Review of "High secondary formation of nitrogen-containing organics (NOCs) and its possible link to oxidized organics and ammonium" by G. Zhang et al.

General Comments:

This study presents the results of single-particle measurements in Guangzhou, China. The focus is on nitrogen-containing organic compounds (NOC) and the role of ammonium in NOC formation. The highly time resolved measurements span four seasons, and thus could offer new insight into NOC chemistry. While the data set is unique and the topic is certainly appropriate for *Atmospheric Chemistry and Physics*, there are quite a few major issues with the study – including analysis methods, assumptions, data interpretations, and conclusions – that prevent me from recommending it for publication at this time. It is possible that these issues could be addressed with a major revision, but not guaranteed. My specific concerns are addressed below.

Specific Comments:

1. The authors come to the conclusion that NOC formation during their study is not likely from gas-phase reactions, but is predominantly from heterogeneous/particle-phase reactions. Their logic for this argument is quite confusing. I do not believe this conclusion is at all supported by the data presented in the manuscript.

2. Similarly, I think the explanations for the role of NO_x and NH_3 in particle NOC formation are extremely muddled. For NH_3, a positive correlation is observed between the number fraction of particles with NH_4^+ and NOC, while a negative correlation is observed between the relative peak areas of these compound classes. There is not a reasonable explanation given for this surprising and apparently contradictory behavior. Further, the manuscript mostly discounts the negative correlation in the relative peak areas, instead assuming that NH_3 drives (or is prominently involved in) NOC formation. NO_x is completely ruled out as a contributor to NOC formation on the basis of poor (or no) correlations between NOC and NO_x. However, this is a misinterpretation of the data. Many factors (different removal processes and lifetimes of particles vs. gasses, primary vs. secondary species, etc.) could contribute to a lack of correlation even if NO_x did contribute to NOC formation. As the data are currently presented and explained, it is completely unclear how NH_3 or NO_x play a role in NOC formation in the present study, although such analyses should be possible with their data set.

3. The assumptions and discussion related to particle acidity (lines 307 – 337, Figure 7) are not correct. Recent studies have shown that ratios of aerosol inorganics (including variations involving NH4-SO4-NO3) are not suitable proxies for particle acidity (Guo et al., 2015; Hennigan et al., 2015; Murphy et al., 2017). Also, the discussion of acid-catalyzed SOA (lines 375-381) is not correct, so the implications of the present study are misstated.

4. Finally, the application, interpretation, and discussion of PMF and multiple linear regression methods need substantial revision. The explanation of the PMF approach is quite confusing, and as it is written, does not add anything substantive beyond the general correlations presented before it. The multiple linear regression also does not support any of the stated conclusions, beyond what was already presented for the individual correlations to NH_4^+ and

oxygenated organics. The discussion of “modeleld NOCs” (e.g., lines 291, 374, Fig. 4) is misleading, especially compared to how this is typically used in atmospheric studies.

Technical Corrections:
The above issues are substantial enough that any technical corrections can be addressed on review of the revised manuscript.

References:

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Hennigan, C. J., Izumi, J., Sullivan, A. P., Weber, R. J., and Nenes, A.: A critical evaluation of proxy methods used to estimate the acidity of atmospheric particles, Atmos. Chem. Phys., 15, 2775–2790, https://doi.org/10.5194/acp-15-2775-2015, 2015.

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