**Response to Referee #1:**

We would like to thank the referee for her/his helpful remarks. Please find below point-by-point reply to your comments.

We agree to your argument that our study focuses on the rate/degree of non-linearity in the system rather the non-linear dependency seen in the variables discussed in this study when DMS emissions are doubled. We, hence, change the title of this paper to 'Rate of non-linearity in the DMS aerosol-cloud-climate interactions'.

**Q.1) What kind of situation would create such doubling in DMS fluxes? Is the doubling possible based on the current levels of knowledge? Comparable climate change induced changes are usually in range of +6.5% (Houghton et al, 2001), +2.4% (Bopp et al, 2004) and -8.0% (Stier et al, 2006).**

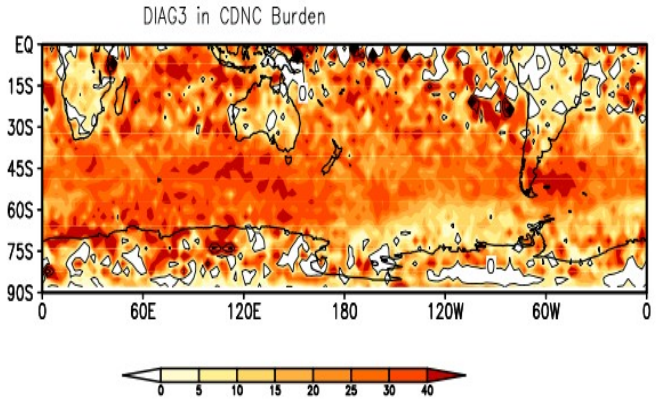
As pointed out by the referee, the doubling of DMS is not estimated in any of the future climate scenarios. However, our study was an idealistic experiment mainly driven by specific geoengineering proposals that were put forward to counteract global warming via stimulating DMS aerosol-cloud-climate interactions through iron fertilization. This is mentioned in the revised manuscript (Section-1; para-2). The present study also, in general, helps in understanding rate of non-linearity in the system. Please note that there is hardly any study that takes into account end to end chemistry-cloud-climate interaction.

**Q.2) Are the simulated SO2, DMS and H2SO4 levels reasonable compared to (similar, not necessarily from the same region) measurements? Are the DMS emissions used in the base-case simulations reasonable (as far as can be determined)?**

In Thomas et al., (2010), the relevant parameters such as the DMS flux to the atmosphere, sulfate distribution and cloud microphysical variables were evaluated. The simulated global annual DMS flux to the atmosphere was estimated to be 23.3 Tg(S)/yr and agrees well with the estimates of Boucher et al., (2003) that used the same gas exchange parameterization of Nightingale et al. (2000) and Kettle and Andreae (2000) DMS climatology. The seasonal variation in modelled $nssSO4^{2-}$ in our baseline simulation (CTRL) over the southern oceans is comparable to those of Gondwe et al. (2003) who estimated a 7-8 times increase in summer DMS emissions compared to winter. This information is included in Section-2 in the revised manuscript.

**Q.3) Are there indications that there is a linear region and non-linear region of the dCDNC/dfDMS or d(CD r)/dfDMS?**

The figure shows the spatial pattern of the percentage change in CDNC burden in 2X_ODMS simulation compared to the CTRL simulation (DIAG3). it can be seen that almost all regions show non-linearity in the system. The oceanic regions south of south America and parts of southern



most Pacific ocean where DMS emissions are highest (Thomas et al., 2010) correspond to regions of high rate of non-linearity.

**Q.4) Are there major sinks of OH not included in the simulations? Such as organic emissions from the oceans? How could they influence the non-linearity?**

The complete tropospheric MOZART2 chemistry (Horowitz et al., 2003) is included in the ECHAM5-HAMMOZ model used here. The oxidants of the pollutants in the atmosphere begins in most cases by photolysis or reaction with OH. However, the organic emissions from the oceans are not included in this study. In a study by Wingenter et al., (2004), it is shown that DMS concentrations are 10 times higher than isoprene during an iron fertilization experiment over a patch in the southern oceans. The author also adds that the increase in NHHCs such as isoprene may increase the local lifetimes of short-lived gases such as DMS by competing for OH. We have mentioned this in the revised manuscript (Section-2; para-3).

