gmd-2017-302: Global sensitivity and uncertainty analysis of an atmospheric chemistry transport model: the FRAME model (v. 9.15.0) as a case study
by Aleksankina et al.

Response to reviewer #1

This paper presents a very useful approach for quantification of the impact of emissions uncertainty on modelled concentrations and deposition of sulphur and nitrogen species. The material is presented clearly and the conclusions are supported by the results presented. I have a few minor comments about the methods section, where I think some further details would be useful.

Response: We thank the reviewer for their very positive comments on the usefulness and presentation of our work.

(1) *The annual average wind rose and wind speed used to calculate trajectories in the FRAME model are generated from WRF - what period was used to generate these trajectories, what resolution was WRF run at, what version of WRF was used and what meteorology was used to drive WRF at the boundaries?*

Response: The following expanded text and additional citation has now been added to the end of the first paragraph of Section 2.1.
"The trajectories are defined by an annual wind rose and annually-averaged wind speed generated for year 2012 from the output of the Weather Research and Forecast model (www.wrf-model.org) (Skamarock et al., 2008) version 3.7.1. The model was run at a 5 km resolution over the UK with boundary and initial conditions initialised by the National Centers for Environmental Prediction Final Global Forecast System (NCEP-GFS-FNL) data (https://rda.ucar.edu/datasets/ds083.2/)."

Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X. Y., Wang, W. and Powers. J. G. (2008) A description of the advanced research WRF version 3. NCAR technical note NCAR/TN-475+STR, 10.5065/D68S4MVH.

(2) *More detail of the inorganic chemistry scheme in FRAME and information on the type of inorganic aerosol module used, with references for both of these.*

Response: The following expanded text and additional citation is now included in Section 2.1.
"The chemical scheme is described in Fournier et al. (2004) and includes gaseous and aqueous-phase oxidation reactions and conversion of the gases NH_3, SO_2, and NO_x to particulate matter (NH_4+, NO_3^-, SO_4^{2-}). NH_4NO_3 is formed by the equilibrium reaction between HNO_3 and NH_3 and nitrate aerosol also arises by the deposition of HNO_3 onto sea salt or large particles. H_2SO_4 reacts with NH_3 to form $(NH_4)_2SO_4$. The aqueous phase reactions include the oxidation of S(IV) by O_3 and the metal catalysed reaction with O_2."

Fournier, N., Dore, A. J., Vieno, M., Weston, K. J., Dragosits, U. and Sutton, M. A. (2004) Modelling the deposition of atmospheric oxidised nitrogen and sulphur to the United Kingdom using a multi-layer long-range transport model. Atmos. Env. 38, 683-694.

(3) The approach taken to the representation of the emission uncertainty (varying the emissions in all grid boxes by the same fraction in each run) is justified in the context of this study. However, it does mean that several important aspects of emissions uncertainty are not included. In particular any uncertainties in the spatial distribution or height of emission are not captured. There are also important sources of emission related uncertainty that FRAME cannot capture such as uncertainty in diurnal or seasonal cycles of input. These limitations should be noted here.

Response: Thank you for these additional limitations we should highlight. The following text has been added to Section 2.2 where we describe the uncertainties in total annual emissions. "It is also acknowledged that a number of other aspects of emissions uncertainty are not included. For example, the FRAME model cannot capture uncertainty in assigned seasonal and diurnal cycles in emissions. Uncertainties in the spatial distributions or in height of elevated emissions are also not included."

An additional reminder of other emissions uncertainties has also been added at the start of Section 3.2 when presenting the results of the uncertainty propagation.

(4) Finally, it would be interesting to see the impact of these uncertainties on the secondary inorganic aerosol mass. This may be beyond the scope of this study, but concentrations of PM2.5 are highly relevant for air quality forecasting and policy relevant research. If the results are available, it would be a valuable addition to this study.

Response: We agree this is an important policy-relevant question. We used the FRAME model in this work as a 'proof of concept' for this global sensitivity approach. We are currently applying our methods to the more sophisticated EMEP4UK atmospheric chemistry transport model (www.emep4uk.ceh.ac.uk), which incorporates simulation of all PM components, including a more advanced formulation of the formation of secondary inorganic and organic aerosols, and VOC-NOx-ozone chemistry, and will be reporting on the findings from this model in other papers being prepared.