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Reply to Comment on "Fossil Fuel Combustion-Related Emissions Dominate Atmospheric Ammonia Sources during Severe Haze Episodes: Evidence from ^{15}N-Stable Isotope in Size-Resolved Aerosol Ammonium"

We appreciate the opportunity to respond to the comments of Chang and Ma[1] regarding our article,[2] and we also hope to further clarify the findings of our work. Their comments on our work focus on the source apportionment of ammonia (NH_3) during haze episodes in Beijing. We do not think that their objections are well founded, and their speculations do not change our conclusions.

1. POWER PLANT NH_3 SLIP

Chang and Ma raised a concern about the major contribution of power-plant NH_3 slip in Beijing, but they have not provided scientifically sound evidence to support their argument. To support their speculations, they cited the changes in energy utilization reformation of power plants since September of 2013. However, our study focused on the severe haze events in early 2013. Thus, it is reasonable that the post-2013 scenarios differ from those in our study. Even so, the point argued by Chang and Ma that "Power plant ammonia slip is not a major ammonia source in Beijing" is very arbitrary. Even in the extreme case of the power plants in Beijing all being shut down, the NH_3 slip from power plants in Hebei and Tianjin (hundreds kilometers away from our sampling site) can be transported to Beijing within 3 days or less. During haze periods, in our study, the atmospheric transport velocity was 50−100 km d^{-1}, as estimated both from the measured surface WS (Figure 1) and by back-trajectory analysis (SI Figure S4).[2]

In fact, 16 coal-fired power utilities in Beijing still are controlled by a selective catalytic reduction (SCR) system and nine other utilities combine SCR and selective noncatalytic NO_x reduction (SNCR) technologies. The NH_3 emissions from these sources in Beijing are expected to be enormous due to the electricity demand and the fact that extra coal is consumed for residential heating in winter. More details can be found in the fourth paragraph in the Discussion section of our paper.[2]

2. POOLING THE "VOLATILIZED NH_3" SOURCES AS ONE ENDMEMBER

For the purposes of simplification, it is common to integrate several sources with overlapping ^{15}N signatures. Therefore, to combine agricultural sources from fertilizers and livestock as a whole is reasonable.[3] In our study, we used a mean value of −39.5‰ to represent a moderate agricultural emission signature that reflected the contribution of agricultural activities to NH_3 in Beijing. It will increase the uncertainty if we were to partition four sources using the "IsoSources" isotopic mixing model when we have only isotope species. That is why we combined the sources from fertilization and waste into one agricultural source.

Although Chang and Ma argued that pooling the agricultural sources (from waste and fertilizers) will inevitably underestimate the contribution of agricultural activities to ambient NH_3 in Beijing, they have not provided supporting evidence for this claim. They provided data showing NH_3 emissions from fertilizer application and livestock waste and stated that these sources have distinct δ^{15}N values. However, these sources are essentially the same "volatilized NH_3" source and if the literature data is taken into consideration, these sources' signatures are observed to overlap (for details, see http://www.atmos-chem-phys-discuss.net/acp-2016-432/#discussion). It is thus a more realistic approach to combine the "volatilized NH_3" sources as one endmember, as we did in our study.

3. UNCONSIDERED SOURCES AND FUTURE DIRECTIONS

Chang and Ma suggested that urban waste-related sources should be taken into account in our source partition. They refer to a Shanghai report of human excreta being stored in septic tanks as a stable and important source of atmospheric NH_3, which contributes over 11% of the total NH_3 emissions in the Shanghai urban area. However, that study area is in a subtropical region and the study period was in summer. We believe that the NH_3 emission from human waste in Beijing was much smaller due to the cold temperature during the study period (around 0 °C, January 24 to February 1, 2013), which is not favorable for NH_3 volatilization.

Due to the paucity of signature data in China, we employed a comprehensive inventory of δ^{15}N−NH_3 values from major emission sources observed in the U.S.,[3] and this point should be considered in the interpretation of our results. However, these isotopic signatures of fossil-fuel-related emissions in China and the U.S. are unlikely to significantly differ as the two countries use similar catalytic technology (for more details, see section 2.4 of our paper[2]). To further reduce the uncertainty in the source apportionment of NH_3 based on δ^{15}N-NH_3 values, we suggest additional field and laboratory experiments to adequately characterize the endmember signatures and a subsequent fractionation process under different air pollution and meteorological conditions.

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Notes
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