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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"

Pan et al.[1] presented the isotopic measurements of size-resolved aerosols in Beijing, summarizing that fossil fuel-related ammonia emissions (including traffic, coal combustion and power plants ammonia slip) have overtaken agricultural activities as the dominant source of atmospheric ammonia during hazy days of 2013. This study is timely given that the important role of ammonia in terms of curbing China's $PM_{2.5}$ pollution. The study provides new and interesting insights into the topic of "missing sources of ammonia in urban atmospheres" and the results generally agree with our earlier studies.[2,3] However, we have to point out that the article has a few severe problems, which may lead to wrong conclusions and therefore could potentially mislead China's policy of ammonia reduction in the future. Different from the conclusions by Pan et al.,[1] we propose our viewpoints as follows:

1. POWER PLANT AMMONIA SLIP IS NOT A MAJOR AMMONIA SOURCE IN BEIJING

Pan et al.[1] claimed that during haze periods, 49% of ammonia in the ambient atmosphere of Beijing derived from power plant NH_3 slip, which can hardly be true. In September 2013, a five-year plan was introduced in Beijing to slash coal consumption, and there were only four coal-fired power plants (CFPP) operating near the city's urban areas during wintertime.[4] In 2016, all CFPP in Beijing will be shutted and replaced with gas-fired power plants to cut pollution. The replacement by the four gas-fired power plants will help cut emissions by 10 000 tonnes of sulfur dioxide and 19 000 tonnes of nitric oxide annually.[4] Although ammonia slip is a common issue with SCR (selective catalytic reduction) technology used in CFPP for removal of nitric oxide, the mass concentration of ammonia (typically 3−5 mg NH_3 m^{-3}) in flue gases is 2 or 3 orders of magnitude smaller than that of nitrogen oxides ($NO+NO_2$).[5] Moreover, we would like to remind that although there are many CFPP surrounded Beijing in the North China Plain, most of which are colocated with intensive agricultural production areas.

2. AGRICULTURAL ACTIVITIES CANNOT BE COMBINED AS A SINGLE SOURCE

There are three ammonia sources, i.e., agricultural source, fossil fuel combustion, and power plant ammonia slip, considered in Pan et al.,[1] and the average δ^{15}N values of these sources were used as isotope signatures (δ^{15}N-NH_3) to estimate their relative contributions to the ambient atmosphere in Beijing. We fully understand the author's consideration in terms of the classification of ammonia sources: agricultural ammonia are emitted at environmental temperature, the process of fossil fuel combustion can directly emit ammonia at high temperature, and ammonia slipped from power plant are the residues of gaseous reductants (typically anhydrous ammonia, aqueous ammonia or urea) that are subjected to medium temperature, and the isotopic signatures of these sources can be separated based on various temperatures.

It is well accepted that agricultural activities-fertilizer application merged with livestock production-are the largest contributor of ammonia emissions at regional or global scale. In fact, recent works[3,6] reveal that ammonia emitted from fertilizer application and livestock waste have distinct δ^{15}N values (Figure 1), which can also be reflected by the large variation

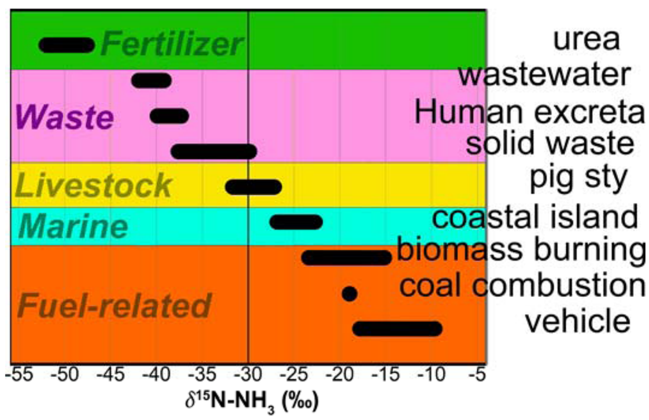

Figure 1. Variation range of δ^{15}N values for the five major NH_3 emission categories and their nine specific sources we developed in China. Except for coal combustion, biomass burning and marine sources, all other data have been reported in ref 3

range of δ^{15}N values for agricultural source in Pan et al.[1] (see Figure 3 in ref 1). This is because that fertilizer application and livestock waste generally represent two totally different nitrogen forms, i.e., inorganic and organic nitrogen, respectively. Situated on the northern edge of the North China Plain, one of the most intensive agricultural regions in China, Beijing is regarded as a receptor of agricultural ammonia from rural areas.[7] In Pan et al.,[1] fertilizer application and livestock waste are combined as a single source, which could inevitably underestimate the contribution of agricultural activities to the ambient NH_3 in Beijing.

3. URBAN WASTE-RELATED SOURCES SHOULD BE TAKEN INTO ACCOUNT

The isotopic signatures of wastewater, solid waste, and human excreta in China were quantified recently[3] (Figure 1). Urban wastewater treatment plants and solid waste are arguably two of the most important ammonia sources in urban China.[8] As a

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case study, we've identified that human excreta stored in septic tanks in Shanghai is a stable and important source of atmospheric ammonia, contributing to over 11% of the total NH_3 emissions in the Shanghai urban areas.[9] Therefore, it is of critical importance to incorporate the isotopic signatures of waste-related sources into isotope mixing model (e.g., IsoSource) in the future.

In addition, the starting point for disentangling agricultural and nonagricultural sources of ambient ammonia is to establish a pool of isotopic signatures that involves major emissions sources. The authors had no attempt to discuss the isotopic signatures used in their study, which are developed out of China and thus could introduce potential bias for the current conclusion in Pan et al.[1] Moreover, Pan et al.'s paper[1] missed two important references in terms of quantifying China's ammonia emission factors of coal combustion[10] and on-road traffic,[11] respectively.

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Notes

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