

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2021-529

Anonymous Referee #1

Referee comment on " Atmospheric measurements at Mt. Tai – Part I: HONO formation and its role in the oxidizing capacity of the upper boundary layer" by Chaoyang Xue et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-529-RC1>, 2021

The manuscript presents results of a comprehensive field campaign at two different altitudes, the foot (150 m a.s.l.) and the summit (1534 m a.s.l.) of Mt. Tai (Shandong province, China). Mt. Tai locates in the middle of the NCP with a relatively high pollution level. The measured HONO diurnal profile shows a daytime peak at 12:30 local time, which is interesting since HONO diurnal profiles would typically peak during the night and early morning in more polluted regions. The topic is of interest to the scientific community and is suitable for publication in ACP after addressing the comments below.

The authors claim that OH+NO gas-phase reaction accounts for only 8% of measured HONO, and that 70-98% of the unknown HONO sources can be attributed to vertical transport from ground surfaces. However, the authors didn't show/present the OH values used to calculate the OH+NO reaction rate, and they didn't consider this reaction when calculating the net production of OH from HONO. The authors used an unjustified circular assumption that OH loss in the OH+NO reaction at the ground will be recycled back to OH at a higher altitude without any valid calculation of HONO lifetime vs transport time from the ground to the summit. The authors claim that they calculated HOx budget, although they only calculated gross HONO photolysis and O₃ photolysis. (primary sources of OH only). I suggest the authors limit their discussions to HONO sources and sinks, and that they should account for NO+OH reaction in calculating HONO_{pss} or assume several OH values around those published earlier to calculate their uncertainties. Otherwise, the manuscript is publishable after addressing these comments.

Specific comments:

Page 15, Line 320: The authors didn't justify the use of OH-j(O¹D) correlation from previous publications to calculate OH in this study. Although some studies showed a good correlation, it still may not be a good proxy for OH given the large variation in the obtained slopes. The authors use a circular argument that OH is not important since NO+OH is not important, to justify the uncertainty associated with their approach. At

which OH levels does the NO+OH reaction accounts for 8%?

Maybe, it is safer to either simulate OH using a box model or use a range of OH levels around those reported previously by Kanaya et al. (2009) to show that it is not important, as they claim This is a major issue that the authors need to address before continuing with their calculations of unknown HONO sources.

The authors used several assumptions to calculate the contribution of different HONO sources to measured HONO levels. Most importantly is the photolysis of pNO₃, for which the authors used a range of enhancement factors (EF) that ranges from 1 to ~15.6, accounting for 0.6 to 9.6%, depending on EF, leaving ~93% of HONO unknown sources unknown. I think. A major uncertainty here is related to HONO_{pss}, which the authors didn't sufficiently address, which affects the unknown fraction HONO.

Page 18, lines 404-412: The authors' argument of OH recycling via HONO photolysis as the source of OH at higher altitude is not justified and is flawed. The authors didn't provide information about the HONO lifetime vs the transport time to this altitude. I think this whole paragraph should be just deleted.

Page 18, line 414: provide a reference...

Page 19, lines 418-420: This long sentence is not clear at all....either provide all relevant information or leave it for the accompanying paper. Otherwise, HONO net photolysis should be used to calculate HONO relative contribution to OH primary sources.

Page 19, lines 428-438: Again, this is all irrelevant if HONO net contribution is not calculated.

Page 19, lines 425-445: replace HO_x with OH since you HONO and O₃ photolysis are sources of OH only, not HO₂.