Authors' response

gmd-2017-126-RC1-supplement (2 August 2017)

We thank the anonymous referee #1 for the comments regarding our paper. We appreciate suggestions for further studies. Nevertheless, we would like to stress, that the main purpose of this paper is providing a proper description and reference regarding the implementation of the bromine explosion mechanism in EMAC. Although we compare modeling results with observational data, we do not study the mechanism, which had been proposed by Toyota et al. (2011), in detail.

- General comments:

  - Examination of TIMING of the bromine release mechanism for better understanding of the proposed/applied mechanism: Get corresponding model BrO VCD according to satellite overpass time for chosen sites. Scatter plot based on whole year data as in Yang et al. (2010). For better comparison, a lead-lag relationship can be used. We much appreciate the proposal of further statistical analysis regarding temporal coincidence of BrO enhancements comparing GOME satellite observed VCD and our modeling results. The modeling data will indeed allow for a variety of studies, e.g. temporal or spatial correlations as proposed by the referee. We would like to address these in follow-up studies. Here, as stated above, we intent to focus on a proper deception of the mechanism and its implementation into EMAC to serve as reference. A detailed validation of the mechanism in comparison to observation is beyond the scope of the present manuscript. Closely following the work of Toyota et al. (2011), we are able to show that the mechanism works astonishingly well without any change of parameter or fine-tuning to our model. We provide here a figure (Fig. 1) of BrO VCD at the sites which had been chosen for ODE. However, while this provides some comparison of timing of BrO enhancements, we acknowledge that this is not a proper validation and choose not to include this figure in our manuscript. To assess the contribution of bromine explosions to the BrO VCD, we subtracted the reference simulation (BrXplo_ref) from model integrations including bromine explosions (BrXplo_fysic, BrXplo_mysic, and BrXplo_mysic_rs). A computed zonal mean BrO VCD has been subtracted from GOME tropospheric VCD to highlight bromine explosion events. Satellite data, however does not allow for assessing the most interesting dates in northern and southern hemispheric winter where model results show a strong enhancement of BrO and ODE not present in surface ozone observation. We find a general but not strict temporal agreement in case of Barrow in spring-time. In late April and early May, we do not find BrO enhancements at Alert. Since our modeling results have not shown the long-lasting 2000s ODE at Alert this was to be expected. As pointed out by Strong et al. (2002), this long-lasting depletion event was related to transport of ozone poor air originating from sea ice. It is not clear whether transport or depletion is too weak in our simulation. Some better agreement is found in case of Zeppelin Mountain. At Neumeyer Station we probably find a coincidental event in late September. If ODE are qualitatively well reproduced in comparison with observation, we do also find coincidental BrO enhancements. But studying these in detail is well beyond the scope of this paper.
  - Missing sea spray acting as a bromine source. Why this kind of source is not included in the EMAC? A discussion covering this issue should be given. In general, an emission of tracers from sea spray could be included in EMAC. Since we focus on the implementation of a simple bromine release mechanism from sea ice, we have not considered sea spray on purpose. However, as pointed out by the referee, we shall include a discussion about this matter in the revised manuscript.

- Specific comments:

  - P2 L1: a review paper by Abbatt et al. (2012) should be cited here. We thank the referee for suggesting to include a reference to the work of Abbatt et al. (2012).
  - P2 L22: removal the pair of bracket in "(boundary)" By putting brackets in "(boundary)" we intended to acknowledge the capability of EMAC treating input data at *any* given level not only at the boundary layer as source of emission. We removed the brackets and made our point clearer: *[...] concentrations of tracers at the boundary layer or any other given level [...]*

