Summary of changes in the revised manuscript

Following the reviewers' suggestions, we are now comparing anomalies of vertical column densities of BrO between GOME and our EMAC simulations in Fig. 4, rather than with the derived GOME-SLIMCAT tropospheric BrO product. In addition to the anomaly plots highlighting the bromine explosion events, we provide comparisons of zonal mean absolute BrO vertical columns densities as supplementary information.

We appreciate the reviewers' suggestions to look at a statistical correlation of observed and measured BrO VCDs on a day-to-day basis, but unfortunately we don't have the daily data at hand to do a meaningful comparison here. Instead, we have extended the comparison between modelled and measured surface ozone including a statistical (lag-)correlation at selected stations.

We have also included a number of specific changes that we list in the following, section by section. The detailed text changes are color coded in the revised manuscript attached. Wording has been corrected where pointed out by the reviewers. Also some typographic mistakes have been corrected.

Section 1

- Following the suggestions of the reviewers additional citations, e.g., Barrie et al. (1988) and Abbatt et al. (2012), have been included.
- Based on the review article by Abbatt et al. (2012), we have extended our introduction regarding similarities between polar boundary layer heterogeneous chemistry and PSCs
- as well as a more detailed description of the prominent bromine release mechanism categories (frost flowers, bulk ice and snow, blowing snow, snowpack chemistry).

Section 2

- We have added a remark on similarities in heterogeneous chemistry between the polar boundary layer and PSCs in accordance with Section 1.
- We provide a list of bromine related heterogeneous reactions included in MECCA as Supplement S.1.
- We have adding more detail about the software structure and a remark on the used heterogeneous chemistry.

Section 3

In accordance to the reviewers' comments we have made major revisions to this section:

- We have extended the description and discussion of the results of both BrO and O_3.
- Due to this extension, we have decided splitting the section into two subsection for comparison of total BrO VCD and ODE, respectively.
- Previously, we compared modeled total VCD with tropospheric VCD of GOME. By courtesy, Andreas Richter (University of Bremen) provided us with monthly averaged total BrO VCD of GOME. To avoid uncertainties due to SLIMCAT retrieved tropospheric columns in case of GOME date as well as additional uncertainties by estimating modeled tropospheric VCD, we have chosen a comparison of GOME and EMAC with respect to a total BrO VCD. This also allows for an analysis as unbiased as possible for the datasets are most similar and can be further processed in the exact same way.
- Spatial comparison (Fig. 4) of observation and model is now displayed as anomalies with respect to the zonal mean emphasizing the BrO hotspots.
- Accordingly, the description of the results has been extended.

– Additionally, for both, observation and model (BrXplo_ref, BrXplo_mysic), we have computed zonal means (Supplement S.5). We show that the overall model performance regarding VCD of BrO is is improved by applying this simple bromine release mechanism.

– The chosen data, however, does not provide a temporal resolution better than 1-monthly. For addressing the reviewers' comments regarding a temporal correlation of events between observation and model, we have chosen our ozone data that is available in 1-hourly resolution.

– We have computed correlation coefficients at Barrow between surface ozone observation and two model integrations. In Fig. 7, the correlation is shown as binned 2D-histogram. We provide additional plots for the stations in the northern hemisphere as Supplement S.7. Text has been added accordingly.

– We have added a remark about the recognized ODE at Alert in 2000, which is absent in our modeling results.

– We have appended the discussion regarding further release mechanisms such as sea spray or blowing snow has been.

– Results of the additional sensitivity study with reduced ozone dry deposition has been moved to this section.

Section 4

– The section has been extended in accordance to the above changes.

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 [...]*
- 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.

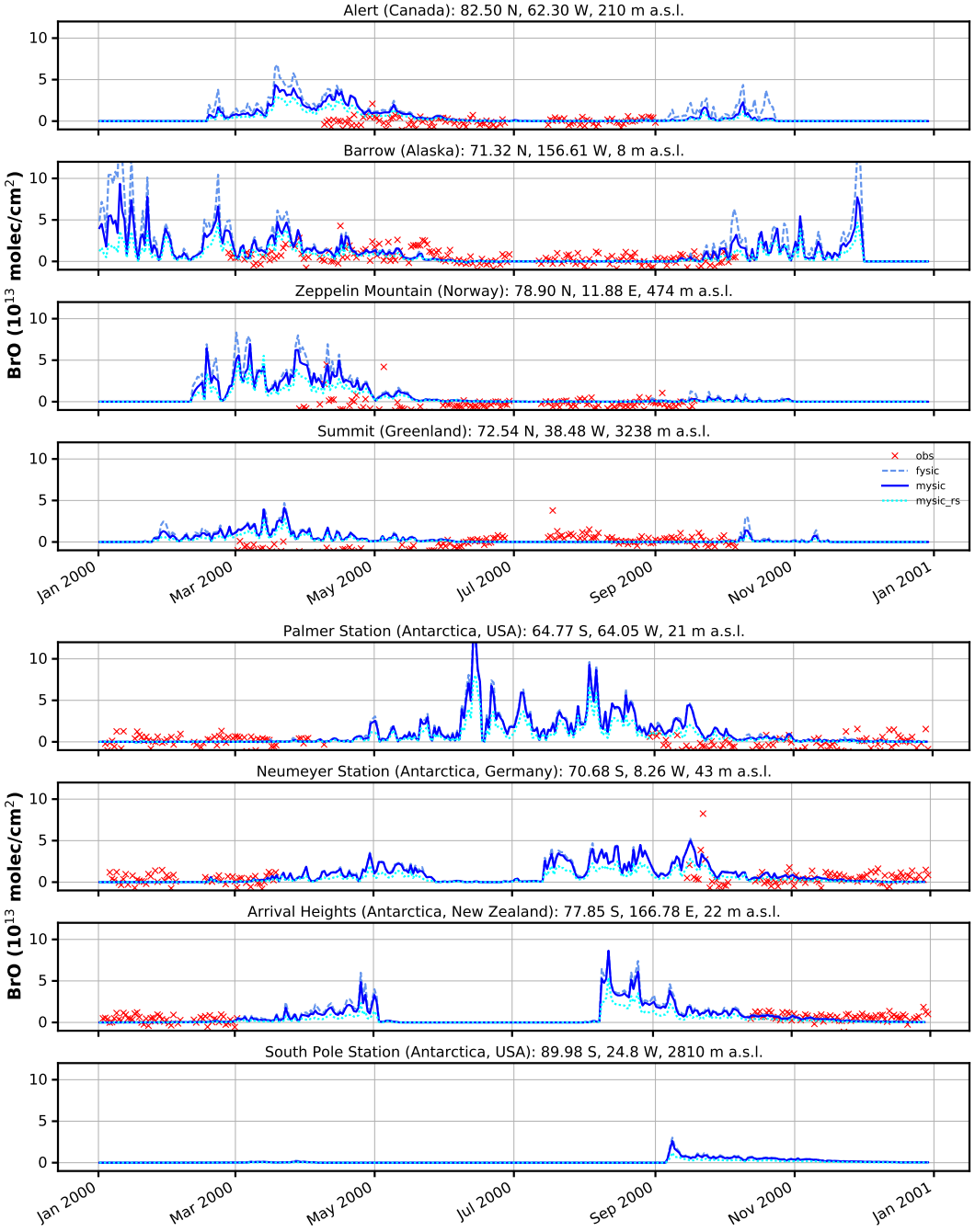

Figure 1. BrO VCD evaluated at observation sites on the northern southern hemisphere. The same as for surface ozone comparison have been chosen. The reference simulation is subtracted from the shown modeling results. GOME tropospheric VCD is shown subtracted by its zonal mean to emphasize BrO enhancements.

