

Interactive comment on “Temporal and spatial characteristics of ozone depletion events from measurements in the Arctic” by J. W. Halfacre et al.

Anonymous Referee #1

Received and published: 13 January 2014

Review of the manuscript (manuscript number acp-2013-780) for ACP, entitled: "Temporal and spatial characteristics of ozone depletion events from measurements in the Arctic" by J.W. Halfacre et al. MS No.: acp-2013-780

The manuscript reports on ozone and BrO measurements made by a series of buoy – based instruments on the Arctic sea-ice and describes a first analysis of ozone data recorded during spring of 2009 through 2011 with one or two buoys being operative simultaneously. These innovative measurements on the arctic sea ice are the first of their kind and give new and interesting information on the related phenomena of ozone depletion and bromine release in the polar lower troposphere. In particular frequencies

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of major and “ordinary” ozone depletion events (ODEs) are reported, also O₃ decay rates at the beginning of the events and correlations of ODE frequency with wind speed and temperature. These results are definitely worth publishing and are well within the scope of ACP. My main criticism with the manuscript is that it is much too long in comparison to the - definitely valuable - new data and findings it presents. Also the clarity of the arguments should be improved (see below). In this reviewer’s opinion the manuscript would benefit from shortening, for instance some of the material could be deleted without affecting the scientific value of the manuscript; other parts could be moved to supplementary material:

1) The ODE definition (section 2.2) could be presented in a much more compact way by saying that ODEs and MODEs are defined by O₃ falling below thresholds of <15 ppb and 10 ppb, respectively, for > 1 hour. Starting times are defined by O₃ levels falling below 90% ... (ODE) or below the threshold level, respectively while stop times ... All remaining information (including the review of ODE definitions by other authors) could be transferred to the supplementary material.

2) Section 2.4 could also be shortened by saying that there are basically two (extreme) explanations for ODEs: (1) Advection of already depleted air (dynamic hypothesis, DH), (2) in-situ chemical destruction (chemical hypothesis CH), of course also combinations are possible (and in fact likely). Throughout the manuscript these two hypotheses are frequently mixed, which is rather confusing. For instance all arguments made about the size of ODEs rest on the DH, while the discussion about measured BrO-levels being too low assumes the CH being correct. The DH and the CH in their pure form are mutually exclusive (unless one assumes some combination, but this is not attempted in the manuscript) and this should be clearly said. Since it may be impossible from the data to decide which hypothesis is correct it is of course warranted to study both under the headlines “assuming the DH being correct we can conclude ...” (e.g. conclusions about the spatial extent of ODEs can be drawn) and “assuming the CH being correct we can conclude ...” (e.g. about the level of BrO and other halogen species), respectively.

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3) Section 2.4, Monte Carlo “Experiment”: The justification and usefulness of the Monte Carlo study (or numerical experiment) does not become clear, in particular, why do the Monte Carlo numerical experiments “provide statistical support” (page 30246, line 6) to the DH? The description of the Monte Carlo numerical experiments could be deleted altogether or moved to the supplementary material. Likewise Fig. 9 does not appear to provide much information and could be deleted or moved to the supplementary material.

4) Section 3.1, On page 30249 the authors state that the measured BrO levels lead to an underprediction of the rate of O₃ loss by a factor of 3.6 (on average). Is this finding not a clear indication that the CH is wrong and the DH correct? This point should be discussed

5) Page 30252 and 1st para of page 30253: The attempt to “potentially test for missing chemistry” should be deleted in view of the fact that the CH is probably not correct (see point 4, above).

6) Section 3.3 describes interesting conclusion, it is convincingly written and should be retained, but shortened. For instance the text on page 30258, lines 14 to 24 could be replaced by saying that the same analysis as for the T-dependence was performed for wind speed.

Minor points: 1) Abstract: The changes in the main body of the manuscript (e.g. DH vs. CH discussion) must be reflected in the abstract

2) Page 30236, lines 9ff: “the prominent regional tropospheric oxidation pathways ... other than OH radicals, notably ...” What is the evidence for this statement?

3) Page 30236, line 21: R4 is not destroying O₃ (the O₃ consumed by Br+O₃ is regenerated by the photolysis of ClO making R4 part of a null-cycle. However the other two product channels of the BrO + ClO reaction lead to O₃ destruction.

4) Page 30243, line 7: Detection limits for BrO between 2-4 E13 molec./cm² are

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quoted, this does not seem to fit with a stated noise level of the measured BrO-column density of 4 E13 molec./cm². The detection limit is usually taken as twice or three times the noise level.

5) Page 30246, line 9: Why are the depletion regions assumed to be circular? The satellite observations clearly show that they are not.

6) Page 30253, section 3.2: Could one not just simply say that the diameter $D_{ode} = v_{wind} \times t_{ode}$ (with v_{wind} = average wind speed, t_{ode} = ODE-duration)? However, this assumes that the (circular) ODE is blown across the measurement site in such a way that the centre of the ODE crosses the buoy. If just a secant crosses, then the above D_{ode} is just a lower limit to the true diameter of the ODE! Likewise, if the ODE is not circular, its area might be overestimated by calculating it as $0.25 \times D^2 \times \pi$. These points should be discussed.

7) Section 3.1: When the DH is correct (which is likely, see above) then not only the O₃ depletion times are interesting but also the O₃ recovery time scales should be analysed.

8) Page 30248, line 13: did Morin et al 2005 really observe O₃ depletion within 3min?

9) Page 30249, Eq. (3): this calculation and the assertion that BrO + HO₂ dominates over BrO + BrO only rests on Stephens et al. 2013b “in prep.” The arguments used by these authors can not be verified by the reader, therefore this part (including Eq. (3)) should either be explained or removed.

10) Page 30256, Sentence starting in line 24 is redundant and should be deleted.

In summary, I feel that this manuscript reports important new data and interpretations relevant to Arctic boundary layer chemistry; however there are a number of points (see above) that need clarification before it can be published, also the manuscript would gain considerably by shortening it.

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