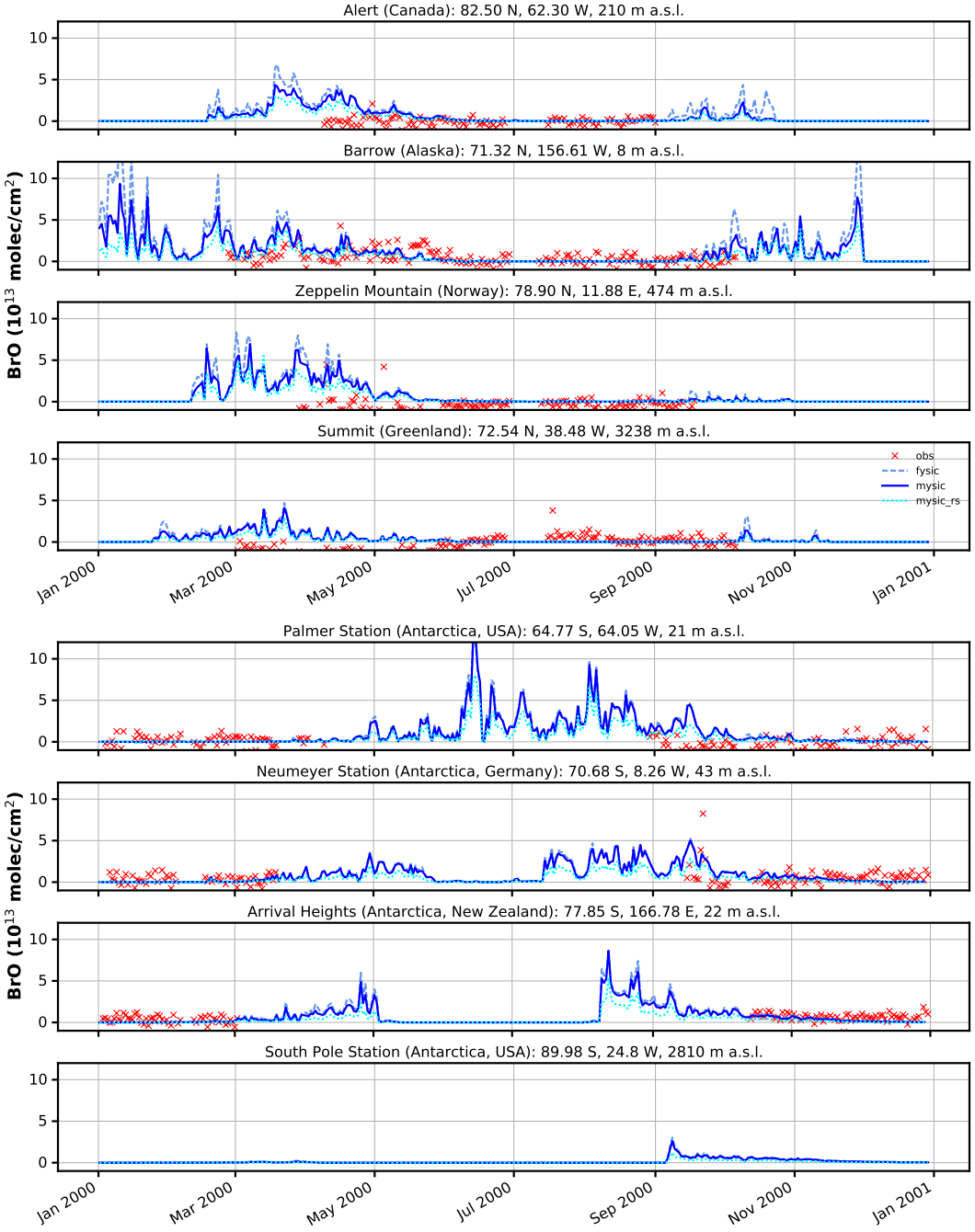

Figure 1. BrO VCD evaluated at observation sites on the northern southern hemisphere. The same as for surface ozone comparison have been chosen. The reference simulation is subtracted from the shown modeling results. GOME tropospheric VCD is shown subtracted by its zonal mean to emphasize BrO enhancements.

- P2 L23: why italic "online" is used here? The comment is absolutely valid. For there is no specific reason, we removed the italic font.

- P3 L20–21: Some discussions should be given to explain why such as a higher value (7.5%) of molar yield at solar zenith angle $> 85°$ (comparing to 0.1% at dark) is introduced in the model, though this number is from Toyota et al paper. This parameter is one critical parameter to allow enough bromine releasing from snow to match the observation. Either a justification, e.g. reference, or a caution must be given to remind readers of what is going on here. It has to be indeed remarked that these values are of importance to the amount of Br_2 released by the mechanism. As pointed out, the values of Φ_1 have been taken from Toyota et al. (2011) and have not been *tuned* to our model. In Section 3.1, Toyota et al. (2011) describe in detail how they obtain the specific value of 0.075 though cross-validation with observed spring-time ozone boundary layer values at Alert, Barrow, and Zeppelin. We add this reminder: *The specific value of Φ_1 has been cautiously obtained as best choice by cross-validating modeling results with observed spring-time boundary layer ozone data at Alert, Barrow, and Zeppelin (Toyota et al., 2011, Section 3.1).*

- P6 figure 2 and P7 L1–2: is the EMAC BrO VCD shown here a total of tropospheric and stratospheric BrO? If so, then a tropospheric column value should be worked out to make a direct comparison with satellite-based tropospheric BrO. We have indeed compared *tropospheric* GOME BrO VCD with *total* BrO VCD of our model simulation. Total GOME BrO VCD have now been provided by courtesy of Andreas Richter (University of Bremen). We update all figures and comparisons in the corresponding section accordingly.

