Atmos. Chem. Phys. Discuss., 10, C4847–C4854, 2010
www.atmos-chem-phys-discuss.net/10/C4847/2010/

Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Vertical structure of Antarctic tropospheric ozone depletion events: characteristics and broader implications" *by* A. E. Jones et al.

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Received and published: 6 July 2010

Response to Reviewer 2

We thank the anonymous referee for comments which we will deal with here in turn.

General comment: "there is a lack of conciseness and I recommend to shorten the manuscript significantly, focussing more on the interpretation of their own (and very interesting) data instead of recapitulating formerly published results."

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Our paper as published on ACPD is 58 pages long, and out of these, 4.5 pages of the Discussion section refer to or discuss other people's previously published work. In terms of balance, we feel that our paper is rightly weighted towards our new results. In terms of "recapitulating" formerly published results, we are building our case by including the very limited number of previous observations as a means of supplementing and extending our observations suite. Had we not done so, we would rightly have been open to criticism. We therefore do not agree that our paper lacks conciseness and warrants a blanket reduction in length. Some adjustments are, however, made in light of the reviewers other comments (see later).

The reviewer comments that it is well known that the frequency and properties of ODEs at coastal sites are distinctly different from offshore conditions, citing work by Bottenheim and Poehler to support his case. We note that both of these studies were carried out over the Arctic Ocean, where meteorological conditions are quite different from over Antarctic sea ice. It is therefore not possible for us to make statements about ODEs within the Antarctic sea ice zone.

He goes on to ask: to what extent the episodic nature of ODEs at coastal sites is determined by transport of bromine activated and ozone depleted air from the ocean to the measurement site, rather than by the stability of the BL. This question is relevant to work reporting ground-based time series observations, but that's not what our paper is doing. The first half of our paper probes the details of boundary layer structure and the link to the vertical structure of ozone depletion evident at low wind speeds. This part really says, if you want to successfully model ODEs observed at a fixed coastal location (which is where the majority of observations are made), you will need to take into account the complex dynamical behaviour of the boundary layer. As our measurements were all made at low wind speeds, they likely reflect local processing (a conclusion based also on previously published work from Halley, Jones et al., 2006). Clearly the stability of the BL, or more correctly, the physical structure of the BL, is highly relevant here. The second part of our paper scales up considerably, and doesn't concern itself with issues of BL stability, rather on air mass transport.

Next question: Is it also possible that low pressure systems do not increase the release of BrO into the atmosphere, but merely increase the probability that air is transported from the sea ice to Halley?

This question encapsulates the reasoning from earlier papers, which argued that a low pressure system associated with loss of ozone at 1 to 3 km was passively transporting air low in ozone towards the observing site. Our view is that low pressure systems can be actively involved in processing air and destroying the ozone. We expand on this in our Discussion section. That low pressure systems increase the release of bromine gases into the atmosphere is substantially supported by the recent paper by Yang et al., ACPD, 2010. In their paper, they model the influence of blowing snow as a bromine source via enhanced production of aerosol. They find considerably improved agreement between their model and tropospheric BrO vertical columns from GOME when this process is included in their model. i.e. the evidence strongly suggests that low pressure systems do not just transport air low in ozone, but that they actively enhance gas-phase BrO. However, as ozone-poor air can of course be advected, and in response to the reviewers concerns, we have included discussion on this in the paper. Specifically, in section 4.2.2 we have added: “Further, ODEs observed under conditions of high wind speeds can arise from transport to the observation site of air masses already depleted in ozone (e.g. Simpson et al., 2007). Thus separating out the role of low pressure systems, into chemically driving depletion as opposed to transporting already-depleted air masses, is no simple task.”

Next comment: It has been shown by Theys et al. (2009) that enhancements in total BrO column can be caused by increases in stratospheric BrO due to a decrease

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in tropopause height at low surface pressure. It is therefore not possible to conclude on tropospheric BrO from satellite measurements of the total BrO column unless the tropopause height and the corresponding variation of the stratospheric BrO column are considered, as done in the study of Yang et al., (2010).

In response to the reviewers concerns, we have been in contact with Nicolas Theys (of Theys et al. 2009) who also derived the tropospheric vertical BrO columns used in the study of Yang et al (2010). Dr Theys derived the tropospheric vertical BrO column for the examples we show in Fig. 19 of our paper (9th Sept 1999, 11th Sept 1999, 23rd Sept 2000), using his climatological data validated using ground-based balloon and satellite limb stratospheric BrO observations. As these examples come early in the satellite record, when pixel sizes were larger, more data coverage was lost during the cloud filtering stage. But it is nonetheless clear that in each case, the structure of BrO enhancements that are evident in the total column are still present in the tropospheric column, with tropospheric column amounts up to 8 x 10^{13} molec.cm^{-2}. The calculated stratospheric column at southern high latitudes is 2.5 to 4.5 x 10^{13} molec.cm^{-2} (Theys et al., 2009), so that the BrO hotspots shown in the BrO total column cannot be attributed to the stratosphere but must indicate a tropospheric source. We have included further discussion in the paper, in Section 5.2, to cover this point:

“Recently it has been shown by Theys et al. (2009) that some enhancement in total BrO column can be caused by increases in stratospheric BrO due to a decrease in tropopause height coincident with low surface pressure. For the cases we have presented here, tropospheric BrO columns (derived using climatological data validated using ground-based balloon and satellite limb stratospheric BrO observations) indicate the same structure of BrO enhancements that are evident in the total column, with tropospheric column amounts up to 8 x 10^{13} molec.cm^{-2} (Theys pers. comm. 2010). Given that calculated stratospheric columns at southern high latitudes range from 2.5

to 4.5 x 10^{13} molec.cm^{-2} (Theys et al., 2009), the BrO hotspots shown in the BrO total column cannot be attributed to the stratosphere and must arise from a tropospheric source."

