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Reply on RC2

Catherine Hardacre et al.

Author comment on "Evaluation of SO_2, SO_4^{2-} and an updated SO_2 dry deposition parameterization in the United Kingdom Earth System Model" by Catherine Hardacre et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-238-AC2, 2021

Reply to Reviewer 2

We thank the reviewer for their careful reading of the manuscript and their suggestions for improvement. The Reviewer's comments and our responses can be found below.

[Specific Comments]

I am a bit skeptical when it comes to the selection of observations to be used for evaluating trends. More details of this and other specific comments in the bullet points:

1. Line 35-38. Note that the unit is TgSOx (as SO2)

=> Thank you for highlighting theis error. Now corrected.

2. Line 69. “Relatively short lifetime”. Relative to what? A lot of species has shorter lifetime than this.
=> We have ammended this statement, please see our response to Comment 3.

3. Line 70. In this general statements it seems like the removal of SO2 is mainly described as deposition of the component. Even though it is described later in the introduction one should mention that oxidation of SO2 to SO4 is important factor for the lifetime of SO2, and that rate is dependent on the oxidation capacity and the acidity of the cloud droplets (which other atmospheric components like NH3 will influence). Maybe here just make it shorter and state that the lifetime of so2 depends on both wet and dry deposition of the molecule and the oxidation rate to SO4?

=> We have reworded this paragraph in line with the reviewer's suggestions. The paragraph is now as follows: '... The lifetime of SO2 depends on both wet and dry deposition of the molecule and the oxidation rate to SO4. The 2 day lifetime is such that much of the loss via oxidation and deposition occurs locally. SO2 loss near sources and the impact of environmental conditions on loss processes have been investigated in a number of studies.'

4. Line 75-90. In this section, the references are really old.

=> We agree that there references in this section are relatively old, these studies having been done from ~1970-1990 when acid deposition over Europe and North America was a

serious problem. While there has been a number of measurement studies measuring SO2 dry deposition, particularly the impact of surface wetness on deposition velocity (e.g. Fowler et al., 1995; Feliciano et al., 2001; Matsuda et al., 2006; Tsai et al., 2011; Myles et al., 2012) there appears to have been less drive for the type of studies we reference here i.e. looking at loss rates close to sources. There has also been considerable work to develop parameterizations for SO2 dry deposition in regional and global scale models over the last 20 years or so (e.g. Zhang et al., 2003; Ganzeveld and Lelieveld, 1995; Ganzeveld et al., 1998). We particularly cite Garland and Branson, 1977; Fowler et al., 1978; Erisman and Baldocchi, 1994 and Erisman et al., 1994 as they highlight that the loss processes are occuring sub-grid scale compared to UKESM1.

5. Line 119 – 122 starting with "Following the increasingly.." seems a bit oddly placed in the introduction. Should maybe be moved to the beginning as the reason why monitoring are being conducted?

=> We have re-worded this sentence as follows: 'In the 1970's and 1980's the increasingly detrimental impacts of rising SO2 emissions on acid deposition, air quality and human health in Europe and North America led to monitoring networks being set up in these regions...'

6. Line 118. I don't really agree with the statement of "the main challenge to capture historical trends" is the lack of observations. Sulfur is one of the species that has been monitored the most in especially Europe and North America. But there are of course regions in the world where this stamen is very valid. I assume one of the challenges is the non-linearity in trends, i.e. the dependence on atmospheric chemistry on the sulfur trends, and the lack of a range of data to detailed process studies on a large scale as well as long term flux measurements and not only atmospheric concentrations?

=> We agree with the reviewer in that sulphur species are relatively well observed compared to many atmospheric constituents, at least in Europe and North America for the period from the 1970's/80's to the present day. However, the point we woudl like to make is that even with these data sets we can still only evaluate part of the model's historical simulation (which runs from 1850 - 2014), and similar data sets are not available for other major source regions such as India, China and the Middle East, or remote regions. While the satellite observations of SO2 can help with spatial coverage they are somewhat temporally limited (with regard to evaluating the historical period) and capture only SO2 through the column. We do also mean to allude to the lack of data available for large scale process studies, i.e. flux measurements and co-located measurements for relevant species. We have ammended the paragraph to capture these points as follows:
'Sulphur species are relatively well observed compared to many atmospheric components as their role in air pollution is well established. In the 1970's and 1980's the increasingly detrimental impacts of rising SO2 emissions on acid deposition, air quality and human health in Europe and North America led to monitoring networks being set up in these regions (Torseth 2012; CASTNET 2004). Rising pollution in Asia also led to the establishment of the The Acid Deposition Monitoring Network in East Asia (EANET) in 2001 (e.g. Wang 2008). However, even with these data sets it is only possible to evaluate model simulations of the recent historical period and similar data sets are not available for other large source regions such as India, the Middle East, or remote regions. Further, the lack of a range of measurements, including flux observations, hinders detailed process studies at large scales.'

