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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Model bias in simulating major chemical components of $PM_{2.5}$ in China" *by* Ruqian Miao et al.

Anonymous Referee #2

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Model bias in simulating major chemical components of PM2.5 in China

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The paper presents a comprehensive investigation into the uncertainties in PM2.5 simulated with the GEOS-Chem model for China and potential sources of errors. PM is a complex pollutant and even after the decades of its modelling, most of air quality/chemical transport models are still often struggling with accurate representation of PM, in particular during pollution episodes. Given large uncertainties in descriptions of aerosol chemical and physical processes, the availability of good quality observations is crucial for models' evaluation and constraining. In the presented work, the authors

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compiled and used for the model evaluation an impressive volume of observational data of non-refractive submicron PM components (sulphate, nitrate, ammonium and organic aerosols), as well as aerosol gaseous precursors in China. They also performed a series of sensitivity tests, modifying multiple parameters, to obtain the best model correspondence with the observations.

The paper is in general fairly well written (though the bounty of technicalities sometimes makes reading somewhat heavy); the figures and tables are quite helpful in visualizing and presenting the results. The topic of the paper is highly relevant, the scientific material and findings are quite interesting, and thus after some minor revisions it can be recommended for publication in ACP.

My first reaction is that given the impressive amount of testing, the conclusions appear somewhat little constructive and of a rather general character. In other words, it is unclear what would be the first priorities the authors plan to improve the GEOS_Chem's performance with respect to PM2.5. Would the authors comment on that?

The 'best combination' of all tested parameters did not yield a satisfactory model agreement with short term observations in Beijing. Has it been tested against the whole 2006-2016 campaign dataset?

In particular, the GEOS-chem is shown to have troubles to reproduce observed concentrations of SO4 and NO3. Is that for China simulations only? Or was that seen also so for other world's regions? I'd suggest to include in Introduction a small paragraph about that if such evaluations are available.

Have the authors seen a paper by H. Bian et al.: Investigation of global particulate nitrate from the AeroCom phase III experiment (Atmos. Chem. Phys., 17, 12911–12940, 2017 https://doi.org/10.5194/acp-17-12911-2017)? It does not appear that overestimating NO3 in Asia is a generic feature among the nine CTM and climate models participating in the paper.

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There are other processes, not been investigated in the paper, which could be sources of e.g. NO3 overestimation, for instance the equilibrium formation of ammonium nitrate. How well does ISORROPIA work for China chemical regime? Has this been studied before? Any reference to the results?

Another source of uncertainties is dry deposition velocities of NO3 and NH4, which were measured to be higher than typically predicted by the models (E. Nemitz et al.: Concentrations and surface exchange fluxes of particles over heathland; Atmos. Chem. Phys., 4, 1007–1024, 2004 www.atmos-chem-phys.org/acp/4/1007/). Would the authors consider to investigate into this process?

A minor general comment: winter haze events are mentioned every now and then, without any clear context. Could the authors explain early in the paper why haze occurrence is an issue in the manuscript?

Other comments and suggestions to the text:

Line 27. suggesting existing inaccuracies in the processes description (or presentation).

Lines 30-31 and 172-174. It's unclear what heterogeneous SO2 oxidation reactions are in the model (by H2O2, ozone?)

Lines 34-35. Again, can the author show that ISORROPIA is working properly? Clarify 'related to removal'. Wet or dry, or both.

Line 42. what is considered to be reasonable?

Line 45. 'the model performance on PM2.5 is component-dependent' sounds strange. Maybe like: even though the model represents well observed PM2.5, it may happen due to compensation errors in model simulated PM2.5 components.

Line 48. I agree to some extend about SOA, but not about sulphate and nitrate (see my Ref. to Bian above). Perhaps the authors mean only specific studies for China.

Line 57. Suggestion: The uncertainties in the emissions of primary PM and gaseous precursors of secondary PM are quite large. . ..

Line 82. Suggested: therefore it is important to evaluate the model for all individual components of PM2.5

Line 83. The measurements' artefacts can also decrease the discrepancies. . . the point is that in such cases, the model evaluation results give a wrong message.

Line 103. Write: Institute of Atmospheric Physics (IAP) – for future use of abbreviation. Could you write here what site type is this (urban/suburban background?)

Line 130. Suggestion: For comparison with observations at IAP. Beijing, the model simulations were performed for the ASCM measurements period. . ...

Line 132: Do I understand right that model simulations for 2012 meteorological conditions were used for comparison with 2006-2016 observations. Could the authors then say how (un)typical the 2012 weather was. Were year dependent emissions used, or also the same for 2012?

Line 150. Suggested: 80% which is considered to be a reasonable assumption (instead of 'for simplicity')

Line 151. However, Hodzic. . . showed that the results were not very sensitive to. . ...

Line 161-162. What is the relative contribution of non-agricultural NH3 emissions compared to the agricultural ones? This is important to know when analysing ammonium nitrate formation in cities.

Lines 172-180. Move up to Chemistry description, above the emissions.

Line 182. Suggestion: Model performance for the individual PM components

Line 188: The modelled ammonium concentrations compare with observations better than simulated sulfate. . ..

Line 191. Further, we find that the model biases. . .

Lines 192 and 208-210 some repetition.

Line 196. Would you expect the model performance to differ over China? Why? How different those regions are (weather, emissions?). Does that mean that the model is insensitive to the differences in meteo and chemical regimes?

Line 208. Suggestion: On a seasonal basis,..

Line 210-11. ..the overestimation of nitrate concentration is largest in spring, summer and autumn.., while the model bias is much smaller in winter. . .

Line 212-213. Are those simulations also with GEOS_Chem and ISORROPIA?

Line 214. In all seasons?

Line 217. The model performs worst in autumn

Line 218. This is about the only time when the correlation is mentioned. Why it's considered important here, but not for the other components? Is the relative importance of ASOA greater in summer?

Line 220. –compared to the 2 years of hourly observations. . ..

Line 223. ..underestimation of sulfate and OA by the overestimation of nitrate. . .

Line 226-27. Explain exclusion of the observations over 150 ug-m3

Line 242. Does that mean: If the evaporation of ammonium nitrate . . .. . .was accounted for in the model, the day time variation.. could be flatter?

Line 254. Semivolatile POA scheme previously used? in GEOS-Chem

Line 267. . . .simulations lead

Line 271. Suggestion: The uncertainties related with emission data 8including their temporal profiles) are considered to be one of the major sources of inaccuracies in

modelled concentrations. . ...

Line 275. It is widely shown that regional models cannot accurately reproduce NO2 at urban sites. Would the authors really expect the model with a resolution of 50-60 km to be capable of managing that?

Lines 282-285. What is the main sources and relative importance of non-agricultural NH3? Why is it especially important during haze events?

Lines 288-290: Unclear what is said here.

Lines 311-12. Should be formulated more clear: The photolysis rate of particle-phase HNO3 was shown in aged air masses to be higher than for the gaseous HNO3. . ..., but in Beijing particulate NO3 may have lower photolysis rates, because. . ...

Line 315. From the factors

Line 319-20. . . .weeks were free of severe haze episodes (with extreme conditions which the model fails to reproduce????)

Line 338. The increased wet deposition if nitrate. . ..

Line 360. Faster photolysis of particulate nitrate? Sounds contradictory to what is written on lines 311-312

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