In the case here where we double the DMS fluxes through a hypothetical fertilization of the oceans, isoprene and organic emissions would also increase and further compete with DMS for oxidation with OH. Even if we cannot quantify it, we can suppose that the rates (2xDMS vs 1xDMS vs noDMS) of SO4 and CDNC formation would further decrease compared to our simulation. However, we have to keep in mind that the marine secondary organic aerosol (SOA) derived by isoprene and other organics oxidation can also act as cloud condensation nuclei (for example, papers by O'Dowd). Isoprene is proposed as the most probable source for SOA (Meskhidze and Nenes, 2006; Roelofs, 2008). More simulations will be needed to quantify the relative contribution of organic emissions to the changes in cloud microphysical properties.

**Q.5) I would also like to see more discussion on other potential non-linearities. Are the overall CDNC, TOA and CD effective radii nonlinearities dominated by the OH? There are many other factors in the the aerosol-cloud processes which could significantly change if the system is perturbed. Examples are: oxidation rates of other pollutants, growth of large particles (resulting in decrease of almost-CCN sized particles), increases in deposition rates of the grown particles, new particle formation, etc. These should be more discussed than actually simulated. Do the authors have a good reason to believe the OH process is the dominating one?**

This is an important comment. Please note that different factors affect rate of non-linearity in different parts in the cycle of chemistry-aerosol-cloud-climate link. For example, in the chemistry part, reactions with OH undoubtedly have the first order impact on the rate of non-linearity. But, for the aerosol-cloud interaction part, the availability of liquid water in the atmosphere critically important. Our results show that the SO4 production from gas phase oxidation of SO2 is highly non-linear and puts a significant limit to its linear behavior. Since the OH availability is not changed in all the three experiments and since the efficient sinks of OH are CO and CH4, the limited availability for gas phase chemistry should undeniably drive the non-linearity in the system. This is already mentioned in the manuscript (Section-3.1; para 2).

**Q.6) The DMS emission rates are strongly dependent on the meteorological conditions. In the simulations done in this work, the model was nudged to ECMWF fields. However, the nudging on my understanding will not produce exactly same meteorological fields due normally every 6h nudging. Do the surface wind fields show strong variability between the simulations (probably not)?**

We would like to point out that monthly DMS sea water concentration climatology is prescribed from the Kettle and Andreae (2000) data base and they are then interpolated for each model time step. As expected by the referee, we investigated surface wind fields between our simulations and found extremely negligible variability. Please note that our 6-hourly wind fields would remain the same in all our three simulations.

**Q.7) Do the authors consider other boundary conditions of their work as a major influence of the results? These include resolution, period of the study (2000), location of**

**comparison region (75-30S), relatively old-fashioned new particle formation routine (Vehkamaki, 2002), sea salt emission rates, etc?**

Referee raises an interesting point. It is important to consider how other boundary conditions might influence our results. For regional scale studies, the model resolution will certainly play an important role, but, the large scale statistical features as studied here, will not be significantly influenced. Kettle and Andreae (2000) showed that the interannual variability in DMS solely due to wind speed is up to 10% which is considerably less than the seasonal variability. So, we expect that the DMS climatology should be a good representative of the current climate scenario. Vast amount of literature demonstrates the dominance of DMS emissions in the chosen latitude band of 30-75S based on ground based measurements and satellite data sets (Charlson et al., 1987; Ayres et al., 1997, Shon et al., 2000 and references therein) . Therefore, this is an ideal region to study impacts of DMS emissions and additionally, this region is free from anthropogenic pollutants. We do, however, understand that, new paramterization schemes for particle formation would provide slightly different estimates, but, the impact of such schemes are not yet evaluated in the framework of ECHAM5-HAMMOZ model.

