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Atmospheric
Chemistry
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Discussions

Interactive comment on "A three-dimensional model study of methanesulphonic acid to non sea salt sulphate ratio at mid and high-southern latitudes" *by* H. Castebrunet et al.

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Reply to Anonymous Referees

We thank both referees for their reading of our article, and are especially grateful to Referee#1 for his encouraging comments. Our confrontation of model results with observations has been read differently by the two referees: Referee#1 suggests emphasizing more our new insights into DMS chemistry at high latitudes, whereas Referee#2 questions our model approximations and performance. We discuss below some specificities of the high Southern latitude sulphur cycle in terms of both uncertainties and locally emphasized processes, as well as how non-modelled processed are discussed.

1- Specific answer to Referee#1

1a - Specific aim of this study (introduction).
A short paragraph was added at the end of the introduction (p.15000, l.25), specifying that we aim at better understanding the major processes responsible for the seasonal and meridional variations of R, with a special focus on the Antarctic continent in relation with ice core data and the past sulphur cycle.

1b - Short summary paragraph on atmospheric and ice core implication of this study (end of conclusions).
We added a short paragraph at the end of conclusions (p.15021 l.12), highlighting the sensitivity of R to DMS oxidation pathways and the impact of atmospheric chemistry and transport on the spatial distribution of R in snow.

1c - Model evaluation/validation: changed (see also 2d1)

2- Specific answer to Referee#2

2a - General comments

2a1- Model performance

This work discusses a "climatological" rather than "meteorological" time scale behaviour of the sulphur cycle. The data used are multi-year average values with different available years for the different stations and species (see captions of Figures 2, 3, 5, 6, 7, and p.15015 l1-5). Thus the transport is freely calculated by the climate model (4 years average, see p.15005 l4-6) rather than constrained with meteorological data from specific years. This is now specified at p.15003 l.13. This model setup, consistent with the nature of the data used, cannot aim at reaching the same precision than a model constrained for simulating highly instrumented measurement campaigns over specific periods of time. Being short lived species, the studied sulphur compounds undergo strong year to year concentration variations (often a factor of two or more) as illustrated by the vertical bars on Figs 2, 3 and 8, as well as high short time variability. This high variability is more finely illustrated by the detailed time-concentration signals in some cited references for the data used (e.g. Preunkert et al., 2007; Weller and Wagenbach, 2007). Variations of DMS, MSA and $nssSO_4$ are not always correlated due to their different lifetimes and the effect of transport (see e.g. Preunkert et al., 2007). In this context, model/data mismatches of a factor two or more do not necessarily mean bad model performance. Concerning the DMS gas phase oxidation scheme, Lucas and Prinn (2002) stress the fact that gas phase MSA production is under-estimated by factors 10^4 to 10^5 by the detailed processes taken into account in their study (see also 2a2). Earlier attempts to simulate year-to-year sulphur species variations with constrained meteorology lead to under-estimated variabilities (Sciare et al., JGR, 2000, p26369-26377, Cosme et al., 2002) and stressed the potential role of year to year variations of sea-to-air DMS fluxes (involving the variabilities of DMS concentrations in sea water, sea-to air transfer and the complex effect of sea ice on both of them). The above examples illustrate the fact that the simplified chemistry scheme is not the only major source of model/data mismatch (see also e.g. p.15003 l.21-23 (DMS emissions) p.15008 l.2-15 and p.15012 l.19-24 (transport and deposition) p.15018 l.21 - p.15019 l.3 (deposition)). We modified our manuscript in order to specify more clearly that we are using a climate mode model setup in relation with heterogeneous multi-annual data (p.15003 l.13).

2a2- Simplified chemical mechanism.

