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**Boundary layer  
ozone in the central  
Arctic**

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# Ozone in the Boundary Layer air over the Arctic Ocean – measurements during the TARA expedition

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## Abstract

A full year of measurements of surface ozone over the Arctic Ocean far removed from land is presented (81° N – 88° N latitude). The data were obtained during the drift of the French schooner TARA between September 2006 and January 2008, while frozen in the Arctic Ocean. The data confirm that long periods of virtually total absence of ozone occur in the spring (mid March to mid June) after Polar sunrise. At other times of the year ozone concentrations are comparable to other oceanic observations with winter mole fractions of ca. 30–40 nmol mol<sup>-1</sup> and summer minima of ca. 20 nmol mol<sup>-1</sup>. Contrary to earlier observations from ozone sonde data obtained at Arctic coastal observatories, the ambient temperature was well above –20°C during most ODEs (ozone depletion episodes). Backwards trajectory calculations suggest that during these ODEs the air had previously been in contact with the frozen ocean surface for several days and originated largely from the Siberian coast where several large open flaw leads developed in the spring of 2007.

## 1 Introduction

Marine boundary layer ozone depletion in the Arctic following Polar sunrise was first reported in the 1980s (Oltmans, 1981; Oltmans and Komhyr, 1986; Barrie et al., 1989; Bottenheim et al., 1986). It is due to fast chemical reactions involving ozone and bromine atoms that are produced by chemical activation of seasalt bromide (for a recent review see Simpson et al., 2007). Ozone is destroyed by the reaction with bromine atoms leading to the production of a bromine-oxide molecule, as first proposed by Barrie et al. (1988):



The presence of BrO during an ozone depletion event in the Arctic was confirmed by Hausmann and Platt (1994). Since this depends on Reaction (R1) it can be considered

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a marker for the occurrence of ozone depletion chemistry. Based on this fact, and with the advent of the GOME satellite measurements of BrO that have shown widespread areas where tropospheric BrO levels exceed anticipated background values (Richter et al., 1998; Wagner and Platt, 1998), it is believed that the surface ozone depletion is an Arctic-wide phenomenon. While it is clear that the Arctic ozone depletion in the boundary layer originates from surface mediated chemical interactions, actual in-situ observations have been limited to those obtained at coastal sites (Simpson et al., 2007), or campaigns of short duration such as on ice islands near the coast (Hopper et al., 1994, 1998; Morin et al., 2005), from aircraft (Sheridan et al., 1993; Leaitch et al., 1994; Jaeschke et al., 1999; Ridley et al., 2003) or icebreakers (Jacobi et al., 2006).

TARA is a privately owned French polar schooner that was specifically built to withstand a journey to the North Pole and resist the forces of the Arctic Ocean ice. During 2006–2008 she drifted frozen in the Arctic Ocean ice, recreating the Nansen expedition of 1893–1895 (<http://www.taraexpeditions.org>). For this purpose TARA sailed in the summer of 2006 from its home port in Lorient, France, to Tiksi, a small town at the mouth of the Lena River in Siberia, Russia, and then headed north. Aided by the Russian Icebreaker the Kapitan Dranytsin TARA reached an ice floe at 80° N/143° E where she let itself freeze in the ice. The subsequent drift lasted about 16 months before TARA was released in the North Greenland Sea in late January 2008 (see Fig. 1).

During this expedition the crew on board of TARA undertook several experiments investigating the physical and chemical state of the environment at its location in the Arctic Ocean ice as part of the European Union project DAMOCLES (<http://www.damocles-eu.org>). Included was the installation and operation of a commercial ozone monitor. As a result, the TARA expedition has provided for the first time a full year of observations of the O<sub>3</sub> concentration over the frozen Arctic Ocean surface hundreds of kilometers from land.

## 2 Experimental

The ozone monitor used during the TARA expedition was a Thermo Electron Industries Model 049 ozone monitor (TEI049) which is based on UV absorption spectroscopy. The operation of the instrument, as well as data collection was 100% controlled by a Campbell Scientific Model CS21X data logger, and the results were transferred daily to an onboard laptop computer. The CS21X was programmed to collect raw data at a 10 s interval, and to compute a five minute average of these data. The 5-min averages were frequently transmitted via satellite communication eventually to Toronto to permit status control during the drift.

The TEI049 did not have extensive electronic processor control. Hence to be able to fully assess the operation of the instrument after the expedition several parameters indicating its operational status were continually logged. These included the temperature and sample pressure of the absorption cells. The sample flow through the instrument, which is controlled by glass capillaries, was logged by installing small electronic flow meters in the flow path following the absorption cells but before the capillaries. An additional pressure sensor determined the vacuum produced after the flow controlling capillaries to gauge sampling pump performance.

Once a day a zero reading was collected by sampling ambient air via a zero trap to remove ozone; every fifth day the zero air was internally photolyzed to produce a span sample containing ozone at approx.  $60 \text{ nmol mol}^{-1}$ . However this procedure was subsequently found to be unreliable and the readings could only be used to confirm that the instrument was operating properly (i.e. responding to the presence of ozone). Rather, the instrument was calibrated against a National Institute of Standards and Technology (NIST) traceable standard before and after the expedition. It was planned to perform an additional calibration midway during the drift but due to logistical problems this calibration did not materialize, and no additional calibrations during the expedition were attainable. The results from the pre and post calibrations showed a 18% drift in the slope of the calibration and negligible drift in the zero. It is debatable whether this drift

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had occurred linearly over time and the daily zero/span data were not reliable to assess this fact. Therefore, the mean of the two calibration slopes was used to adjust the raw data.