- P7 L1–9: as mentioned in the general comment, just a spatial comparison for BrO is not good enough, a temporal comparison between daily satellite BrO VCD and corresponding model BrO should be given here to allow a further examination of the bromine releasing mechanism applied. The purpose of our current paper is not to examine the release mechanism originally proposed by Toyota et al. (2011), but to implement the mechanism in our model. The suggested comparisons may be subject to further, more detailed studies.

gmd-2017-126-RC2-interactively (4 August 2017)

We would like to thank the anonymous referee #2 for suggesting further important literature and elaboration of sections.

– Major comments:

- How is bromine recycling on aerosol treated? Is this important to sustain halogen activation and does it contribute to ozone depletion events? Bromine recycling on aerosols is treated in the same way as it is for polar stratospheric clouds (PSCs). In these cold regimes, icy surfaces allow or accelerate reactions which are impossible or rather slow in the gas phase. For sustaining catalytic ozone depletion, the activation of halogens through heterogeneous reactions is very important. The set of heterogeneous reactions involving bromine and chlorine used in our simulation is given in the updated supplement.

- The authors should look deeper into the literature as to how the understanding of halogen chemistry in the Arctic & Antarctic has developed over time. Papers such as Barrie et al. (1988) and Abbatt et al. (2012) should not be omitted from the reference list. In addition, Simpson et al. (2007) provides an excellent overview of how our understanding of halogen chemistry and ODEs has developed. We much appreciate the suggestion of these important papers and take them into consideration in our revised introduction.

- A clearer discussion of how snow contributes to halogen activation is needed, as discussed by Pratt et al. (2013) and Thomas et al., 2011. Thank you for pointing this out. We will include a discussion of halogen activation on snow in our revised manuscript.

- A list of the reactions that are included to describe the halogen cycle is needed either in the paper or in the supplement, including a short discussion of how heterogeneous reactions on aerosols are treated. A list of heterogeneous reactions as implemented in the model have been added as supplement. A discussion will be included in the revised introduction.

- In general, I find the discussion of the results too short. Major features of the figures are not really described, which leaves the reader a bit lost as to what the model validation section means. For example, why is the surface ozone so low in the model compared to the measurement sites in Antarctica (Neumeyer and South Pole Stations)? If the model is so poor at predicting background ozone, does it make sense to evaluate the contribution of halogens to ozone depletion events in this region? Since the description and discussion of the figures and results may be indeed slightly too brief, we will add a more thorough description of the plots and their features. Regarding the prediction capabilities of surface ozone in Antarctica, although the model prediction is systematically below observation in the southern hemisphere and Greenland, it is appropriate to qualitatively look at the occurrence of ODEs there and how Antarctic ODEs are reproduced by this simple mechanism. There may be missing sources of ozone emission from the snowpack itself which are currently not implemented in EMAC. However, the intention of this manuscript is to describe the implemented bromine release mechanism, not a general validation of the model performance.

- In the Antarctic, another source of bromine activation that has not been included here may be more important (from sea-salt aerosols formed form blowing snow, (Yang et al., 2010)). The authors should discuss more clearly the implications for not included this mechanism, which may be included in a future study. We acknowledge the work by Yang et al. (2010) and the importance of the blowing-snow that has been neglected in our model so far. We will include this in the revised discussion. Bearing in mind that the release of sea salt aerosols is not included in our model simulations, we believe that it is nevertheless instructive to test by how much the Toyota et al. (2011) mechanism can explain bromine enhancements in Southern Hemisphere high latitudes.

– Minor comments:

- Abstract – "Most likely, they are related to events of boundary layer enhancement of bromine." This statement doesn't accurately reflect our understanding of boundary layer ozone depletion events, suggest to take out "Most likely". We follow the suggestion of the referee.

- P1 L13: "Events of near-complete depletion of polar boundary layer ozone are observed frequently during spring-time over both hemispheres (Oltmans, 1981; Bottenheim et al., 1986, 2002, 2009)". I expect to see Barrie et al. as a main reference in this reference list. We have included a citation of the important work by Barrie et al. (1988).