– 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 springtime 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.

Polar boundary layer bromine explosion and ozone depletion events in the chemistry-climate model EMAC v2.52: Implementation and evaluation of AirSnow algorithm

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Abstract. Ozone depletion events (ODE) in the polar boundary layer have been observed frequently during spring-time. ~~Most likely, they~~ They are related to events of boundary layer enhancement of bromine. Consequently, increased vertical column densities (VCD) of BrO have been observed from satellites. These so called bromine explosion events have been discussed serving as source of tropospheric BrO at high latitudes. We have implemented a treatment of bromine release and recycling on sea ice and snow covered surfaces in the global chemistry-climate model EMAC (ECHAM/MESSy Atmospheric Chemistry) based on the scheme of Toyota et al. (2011). In this scheme, dry deposition fluxes of HBr, $HOBr$, and $BrNO_3$ over ice and snow covered surfaces are recycled into Br_2 fluxes. In addition, dry deposition of O_3, dependent on temperature and sunlight, triggers a Br_2 release from surfaces associated with first-year sea ice. Many aspects of observed bromine enhancements and associated episodes of near-complete depletion of boundary layer ozone, both in the Arctic and in the Antarctic, are reproduced by this relatively simple approach. We present first results from our global model studies extending over a full annual cycle, including comparisons with GOME satellite BrO VCD and surface ozone observations.

1 Introduction

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)~~(Oltmans, 1981; Barrie et al., 1988; Bottenheim et al., 1986, 2002, 2009). Individual events typically last between several hours to a few days. The boundary layer ozone depletion events (ODE) are almost certainly related to events of strongly enhanced bromine, so called bromine explosion events. Enhanced bromine monoxide (BrO) column densities are regularly observed from satellites over both hemispheres, predominantly over the marginal sea ice zone, but sometimes also over inland ice and snow covered regions (e.g., Richter et al., 1998). In addition to their impact on boundary layer ozone, bromine explosion events play an important role in mercury deposition and corresponding environmental impacts (Lindberg et al., 2002; Stephens et al., 2012). Proposed mechanisms for bromine explosion events involve frost flowers on thin sea ice (Kaleschke et al., 2004) and blowing of saline snow on sea ice (Yang et al., 2010). Carbonate precipitation in brine at low temperatures has been suggested as efficient release trigger of ~~sea-salt~~ sea salt bromine to the atmosphere (Sander et al., 2006). However, measurements of Br_2 release in dependence of illumination and ozone volume mixing ratio (VMR) from

various types of snow and ice indicate that neither sea ice itself nor brine icicles are a major source for Br_2 but in addition to snow on sea ice also snow on land surfaces has to be taken into consideration (Pratt et al., 2013). Recent reviews on the subject are provided by Simpson et al. (2007), Saiz-Lopez and von Glasow (2012), Abbatt et al. (2012), and Custard et al. (2015). ~~In spite of~~ There has been considerable progress in describing the mechanisms involved in bromine release and boundary layer ODE ~~,~~ based on field measurements and laboratory experiments. Regarding the underlying heterogeneous chemical reactions, many similarities can be drawn between the very cold and hostile polar boundary layer and the polar upper troposphere - lower stratosphere (UTLS), where polar stratospheric clouds (PSCs) play a major role in halogen activation. In these cold regimes, icy surfaces allow or accelerate reactions which are impossible or rather slow in gas phase chemistry. For sustaining catalytic ozone depletion, the activation of halogens through heterogeneous reactions is very important. While mainly chlorine is activated in PSCs, bromine activation is favored by the processes taking place in the polar boundary layer. In the following we will give an account of the review article by Abbatt et al. (2012). Accordingly, the existing modeling approaches ~~still rely on a number of semi-empirical assumptions.~~ can be grouped into four categories:

- Frost flowers ($\rightarrow$ sea salt aerosol formation),
- bulk ice and snow ($\rightarrow$ Br_2 release),
- blowing of saline snow ($\rightarrow$ uplifting of sea salt and aerosol formation), and
- snowpack (photo)chemistry ($\rightarrow$ Br_2 release).

Frost flowers covered in high saline brine, are sturdy while fragile in appearance and contribute less to saline aerosol formation and bromine explosion events than originally anticipated (Domine et al., 2005). Br^- enriched brine is formed on sea ice through drainage and precipitation of hydrohalite ($NaCl \cdot 2H_2O$) at temperatures below 251 K (Abbatt et al., 2012, and references therein). In the course of summer, most salt is washed out from sea ice. Therefore, multi-year sea ice can be discarded as source of bromine explosion events. In contrast to solutions, acidity is not important on icy surfaces (Adams et al., 2002). Since HOBr is rather rapidly reacting forming Br_2, the rate of Br_2 release is mainly limited by mass transfer from the atmosphere to snow or ice (Huff and Abbatt, 2000). Apart from the complex heterogeneous photochemistry taking place in a quasi-liquid phase on ice grains in the snowpack (Thomas et al., 2011; Pratt et al., 2013), ozone itself has the capacity of triggering auto-catalytic reactions by oxidizing bromine already before polar sunrise.

On the basis of empirical and modeling results, Toyota et al. (2011) presented a ~~parametrization~~ parameterization of Br_2 release from bulk ice and snow within the Global Environmental Multiscale model with Air Quality processes (GEM-AQ). GEM-AQ is based on Canada's operational weather prediction model developed by the Meteorological Services of Canada (MSC) for the interaction of atmospheric chemistry with sea ice and snow surfaces. This ~~parametrization~~ parameterization reproduces many aspects of observed bromine enhancements and boundary layer ODE.

Here we present an implementation of a mechanism based on the work of Toyota et al. (2011) into the ECHAM/MESSy Atmospheric Chemistry (EMAC) model (Jöckel et al., 2010). The mechanism and its integration into the existing submodel ONEMIS (Kerkweg et al., 2006) are described in Sect. 2. In Sect. 3, results from several one year long integrations of the

model with and without bromine release are presented and compared to surface ozone observations as well as observations of ~~tropospheric~~ BrO vertical column density (VCD) from the Global Ozone Monitoring Experiment (GOME) satellite instrument on board ERS-2 (Richter et al., 1998, 2002). We show that many aspects of observations regarding BrO enhancements and ODE are reproduced by this mechanism without any further tuning of parameters. Unlike most previous modeling studies, we do not focus on Arctic spring time only but investigate a full annual cycle on both hemispheres.