The TEI049 and CS21X were installed in a storage compartment at the aft of the vessel, approximately 10 m behind the communications room where the laptop computer was placed. This storage compartment was not heated and hence was expected to experience variable, and on occasion quite low ambient temperatures. Therefore, a thermocouple was installed to control the power to the cooling fan of the TEI049 and the fan was programmed to operate only when the ambient temperature inside the instrument casing was higher than +20°C. Prior to installation of the thus modified instrument, its operation at low ambient temperatures was inspected in a cold room and found to be acceptable down to -15°C, well below the official instrument specifications. During the drift the temperature in the storage compartment rarely dropped below -10°C; data collected at lower temperature were deleted from consideration. Ambient air was sampled via a weekly changed Teflon intake filter (47 mm diameter, 5 µm pore size) and approx. 10 m of 3/8" ID PFA Teflon tubing. The filter was mounted in a PFA filter holder housed in a stainless steel hood (to protect against inclement weather) which was mounted at the back railing of the vessel.

While one objective of the expedition was to use as much as feasible green energy, it was unavoidable to operate a diesel power generator for several hours per day to produce electrical energy, especially during the cold, dark winter. The exhaust of the generator was not in close proximity to the air intake for the ozone instrument, but nevertheless some impact was anticipated, especially under conditions of low local wind speed. For this reason all data collected during the actual operation of the diesel generators were filtered from the final data set. Filtered data are indicated in light grey in Fig. 2.

In order to obtain insight into the origin and destination of the air mass that was measured during the campaign, daily trajectories were calculated with the Environment Canada 3-D trajectory model which is similar to the well known Hysplit model (Draxler

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and Hess, 1998). The calculations started for each day at 12:00 UTC and an altitude of 10 and 500 m above sea level, and were calculated for a 10 day period backwards as well as forwards from the (variable) location of TARA.

### 3 Results

Figure 2 shows a one year record of ozone measurements made on board of TARA. It has to be realized that these measurements were not obtained at one fixed location, but rather during the drift of TARA shown in Fig. 1. However, they were all obtained at a location higher than 80° N latitude and hence represent a full year of observations of surface layer air over the frozen Arctic Ocean. The red curve is a smoothed curve of the 5 min averaged valid observations as discussed above; the curve in light grey shows measurements filtered from consideration since the diesel power generator was operating during that time. The data were smoothed since even the 5 min record was often quite noisy with an amplitude occasionally as large as 5–10 nmol mol<sup>-1</sup>. Closer inspection of the full data record indicates that this noise was probably caused by fluctuations in the electrical power at the times that the diesel generator was not in operation. When the generator was operating, the noise decreased to less than 2 nmol mol<sup>-1</sup> (but as mentioned, these data were filtered from the final record).

Figure 2 shows an annual pattern that is to a large extent similar to observations at Arctic coastal observatories (Alert, Barrow and Zeppelin Fjellet (Ny-Ålesund, Svalbard), see Helmig et al., 2007); for comparison purposes we include concurrent observations at Alert. The similarity between the TARA and Alert data is striking. At both locations, typical wintertime ozone mixing ratios (October 2006 to March 2007) ranged between 30 and 40 nmol mol<sup>-1</sup>. During summer (mid-June to mid-September 2007), ozone levels were lower than in winter, with average values on the order of 15–20 nmol mol<sup>-1</sup>. October plays the role of a transitional month between the generally lower mixing ratios in summer and the higher ozone levels in winter. Thus, the seasonal trend of ozone levels in the middle of the Arctic Ocean appears consistent with

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coastal records for winter, summer and fall.

In Fig. 3 we present a closer look at the spring data for 2007. We have included the concurrent observations at the coastal observatories Alert, Zeppelin Fjellet and Barrow. At this time of year the difference between coastal sites and the central Arctic (TARA) appears much more contrasted, with longer and more pronounced ozone depletion episodes at TARA. Remarkably, at TARA the “ozone depletion season” (as defined in the next section) appears to start quite abruptly around mid March, close to equinox, continuing into early June. Coincidentally, at this period in the drift TARA was at its closest to the North Pole (latN/lonE 86.3/133.0, 87.1/130.6, 88.4/108.4 and 88.3/63.2 on 15 March, 15 April, 15 May and 15 June, respectively). Consequently these data were not only obtained exclusively over the frozen Arctic Ocean, but also represent geographically the most Northerly observations to date. In Figs. 3 and 4 we have added the actual 5 min data as open circles. As can be seen the ozone values drop quite rapidly with the advent of sunlight. There are occasional short periods when the ozone mole fraction increases almost to normal background levels of 30–40 nmol mol<sup>-1</sup>, but the prevailing levels are close to zero. In particular between 20 April and 10 May ozone is virtually completely absent. While the detection limit of the TEI049 is nominally 0.5–1 nmol mol<sup>-1</sup>, it is not possible to obtain an accurate number in this case, given the noise in the actual signal. Figure 4a and 4b show concurrent meteorological observations, temperature, wind speed and wind direction, as measured from a 10 m tower. Figure 4a summarizes data from daily back trajectories. In this case each entry pertains to the history of an air parcel arriving at TARA on the date indicated.

