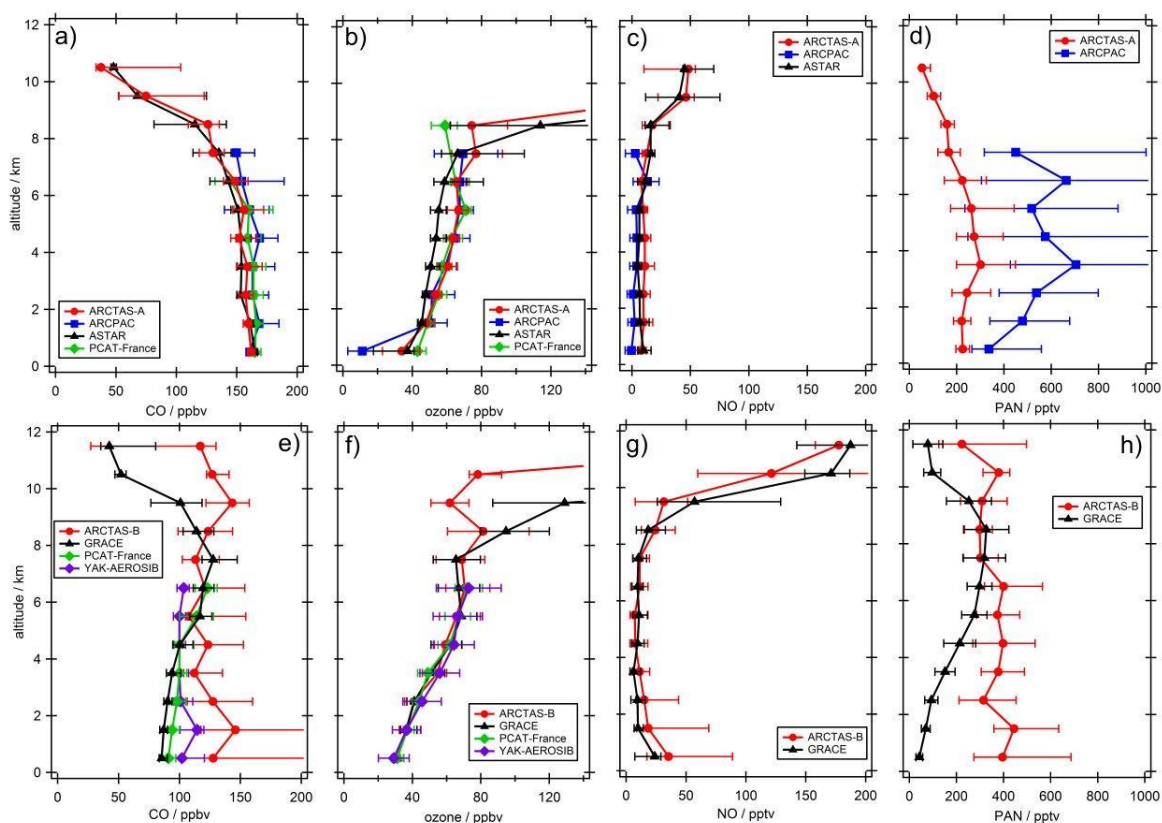
3 **Figure 7:** Track of the R/V Knorr ship during the ICEALOT cruise (19 March to 24 April
 4 2008) colored by regions of air mass origin including Long Island (purple), North America
 5 (pink), North Atlantic (dark blue), Europe (bronze), and Arctic (teal). Gray represents air
 6 masses with no distinct origin or periods without sampling. The pies represent the submicron
 7 particle composition (left) and organic functional group composition (right) for the European
 8 and Arctic source regions during ICEALOT and Barrow, Alaska (see Fig. 1) (dark purple).
 9 The submicron particle composition excludes the elemental carbon (estimated on average less
 10 than 3% during ICEALOT and at Barrow). OM is submicron organic mass. IOM is inorganic
 11 oxidized material (dust and fly ash). Note that the Barrow data is an average of 2 years (2008
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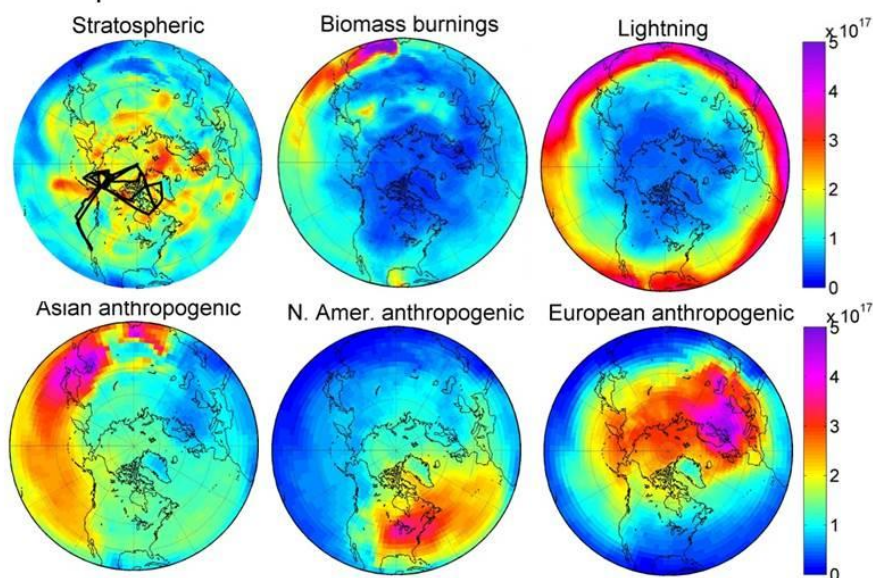
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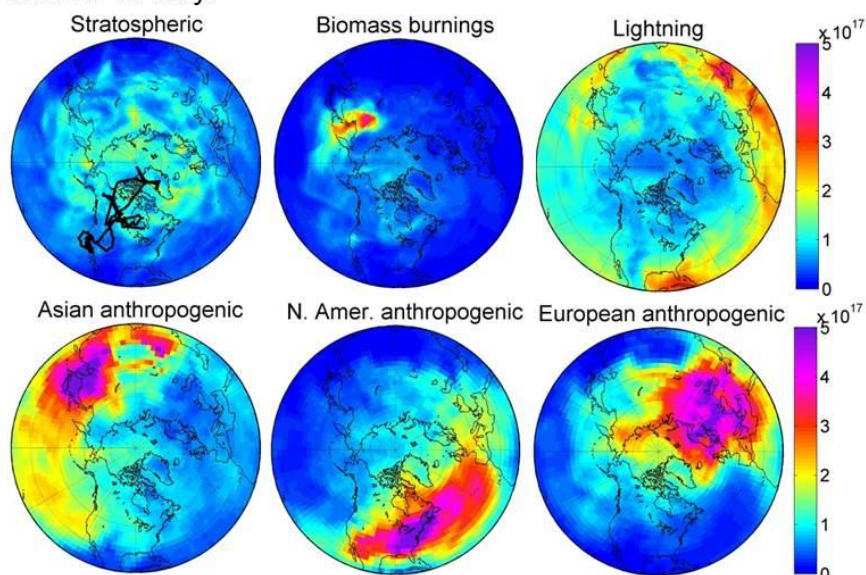
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O₃ contributions