We describe the complexity of DMS oxidation chemistry in Section 2 (p 15001- 15002) and explain why two important processes: heterogeneous loss of DMSO and DMS oxidation by BrO were not modelled at p15004 l21 - p15005 l22. In the case of DMS+BrO, specifically pointed by Referee#2, it is due to the difficulty of generating reliable time dependent 3D BrO fields linked to the incomplete understanding of it source processes on snow and sea-ice (BrO is further discussed below in subsection 2b). The effect of the missing DMSO sink is evaluated indirectly through the comparison of MSA/nssSO_4 and (MSA+DMSO)/nssSO_4 ratios, and the effect of BrO is discussed in terms of model/data differences using its specific seasonality (p.15010 l.9-11, p.15010 l.26 - p.15011 l.5). We also stress that the strong effect of air-snow interactions on polar boundary layer oxidant chemistry (HOx, NOx) is not represented (p.15007 l.10-18). To our knowledge, no global model takes into account these complex processes yet, although they are very important in the Arctic and Antarctic boundary layers. We discuss their potential impact in terms of model/data differences at inland Antarctic sites less influenced by marine chemistry than coastal sites (p.15007 l.10-18). Concerning the simplifications of the gas phase DMS oxidation scheme (with OH and NO_3), they aim at reducing the number of model species as 3D chemistry/transport modelling of a tracer is computer time expensive. When dealing with seasonal (i.e. somewhat long term) variations of a few major species, some low concentration short lived intermediate species can be ignored safely if the branching ratios leading to their oxidation products are correctly taken into account. Note that Karl et al. (2007) start their summary section by stressing the good performance of the simplified oxidation scheme tested (NI, four species and six reactions) in predicting end product concentrations (MSA, nssSO_4 and their ratio). This is further discussed below in relation with the study of Lucas and Prinn (2005). We discuss mismatches between (always imperfect) models and (always incomplete) data, using their specificities (seasonality, location, speciation, etc.) in order to provide insights about which missing processes are most likely important. Looking at it the other way, a perfect model/data match would mean that there is no more research work to be done on the subject (assuming that it is not due to artificial tuning and/or compensation of errors). In the middle, we think that the best way to tackle incompletely understood processes in a model study is ambiguous, dependent on the objectives of the study and the long-term model development effort. The sulphur cycle is especially complex, with its multi-phase aspects and numerous chemical reactions. Despite its under-constrained aspect, we believe that our study provides new useful

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insights for the understanding of MSA/nssSO_4 ratios in ice cores and DMS chemistry at high latitudes as stressed by Referee#1.

2a3- Performance of other models.
A wide range of models (e.g. mechanistic chemistry box models to 3D chemistry-transport, chemistry-climate or even earth-system models including biological processes, with various forms of reduced complexity models) find their specific applications without excluding one another. The comments below do not aim at discrediting any of the mentioned studies, but only at pointing their different natures. We characterize the references quoted by Referee#2 the following way:

- Barnes et al. (2006) is mostly an in depth review of the DMS oxidation scheme (reaction rates and pathways) providing recommendations for modelling studies, it is quoted several times in our article (p.14977 l.17, p.15001 l.10, p.15002 l.4). Barnes et al. (2006) mention twice in a positive way our model's initial chemistry scheme (Boucher et al., 2003 - see also subsection 2c6).

- Karl et al. (2007), Lucas and Prinn (2002) and von Glasow and Crutzen (2004) use 0D or 1D mechanistic models applied to marine boundary layer chemistry. This family of models is unable to represent seasonal or large scale meridional variations due to the absence of (horizontal) transport. This problem is amplified at high latitudes as polar night chemistry is slow (see p.14999 l.24 - p.15000 l.1). Due to this limitation these models are run over periods of only a few days, and aim at testing specific or complex short term chemical processes. We quote similar studies, trying to focus on those applied to Antarctica (e.g. Jefferson, 1998) or analyzing the MSA/nssSO_4 ratio (e.g. Kerminen, 1998, Koga and Tanaka, 1999).

- Lucas and Prinn (2005) use a 3D chemical-transport model run for 1 year with constrained meteorology in relation with measurement campaigns. A major difference between this study and ours is that Lucas and Prinn (2005) discuss gas phase MSA and sulphate concentrations, whereas we discuss total (mostly aerosol) MSA and sulphate. Another important difference with our study is the absence of aqueous phase sulphur oxidation processes in the Lucas and Prinn (2005) study, which concentrates on gas phase chemistry. As in our study, DMS+BrO is not taken into account. Lucas and Prinn (2005) compare two detailed and two simplified chemistry schemes. They recommend the use of temperature dependent branching ratios and explicit modelling of DMSO. This is the case in our study. This was implemented in an earlier version of our chemistry scheme (e.g. Cosme et al., 2002; Boucher et al., 2003). The comparison of this earlier oxidation scheme with the two simplified mechanism tested by Lucas and Prinn (2005) is discussed in Boucher et al. (2002). In our study, we further updated the temperature-dependent reaction rates and branching ratios (see p.15003 l.27 - p.15004 l.18 and Table 1) following e.g. recommendations from Atkinson et al. (2006) and Sander et al. (2006). The two detailed oxidation schemes tested by Lucas and Prinn (2005) differ by their MSA production pathways: directly from DMS (C1) or through DMSO and MSIA (C2). The resulting (gaseous) MSA concentrations are within a factor of five in most regions, with higher differences in the high latitudes lower and middle troposphere (Lucas and Prinn, 2005, p9). An overall better consistency with measurements is obtained when MSA production occurs through DMSO and MSIA, which is the case in our study.