2 Model and experiments

The EMAC model is a numerical chemistry-climate model, based on 5th generation European Centre Hamburg general circulation model (ECHAM5) (Roeckner et al., 2006) as dynamical core. Various submodels describe atmospheric and Earth system processes and are coupled via the Modular Earth Submodel System (MESSy) (Jöckel et al., 2005). MESSy provides an infrastructure with generalized interfaces for control and coupling of components. Further information about MESSy and EMAC is available from the MESSy project homepage. MESSy enables for a flexible handling of emissions in EMAC, e.g., prescribed fluxes, ~~(boundary) layer~~ concentrations of tracers at the boundary layer or any other given level, and emissions dependent on dynamical atmospheric fields. Latter are treated as ~~*online*~~ online emissions using the submodel ONEMIS (Kerkweg et al., 2006). ONEMIS provides facility functions for flux to tracer concentration conversions. According to the MESSy philosophy, ONEMIS is separated into a submodel interface layer (~~smil~~SMIL) for unified data handling among different submodels and an implementation layer of the actual emission mechanisms (~~smcl~~submodel core layer, SMCL). A recap of the mechanism proposed by Toyota et al. (2011) (Sect. 2.1) and details about its integration into the EMAC model (Sect. 2.2) are given in the following. In Sect. 2.3, scope and setup of ~~the~~ a set of test experiments are summarized.

2.1 Description of the mechanism

It is assumed that at least part of the observed Br_2 flux originates from heterogeneous reactions on snow grains in the surface layer of a snowpack (Pratt et al., 2013). These snow grains are considered coated by a Br^- enriched film of liquid water and show a distinct acidity. In this quasi-liquid phase, heterogeneous reactions of $HOBr$ and $BrNO_3$ with either Br^- and Cl^- can take place:

$$HOBr + Br^- \xrightarrow{H^+} Br_2 + H_2O, \quad \text{(R1)}$$

$$BrNO_3 + Br^- \rightarrow Br_2 + NO_3^-, \quad \text{(R2)}$$

$$HOBr + Cl^- \xrightarrow{H^+} BrCl + H_2O, \quad \text{(R3)}$$

$$BrNO_3 + Cl^- \rightarrow BrCl + NO_3^-. \quad \text{(R4)}$$

Interhalogene reactions may convert $BrCl$ into Br_2:

$$BrCl + Br^- \leftrightarrow Br_2Cl^- \leftrightarrow Br_2 + Cl^- \tag{R5}$$

$BrCl$ is partly released to the atmosphere before undergoing this last reaction. In addition, various photochemical gas-, aqueous-, and heterogeneous-phase reactions are taking place in the top layer of a snowpack (for details see, e.g., Pratt et al., 2013, Fig. 2), which are rather similar to heterogeneous reactions occurring on PSCs. A list of heterogeneous reactions involving bromine included in MECCA is provided as Supplement S.1. Another reaction pathway ~~is actually~~ oxidizing bromine is triggered by ozone dry deposition ~~under~~ without the influence of sunlight. ~~This pathway accumulates various gas, aqueous, and heterogeneous phase reactions (for details see Pratt et al., 2013, Fig. 2). Toyota et al. (2011) have parametrized these heterogeneous reaction pathways ($HOBr$ / $BrNO_3$ / $O_3 \rightarrow Br_2$) in a simple way.~~Three surface types, first-year sea ice (FY), multi-year sea ice (MY), and snow on land (LS) are differentiated. In any case, the respective surface temperature has to be below a temperature threshold T_{crit}. The critical conversion of a dry deposition flux of ozone (Φ_{O_3}) into an emission flux of Br_2 (or $BrCl$) is moderated by an ad hoc molar yield Φ_1, dependent on surface type and illumination. Toyota et al. (2011) have parametrized these heterogeneous reaction pathways ($HOBr$ / $BrNO_3$ / $O_3 \rightarrow Br_2$) in a simple way taking state-of-the-art knowledge into account:

$$\Phi_1 = \begin{cases} 0.001 & \text{if } \textit{dark} \text{ FY}, \\ 0.075 & \text{if } \textit{sunlit} \text{ FY}, \\ 0 & \text{if MY or LS}. \end{cases} \tag{1}$$

I.e., on FY sea ice, only 0.1% of the dry deposition of O_3 will be converted into Br_2 ~~if the~~ in case the surface is not sunlit (sun's zenith angle ~~is~~ above $\theta_{\mathrm{crit}} = 85\,^\circ$), otherwise 7.5% is converted. No release of Br_2 from MY sea ice or LS is assumed. The specific value of Φ_1 has been 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).

The conversion of dry deposition fluxes of $HOBr$ (Φ_{HOBr}), $BrNO_3$ (Φ_{BrNO_3}), and HBr (Φ_{HBr}) is considered independent of illumination. In case of FY sea ice, the ~~snow pack~~ snowpack on top is regarded as an infinite pool of Br^- and Cl^-. The sum of $HOBr$ and $BrNO_3$ dry deposition fluxes ($\Phi_{HOBr} + \Phi_{BrNO_3}$) is fully recycled into Br_2. In case of MY sea ice, only the Cl^- pool remains infinite, for Cl^- is about 2 to 3 orders of magnitude more abundant in snow than Br^- (Toyota et al., 2011). The release of Br_2 depends on $\Phi_{HOBr} + \Phi_{BrNO_3}$ in comparison to the dry deposition flux of HBr. If $\Phi_{HOBr} + \Phi_{BrNO_3}$ was less than Φ_{HBr} a full conversion of $\Phi_{HOBr} + \Phi_{BrNO_3}$ to Br_2 is assumed. Otherwise, only half of the difference $\Phi_{HOBr} + \Phi_{BrNO_3} - \Phi_{HBr}$ is recycled to Br_2, the other half is converted to $BrCl$. For LS, neither Br^- nor Cl^- is available unlimited. Hence, only the smaller of $\Phi_{HOBr} + \Phi_{BrNO_3}$ and Φ_{HBr} is converted to Br_2. The resulting *yield* is summarized in Φ_2:

$$\Phi_2 = \begin{cases} 1 & \text{if FY}, \\ 0.5 - 1 & \text{if MY}, \\ 0 - 1 & \text{if LS}. \end{cases} \tag{2}$$

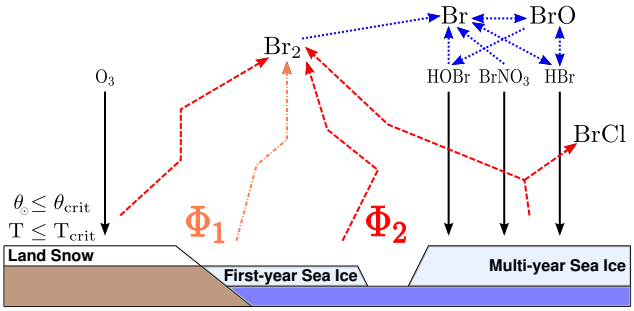


Figure 2. Schematic scenario of bromine release from first-year sea ice, multi-year sea ice, and land snow adapted from Toyota et al. (2011) for a temperature threshold T_{crit}. Black arrows denote dry deposition of HOBr, $BrNO_3$, HBr, and O_3. Blue doted arrows indicate gas-phase photochemistry. Dry deposition fluxes are recycled into Br_2 with respect to a molar yield Φ_1 in case of O_3 (dashed orange) and Φ_2 in case of the brominated species (dashed red).