1-19 April:



18 June-13 July:



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3 **Figure 10.** Principal contributions to O₃ columns (molecules cm⁻²) from the ground to 300

4 hPa during spring (1 to 19 April 2008) and summer (18 June to 13 July 2008) ARCTAS

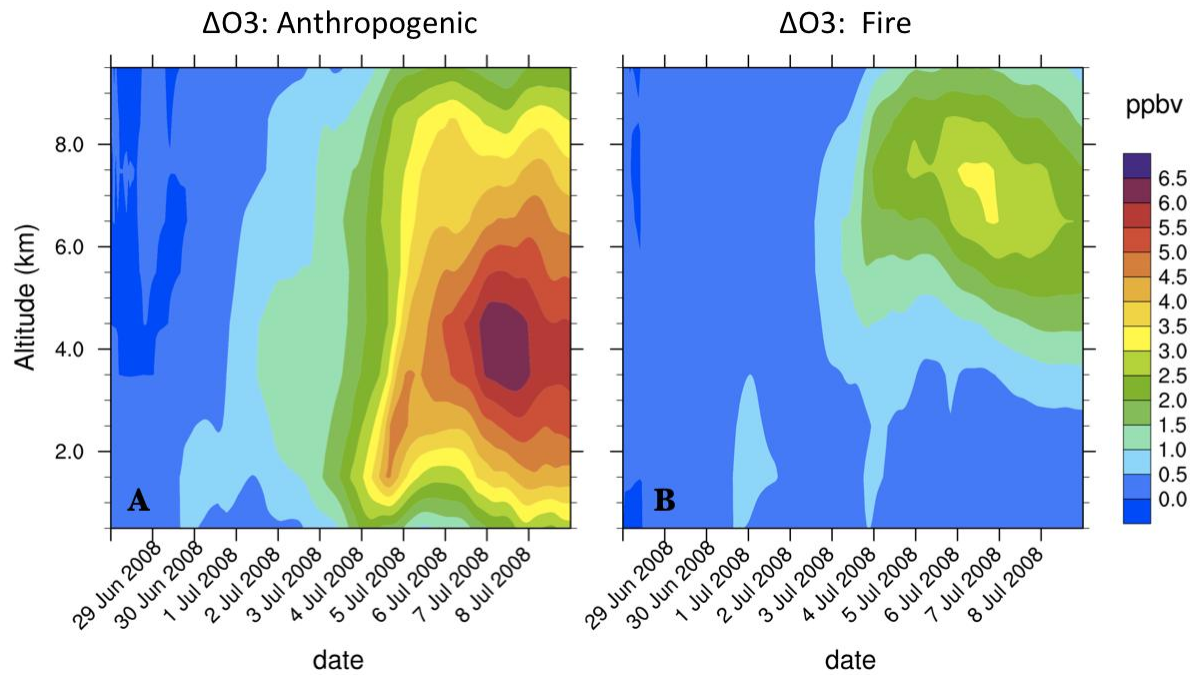
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6 these periods are superimposed in black on the stratospheric contribution plots. From Wespes

7 et al. (2012).

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6 emissions. From Thomas et al. (2013).

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