- von Glasow and Crutzen (2004) focus on halogen chemistry (not taken into account by the above three other model studies) using a 1D model and a complex specific mechanism involving several halogen species, gas phase and pH dependent aqueous phase sulphur-halogen reactions, and interactive oxidants. This study mentions that so far, no global information about BrO concentrations in the marine boundary layer (MBL) is available. von Glasow and Crutzen (2004) stress the fact that gas phase DMS+BrO chemistry strongly emphasize the effects of the uncertainty on end products of DMS oxidation, due to the uncertainty on the products of the MSIA + OH reaction (see e.g. second sentence of their abstract). We also stress this uncertainty in our abstract, conclusions and text. As oxidation processes by halogens have both MSA and nssSO_4 as end products in the gas and aqueous phase, with incompletely constrained branching ratios, their overall effect on MSA/SO_4 ratios is difficult to predict. In summary, these studies involving widely different modelling tools and objectives are somewhat difficult to compare with our paper. We found no evidence that they lead to overall smaller model/data mismatches than in our work. We now quote Karl et al. (2007) and Lucas and Prinn (2002) p.15001 l.26; Lucas and Prinn (2005) p.15004 l.11; and von Glasow and Crutzen (2004) p.15002 l.20 and p.15005 l.3.

2b - Halogen chemistry.

The fixed BrO test suggested by Referee#2 was performed with a previous version of our model (Boucher et al., 2003) as mentioned in the manuscript (p.15004 l.28-29). The major reason why we did not perform full implementation is explained in the model description section (p.15005 l.1-3): it would require to generate 3D time dependent BrO fields, and the current knowledge of BrO production on snow and on sea-ice is insufficient to permit full modelling. This is introduced p.15002 l.13-18. For example, Simpson et al. (ACP, 2007) indicate that in the Arctic BrO more likely originate from first year sea ice than frost flowers. Wagner et al. (ACP, 2007) confirm a BrO - first year sea ice correlation around Antarctica and point some inconsistencies of their results with satellite measurements. On the other hand, Jones et al. (ACP, 2009) observed enhanced BrO levels in high wind speed and saline blowing snow conditions in coastal Antarctica. Saiz-Lopez et al. (2007) observed elevated IO and BrO levels (up to 6 ppt) in air masses over Coastal Antarctica that had not been over sea ice for at least three days, and suggest that halogen activation occurs on Antarctic snow. The anti-correlation mentionned p.15002 l.19-20 (Introductory section on DMS oxidation) mostly aims at pointing that field data are not completely understood, and should not be isolated from the previous introduction on BrO emissions and DMS lifetime. We suppressed the following sentence (p.15002 l.20-21), which was confusing. The test performed by Boucher et al. (2003) indicates that 1ppt BrO in the lower 1.4km of the atmosphere induces a 40 to 60% decrease in DMS concentrations at high Southern

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latitudes. The DMSO production rate from oxidation of DMS reaches more than 60% at high Southern latitudes. The observed average BrO levels in the Antarctic boundary layer exceed 5ppt in October-November and are of about 3 ppt in December-March (Saiz-Lopez et al., 2007). Extrapolated at the regional scale, such BrO levels would drastically reduce DMS levels in the atmosphere and create a strong inconsistency between the commonly used DMS emission estimates from Kettle et al. (1999) and observed atmospheric DMS concentrations. It would also produce DMSO levels much higher than the observed values. Based on observations of DMS, BrO and MSA at Halley Bay, Read et al. (2008) calculated that assuming a 80% DMSO production from DMS+BrO with 3ppt of BrO increases MSA production from DMS by a factor 9. Thus including similar BrO levels and oxidation rates would likely strongly increase MSA/nssSO_4 ratios and the discrepancies with observed levels. Due to large uncertainties on several aspect of the DMS cycle, we could argue that other processes might compensate for these discrepancies. However, the DMS-BrO interactions should be better constrained before putting forward such arguments. These discrepancies are summarized p.15010 l.24 - p.15011 l.5, and we suggest that DMSO measurements at Halley Bay would help solving these issues. We now mention DMS+BrO chemistry in the abstract and conclusion. We also modified Section 2 (p.15002 l.20, see also 2a3). We further discuss interactions of BrO with other oxidants below (2c8).