- P5 L28: This sentence should be combined with next paragraph to avoid having a one sentence paragraph. We follow the suggestion.

References

Abbatt, T. P. D., Thomas, J. L., Abrahamsson, K., Boxxe, C., Granfors, A., Jones, A. E., King, M. D., Saiz-Lopez, A., Shepson, P. B., Sodeau, J., Toohey, D. W.and Toubin, C., von Glasow, R., Wren, S. N., and Yang, X.: Halogene activation via interactions with environmental ice and snow in the polar lower troposphere amd other regions, Atmos. Chem. Phys., 12, 6237–6271, doi:10.5194/acp-12-6237-2012, 2012.

Barrie, L. A., Bottenheim, J. W., Schnell, R. C., Crutzen, P. J., and Rasmussen, R. A.: Ozone destruction and photochemical reactions at polar sunrise in the lower Arctic atmosphere, Nature, 334, 138–141, doi:10.1038/334138a0, 1988.

Bottenheim, J. W., Gallant, A. G., and Brice, K. A.: Measutements of NOy Species and O-3 at 82-Degrees-N Latitude, Geophys. Res. Lett., 13, 113–116, doi:10.1029/GL013i002p00113, 1986.

Bottenheim, J. W., Fuentes, J. D., Tarasick, D. W., and Anlauf, K. G.: Ozone in the Arctic lower troposphere during winter and spring 2000 (ALERT2000), Atmos. Environ., 36, 2535–2544, doi:10.1016/S1352-2310(02)00121-8, 2002.

Bottenheim, J. W., Netcheva, S., Morin, S., and Nghiem, S. V.: Ozone in the boundary layer air over the Arctic Ocean: measurements during the TARA transpolar drift 2006-2008, Atmos. Chem. Phys., 9, 4545–4557, 2009.

Oltmans, S. J.: Surface Ozone Measurements In Clean-Air, J. Geophys. Res.-Oceans Atmos., 86, 1174–1180, doi:10.1029/JC086iC02p01174, 1981.

Pratt, K. A., Custard, K. D., Shepson, P. B., Douglas, T. A., Pöhler, D., General, S., Zielcke, J., Simpson, W. R., Platt, U., Tanner, D. J., Huey, L. G., Carlsen, M., and Stirm, B. H.: Photochemical production of molecular bromine in Arctic surface snowpacks, Nat. Geosci., 6, 351–356, doi:10.1038/NGEO1779, 2013.

Simpson, W. R., von Glasow, R., Riedel, K., Anderson, P., Ariya, P., Bottenheim, J., Burrows, J., Carpenter, L. J., Friess, U., Goodsite, M. E., Heard, D., Hutterli, M., Jacobi, H.-W., Kaleschke, L., Neff, B., Plane, J., Platt, U., Richter, A., Roscoe, H., Sander, R., Shepson, P., Sodeau, J., Steffen, A., Wagner, T., and Wolff, E.: Halogens and their role in polar boundary-layer ozone depletion, Atmos. Chem. Phys., 7, 4375–4418, 2007.

Strong, C., Fuentes, J. D., Davis, R. E., and Bottenheim, J. W.: Thermodynamic attributes of Arctic boundary layer ozone depletion, Atmos. Environ., 36, 2641–2652, doi:https://doi.org/10.1016/S1352-2310(02)00114-0, http://www.sciencedirect.com/science/article/pii/S1352231002001140, Air/Snow/Ice Interactions in the Arctic: Results from ALERT 2000 and SUMMIT 2000, 2002.

Toyota, K., McConnell, J. C., Lupu, A., Neary, L., McLinden, C. A., Richter, A., Kwok, R., Semeniuk, K., Kaminski, J. W., Gong, S. L., Jarosz, J., Chipperfield, M. P., and Sioris, C. E.: Analysis of reactive bromine production and ozone depletion in the Arctic boundary layer using 3-D simulations with GEM-AQ: inference from synoptic-scale patterns, Atmos. Chem. Phys., 11, 3949–3979, doi:10.5194/acp-11-3949-2011, 2011.

Yang, X., Pyle, J. A., Cox, R. A., Theys, N., and Van Roozendael, M.: Snow-sourced bromine and its implications for polar tropospheric ozone, Atmos. Chem. Phys., 10, 7763–7773, doi:10.5194/acp-10-7763-2010, 2010.