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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Surface ozone depletion episodes in the Arctic and Antarctic from historical ozonesonde records" *by* D. W. Tarasick and J. W. Bottenheim

H. Roscoe (Referee)

h.roscoe@bas.ac.uk

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General comments: This excellent contribution to the body of work on polar tropospheric ozone loss should clearly be published after some revisions. The authors are to be congratulated on their innovative thinking about critical level data as a compression technique rather than a reduction in resolution. Their demonstration of the point with ozone data in their comparisons of their Figures 1 and 2 are particularly compelling.

Specific comments: (note that some of these rely on presentations from EGS-2002 of which only the abstract will be available to the authors; hopefully they are nevertheless useful contributions to the discussion).

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1. The smaller frequency of events at more southerly Canadian sites may be well explained by new measurements and models of BrO and O3 from Hudson Bay (Hoenninger et al. 2002), who found that ozone loss did occur but was rarely complete, and their model showed that the BrO cycled to Br2 in the dark hours that existed their in spring. Typical ozone destruction rates were 0.2 to 0.5 ppbv/hour in daylight, so that local sources of BrO could never destroy all the ozone in the available daylight. Complete destruction could occur much further north, and when this air blew over their station it appeared as an event of complete ozone loss. Further evidence for these slow destruction rates comes from measurements of BrO and O3 from the Dead Sea (Zingler et al. 2002), which showed the amount of ozone lost during BrO enhancements to be about 5 to 10 ppbv. The complete exchange of air masses that occurred during most nights in the Dead Sea valley meant that the process had to restart the following day, with no residual Br2 to be photolysed at sunrise. This was unlike several occasions at Hudson Bay, where despite some mixing, enough residual Br2 remained that some BrO formed within an hour of sunrise and small amounts of further ozone loss took place during a second day of the episode.

2. In Antarctica, GOME data shows that BrO is still present over ice in the Weddel Sea and over the continent. Care is needed, as Fayt & Roozendael (2002) have shown that much of the apparent large amounts of BrO measured by GOME over Antarctica in December is probably in the stratosphere. However, sufficient BrO remains after subtraction of calculated stratospheric amounts that the same mechanism of ozone loss could easily occur in December. If the episodes in Antarctica in Table 1 could be separated by individual months rather than a 3-month average, this would become apparent, as much less excess BrO remains by February, according to GOME measurements.

3. A common ozonesonde malfunction is excess background current, significantly above zero when measuring zero-ozone air during pre-flight tests. If such a sonde is left with zero-ozone air in the laboratory, its output will often decrease to small values during the subsequent 30 minutes to 2 hours. Hence if such a sonde is launched

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and the large value of background current at launch is subtracted from flight data, the ozone near the ground will be approximately correct, but the ozone aloft will be too small, often leading to negative values at the tropopause. Such a malfunction cannot produce false episodes of near-zero surface ozone. Malfunctions which might lead to small amounts at the surface would have to involve loss of response of the sonde element and recovery during flight, but this is again unlikely to lead to sudden steps in the ozone profile. If the authors examined some of the possibly false episodes from Brewer-Mast sondes, perhaps their details would shed light on this. But in fact Table 1 lends little support for the assertion in the text (p344 line 25) that BM sondes produce more episodes outside the spring period, which must therefore be presumed false. For example, outside MAM, Resolute has 16 episodes out of 625 flights from ECC sondes, and 11 episodes out of 433 flights from BM sondes. Churchill has a rather more convincing difference, but the assertion is overstating the case and should surely be toned down.

4. Freiss et al. (2002) showed that at Neumayeur there were a large number of ozone-loss episodes in 1999 in surface-ozone data and a significant number in 2000, by examining the continous record of the surface ozone monitor, as opposed to the ozonesonde flights which were about twice a week.. The fact that there are so few episodes in Table 1 suggests that either there is a sampling problem in the sondes, unlikely as their frequency is increased in the spring during the ozone hole period, or there is something amiss in the identification of episodes in the Neumayeur data. Is it possible that the identification of critical levels by hand in ozonesonde output is being done poorly there, because the operators normally rely on the high resolution digitised data? Is it possible that the operators are recording ozone at critical levels in temperature, rather than at critical levels in ozone?

5. In the Canadian Arctic, the fact that short-term ozone loss episodes occur coincident with short-term colder temperatures is surely the result of transport of ozone-poor air from higher latitudes, as sometimes found by Hoenninger et al. (2002). If so, it is not

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relevant to the discussion of whether or not the frequency of episodes might increase if there is a long term increase in temperature due to increased greenhouse gases. We could speculate about the cause of the observed increase in frequency of ozone loss episodes. For example, increased greenhouse gases have very likely increased the number of open water leads within the Arctic pack ice at the important sunrise period. It is the freezing process within these leads, when winds are light, which forms the frost flowers that are likely to be the source of the enhanced-sea-salt aerosols responsible. It would be difficult to confirm such speculation now, as current climate models lack sufficient accuracy in sea-ice description to allow a firm conclusion, and they contain no description of frost-flower formation; but future models will hopefully include these processes.

6. The equilibria of sea water with brine continue to change all the way down to -52°C, when about 0.2 % remains as liquid (Richardson 1976). There is no threshold at -20°C, as the authors seem to claim in the last paragraph of Section 3. There is a different regime at about -22.5°C (Richardson 1976, Figure 7), but this seems to be related to the temperature at which freezing of Na2SO4.10H2O is almost complete (Richardson 1976, Table I). Certainly there is nothing special about Na or Cl ions at -20°C; sadly Br ions are not listed in this work. An alternative possibility is that the enhanced sea-salt aerosol comes from frost flowers as the authors suggest, that it takes about 48 hours for frost flowers to grow at temperatures below -20°C, that their growth is much slower at higher temperatures (Martin et al 1995, 1996), and that storms which close up the open water happen every few days. Furthermore, although the details of frost flowers are far from conclusive, there is evidence that they are formed direct from water vapour as hoar frost (Perovich & Richter-Menge 1994), so that when young they are pure water, not saline at all. But because sea water has no density minimum above the freezing point, ice forms below the surface before trying to float, which creates pools of slush, and as more freezing occurs brine is expelled to the top of the pool as well as downwards (Martin et al 1995). It is thought that the porous frost flowers then wick up the brine, eventually becoming much more saline than sea water and

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very much more saline than equilibrium sea ice (Rankin et al. 2000). This scenario is supported by measurements by Rankin (2001), who found one frost flower sample with less salinity than sea water, others with more, but all were equally fractionated relative to sea water. The whole process may therefore take 2 to 3 days below -20°C, and much longer above. Hence it is the time that is critical: when warmer the process takes longer, and when warmer there are more storms in the vicinity so there is less time to complete the process.

7. Section 1 ignores my own modest contribution to the study of polar tropospheric ozone loss (Roscoe et al 2001), and that of Kreher referenced therein. More recently, Udo Friess has defended a PhD thesis which has substantially advanced our knowledge of episodes of Antarctic tropospheric ozone loss - hopefully he will be able to contribute to the discussion of this paper via ACPD.

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Technical corrections:

P343 l25&26 - should state the nature and size of the correction for known bias, so we can assess the importance of its being done accurately.

Section 3, last paragraph, last sentence - given my comment in point 6 above, this is overly simplified, and by linking the phrases "highly saline" with "young sea ice" it may be confusing, even if technically correct by comparison to 1 month or 1 year old sea ice.

Throughout - capitalisation of Arctic and Antarctic seems to be a little random.

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