Schematically, all release scenarios are shown in Fig. 2 (adapted from Fig. 1 of Toyota et al. (2011)). Herein, black arrows denote dry deposition of HOBr, $BrNO_3$, HBr, and O_3. Blue doted arrows indicate gas-phase photochemistry. The recycled fluxes are displayed by dashed orange (O_3) and red (HOBr, $BrNO_3$, HBr) arrows.

2.2 Implementation

In accordance to the described scheme, submodel interface layer (SMIL), submodel core layer (SMCL), and namelist of ONEMIS have been extended based on EMAC version 2.52. Channel objects, ~~which are~~ used by a subroutine `airsnow_emissions` (implemented in SMCL), include surface temperature (`tsurf`), fraction of snow cover on land (`cvs`), fraction of ice cover on ocean (`seaice`), cosine of sun's zenith angle (`cossza`), and dry deposition fluxes of HOBr, $BrNO_3$, HBr, and O_3 (`drydepflux_<HOBr, BrNO3, HBr, O3>`). Dry deposition is computed by submodel DDEP (formerly DRYDEP, Kerkweg et al., 2006, b). In the SMIL of ONEMIS, these channel objects are defined and initialized and the subroutine `airsnow_emissions` is called. Additional information about multi-year sea ice cover (MYSIC) has to be provided through data import. Currently, we are using a MYSIC estimate based on mean SIC from ERA-Interim (see Section 2.3). Steering parameters, Φ_1, T_{crit}, and θ_{crit}, can be changed in the corresponding control sequence within the ONEMIS namelist file. However, the parameter relevant to MY sea ice and LS in Φ_1 is currently not used, since no ~~parametrization~~ parameterization has been provided by Toyota et al. (2011). New output channels `snow_air_flux_br2` and `snow_air_flux_brcl` have been defined ~~. Instead of actual code, a Nassi-Shneiderman diagram displaying~~ in the SMIL of ONEMIS. More detail of the algorithm implemented in the subroutine `airsnow_emissions` ~~in more detail is provided~~ is provided as Nassi-Shneiderman diagram in Supplement S~~.1~~.2. The new emission mechanism has been named *AirSnow* and can be switched on in the ONEMIS namelist – an example excerpt has been added as Supplement S~~.2~~.3. After Br_2 has been released, we make use of atmospheric bromine chemistry that is identical to EMAC's standard stratospheric bromine chemistry (Supplement S.1).

2.3 Validation Experiments

Three experiments have been performed using EMAC version 2.52 (see Table 1 for a summary). The basic model setup has been adapted from RC1SD-base-08, which is part of a ~~Chemistry-Climate~~ Chemistry Climate Model Initiative (CCMI) recommended set of simulations by the Earth System ~~Chemistry-Climate~~ Chemistry integrated Modelling (ESCiMo) consortium (Jöckel et al., 2016). The model integrations use specified dynamics nudged to ERA-Interim for the year 2000. Accordingly, ERA-Interim sea ice cover (SIC) has been used. The chosen spatial resolution is T42L90MA corresponding to a $2.8° \times 2.8°$ grid, with a top level at 0.01 hPa and distributed to 90 levels. Output has been saved with ~~one hourly~~ 1-hourly temporal resolution. In contrast to RC1SD-base-08, fluxes of brominated very short-lived substances (VSLS), CH_2Br_2 and $CHBr_3$, are computed online from sea water concentrations (Ziska et al., 2013) using the EMAC submodel AIRSEA (Pozzer et al., 2006) as described by Lennartz et al. (2015). In this scheme, sea ice acts as a lid blocking the emission of VSLS to the atmosphere. Comprehensive tropospheric and stratospheric chemistry as well as heterogeneous reactions within MECCA (Sander et al., 2011) have been activated for an aerosol surface area concentration climatology.

The basic parameter setup has been adopted without changes as proposed by Toyota et al. (2011). The temperature threshold for all simulations has been $T_{\mathrm{crit}} = -15\,^{\circ}\mathrm{C}$, accordingly.

In EMAC no discrimination is made between FY sea ice and MY sea ice, therefore we initially assume all ice to be first-year (BrXplo_fysic). A multi-year sea ice cover has been computed from RC1SD-base-08 ~~10 hourly~~ 10-hourly SIC output based on ERA-Interim. We regard ice at a fixed location that survived one melting season as multi-year. Hence for simplicity, we assume no drift of ice masses. SIC has been integrated for respective summer months on northern (August/September) and southern (February/March) hemisphere. The SIC at the minimum of the integrated SIC has been chosen as MYSIC for the respective year after. The resulting MYSIC for the year 2000 are shown in Fig. 3 together with monthly mean SIC for April (northern hemisphere) and September (southern hemisphere). The result is very similar with regard to patterns and extend of MYSIC on maps retrieved from satellite observation (US National Snow & Ice Data Center (NSIDC), 2017). Based on the MYSIC estimate, a second model integration (BrXplo_mysic) has been conducted. For comparison, a reference simulation with bromine release mechanism switched off has been done (referred to as BrXplo_ref). In a further sensitivity simulation, we have decreased the dry deposition of ozone over snow covered regions as proposed by Helmig et al. (2007) by changing the surface resistance in DDEP for ozone on snow and ice surfaces from the value of $r_{\mathrm{O_3}}^{\mathrm{ice-snow}} = 1/2000\,\mathrm{sm}^{-1}$ (Wesely, 1989) to $r_{\mathrm{O_3}}^{\mathrm{ice-snow}} = 1/10000\,\mathrm{sm}^{-1}$ (Helmig et al., 2007).

3 Results

In this section, we ~~qualitatively compare our simulation~~ compare our simulations' results with observational data~~regarding VCD for both northern and southern hemisphere and depletion events of surface ozone. Since~~ . For Br_2, which has been released from ice and snow, is transformed into BrO photolytically, enhancements of Br_2 ~~result in~~ lead to an increase of the BrO vertical column density~~which is observable~~ . These enhancements have been observed by satellite instruments ~~. We use GOME tropospheric VCD~~ (e.g., Richter et al., 1998). At first, we compare the spatial distribution of BrO ~~which has been~~

Table 1. EMAC model experiments used in this study. All experiments have been done using specified dynamics nudged to ERA-Interim. Accordingly, ERA-Interim SIC has been used. The setup is based on the consortial ESCiMo simulation RC1SD-base-08. Experiments have been performed for an assumption of first-year sea ice only (FYSIC) and for a multi-year sea ice cover (MYSIC) estimated from SIC. The temperature threshold for all simulations has been $T_{\mathrm{crit}} = -15\,^{\circ}\,\mathrm{C}$, accordingly.