Next comment, with regards the reviewers concerns (page C2892, paragraph 2) of how we dealt with the Friess et al paper: We were in no way seeking to ignore or insufficiently refer to the results and conclusions of the Freiss et al paper, which, to the contrary, we have always held in high regard. However, we feel that it is interesting to revisit earlier work when more information can be gleaned from it. Perhaps we did not refer to all the details mentioned by the reviewer, but an introduction to the paper and some of its key findings is given in our manuscript on page 8215, starting line 14. Our aim, however, was very simple – to see whether elevated BrO events in the Neumayer region, which had associated ozone profile depletion above 1 km altitude, were associated with low pressure systems during which it was sufficiently windy to achieve lofted snow. These issues were not discussed by Friess et al., and in this way we are building on their work. The point of showing the satellite maps in our paper is to enable easy comparison between the satellite data and the regional meteorological charts. Given the current debate about BrO hotspots as seen in BrO total vertical columns, there is now also an opportunity to explore whether the enhancement discussed also in the Friess et al paper really is in the troposphere. We would also like to point out that the ODE starting on 23rd Sept 2000, shown in Fig 8 of Friess et al, did not correspond to long sea ice surface contact time, so perhaps a different mechanism was active.

Next point, the issue of showing satellite measurements of sea ice: Certainly this was the luxury diagram in the paper. But the visual image makes a much greater impact than a description of X thousand km^2. However, if the editor feels that this figure ought to be removed then we are happy to do so.

Re citation of Saiz-Lopez et al. The reason we didn't include citation to these papers was because we didn't see them as strictly relevant, and didn't want to be accused of over-citing our own work. However, we are of course happy to include them, and have inserted a few additional sentences at the end of section 2.

Specific comments:
Reference to Neuman et al is now included in the Introduction.

P8191L11: We have included the following: The BrO thus formed can then react with HO2 to form HOBr.

P8191L25: actually we are talking here about surface ozone measurements which is implicit in the context. However, we have included an additional sentence explaining that ship-board measurements of halogens have also been carried out.

P8192L12 Reference to Freiss et al is now included.

P8199L22 done

P8202L7 original text: " The strongly positive $\frac{\partial \phi}{\partial z}$ indicates that heat was flowing from the atmosphere into the radiatively-cooling snow surface below. "

New text, " Heat was flowing down a thermal gradient from the atmosphere into the radiatively-cooling snow surface below at about 5 Wm^{-2} as measured by a sonic anemometer at 4 m (not shown). "

Section 4.2.2 In response to the reviewers comment we have included additional information:
Back trajectories were calculated for both of the dislocated events using the online trajectory model of the British Atmospheric Data Centre (fully described at

http://badc.nerc.ac.uk/community/trajectory/). The trajectories were calculated using meteorological fields from the ECMWF at a resolution of 2.5^o, the maximum available for this time period, and were run backwards from Halley for 5 days. In each case trajectories ending at different pressure levels were calculated in order to provide information on the history of air mass at different heights in the ozone profile.

With regards showing the altitude of the back trajectories in Fig 10, because bromine activation should only be expected if the air parcel is in contact with the surface... The evidence presented in the latter half of this paper, together with the results of Yang et al 2010, strongly suggest that ozone depletion does not have to be restricted to reactions occurring at the ground, but can occur on lofted surfaces. In this case, contact with the ground is not a prerequisite. In response to the reviewers request, we now show the altitude of the back trajectories in Figure 10. For 21st September 1987 (Fig 10a, lower panel) the green, turquoise, and blue lines are the trajectories for air parcels arriving at Halley at 900, 850 and 800 hPa respectively (i.e. between $\sim$ 1 km and $\sim$ 2 km above ground level – the altitude region of low ozone air in the profile on this day). None show a trajectory path lower than $\sim$ 900 hPa ($\sim$ 1 km). For 28th September 1987 (Fig 10b, lower panel) the green, turquoise, and blue lines are the trajectories for air parcels arriving at Halley at 900, 850 and 700 hPa respectively (i.e. between $\sim$ 1 km and $\sim$ 3 km above ground level – the altitude region of low ozone air in the profile on this day). Only one shows a trajectory path slightly lower than 900 hPa for a short period of time. In neither case do the trajectories follow a path below the threshold of 100 m used by Friess et al.

P8216L10: In Friess et al, the authors write “Besides snow drift that could provide scattering particles, it is very likely that the light path enhancements are at least partially caused by sea-salt aerosols which are transported from the sea ice surface to Neumayer station.” We have amended our text to read: “The authors suggest that, as well as snow drift, it was very likely that the light path enhancement was at least

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partially caused by sea salt aerosol.” We hope that this is both accurate and not too close to plagiarism.

Figures 3-8: We originally did plot all these figures on the same scale, but found that in so doing, we suppressed the most significant features. In order to glean the relevant information, they need to be plotted on different scales. Similarly, extending the x-axis to zero squashes the data and makes it harder to see the features. To address the reviewers concern, we have included a note in the figure captions of Figs 4-8 that the scales are different.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 8189, 2010.

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