## 4 Discussion

Several environmental conditions are necessary for Arctic ozone depletion in the (marine) boundary layer to occur. Although many of the details are still subject of ongoing debate they can be summarized as follows:

1. Presence of bromine atoms, predominantly produced from sea salt bromide ions.

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2. Presence of sunlight. This condition derives from the fact that Br atoms have to be produced from reactions such as



3. Presence of a suitable surface for stimulating the so-called “bromine explosion” (Platt and Janssen, 1995; Wennberg, 1999), translating into ice and snow surfaces highly concentrated in sea salt, such as ice covered by frost flowers. Whether the presence of frost flowers is a necessary condition is a hotly debated topic (e.g. Rankin et al., 2002; Kaleschke et al., 2004; Domine et al., 2005; Simpson et al., 2005; Piot and von Glasow, 2008).

4. Low temperatures. Tarasick and Bottenheim (2002) deduced from ozone sonde records that ozone depletion events seemed to be associated with low ambient temperatures. Based on chemical considerations they hypothesized that temperatures below  $-20^\circ\text{C}$  would favor ozone depletion. Low temperatures may be required to modify the chemical properties of reactive surfaces in a manner able to speed up halogen activation. This includes increased bromide/chloride ratio in seawater-derived media due to precipitation of  $\text{NaCl} \cdot 2\text{H}_2\text{O}$  below  $-22^\circ\text{C}$  (Koop et al., 2000; Morin et al., 2008), and potential reduction in alkalinity of the surface due to carbonate precipitation (Sander et al., 2006; Piot and von Glasow, 2008), although recent investigations have cast doubts on this hypothesis (Morin et al., 2008).

5. Presence of a strong stably stratified boundary layer (Lehrer et al., 2004).

For an extensive review of these aspects we refer to Simpson et al. (2007).

A generally accepted definition of what constitutes an “ozone depletion episode” does not exist. A working definition of a “severe ozone depletion episode” was proposed by Ridley et al. (2003), subsequently adopted by Piot and von Glasow (2008). In this paper we will follow their suggestion and define the “ozone depletion season”

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as the period of the year when severe ozone depletions are observed at a site, with “severe” implying ozone mole fractions below  $5 \text{ nmol mol}^{-1}$  (or 80% depleted from normal, ca.  $40 \text{ nmol mol}^{-1}$  in spring), and with a transition period where ozone is between  $5\text{--}20 \text{ nmol mol}^{-1}$  (>50% deviation from normal). Occasional decreases in ozone of ca.  $5\text{--}10 \text{ nmol mol}^{-1}$  were observed at TARA throughout the year, especially in the winter. Such episodes are also observed at the Arctic coastal observatories. They are due to rapid transport of polluted air masses from mid latitude regions (Harris et al., 2000; Worthy et al., 1994). In fact, at the Alert observatory occasionally an increase in the gaseous mercury concentration has been observed during such episodes which excludes halogen chemistry as the cause for the decrease in ozone (Bottenheim and Steffen, unpublished results, 2003).

As can be seen in Fig. 4, sunrise over TARA occurred on 14 March, and by 29 March there was 24 h sunlight. The late date of polar sunrise was due to the fact that at this time TARA was at almost  $87^\circ \text{ N}$ . Within a week of sunrise the first ozone depletion episode was registered, with a decrease of the ozone mole fraction to almost  $5 \text{ nmol mol}^{-1}$  (21–22 March) and the ozone depletion season had started. This episode lasted about two days and terminated when both ambient temperature and wind speed increased drastically (to  $-8^\circ \text{ C}$  and  $12 \text{ m s}^{-1}$  respectively), and the air mass origin veered towards the South, all indicative of a change in synoptic weather patterns. It is tempting to suggest that there is a correlation and that the depletion was occurring locally with the first rays of sunlight. On March 21 there was already 12 h sunlight, the local wind speed was almost zero, the temperature was ca.  $-30^\circ \text{ C}$  and the air mass origin was from the North (i.e. from the North Pole and beyond to the Canadian Arctic). Trajectory data suggest that an air parcel arriving at 10 m above sea level (a.s.l.) had been close to the surface (arbitrarily defined as less than 200 m a.s.l.) for about 3 days, a relatively short but sufficient period of time for rapid ozone depletion chemistry in a shallow boundary layer (Lehrer et al., 2004). Since we have no additional chemical information it is not possible to be certain about local production. We note that the conditions during this first episode were somewhat unusual in com-

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parison with subsequent observations (temperature at  $-30^{\circ}\text{C}$ , origin from the North). The next depletion started around 29 March, lasting until about 3 April. Subsequently for 2.5 weeks the ozone mole fraction was variable with the occasional recovery from total depletion. Each time the ozone mole fraction recovered there was a concurrent substantial increase in ambient temperature and often wind speed (e.g. 5, 14, 17 and 19 April). Trajectories do not indicate a major change in air mass origin, but suggest that the air mass observed had remained below 200 m a.s.l. for at least 4 days. Note that at this time there was already 24 h of sunlight, and hence these variations can not be due to a day-night effect. On 21 April the longest uninterrupted total ozone depletion period started continuing to 7 May when a partial ozone recovery was observed. The wind speed was quite variable ranging from greater than  $10\text{ m s}^{-1}$  on April 24 to below  $2\text{ m s}^{-1}$  in early May without any indication of a variation in the ozone mole fraction. From 8 to 25 May, ozone remained generally well below  $10\text{ nmol mol}^{-1}$  but was more variable with occasionally total depletion, except for another partial recovery on 11/12 May. Temperature was on average slowly increasing from  $-17^{\circ}\text{C}$  on 21 April to  $-13^{\circ}\text{C}$  on 25 May (Vihma et al., 2008). Trajectory data suggest an average contact with the surface of at least the last 4 days before reaching TARA, and the origin is mostly from the South. The ozone depletion season as defined above came to a close on 25 May. For another two weeks ozone mole fractions varied between  $10\text{--}20\text{ nmol mol}^{-1}$ , until on 11 June ozone increased to  $30\text{ nmol mol}^{-1}$  and no further ozone depletions were observed. During this time the ambient temperature was on average at  $-6^{\circ}\text{C}$ , increasing to about  $0^{\circ}\text{C}$  on 11 June.