Experiment	Model Version	Resolution	Time-Span	Chemistry	VSLS Emission	~~Polar Bromine Release~~AirSnow	$r_{\mathrm{O_3}}^{\mathrm{ice-s}}$
BrXplo_ref	2.52	T42L90MA	Jan–Dec 2000	full	AIRSEA	no	1/2000
BrXplo_fysic	2.52	T42L90MA	Jan–Dec 2000	full	AIRSEA	FYSIC	1/2000
BrXplo_mysic	2.52	T42L90MA	Jan–Dec 2000	full	AIRSEA	MYSIC	1/2000
BrXplo_mysic_rs	2.52	T42L90MA	Jan–Dec 2000	full	AIRSEA	MYSIC	1/10000

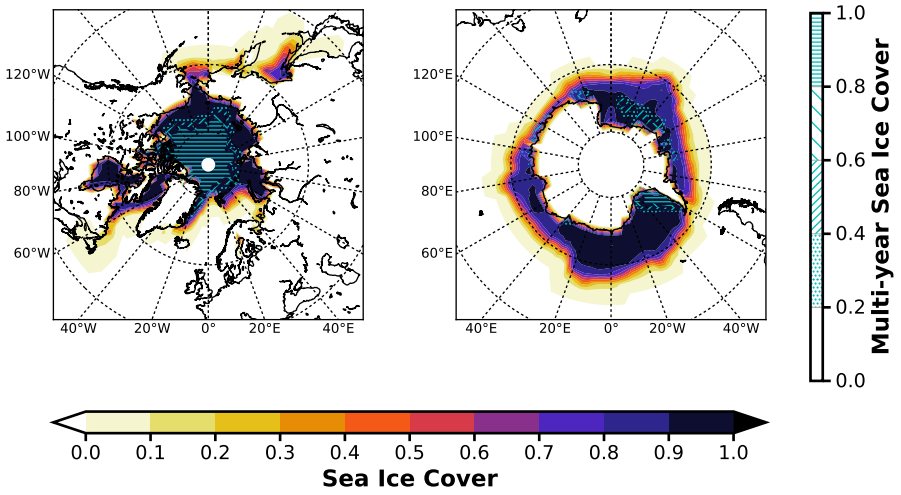


Figure 3. Sea ice cover fraction and estimated multi-year sea ice cover fraction for the year 2000. Mean SIC are shown for April on the northern hemisphere and September on the southern. MYSIC has been computed from RC1SD-base-08 ~~10 hourly~~ 10-hourly SIC based on ERA-Interim. For simplicity, we assume ice that survived one melting season as multi-year. (left) Northern hemisphere; (right) Southern hemisphere.

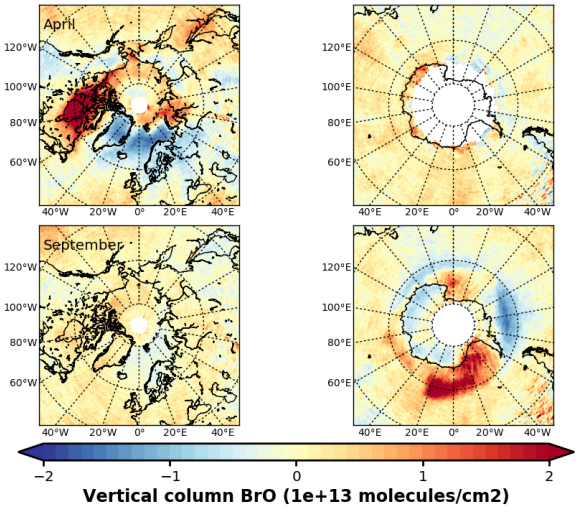
~~computed from total VCD for solar zenith angles less or equal to 80° by subtracting SLIMCAT modeled stratospheric VCD . Monthly mean tropospheric VCD from GOME-SLIMCAT retrievals~~ total VCD as simulated with EMAC (BrXplo_mysic) with GOME retrieved total VCD in both hemispheres. Implications on depletion events of surface ozone will be drawn in comparison to observational data at several ground-based sites in both hemispheres.

3.1 Total BrO vertical column density

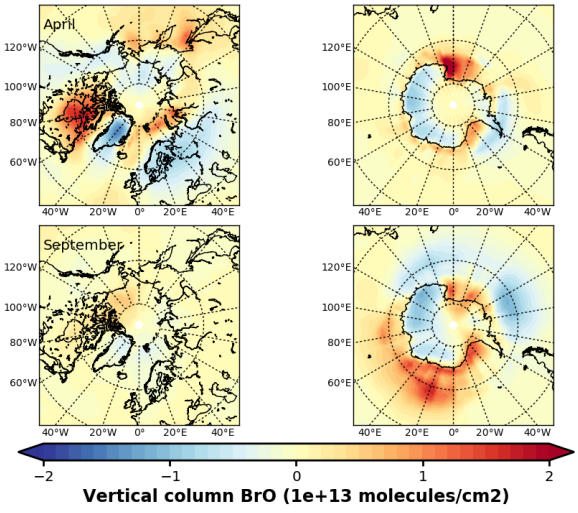
Monthly mean GOME VCD retrieval of BrO subtracted by corresponding zonal means are shown in Fig. 4a) for both, northern and southern polar regions in April and September, respectively. The associated zonal means are available as Supplement S.5.1. In April, GOME data display a strong enhancement of BrO VCD across the whole coastal region of the Arctic ocean~~down to Hudson Bay~~, except for the coast of Greenland. Hotspots can be found down the Hudson Bay, east of Novaya Zemlya, and around Hokkaido. There are ~~signs of~~ only slight enhancements in the Antarctic coastal regions, ~~where data are available~~but data are sparse. In September, enhancements ~~above Antarctica are in particular found~~ around Antarctica can be in particular observed in the Ross and Weddell sea areas.

From ~~hourly~~ 1-hourly BrO profiles of the EMAC model output, a total VCD has been ~~computed~~ integrated and re-sampled ~~in accordance to~~ to 10–11 am local solar time~~10 UTC, for~~ , according to the ERS-2 equator crossing time ~~had been~~ of 10.30 am local time. In general, transition times at high latitudes differ from the equator crossing time due to ~~the satellite orbitand total~~ satellite orbit. Differences in local local time may account for part of the differences seen in the BrO comparison. The re-sampled data has been averaged monthly. As there is an offset between EMAC and GOME BrO VCD~~is not directly comparable to tropospheric columns. However, stratospheric columns can be considered constant over space and time (e.g., Richter et al., 19~~ ~~Hence, a qualitative comparison between the two sets of data is sufficiently unaffected by these concerns. The monthly averaged~~ , we are showing anomalies here. The zonal mean BrO VCD has been subtracted to highlight the bromine explosion events. The associated zonal mean BrO data are show as Supplement S.5.2. The resulting EMAC total BrO VCD are shown in Fig. 4b). ~~Spatial~~ In comparison to GOME data, spatial patterns of BrO VCD are reasonably well reproduced by EMAC in ~~Northern~~ the northern hemisphere in April. ~~Compared to GOME, VCD may be underestimated~~ Only westward from Hudson Bay ~~respectively eastward from the Laptew sea. Regarding the Southern hemisphere in September,~~ and eastward from Novaya Zemlya, respectively, no BrO ~~VCD is likely overestimated in the modelbut spatial patterns are rather similar.An~~ enhancement is found in our simulation. The Hokkaido hotspot appears slightly shifted northward. In September, both observation and model, agree well in both hemispheres. In April, satellite data of Antarctica are too sparse to identify the hotspots that occur in the simulation.