**Specific comments:**

**Regarding tables: Is there a specific reason why in e.g. Table 1, the row title explanation is given above the column title explanation. This is confusing (and the reason for the arrows?) as the column titles are aligned on the top of the table cell. I would either change the order (parameters // diagnostics) or remove the explanations altogether and explain in the figure caption.**

The first box in the first column now reads as 'Diagnostics' only.

**Figures have (at least in my PDF) atrocious resolution. They are readable, but should really be given in better format for publication.**

I have submitted the figures in very good resolution in the revised manuscript.

**References:**

Thomas, M. A., Suntharalingam, P., Pozzoli, L., Rast, S., Devasthale, A., Kloster, S., Feichter, J., and Lenton, T. M.: Quantification of DMS aerosol-cloud-climate interactions using the ECHAM5-HAMMOZ model in a current climate scenario, Atmospheric Chemistry and Physics, 10, 7425–7438, 2010.

Boucher, O., Moulin, C., Belviso, S., Aumont, O., Bopp, L., Cosme, E., von Kuhlmann, R., Lawrence, M. G. ., Pham, M., Reddy, M. S., Sciare2, J., and Venkataraman, C.: DMS atmospheric concentrations and sulphate aerosol indirect radiative forcing: a sensitivity study to the DMS source representation and oxidation, Atmos. Chem. Phys., 3, 49–65, 2003.

Nightingale, P. D., Malin, G., Law, C. S., Liss, A. J. W. P. S., Liddicoat, M. I., Boutin, J., and Upstill-Goddard, R. C.: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, Global Biogeochemical Cycles, 14, 373–388, 2000.

Kettle, A. and Andreae, M.: Flux of dimethylsulfide from the oceans: A comparison of updated data sets and flux models, Journal of Geophysical Research, 105, 26,793–26,808, 2000.

Wingenter, O. W., Haase, K. B., Strutton, P., Friederich, G., Meinardi, S., Blake, D. R., and Roland, F. S.: Changing concentrations of CO, CH4 , C5 H8 , CH3 Br, CH3 I, and dimethyl sulfide during the Southern Ocean Iron Enrichment Experiments, Proc. Nat. Acad. Sci., 101(23), 8537–8541, 2004.

Horowitz, Larry W.; Stacy Walters, Denise L. Mauzerall, Louisa K. Emmons, Philip J. Rasch, Claire Granier, Xuexi Tie, Jean-François Lamarque, Martin G. Schultz, Geoffrey S. Tyndall, John J. Orlando, Guy P. Brasseur, A global simulation of tropospheric ozone and related tracers: Description and

evaluation of MOZART, version 2. Journal of Geophysical Research, 108 (D24), 2003.

Meskhidze, N. and A. Nenes, Phytoplankton and cloudiness in the Southern Ocean, Science, 314, 1419-1423, 2006.

Roelofs, G.J., A GCM study of organic matter in marine aerosol and its potential contribution to cloud drop activation, Atmos. Chem. Phys., 8, 709-719, 2008.

Charlson, R. J., J. E. Lovelock, M. O. Andreae, and S. G. Warren, Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate, Nature, 326, 655–661, doi:10.1038/326655a0, 1987.

Ayers, G.P., Cainey, J.M., Gillett, R.W., Saltzman, E.S., Hooper, M., Sulfur dioxide and dimethyl sulfide in marine air at Cape Grim, Tasmania. Tellus B 49, 292-299, 1997.

Shon, Zang-Ho, D. Davis, G. Chen, G. Grodzinsky, A. Bandy, D. Thornton, S. Sandholm, J. Bradshaw. R. Stickel, L. Mauldin, D. Tanner, and F. Eisele, Evaluation of the DMS Flux and its conversion to SO2 over the Southern Ocean, Atmos. Environ., 35, 159-172, 2001.