Ambient temperature is one parameter that showed qualitatively a good correlation with ozone depletion, often substantially increasing during a temporary recovery of ozone. We do not have vertical profile data, but suspect that these observations are indicative of a temporary break-up of the lower boundary layer which is usually only a few hundred meters deep (Bottenheim et al., 2002). Remarkably, on average the ambient temperature starting mid April was well above  $-20^{\circ}\text{C}$ . Tarasick and Bottenheim (2002) observed that ozone depletions, recorded by ozone sondes at Arctic coastal

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observatories, appeared to be predominantly associated with ambient temperatures below  $-20^{\circ}\text{C}$ . These authors argued on thermodynamic grounds that temperatures below  $-20^{\circ}\text{C}$  would promote ozone depletion chemistry. Piot and von Glasow (2008) in their modeling study did not find such a marked temperature dependence. Clearly more research is required to resolve this issue.

Another way to look at the TARA data is to inspect the associated back trajectory data. One might question the veracity of such data, and we certainly would not want to overstate their use, especially after the first couple of days (Kahl, 1993). Figure 4 indicates that according to the trajectory data air parcels had traveled at a rate of approximately  $300\text{--}400\text{ km day}^{-1}$  or about  $3\text{--}4\text{ m s}^{-1}$  before reaching TARA. It is encouraging to note that this number is in very good agreement with the locally observed wind speed. Furthermore, a cursory inspection of Fig. 4 shows reasonably good agreement between the origin of the observed air mass as suggested by the trajectories and the local wind direction. We therefore feel confident that certainly qualitatively we can use the trajectory data to investigate their recent past of the air observed at TARA.

In Fig. 5 we present a polar plot of the origin of air that contained less than  $5\text{ nmol mol}^{-1}$  of ozone. It can be seen that the majority of severely ozone depleted air originated from the South to Southeast. As mentioned earlier the trajectory data suggest that these air masses had been in contact with the surface for at least the last four days, sufficient time to permit development of the bromine explosion and hence ozone depletion to occur. We inspect satellite data to explore whether they can shed more light on conditions that preceded the observed ozone depleted air. In particular we wish to ascertain whether conditions (1) presence of bromine atoms, and (3) presence of suitable surface conditions were fulfilled.

For the presence of bromine we consulted the daily distribution of BrO over the Arctic derived from SCIAMACHY data, available on the internet at ([http://www-iup.physik.uni-bremen.de/doas/scia\\_data\\_browser.htm](http://www-iup.physik.uni-bremen.de/doas/scia_data_browser.htm)). These images represent the tropospheric column of BrO and hence it is somewhat debatable whether they are a true representation of BrO at the surface (see e.g. McElroy et al., 1999; Hönninger and

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Platt, 2002; Morin et al., 2005). Nevertheless, the SCIAMACHY images for the period between 20 April–10 May 2007 show the presence of BrO clouds with vertical column densities of ca.  $1.0 \times 10^{14}$  molec  $\text{cm}^{-2}$  over the East Siberian Sea, Southeast of the TARA location. While the specific location of the maximum is variable, given the inherent uncertainty in trajectory data, we conclude that the required condition of presence of BrO was likely fulfilled. The maxima of column BrO were in general not coinciding with the location of TARA itself, in particular during the 20 April–6 May total depletion episode. This is in fact as should be expected. If the surface air contains about  $1 \text{ nmol mol}^{-1}$  of ozone (or less), then the BrO concentration should be expected to be low too since ozone is required to sustain elevated BrO concentrations.

The ice surface conditions were probed using sea ice maps derived from QuikSCAT satellite scatterometer data (Nghiem et al., 2005, 2006; Nghiem and Neumann, 2007). These maps show an immense region of first year ice on the Russian side of the Arctic Ocean during the spring of 2007. In 2007, the acceleration of the Transpolar Drift, known as the “Polar Express”, excessively transported sea ice toward the Atlantic sector and finally out of the Arctic across Fram Strait, leaving the Russian side dominated by first year ice (Nghiem et al., 2007). The Transpolar Drift acceleration was actually verified by none other than the drift of TARA itself (Gascard et al., 2008). These conditions suggest that there was an abundance of leads, polynyas, recently refrozen surfaces, thin ice, and frost flowers. Notable events can be seen in these maps:

1. 10 April (Fig. 6a): An elongated feature of open water opened up in the Laptev Sea along the east of the Taymyr Peninsula and stretched to the north all the way to the northernmost tip of the North Land.
2. 18 April (Fig. 6b): The open water feature started on 10 April became a vast polynya, more than 260 km at the largest east-west extent and about 590 km in the north-south direction. Several small polynyas were also detected in the Laptev Sea to north of the Russian coast between the Taymyr Peninsula and the Lena River basin.