A full overview of monthly mean total BrO VCD for both, observation and model, including all months ~~can be found in~~ has been added as Supplement S~~.3. Apparently~~.4. In the northern hemisphere, the implemented mechanism is prone for ~~increased~~ BrO VCD enhancements shifted to early winter compared to GOME retrievals. ~~A comparison of the~~ In late spring and early summer, however, too few BrO ~~VCD spectra~~ is formed in the model. This may hint to sources of BrO in the Arctic which are not represented by this mechanism or an adherence to the chosen parameters. Further studies would be needed to resolve the source of this discrepancy. In the southern hemisphere, the modeled BrO enhancements in, e.g., August and September are similar in their occurrence, while the sparseness of GOME data ~~and simulation implies a good agreement of the first order of magnitude while there are probably missing sources of higher magnitude in the implemented mechanism . But this will be subject to further studies.~~ in austral winter does not permit further conclusions regarding the quality of the parameterization in this region. Taking a look at the zonally averaged total BrO VCD (Supplement S.5), we find, that the modeled BrO VCD is generally too small in polar summer compared to observation by about $(1-4)\cdot 10^{13}\,\mathrm{molecules\,cm^{-2}}$ in both hemispheres, respectively. A better agreement between observation and model is achieved in winter. This is due to the implementation of the



(a) GOME



(b) EMAC

Figure 4. ~~Monthly~~ Anomalies of monthly mean ~~vertical column density~~ VCD of BrO for the Arctic and Antarctic ~~spring~~ (April and ~~austral spring~~September) with respect to monthly averaged zonal mean (see Supplement S.5), respectively. EMAC data have been sampled in accordance to local solar time ~~10~~10–11 ~~UTC~~am. (a) ~~GOME-SLIMCAT tropospheric~~GOME; (b) EMAC ~~total~~(BrXplo_mysic).

bromine release mechanism (doted lines indicating the reference simulation). Hence, taking the bromine released from ice and snow into account the overall model performance is enhanced with respect to polar BrO observation.

Table 2. Observation sites for surface ozone comparison. However, for Palmer station and Arrival Heights no observations of surface ozone are available for the year 2000, so that we present model results only for these two stations.

Site	Location	Latitude (°N)	Longitude (°E)	Altitude (m a.s.l.)	Data Provider
Alert	Canada	82.50	-62.30	210	EBAS (NILU)
Barrow	Alaska	71.32	-156.61	8	ESRL/GMD (NOAA)
Zeppelin Mountain	Spitsbergen	78.90	11.88	474	EBAS (NILU)
Summit	Greenland	72.54	-38.48	3238	ESRL/GMD (NOAA)
Palmer Station	Antarctica	-64.77	-64.05	21	*ESRL/GMD (NOAA)*
Neumayer Station	Antarctica	-70.68	-8.26	43	EBAS (NILU)
Arrival Heights	Antarctica	-77.85	166.78	22	*ESRL/GMD (NOAA)*
South Pole Station	Antarctica	-89.98	-24.8	2810	ESRL/GMD (NOAA)

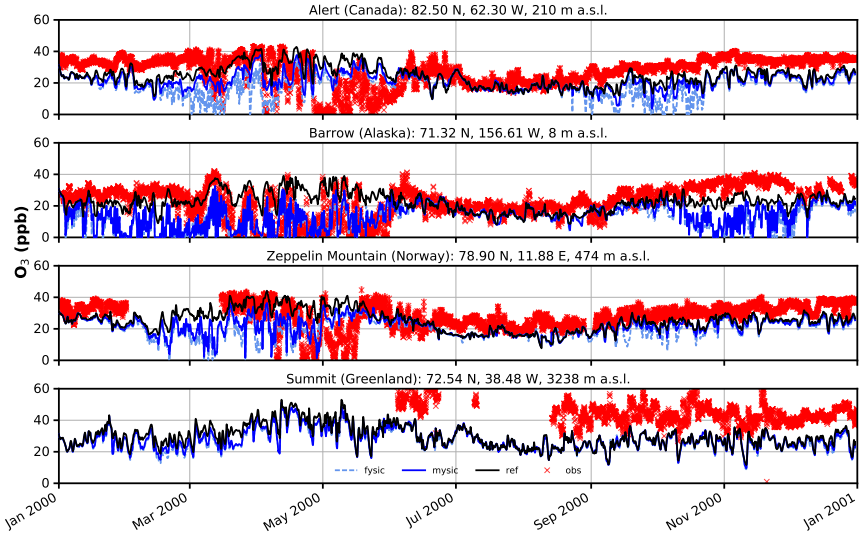
3.2 Ozone depletion events

Regarding depletion events of surface ozone, four different observation sites have been chosen on each hemisphere for comparison (Table 2). ~~However, no~~ No data for Arrival Heights and Palmer Station have been available ~~for~~ in 2000. For these stations, we show model results only. Time series of surface ozone VMR are shown in Figures 5–6 including both in situ observations (where available) and model simulations. For each simulation, the nearest grid point has been chosen as representative. In general, we find a good agreement between BrXplo_ref and observations for seasons without bromine release from ice and snow, except for Summit, South Pole station, and Neumayer station in austral winter, where model results are systematically lower compared to observations. In case of BrXplo_fysic all northern hemispheric sites display depletion events in spring as well as in fall. While the depletion events are not entirely in temporal coincidence with observed events, their frequency is generally well reproduced. However, events of ozone depletion in fall are not present in observation data. For Zeppelin Mountain and Alert, these *fault events* are due to the FYSIC assumption. For a decent multi-year sea ice cover is implemented in BrXplo_mysic, they vanish. In case of Barrow, a closer look into spring reveals an astonishing temporal as well as quantitative coincidence of surface ozone VMR especially in April (Fig. 6). The apparent *wiggles* are partly due to hard trigger thresholds T_{crit} and θ_{crit}, but similar structures are in fact apparent in the surface ozone observations at Barrow implying a diurnal variation of O_3 depletion. At Alert, our model does not capture the 2000s ODE that inflicted continuously low surface ozone levels for several days from late April until early May. As pointed out by Strong et al. (2002), this long-lasting depletion event was related to transport of ozone poor air originating from a region north of Ellesmere Island and the eastern arctic ocean, respectively. It is not clear whether transport of ozone depleted air masses or depletion itself is too weak in our simulation. At about the same time (late April, early May) observation displays a series of ODEs at Zeppelin mountain, which is also only partly reproduced by the model (e.g. on April 28th). Comparing observation and simulation in the southern hemisphere and Greenland, we find in general less ozone in BrXplo_mysic as well as in BrXplo_ref. This may hint