2c - Model description.

Describing a zoomed GCM-tracer model in a few pages requires some choices. We focus on recent model configuration changes and try to avoid repeating previous publications. Note that additional information is provided in result sections, when directly used in the discussion (see 2c4). We completed Section 3.1 taking into account comments by Referee#2.

2c1 - Questions related to model grid.

We briefly describe our model grid at p.15003 l7-13 and refer to Krinner et al. (1997) and Cosme et al. (2002) for complete setup. The grid covers the whole globe, and we now specify that the model is global at p.15003 l3. The vertical grid was adapted to better represent the Antarctic boundary layer (Krinner et al., 1997). The mean elevation of the first 6 layers above the surface is 3, 9, 21, 44, 82 and 141m (this is now specified p.15003 l.11). The number of vertical levels representing the boundary layer is not easily defined due to diurnal, seasonal and geographic variations of the boundary layer height (see e.g. Handorf et al., Boundary-Layer Meteorology, 2009 and Argentini et al., Boundary-Layer Meteorology, 2005).

2c2 - Online/offline and meteorology.

Winds and temperature are fully generated by the climate model, this difference with our previous model setup is commented p15005 l.23-25. The only offline element is oxidant fields as specified at p.15003 l.24-26. We now further specify these model configuration aspects p.15003 l.13 and l.16 (see also 2a1).

2c3 - Spin up and number of modelled years.

As described p.15005 l.4-6, 5 years simulations were performed, the first year is discarded for model spin-up.

2c4 - Deposition velocities for MSA and nssSO_4.

Our general scheme for wet and dry deposition is described in Boucher et al. (2002) (see p.15003 l.14-18). MSA and nssSO_4 dry deposition velocities on snow were modified (p.15008 l.5-6) and are equivalent for MSA and nssSO_4 (p.15012 l.21-22). We now introduce this dispersed information in the model description section (p.15003 l.17).

2c5 - Volcanic / Anthropogenic sources.

Sulphur sources other than oceanic DMS are described in Boucher et al. (2002). Concerning the anthropogenic sources, we also refer to the Cosme et al. (2005) study which quantified their impact (as well of the impact of other sources) in Antarctica (e.g.

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p.14997 l. 14-16, p.15009 l.4-5). REF-ANTH is defined p15005 lines 4-10 together with other simulations. We added more information about sulphur emissions in the model description section (p.15003 l.16). The Korhonen et al. (JGR, 2008) study cited by Referee#2 deals with the roles of DMS, sea-salt and continental sources on CCN number density. We found no direct implication of non-linearities obtained for CCN number density on more mass conservative quantities such as MSA and nssSO_4 concentrations. Isolating specific sources is a commonly used way of testing their influence (see e.g. Granier et al., Atmos. Environ., 2000), although their effects are not always strictly additive due to e.g. non-linear effects of chemical processes when oxidant concentrations are affected.

2c6 - DMS+O_3 reactions in the model.

We use the aqueous chemistry scheme of Boucher et al., (2002) and the same DMS + O_3 reaction rates as Boucher et al. (2003), as specified p.15003 l.14 and p.15004 l.13-18. Note that Barnes et al. (2006) equally quote Boucher et al. (2003) and von Glasow and Crutzen (2004) for their estimates of the global impact of DMS+O_3 aqueous reaction. Short summary of the aqueous chemistry scheme: cloud parameters (e.g. liquid water content) are calculated by the climate model. Henry's Law is assumed for gas-liquid transfers of chemical species, and a simplified pH calculation is performed using aqueous phase sulphur species concentrations and imposed [NH_4^+]/[SO_4^{2-}] ratios. Barnes et al. (2006) recommend the Boucher et al. (2003) approach as a simplified (Henry's law equilibrium) way of implementing multiphase chemistry in 3D models. We now provide more indications about our treatment of aqueous chemistry (p.15004 l.13).

2c7 - DMS flux.

The NOAA-PMEL database contains regularly updated measurements of DMS concentrations in sea surface water. However, it cannot be used as model input because the important work of extrapolation to (largely dominant) areas where no data are available, performed by Kettle et al. (1999) and Kettle and Andreae (2000) is not performed.

