

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-330

Anonymous Referee #2

Referee comment on "Nonlinear responses of particulate nitrate to NO_x emission controls in the megalopolises of China" by Mengmeng Li et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-330-RC2>, 2021

This paper investigates the secondary inorganic aerosol formation in cities in China focusing on nitrate aerosol, using observed data from a Chinese network of station and a WRF-Chem mesoscale model to analyze the response of PM_{2.5} to NO_x emission changes on a seasonal basis. The main findings are the almost linear response of secondary inorganic nitrogen to NO_x emissions changes during summer while in winter the system is highly non-linear. Sensitivities to ammonia and sulfur dioxide emissions, which have been shown to be important, are discussed only based on existing literature. The paper is within the scope of Atmos. Chem. Physics and could be suitable for publication in the journal after a number of improvements suggested below.

My major comment is that the authors miss to show a clear view of the interplay between O₃, SO₂, NO_x, NH₃ and PM_{2.5}. The authors provide some information in this respect in the conclusion but the manuscript can be largely improved in this respect. Therefore, I would suggest that the authors check on relevant references and integrate some of them in the introduction or discussion session. Some are listed below but more might be found by a literature search.

For instance, they could take notice of the following relevant publications for NO₂, NH₃, SO₂ interactions:

Yu, G. et al. Nat. Geosci. <https://doi.org/10.1038/s41561-019-0352-4> (2019) and relevant News and Views comment: <https://doi.org/10.1038/s41561-019-0358-y>

Li, K. et al. PNAS, 2019, 116(2), 422.

For O₃ in China the paper by Liu and Wang, ACP 2020
<https://doi.org/10.5194/acp-20-6305-2020>

The paper by Nenes et al ACP in ACP on aerosol's sensitivity

Nenes et al., ACP, 2021 <https://doi.org/10.5194/acp-21-6023-2021> on the impact of pH on the partitioning and therefore the lifetime of nitrate and ammonia aerosols

Nenes et al., ACP, 2020; <https://doi.org/10.5194/acp-20-3249-2020>, 2020 on the sensitivity of PM mass to ammonia and nitrate availability

The papers on HNO₃ formation from other pathways than the NO₂+OH reaction (there might be more)

Brown, S. S., et al. 2004, Geophys. Res. Lett., 31, L07108, doi:10.1029/2004GL019412.

Vrekoussis et al 2005, Geophys. Res. Lett., 33, L05811, doi:10.1029/2005GL025069

Karydis et al., ACP, 2015, www.atmos-chem-phys.net/16/1491/2016/

Specific comments:

Line 189: capture

Line 194: MB

Line 211-212: that the consideration of the optimized... reduces...

Lines 228: was more than ...

Line 232: (because OM has more than C, O and H) I suggest adding 'mainly' for the hydrogen and oxygen

Line 235: could you provide standard deviation for percent fractions ?

Lines 236: SIA are the dominant components of PM_{2.5} – 48.6% is not the majority – it is below 50%

Lines 237-242: please explain the divers, are temperature, liquid water content of the aerosol impacting these ratios

Lines 258-259: discussion could profit from additional literature – also heterogeneous reactions on dust can produce nitrate aerosol (see references above)

Line 267: nonlinear behaviour in winter

Section 3.3.1 & 3.3.3, I miss here the discussion on the involvement of SO₂ emission changes to PM_{2.5} aerosols and thereby to O₃ and nitrate aerosol

Line 310: have minor changes

Lines 312, 313,345-348: How significant are these correlations?

Line 320: do not forget acid replacement reactions (e.g. reactions on dust or sea-salt aerosol)

Lines 324-325: do not forget here the reduction of NO + O₃ titration reaction

Line 337: reactions

Line 391 move 'during summer-autumn' to the end of line 392

Lines 408-415 and 419-427 would fit better in introduction of discussion. Please move appropriately.