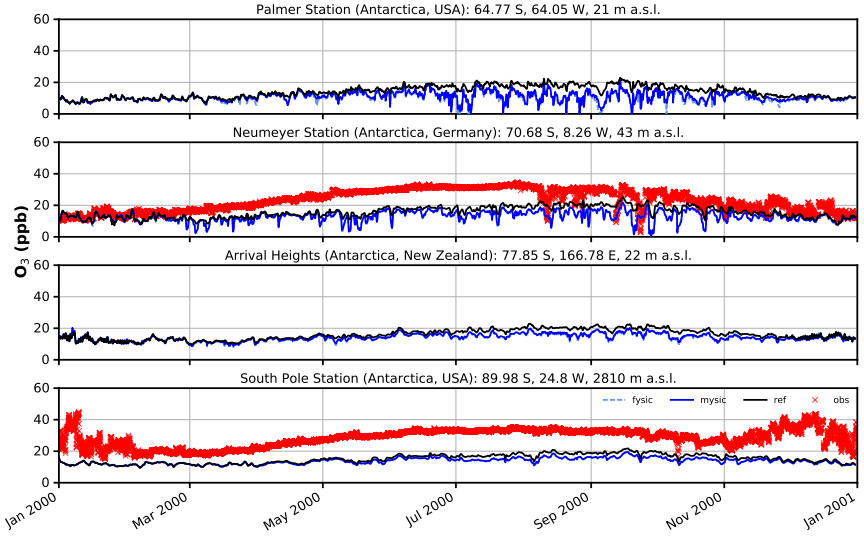
to missing sources of polar ozone released from ice and snow in the model. Any analysis regarding the modeled occurrence of ODEs in the southern hemisphere is not affected by this. Despite the original mechanism's validation for northern hemispheric spring (Toyota et al., 2011), comparison of time series for the southern hemisphere do display ozone depletion events in a similar frequency as found in observational data. ~~Observation sites for surface ozone comparison. Providers typeset in italic refer to unavailable data for 2000. Site Location Latitude Longitude Altitude Data Provider() () () Alert Canada 82.50 -62.30 210 EBAS (NILU)Barrow Alaska 71.32 -156.61 8 ESRL/GMD (NOAA)Zeppelin Mountain Spitsbergen 78.90 11.88 474 EBAS (NILU)Summit Greenland 72.54 -38.48 3238 ESRL/GMD (NOAA)Palmer Station Antarctica -64.77 -64.05 21 *ESRL/GMD (NOAA)*Neumayer Station Antarctica -70.68 -8.26 43 EBAS (NILU)Arrival Heights Antarctica -77.85 166.78 22 *ESRL/GMD (NOAA)*South Pole Station Antarctica-89.98 -24.8 2810 ESRL/GMD (NOAA)~~At Neumayer station, we find some events in late October and early November that might be coincidental, but in most cases simulated ODEs show up later than actually observed ODEs. In summary, while some aspects of ODEs are reproduced remarkably well by the implemented mechanism, especially the long-lasting event at Alert is not reproduced at all. This strongly hints to the involvement of further mechanisms, e.g., blowing snow and sea spray, in the depletion of polar surface ozone which have not yet been modeled in EMAC. In BrXplo_mysic_rs with the reduced dry deposition, ozone depletion events in fall and midwinter are suppressed and the agreement with observed ozone is generally improved (see Supplement S.6). Reducing the ozone dry deposition over snow and ice slightly increases boundary layer ozone at all discussed sites, but even with the reduced dry deposition the model significantly underestimates observed boundary layer ozone in Antarctica, indicating that other mechanisms exist that increase boundary layer ozone under these conditions (e.g., Oltmans, 1981; Helmig et al., 2007). A correlation between observed and modeled surface ozone at Barrow is shown in Fig. 7. (The supplementary information provides additional correlation plots for the other stations in the northern hemisphere, as well as additional plots for the sensitivity simulation with reduced ozone dry deposition.) As already evident from the time series in Fig. 6, low surface ozone values largely absent in the reference simulation are reproduced by the EMAC simulation including bromine explosion events, while some *fault events* are also generated, not present in the observations. Overall the linear correlation coefficients between modeled and observed ozone are improved by inclusion of the bromine explosion mechanism (from 0.008 to 0.21). A corresponding lag correlation analysis shows that largest correlations are found for zero lag, with correlations falling to half of the maximum at about ± 2 days.

4 Discussion and conclusions

Many approaches describing bromine release in the polar regimes rely on modeling of complex micro physical processes which are too detailed for integration in a global chemistry-climate model. We have implemented a bromine release mechanism from sea ice and snow covered land surfaces based on the relatively simple ~~parametrization~~ parameterization suggested by Toyota et al. (2011) in the global chemistry-climate model EMAC. While the original study of Toyota et al. (2011) focused on Arctic spring time only, we extend the simulations to the global scale and a full annual cycle. We show that without any further tuning of the parameters, many aspects of observed polar bromine enhancements and boundary layer ozone depletion events are well reproduced by this mechanism within the EMAC model. Resulting spatial patterns of BrO total VCD and the temporal occur-



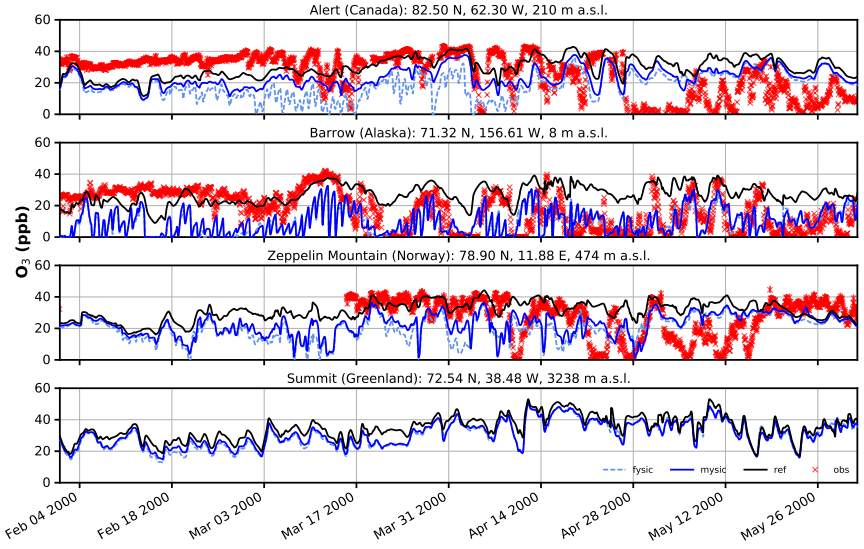
(a)



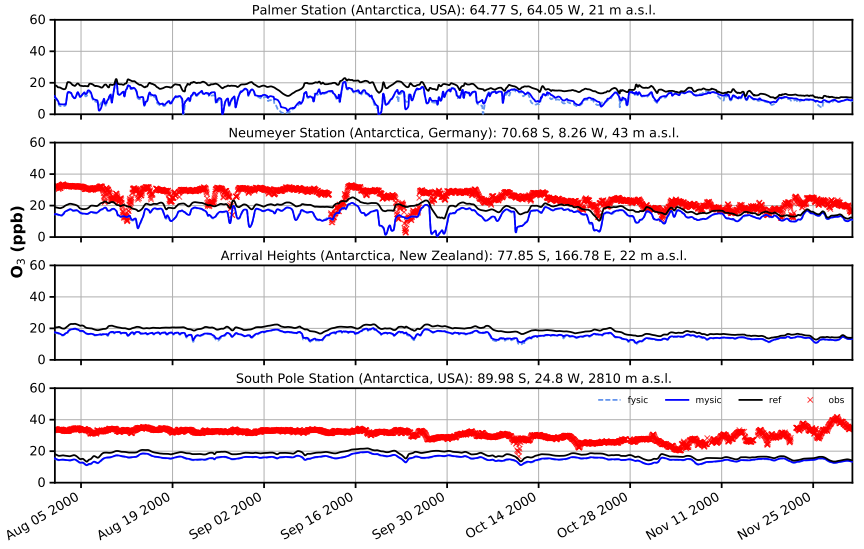
(b)

