# *Interactive comment on* "Role of ambient ammonia in particulate ammonium formation at a rural site in the North China Plain" *by* Zhaoyang Meng et al.

**Anonymous Referee #2**

Received and published: 31 May 2017

This manuscript presents a comprehensive ambient measurement dataset, including various trace gases and particulate species, for over four months at a rural site in the North China Plain (NCP). Ammonia (NH3) is the focus of this study for its role in the formation of secondary inorganic aerosols, which accounts for a major fraction of PM2.5 in NCP. The hourly resolution, higher than many of the previous ambient ammonia measurements, enables detailed studies on individual pollution events and the diurnal variations. However, I hope the authors can take better advantage of this dataset, and go deeper into Atmospheric Chemistry and Atmospheric Physics, as indicated by the journal name. For example, this work aims to understand the impact of ammonia on secondary ammonium aerosols (page 1, line 20), facilitate developing future ammonia emission control policies (page 1, line 32), and examine the sources of ammonia and ammonium and their chemical conversion mechanism (page 3, line 8). These are all

important issues, but I am not convinced that this article has advanced our current knowledge and understanding about these issues after reading it.

Major comments

1. It takes a significant part of this manuscript to explain the observed concentrations. However many of the explanations are qualitative and even speculative. Further quantitative evidences are needed. To name a few:

Page 8, lines 5-6, "the monthly concentration of SO2, NOx, and CO in July and August decreased because of raid photochemical reduction, additional removal by rainfall, and excellent vertical mixing." What are the evidences of photochemistry, wet scavenging, and vertical mixing?

Page 8, lines 8-9, the ozone was highest in June because of "photochemical production, intense burning of biomass, and transport of regional pollution". What are the evidences of more photochemistry, biomass burning, and regional transport in June? Shouldn't July have larger photochemical rates?

Page 8, lines 26-27, the downward mixing of the residual layer containing higher ammonia concentration could lead to an increase of ammonia in the morning. This would require a very large pool of ammonia in the residual layer. Why did it not happen in other months?

Page 9, line 5, the author explains the earlier ammonia morning peak in July by increased emissions. Further evidence?

2. A high observed concentration can always be explained by more emission, less mixing, or less removal. I think a publication in ACP should go beyond reporting the concentrations of these short-lived species, as the concentrations are highly variable. This study used the ratio between ammonium and NHx to infer the gas-particle conversion of ammonia. However the ammonium and ammonia may be from different sources, where ammonium is formed in the city with NOx and SO2, and ammonia is

emitted locally. In other words, what if NHx and NH3 are decoupled?

At page 10, line 13, it is summarized that "This observation emphasizes the important role of NH3 in the formation of secondary SO4, NO3 and NH4 aerosols, which should be further explored ...". The title of this manuscript is about the role of ammonia on secondary inorganic aerosols, but what exactly is this role? It is not satisfying to only know it is important and needs further exploration.

Specific comments

Page 1, line 29: please define "transport of air mass from the North China Plain region", as the site itself is in the middle of NCP.

Page 3, line 22: how large is the "surrounding area" that impacts the measurements of this site?

Section 2.2: what's the response time of the Los Gatos instrument? What is the concentration and accuracy of the calibration gas?

Page 6, line 3: please define "human activity" as it seems a very broad concept.

Page 6, line 5: there is no "Zhng et al., 2010" in the reference list.

Page 6, line 8-9: please clarify how can these results "be used in improving NH3 emission inventory and making future emission control policies".

Figure 2: I understand ammonia is shown in log scale because its concentration spanned three orders of magnitude. However I suggest add a sub plot with linear scale so that the variability is comparable at different concentration levels and the individual spikes from pollution episodes are clearer.

Page 7, line 6: where the urea was applied and how large was the applied area?

Page 7, line 29: what are these "trace gases"?

Page 8, lines 14-15 and page 8, lines 30: these two sentences seem contradict each

other.

Page 10, line 1: higher NO3 level than what?

Page 10, line 20-25: I suggest add a figure showing the slope and correlations. The SO4 should be normalized with its number of charge. What is the evidence for the existence of NH4HSO4?

Page 11, line 17: again, it is better to have more evidences showing that NH3 dry deposition dominates NHx deposition.

Page 12, line 2: where is the reference Meng et al. 2017?