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3. 22 April (Fig. 6c): The vast polynya had been refrozen where the satellite scatterometer recorded very high backscatter, even as high as or higher than backscatter of multi-year ice, indicating the formation of a large region of frost flowers in the gigantic polynya.

5 4. 26 April (Fig. 6d): The backscatter in the polynya reduced somewhat but still remained high (as high as backscatter of mixed ice), indicating the frost flowers had been proceeding into the waning phase.

10 5. 2 May (Fig. 6e): The backscatter in the polynya further reduced to the level of backscatter of first year ice. The increase and then decrease of backscatter in the vast polynya is the classic telltale of the frost flower formation and waning process (Nghiem et al., 1997). The backscatter change corresponding to frost flower formation is observable with the low-resolution QuikSCAT data in this case thanks to the enormous size of the polynya.

15 6. 20 May (Fig. 6f): Another vast polynya was well developed in the Laptev Sea from a series of separated polynyas, located to the east of the Taymyr Peninsula, which started to develop on 2 May as seen in Fig. 6e. The high backscatter area to the south of this polynya was caused by larger ice scatterers formed from refrozen ice that underwent a melt event started around 11/12 May, corresponding to the sudden warming episode with a sharp increase in air temperature and a  
20 concurrent ozone rebound as presented earlier.

In summary, this analysis shows that the conditions were highly favorable for the occurrence of bromine explosions and hence fast ozone depletion (Simpson et al., 2007). The timing of the development of the two vast polynyas in April-early May and in late May coincides well with the two major ozone depletion episodes as seen in  
25 Fig. 3. Also noted is the curving feature of mixed ice (marked by “m” in Fig. 6a) at the top of each ice map in Fig. 6, which was a remnant of older sea ice that survived the 2006 summer. This feature drifted northward along the direction of the Transpolar Drift

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Stream in the upstream of the TARA. It is interesting to note that this potential area of origin for the ozone depletion is in reasonable agreement with the so-called “cold spot” area derived from a climatology of 9 years of ozone and concurrent trajectory data from the three main Arctic observatories, in particular Alert and Zeppelin Fjellet (Bottenheim and Chan, 2006). It is therefore tempting to use forward trajectory data to see whether any of the ozone depleted air observed at TARA might have been transported towards these observatories and ozone depletion was observed. The large majority of forward trajectories did not indicate transport to the Arctic observatories. Matching data are suggested for one day transport from TARA on 23/24 April to Alert on 24/25 April. Inspection of Fig. 2 shows that severe ozone depletion was in fact observed for these days at both TARA and Alert. No matching data were indicated for transport to Zeppelin Fjellet.

Bottenheim and Chan (2006) raised the possibility that since the time required for an air parcel to travel from source to receptor point was on average about 5–6 days, the ozone depletion process might not necessarily be fast. The TARA data do not agree with this scenario. Rather, they reinforce the alternate scenario that extensive air masses devoid of ozone are roaming the Arctic Ocean. In other words, as first speculated by Hopper et al. (1998), over the Arctic Ocean surface the norm is more or less complete absence of ozone, alternated by short episodes where increased turbulence results in the breakup of the surface inversion and mixing of ozone containing air with the surface layer air. Such a scenario does fit well with the TARA data, in particular the extended period of virtually no ozone in the surface air. While as we have discussed above there was a preference for the air observed to originate from the Southeast, and this area appears to have been favorable for depletion chemistry, this was not uniformly the case. Also wind speeds and ambient temperature were not at levels that are believed to be conducive towards local ozone depletion chemistry.

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## 5 Summary and conclusion

The TARA expedition to the Arctic in 2006–2008 has for the first time provided a year long record of ozone mole fractions in the surface boundary layer air over the frozen Arctic Ocean. It has confirmed the occurrence of large periods of substantial to total depletion of ozone in the spring after polar sunrise, long predicted to be the case as derived from remote sensing of BrO. Overall the seasonal cycle was very similar to long term observations at Arctic Ocean coastal observatories, but with more prolonged periods of total depletion. Occasional reappearance of ozone was usually associated with a rise in ambient temperature, suggesting mixing downwards of warmer, ozone containing air from above the surface inversion. Severe ozone depletion was only observed when the temperature returned to levels well below  $-10^{\circ}\text{C}$ . With the data in hand it is not possible to distinguish whether locally a depletion process was taking place. However a combination of back trajectory calculations and remote sensing data suggested that frequently favorable conditions for a bromine explosion and hence ozone depletion existed upwind of TARA. If indeed this area was the genesis of the ozone depletion, then the process would have to be fast, as is generally believed to be the case (Hausmann and Platt, 1994). This reinforces the concept that Arctic Ocean surface air will predominantly contain unusually low levels of ozone in the spring. The collection of long term surface data over more areas of the Arctic Ocean, in particular in the spring is required to verify the concept of a largely ozone free surface air Arctic. In the near future the Arctic Ocean will be covered in the spring largely with first year ice, facilitating the formation of leads and polynyas and thus enhancing the out flux of sea salts. This appears to be the likely surface for producing Br and hence leading to ozone depletion. The scenario then emerges that surface air over the frozen Arctic Ocean will be almost completely devoid of ozone in the spring. Implications for the oxidizing capacity of the surface air, surface exchange processes, and the underlying ocean including the marine biology should be expected but are unknown at this time.