7. Chapter 2.4. It seems like it is not a criterion to have co-located SO2 and SO4 observations and then you could have benefited from also using SO4 aerosol data from IMPROVE. Has that been considered since you state in the beginning that too little data is hampering the comparison with models?

=> The reviewer makes a valid point regarding the IMPROVE data set. However a clear advantage of the CASTNet data for this study is the co-location of the SO2 and SO4 observations, and SO2 dry deposition data. While we are not quite doing a full process analysis here, we are trying to understand bias in UKESM1's representation of sulphur cycle, including processes. In addition we seek to improve the model by modifying the dry deposition process and it is valuable to be able to link the impact of those changes through comparisons of dry deposition, SO2 concentration and sulphate. Please note that we have also ammended our EMEP data set, limiting the measurement sites to those which have longer term data sets and both SO2 and SO4 available (see also the response to Comment 9). We have updated Section 2.4 to include this information.

8. Line 251. SO2 and SO4 are measured with filter pack sampler and weekly sampling intervals, not hourly measurements as stated.

=> Thank you for highlighting theis error. Now corrected.

9. The number of sites has varied through the period and it seems like you have used all the sites without considering the length of the time series? If so, have you compared the trends using only sites with observations for the whole period? Especially in Europe the differences in site density throughout the period may influence the trend. In the beginning it was less sites in Southern Europe. Information about the number of sites should be included in the figure (and in table 3).

=> In updating the EMEP data set to include the data from 2010-2014 we also revised the sites we used in the study. We limited the EMEP sites to those which had at least 10 years of continuous measurements (and in general where SO2 and SO4 measurements were co-located, see also the reply to Comment 7). In this study we have only considered the European region as a whole. Figure 6 shows that the model bias is indeed variable across Europe, and similarly to the USA, the biases are larger in the more polluted regions. As part of our wider analysis we did produce the plots shown in Figure 6 for the two time slices (1990-1995) and 2009-2014). However, the results were not substantially different from those for the whole time period, i.e model bias in SO2 and SO4 concentrations was lower at cleaner sites compared with polluted sites, (see also the response to Comment 12). Therefore we did not see any clear benefit to the manuscript. We have now included the number of sites used to produce Figures 2, 3, 4, 5, 6, 7 and Tables 3, 4, 5, 7 (see also the response to Comment 13).

10. Fig2 and Fig 4 (and fig 11) Why does SO4 in Europe only include data up to 2010? Surely there are observations after 2010 in EMEP, found in http://ebas.nilu.no/ (this database should also be included in the section of data availability). In Figure 6 it seems like you have used data up to 2014?

=> We have now included the full EMEP data set up to 2014 for SO2 and SO4 and the relevant plots and statistical data have been modified (Figures 2, 4, 6, 7, 11 and Tables 3, 4, 5, 7). We have also included the website for the EMEP observations in the data availability section.

11. Fig 3 and Fig 4. For the average annual concentrations for the different 5 years period. Have you used a criteria for the data capture needed to make an average. E.g 75%, 3 out of 5 years etc?

=> Yes, for the 5 year time slice statistics we have only used sites for which there was at least 3 out of 5 years. We have now added this information to Section 2.5.

12. Chapter 3.3. Have you calculated the per cent bias? That may give a different geographical distribution of the bias than absolute concentrations. In addition, it would be

interesting to know whether the model is able to capture the per cent changes (trends) at the different sites, that will give further insight if the model and observations are responding similar to the emission changes.

=> As part of our analysis we did look at percentage bias in addition to normalised mean bias (NMB) for the plots in Section 3.3 (Figures 5 and 6). However the percentage bias plots do not look substantially different from the NMB plots. The model bias (absolute or normalised) is greatest at the sites where the surface concentrations are highest. Given that the bias is very large at some sites, we felt that the NMB plots were easier to interpret than the plots of percentage bias, which could be much greater 100% in some cases. We also looked at the bias during the two time slices. Again, these plots did not show substantial difference from the plots for the full time period, i.e. the sites where the surface concentrations of SO2 ansd SO4 were greatest also had the hihgest bias. We have chosen not to focus on individual sites in this study as comparisons between point observational data and model grid cell output is difficult to justify in isolation, particularly for SO2 where loss processes (e.g. deposition/oxidation) are occuring within a single grid cell. We have summarised the trends in the modelled and observational data in Tables 3 and 4 where we report the trend for the two time slices and over the full period. While we acknowledge that some sites may be very different, we believe these results are indicative of the model's behaviour in the different regions.

13. Fig 7. I assume the blue shaded areas for the modelled and the black variations in the observations indicate the standard deviations between the sites? Should be mentioned in the caption in addition to how many sites are included in the analysis (also in Table 5).

=> In Figure 7 we have now added a description for what the blue shaded region and the black bars represent. We have also included the number of observational data sets used for each data set in the caption for Figure 7, and in Table 5.

13. Fig 12. In the figure caption you should include the time period you are looking at.

=> We have included the time period (2005-2014) in the caption.

[References]

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