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Comment on acp-2021-238

Anonymous Referee #1

Referee comment on "Evaluation of SO_2, SO_4^{2-} and an updated SO_2 dry deposition parameterization in UKESM1" by Catherine Hardacre et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-238-RC2, 2021

Hardacre et al. evaluate the earth system (ES) model UKESM1 before and after update of the model parametrization by comparing with long term sulfur trends in in-situ observations in Europe and North America as well as observations from satellites. For me it does not seem like the new parametrization really improved the model bias. In some areas yes, in others not. Since the main purpose of this paper is to evaluate model performance and not primarily to assess the sulfur trends, I do miss quantifying (or more discussion of the relative importance) of the main errors and not a list of all the potential contributions to it.

Too high emissions and uncertainties in the injection height may surely influence the bias in the model predictions. But the bias is too high that these can be the main reasons. Neither the DMS chemistry, which is not that important, especially not for the earlier period when anthropogenic sources dominate. As the authors also state, there is a need to further assess the parameterization of sulfur chemistry in the model and include cloud water pH (which is very important) and other species which influence the non-linear trends in sulfur. If possible wet deposition should also be included to make it possible to look at the whole sulfur budget in the model output.

Has the model been compared to other ES models? If so do you have any references of that? In Aas et al(2019) there are 6 different global models and they seem to better capture the trends and absolute values better than the UKESM1 model?

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:

- Line 35-38. Note that the unit is TgSOx (as SO2)
- Line 69. "Relatively short lifetime". Relative to what? A lot of species has shorter lifetime than this
- 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?

- Line 75-90. In this section, the references are really old.
- 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?
- 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?
- 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?
- Line 251. SO2 and SO4 are measured with filter pack sampler and weekly sampling intervals, not hourly measurements as stated.
- 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).
- 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?
- 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?
- 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.
- 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).
- Fig 12. In the figure caption you should include the time period you are looking at