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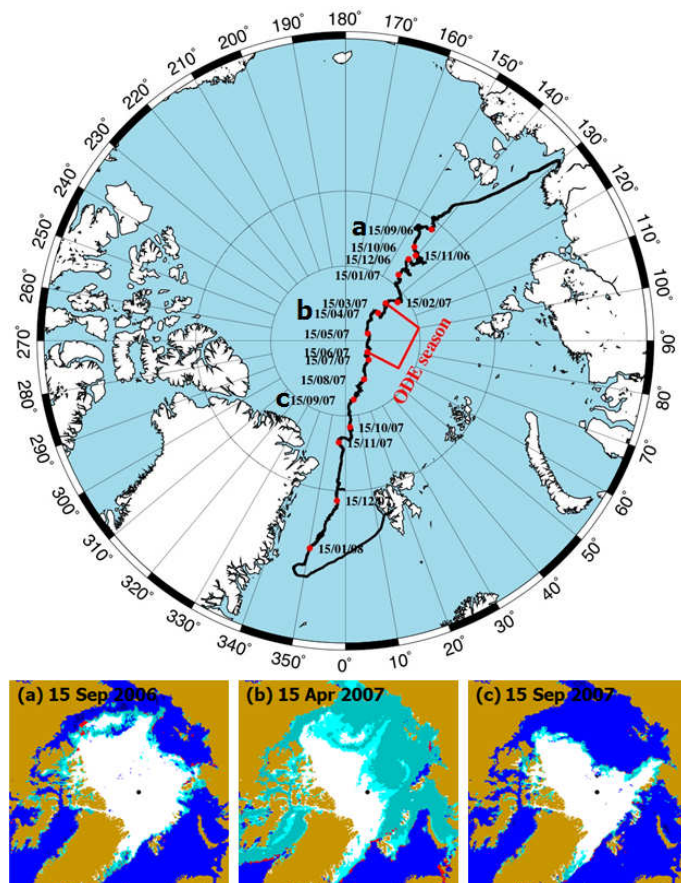
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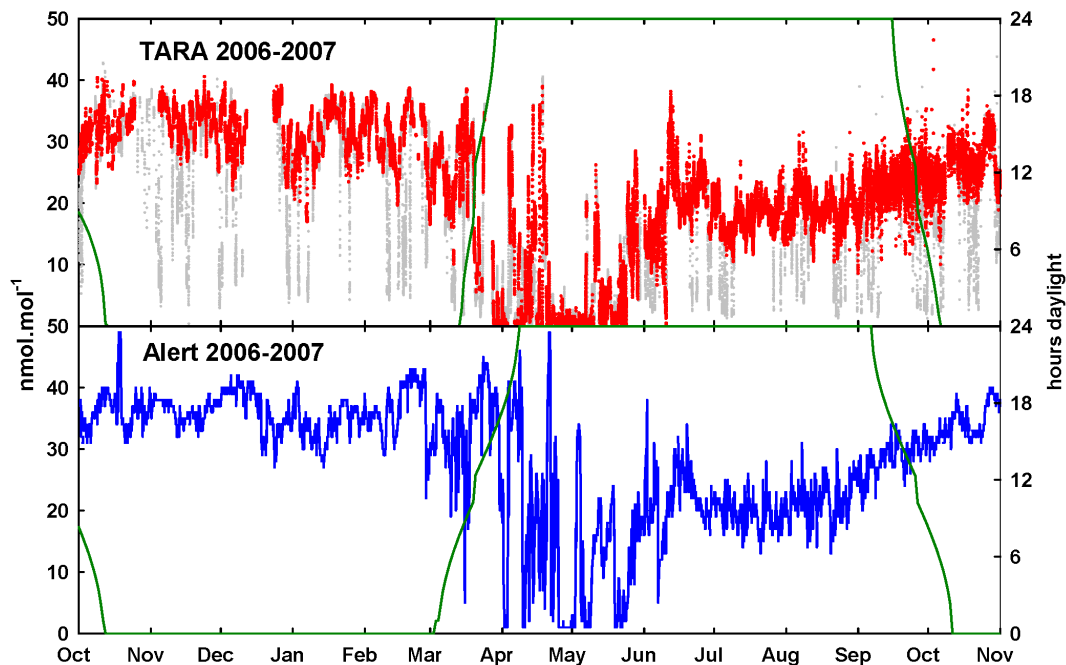


**Fig. 1.** Track of the TARA drift (September 2006 – January 2008). Also shown below is the corresponding sea-ice cover for three relevant dates of the TARA drift (**a** – 15 September 2006, **b** – 15 April 2007 and **c** – 15 September 2007) inferred from QuickSCAT scatterometer measurements.