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The reason why we use Kettle et al. (1999) rather than Kettle and Andreae (2000) is that Kettle and Andreae (2000) lead to too high DMS concentrations around Antarctica in summer. Note that Lucas and Prinn (2005) and von Glasow et al. (ACP, 2004) also use Kettle et al. (1999). In order to evaluate the recent improvements of DMS oceanic concentrations database for the extra-tropical South hemisphere, we compared the data from years 1972-1998 and 1972-2009 extracted from the NOAA-PMEL database, for three latitude bands: 79-65, 79-60 and 79-30 degree South. This is now included in the Supplementary material and mentionned p.15003 l.23. Winter measurements are still very scarce: no data were added for the June to September months, even at mid-latitudes. We found an interesting feature in the summer season data: observations from the highly productive Ross sea sector are included in the 1972-1998 data for the December to February period, whereas they were added after for November. With the addition of new data from other sectors, mean DMS concentrations at high latitudes in the overall database are lower than in the 1972-1998 data in December to February, whereas mean values nearly double in November. Thus the mean summer DMS concentrations around Antarctica may be very sensitive to the weight attributed to the Ross sea sector. Note that sensitivity tests to different DMS oceanic emissions were performed in earlier studies from our group (Cosme et al., 2002, Boucher et al., 2003).

2c8 - By using offline oxidant concentrations you are assuming infinite supply of oxidants.

This is somewhat common in 3D global models, for example Lucas and Prinn (2005) also use fixed oxidant fields. Neglecting oxidant consumption by sulphur chemistry introduces a bias only if sulphur chemistry has a major impact on oxidant destruction. In the case of OH, a quick comparison of Kco+oh[CO] with Kdms+oh[DMS] (where K denotes reaction rates) indicates that OH is destroyed about 20 times faster by CO than DMS in Antarctic summer conditions. The fact that global chemistry models not taking into account sulphur chemistry simulate realistic tropospheric ozone fields provides a similar argument for O_3. However, our fixed oxidant fields do not take into account specific conditions of elevated halogen concentrations implying tropospheric ozone depletion, which induce a reduction of OH production (see e.g. von Glasow and Crutzen, 2004). Thus a complete simulation of DMS interactions with halogen oxides would involve interactive oxidant calculation and may require interactive calculation of photo-dissociation rates including representation of the complex polar insolation conditions in very dim light conditions, as elevated BrO levels were observed at 92 degree solar zenith angle by Saiz-Lopez et al. (2007). These calculations are not impossible, but very expensive in terms of computing time in 3D global models. We now mention the BrO interaction with other oxidants (p.15005 l.3).

2c9 - Intermediate products of the DMS oxidation, why have you not improved this description in the model?

Summarizing the above discussion (2a2 and 2a3), we doubt that we would obtain more reliable or better results (in terms of model/data consistency) as the pathways and rate of MSA production are still very uncertain.

2d1 - Model "validation".

Our French mother language group perceives "model validation" and "model evaluation" as both commonly used and nearly equivalent. We may have missed an English language subtlety and modified this term (p 15000 l.21 and p.15005 l.11). Our intention was to use an appropriate technical term rather than being pedantic.

2d2 - Mismatches of very often a factor of 2x or 3x but up to 6x.

To our knowledge, this is not a poor performance, considering the complex multiphase sulphur cycle and uncertainties on many aspects of it. We are not aware of comparable model studies with much better performances (see also the discussion at 2a1, 2a2, 2a3).

2d3 - Cape Grim discussion.

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The Cape Grim discussion (p.15013 l.10-28) appears in the results section and mostly aims at analysing the oceanic versus anthropogenic impact at this mid-latitude site. We modified our text to make this more clear (p.15013 l.15, and l.24).

2d4 - R at coastal sites - how many data points to you actually refer to?

We use seasonal R data from 5 different long term sulphur measurement stations in coastal Antarctica (see Figs 4 and 5 and p.15009 l.8-11).

2d5 - Found it rather bold to use this model with all its limitations to try and model glacial cycles, what meteorology? oxidant levels?

This work is published (Castebrunet et al., 2006) as mentioned in the paper (p.15017 l.4-5). The LMD climate model has been used for paleo-climate modelling for decades and is involved in international paleo-climate model inter-comparison studies (see e.g. Guiot et al., Clim. Past., 2007, preface of the special issue: Modelling Late Quaternary Climate). Oxidant fields are altered using results from a 2D model constrained with specific LGM boundary conditions (Martinerie et al., JGR, 1995). Recently, 3D climate-chemistry models also started to be used to simulate LGM oxidants (e.g. Valdes et al., GRL, 2005, Kaplan et al., GBC, 2006).

