

Interactive comment on “Rapid SO₂ emission reductions significantly increase tropospheric ammonia concentrations over the North China Plain” by Mingxu Liu et al.

Anonymous Referee #2

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This manuscript addresses an important topic that is ultimately related to the air quality issues in China. The methodology is sound, as similarly done for the US regions by Yu et al. (2018). I believe it should be published after addressing the following major and minor issues.

My major issue is how authors “claim” their results. Their sensitivity studies of (quote) “the SO₂ emission reduction of 50% from 2012 to 2016 could results in a 55% increase in the NH₃ columns, compared to that of 30% recorded by IASI observations.” . . . “the increasing trend of NH₃ can be entirely attributable to the SO₂ emission reductions.” (page 8, line 6-12). I do not believe such a conclusion can be drawn, unless the

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authors have performed and show quantitatively that all other mechanisms (NOx, NH3 emissions, temperatures, precipitations, etc.) do not contribute to the NH3 increase (see more below). The estimated increase of 55% being larger than the observations of 30% only indicates uncertainties.

The last paragraph before Conclusion (page 9, line 14-22) is ambiguous and hand-waving. These “other” mechanisms that are very likely to have also caused the gaseous NH3 to increase, but were dismissed without sufficient quantitative data or figures to back it up. (quote) “...particulate nitrate... concentrations appear to increase in the North China Plain between 2008 and 2016 despite a 23% reduction in NOx emission (Fig. S4). The in situ measurements in Beijing indicated that the NO₃— concentrations fluctuated during 2013–2016. It implied that the NOx emission reduction could not be responsible for the increase in NH3.” Should not “imply” a mechanism that “could not be” responsible... The same process for the SO₂ should be repeated for the NOx, if any conclusions were to be drawn about how NOx reduction affects the gaseous NH3 concentration change. The in situ measurements in Beijing was used to make an argument, but no evidence was shown in the manuscript, additionally, the where about of the data is not included, which does not follow the ACP data policy.

Similarly, for meteorological effects, quote “We also tested the effects of meteorological conditions on NH3 variations by a simulation with meteorological fields in 2016 and anthropogenic emissions in 2012 (Run_16_E12). Compared to the Run_12 case, we found the change in meteorological fields (2012 vs. 2016) had a negligible influence on NH3 concentrations in most of North China Plain.” None of these were shown quantitatively! Can’t make statements like these without any evidence. The following statement “Although temperature increase was reported to partly contribute to the positive trend of NH3 (Warner et 20 al., 2017; Fu et al., 2017), our simulations indicated that the overall meteorological factors could not explain the observed significant increase tropospheric NH3 concentrations over North China Plain.” This sentence is misleading, as if the quoted studies were trying to explain the observed significant increase in tro-

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pospheric NH₃ concentrations by meteorological factors. In fact, Warner et al. (2017) emphasized the leading cause of the NH₃ increase was the reduction of SO₂ in China, I quote "Over China, a combination of expanded agricultural activities, nascent SO₂ control measures, and increasing temperatures cause the observed increases in ammonia."

My minor issues are mainly related to language and choice of words. I believe this manuscript needs to go through English editor at ACP. Also, many word choices are not appropriate for concise scientific publications, and somewhat wishy-washy, e.g., "appear to", "could not be", "may be a potential", "could be responsible", "would bias", "... concentrations disappeared", "... is practically zero...", "could result", "were almost consistent", "could make", "implied", "for almost the entire...", "not well-regulated", "can increase...", "may alter"...

Page 2 line 11: "As a major agricultural country, China is the world's largest emitter of NH₃..." what about India?

Page 2 line 15: "...may be potentially important contributor to haze..." It's a known fact!

Page 2 line 17-19: "Interestingly, satellite observations over the past decade have shown an increase in tropospheric columns of gaseous NH₃ in this area (Warner et al., 2017). But no quantitative studies have been performed to explain it." Warner et al. (2017) was a quantitative study using observations. Should be "But no sensitivity studies..."

Page 2 line 19-20: "Along-term bottom-up inventory indicated that NH₃ emissions in China have displayed a slightly decreasing tendency." Needs references!

Page 3 line 10: "Here, we hypothesize that the rapid SO₂ emission reduction is the reason for the increase in tropospheric NH₃..." Several studies have published the fact that the SO₂ emission reduction is the reason..., not a hypothesis anymore. Should

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reference others' publications here, for global studies or in other regions, than in the North China.

Page 4 line 9: Please pay attention to the order when acronyms are introduced and used throughout the paper.

Page 4 line 15: MEIC should be defined on Page 3 line 7.

Page 4 line 15: "were cut" use reduced.

Page 4 line 19: remove "by our research group"

Page 4 line 21: "in our previous studies..." should be "studies by..."

Page 5 line 5-7: "Meanwhile..." needs references.

Page 5 line 11: use IASI.

Page 6 line 22: "which could be responsible", add partially responsible...

Page 6 line 23-24: bad sentence, rewrite.

Page 7 line 3: "Moreover, we also...", remove also.

Page 7 line 18-19: "These tests support..." Too absolute! No other mechanisms?

Fig. 2: use whole words for Sim., Obs., Sep., and Aug.

Yu et al. (2018) "Long-Term Trend of Gaseous Ammonia Over the United States: Modeling and Comparison With Observations" DOI: 10.1029/2018JD028412

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