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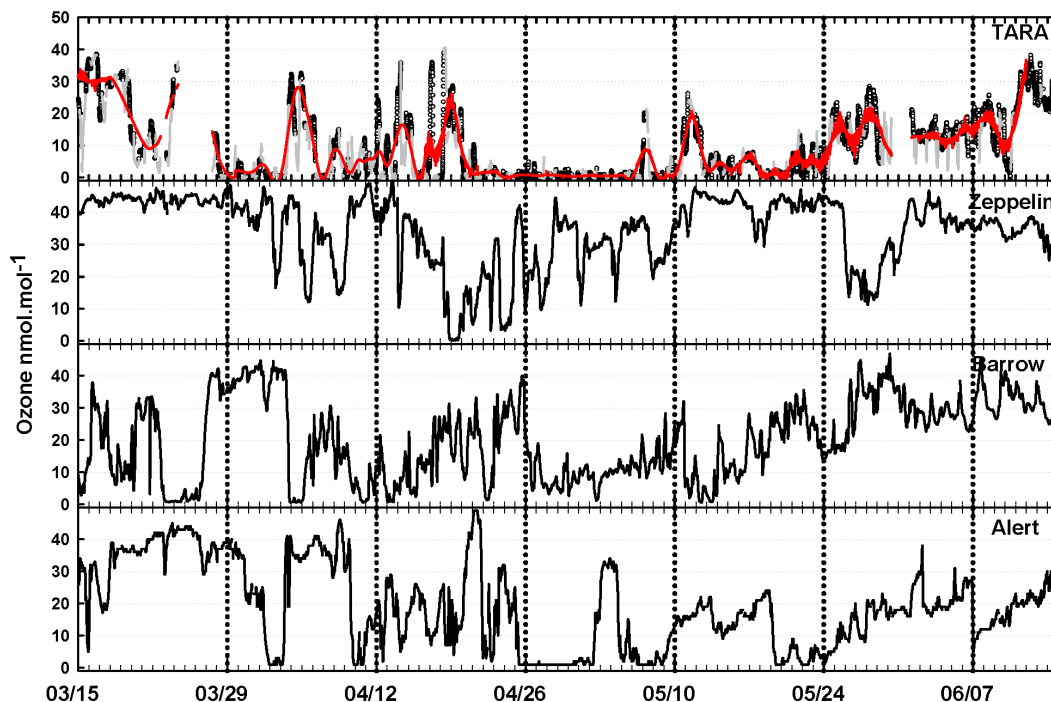


**Fig. 2.** One year of ozone measurements observed over the Arctic Ocean (TARA) and a coastal observatory (Alert, Nunavut, Canada). Top panel: TARA data: red – observations (smoothed) when the power generators were off; grey – observations when a power generator was on; green – hours of daylight. Bottom panel: blue: Alert data, hourly averaged; green – hours of daylight.

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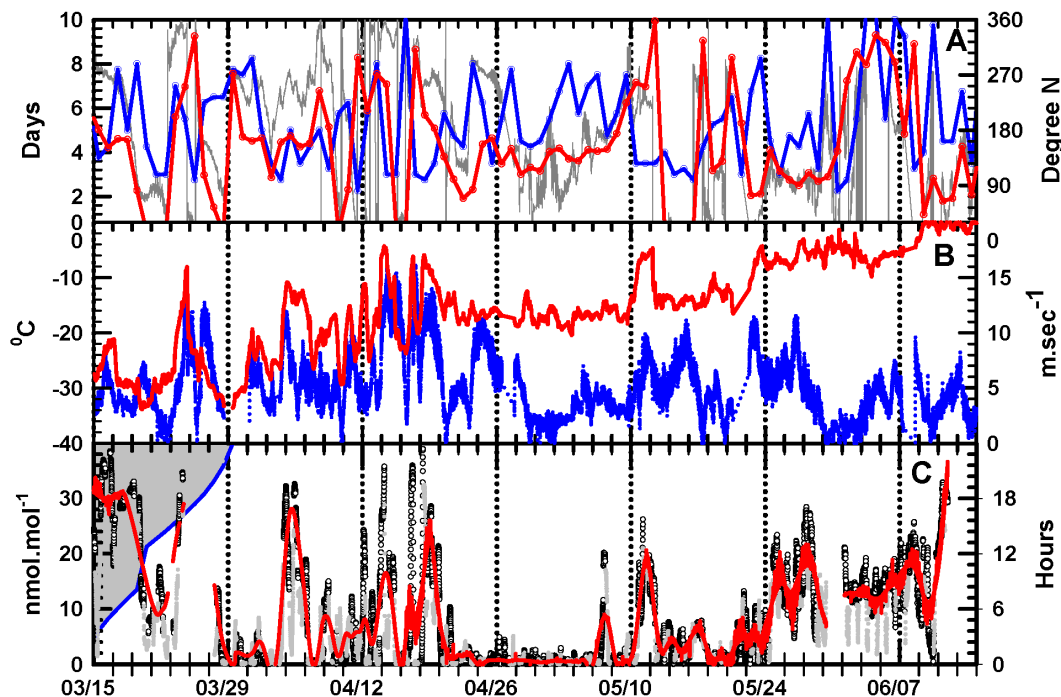


**Fig. 3.** Ozone observations during the “ozone depletion season” (15 March – 15 June 2007). Top panel **(A)**: TARA: red – observations (smoothed) when the power generators were off; black circles – 5 min averaged data when the power generators were off; grey circles – 5 min averaged data when a power generator was operating. Second panel **(B)**: 30 min averaged data at Zeppelin Fjellet (Ny-Ålesund, Svalbard). Third panel **(C)**: 30 min averaged data at Barrow (AK, USA). Bottom panel **(D)**: hourly averaged data at Alert (Nunavut, Canada).

