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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Northwestward Cropland Expansion and Growing Urea-Based Fertilizer Use Enhanced NH_3 Emission Loss in the Contiguous United States" *by* Peiyu Cao et al.

Anonymous Referee #2

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The manuscript by Cao et al. estimates NH3 emissions from fertilizer in the US over the past century. By tracking different types of fertilizer and crops, they identify variability in the spatial distribution of fertilizer emissions and emissions factors. Their results are consistent with previously noted studies of shifting spatial distribution in NH4 deposition, for example, but provide additional valuable levels of detail. My main suggestion would be to provide some quantitative assessments of uncertainty, which I think may constitute minor revisions, as they have at least qualitatively identified the key sources of uncertainty. This and a few other minor comments are included below.

Comments: 57-58: It's not clear to me what land and fertilizer use is being referred to

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Discussion paper

here as distinct from the studies cited in the preceding lines.

107-119: I realize this lies somewhat outside the present paper and is likely within the work of Yu 2018, but could the authors briefly comment on how such spatial resolution was known for these distributions prior to the satellite era? Here they mention how satellites were used to determine spatial reconstructions but do not comment on any other method, which presumably would be necessary for the first half of the century, nor how such different methods have been harmonized into a single consistent dataset.

173: Here emission growth just refers to fertilizer emission, right? Not emission from livestock, which is the larger component of total emissions.

Fig 1 (a): I think it would be more clear to refer to this as loss "from" N fertilizer, not loss "to" fertilizer, since the process being described here is NH3 from fertilizer to the atmosphere, correct?

Fig 2: What drives the drop in RF from the 40s to 50s in NW, NGP and SW?

197: What do the authors mean by "reportedly" here?

Fig 4: Are the stark transition at state boundaries (e.g., west Virginia in 2005, or South Dakota in many years) something that we should interpret as physically meaningful, or is this an artifact in how some of the underlying data used for these estimates is collected at the state level? If the latter, how could this information be interpolate spatially to be more useful for geophysical modeling (such as for input to an atmospheric model)?

Fig 5: to clarify, this is NH3 emission from fertilizer, not total NH3 emission. I think it is important to re-iterate this where possible, to prevent casual readers from taking these figures out of context.

320-340: This is a very valuable list and set of discussion. Still, I wish the consideration of uncertainty could be more quantitative, even if in an approximate manner. What are the contribution of each of these factors to the total uncertainty? Do the authors believe

their total estimates are accurate to within 1%? 10%? 100% or factor of two? Please elaborate.

315 - 319: Interesting, yet a bit speculative. In the NW, biomass burning may have driven some of these trends. Without more careful study, it may be prudent to remove these speculations.

Corrections:

39: Liu 2019 was not an inverse modeling study. They estimated NH3 concentrations but not emissions. The first satellite-based top-down estimate of NH3 emissions was Zhu et al. (JGR, 118, 3355-3368, 2013).

39:))

40: containing

73: Is “spatialized” a word? Perhaps “spatially distributed”

158: I think it’s worth noting that this “data” is an estimate, which will be quite uncertain at great distances between NADP sites, particularly in the western US.

170: Northwest, whereas. . .

173, 187,. . .check throughout: NH3 subscript

220: Our

227: two

317: is heavily involved in the format of PM2.5

323: application

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