Figure 5. Surface ozone mixing ratios at four different observation sites. Comparison of in situ measurements (red crosses) with results from simulation (solid black – EMAC v2.52 default (no bromine explosions); light blue dashed – FYSIC; solid blue – MYSIC). Representatively, the nearest grid point has been chosen. (a) Northern hemisphere; (b) Southern hemisphere.

rence of surface ozone depletion events are comparable to BrO ~~tropospheric~~ VCD retrieval of the GOME satellite instrument ~~, respectively~~ and in situ observation at different sites in both the Arctic and Antarctic, respectively. EMAC provides a wide range of Earth system related submodels and allows for simulations with full tropospheric and stratospheric (heterogeneous) chemistry in a selfconsistant manner. In our model integrations, inorganic bromine species (HBr, HOBr, $BrNO_3$) are provided



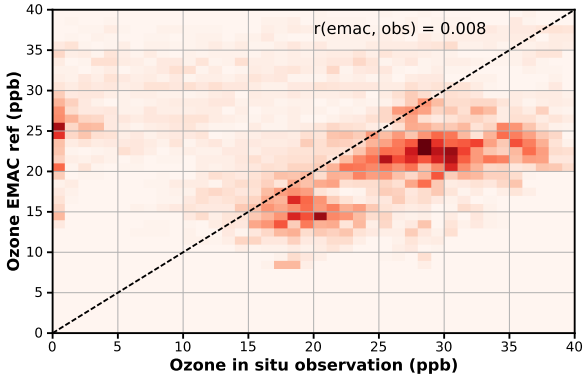
(a)



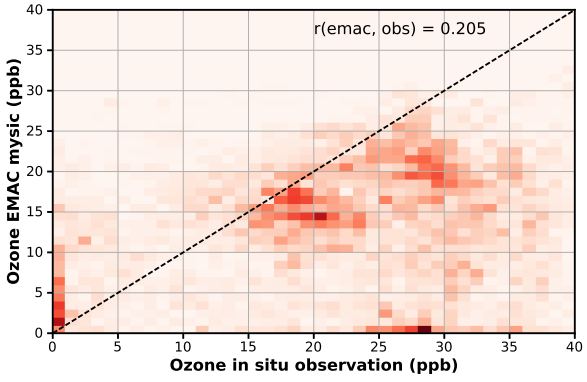
(b)

Figure 6. Surface ozone mixing ratios at four different observation sites for spring and austral spring, respectively. Comparison of in situ measurements (red crosses) with results from simulation (solid black – EMAC v2.52 default (no bromine explosions); light blue dashed – FYSIC; solid blue – MYSIC). Representatively, the nearest grid point has been chosen. (a) Northern hemisphere; (b) Southern hemisphere.

in two ways: through photochemical transformation of organic source gases of natural and anthropogenic origin and through descending stratospheric air containing inorganic bromine. The emission of bromine from very short-lived substances (CH_2Br_2, $CHBr_3$) is consistently computed online from sea water concentrations (Lennartz et al., 2015). ~~However, the~~ The implemented



(a) BrXplo_ref



(b) BrXplo_mysic

Figure 7. Temporal correlation of modeled surface O_3 with observation at Barrow. Data have been binned in bins of 1 ppb width. While observed low ozone events at Barrow are absent in the reference simulation, in the BrXplo_mysic simulation there is now a population where both observation and model simultaneously show low ozone values, which is also reflected in the improved linear correlation coefficient from 0.008 to 0.21. (a) BrXplo_ref; (b) BrXplo_mysic.

bromine release mechanism relies on various assumptions which ~~are not sufficiently well constrainedby observations~~have been cross-validated with observations and are not entirely constrained. In particular, the dry deposition, which is one of the key factors in this bromine release mechanism, is still highly uncertain and hard to measure explicitly.~~In a further sensitivity simulation, we have decreased the dry deposition of ozone over snow covered regions as proposed by Helmig et al. (2007) by increasing the surface resistance in DDEP for ozone on snow and ice surfaces from the value of $r_{O_3}^{ice-snow} = 1/2000$ (Wesely, 1989) to $r_{O_3}^{ice-snow} = 1/10000$ (Helmig et al., 2007). With the reduced dry deposition, ozone depletion events in fall and midwinter are suppressed and the agreement with observed ozone is generally improved (see Supplement S.4). Reducing the ozone dry deposition over snow and ice slightly increases boundary layer ozone at all discussed sites, but even with the reduced dry deposition the model significantly underestimates observed boundary layer ozone in Antarctica, indicating that other mechanisms exist that increase boundary layer ozone under these conditions (e.g., Oltmans, 1981; Helmig et al., 2007).~~

~~Although our~~ Our model simulations with this relatively simple mechanism successfully reproduce many observed features of bromine enhancement and ODEs (spatially as well as temporally), improving the overall model performance regarding BrO VCD and surface ozone concentrations at high latitudes. Although a lag analysis shows highest temporal correlation at zero lag between observation and model data at Barrow, there are still notable differences to other observations. In particular~~there is the~~ , there is a tendency to generate too high BrO columns and too many ODEs in ~~autumn and mid winter . In addition, some of the parameters like the critical temperature, fixed at −15 , are rather ad hoc and not well constrained by observations~~mid winter and spring, which is reduced by decreasing the ozone dry deposition. The recognized ODE observed at Alert in 2000 is not at all reproduced by this bulk-snow-based mechanism. It is ~~possible~~plausible, that in reality different processes, such as ~~snow-pack~~ snowpack chemistry as well as bromine activation by blowing snow and sea spray, all play a role and contribute to the bromine explosion events at different sites. With the present work we have now a framework to further test these mechanisms in a global chemistry climate model.

Code availability. The Modular Earth Submodel System (MESSy) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licensed to all affiliates of institutions, which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium Web-site (http://www.messy-interface.org). The modified code of the submodel ONEMIS described here will be made available with the next official release of the MESSy source code distribution.

Data availability. For any party interested, data can be made available on request.

Author contributions. Stefanie Falk has implemented the described mechanism, run and validated the simulations with observational data. Björn-Martin Sinnhuber suggested this study and took part in the analysis. Both authors contributed to the writing of the paper.

Competing interests. The authors declare that they have no conflict of interest.

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Ozone in situ data for Alert, Neumayer station, and Zeppelin Mountain have been made available by the Norwegian Institute for Air Research. Database of observation data of atmospheric chemical composition and physical properties, EBAS. http://ebas.nilu.no. Data of Alert are provided by Environment Canada / Atmospheric Environmental Service (EC/AES), data of Neumayer station by Helmholtz-Zentrum

Geestacht (HZG), and data of Zeppelin Mountain by Norwegian Institute for Air Research (NILU).

Ozone in situ data for Barrow, Summit, and South Pole station have been provided by U.S. Department of Commerce/National Oceanic & Atmospheric Administration (NOAA) – Earth System Research Laboratory – Global Monitoring Division. https://www.esrl.noaa.gov/gmd/ozwv/surfoz.

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