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**Fig. 4.** TARA observations during the “ozone depletion season” (15 March – 15 June 2007). Top panel (A): grey – wind direction at 10 m, blue - number of days back trajectory was below 200 m; red – bearing of back trajectory at 10 m over 1 day. Middle panel (B): blue – wind speed at 10 m; red – temperature at 10 m. Bottom panel: red – ozone smoothed; black circles – 5 min averaged ozone, generator off; grey circles – 5 min averaged ozone, generator on; blue – hours of daylight.

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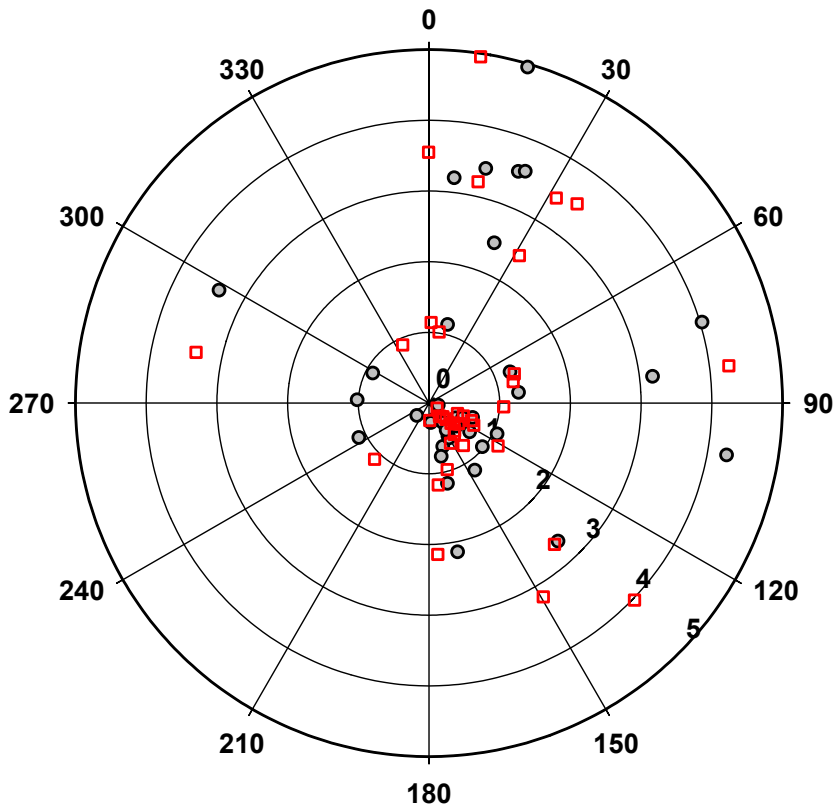
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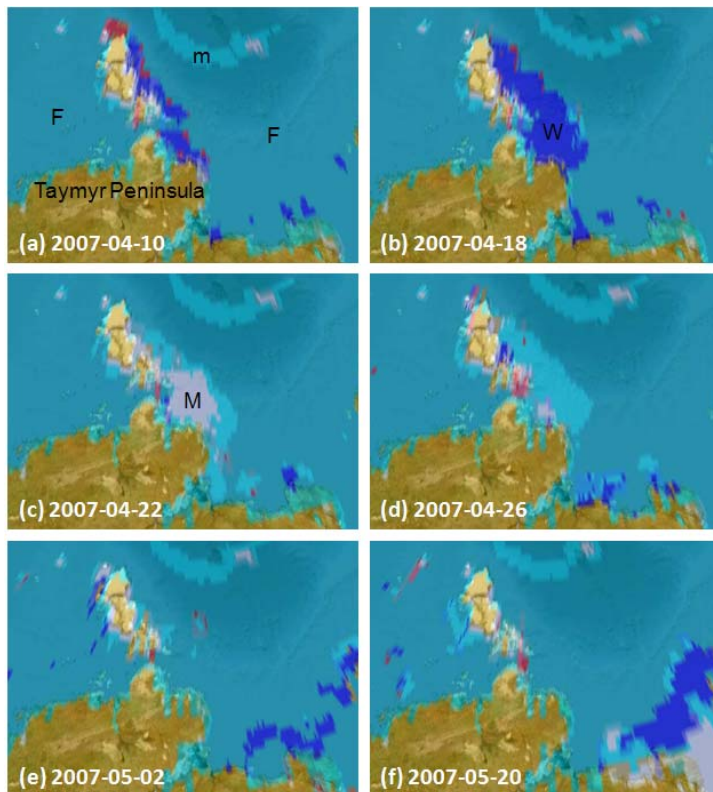


**Fig. 5.** Origin of the observed air during the “ozone depletion season” (15 March – 15 June 2007), from back trajectory calculations. Ozone mole fraction between 0–5 nmol mol<sup>-1</sup>. Black circles – 24 h back; red squares – 48 h back.

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**Fig. 6.** QuikSCAT observations of sea ice in April and May 2007. The color code is: brown for land, blue for open water (W), different shade of cyan for first year ice (F), turquoise for mixed ice (m), and pink for area with backscatter as high as that of multi-year ice (M). The results show the development of a vast polynya along the east coast of the North Land in the north of the Taymyr Peninsula in April and another extensive polynya to the east of the Taymyr Peninsula in May. The polynya seen on 18 April 2007 **(b)** was 84 200 km<sup>2</sup>, which is about the size of Lake Superior (USA, 82 400 km<sup>2</sup>).

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