2e - Specific comments

2e1 - p. 14998 l.13-14 rephrased as: "This has been interpreted as a decrease of MSA production in winter related to lower HO_2 levels (Ayers et al., 1996)".

2e2 - p.14998 l.14, p.14999 l.28, p.15000 l.9: corrected, thanks.

2e3 - p.15002 l.4, "If NO_3" changed to "Although NO_3"

2e4 - p.15007 l.2-3: model $nssSO_4$ results; "dispersed" changed to "variable" Section 4 and elsewhere: R_{obs}, R_{sim}, R_{psim} changed; "than in fall" changed.

2e5 - "surprising that the better correspondence to measured data for Rpsim only holds when the aqueous phase reaction of ozone with DMS is ignored." This reaction likely plays a more important role at high southern latitudes than on the global sulphur cycle because other oxidant levels are smaller, especially during polar night, as explained p.15006 l.11-17. For example Cosme et al. (2002) found that the SO_2 sink over Antarctica is dominated by aqueous oxidation by O_3 rather aqueous oxidation by H_2O_2. In these unusual oxidant conditions with increased relative importance of O_3, our results suggest that the DMS+O_3 aqueous sink might be over-estimated. We now further discuss this p.15010 l.25.

2e6 - Gondwe (p.15011 l.6-17). This discussion focuses only on coastal Antarctica, the Gondwe et al. (2004) global scale study of MSA/SO_4 is referred to in more general terms p.1499 l.17, p.15000 l.1-8, p.15014 l.21-25. For coastal Antarctica, observed values of the MSA/SO_4 ratio in summer range between about 0.15 and 0.45, except at Palmer station: 0.55-0.8 (see Fig.5). Our model results range between about 0.2 and 0.45, except at Palmer (0.3-0.7), whereas Gondwe et al. (2004) obtain about 0.7-1.0 (see their Tables 1 and 2, no results for Palmer). Gondwe et al. (2004) obtain very high values (frequently exceeding 1) in March and September-November, this would be out of scale on our Fig.5. Gondwe et al., 2004 comment this specific model bias in their paragraph [17].

2e7 - p.15014 l.19 "linked with" changed to "linked to".

2e8 - p.15016 l.4: R_{sim} decreases South of 40°S. R_{psim} has a different behaviour (see Fig.8). The model/data discrepancies are discussed p.15015 l.14-26, our most important comment is the high dependence of model results (bracketing the data at extra-tropical latitudes) on the adopted DMS gas phase chemistry scheme. The second part of the discussion (p.15016 l.7-25) comments our sensitivity tests isolating different processes. The limited time and space coverage of the data (p.15015 l.2-4) also has to be taken into account (p.15016 l.18-22).

2e9 - p.15018 l.4: R_{psim} deteriorates the results (in snow). Wording changed. The

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atmosphere-snow transfer of MSA is highly debated in the glaciological community and has important implications for ice-core data interpretation (e.g. p.15018 l.26 - p.15019 l.1, and Castebrunet et al., 2006). In this context, and taking into account other uncertainties (especially on deposition rates on snow), model/data discrepancies are not unexpected, and the fact that our model results present some common features with the observed spatial variations of R is of interest (see comment two of Referee#1).

2e10 - p.15018 l.13: "twice larger than" changed.

2e11 - Web link for PhD thesis of H. Castebrunet added (http://tel.archives-ouvertes.fr/docs/00/18/79/47/PDF/these_castebrunet.pdf)

2e12 - Acidity, liquid water content, gas-liquid phase transfer: see 2c6.

2e13 - Technical corrections of Table 1, Figures and Supplement done.

2e14 - References b-d appear on the continuation page (p.15034).

2e15 - Figure 8: Bates et al. (1992) data originate from a single ship cruise (p.15015 l.2-4) and cannot reflect the natural variability of R. The data from Amsterdam Island and Cape Grim are presented in the caption of Fig.7. Simulated year: see 2a1.

2e16 - Supplementary Fig. 1 (line for case REF-ANTH for MSA missing): MSA has no anthropogenic sources, thus the two lines are superimposed. We now mention it in the caption.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 14995, 2009.