We thank two referees for their careful considerations of the manuscript and their well thought-out comments. These certainly helped to significantly improve the paper. We've addressed all comments and questions below in the form of point-by-point responses. The referees' comments are in *Italic* and our responses are in normal font. Text changes in the revised manuscript are highlighted in color (including a mistake we mentioned in our short comments).

Referee 2:

The authors have presented data which increases the inventory of isotopic signatures of ammonia emission sources, an area lacking in data. The authors have characterized isotopic signatures of NH_3 *emissions sources in China, a region of the world where this has not been done, and the data agrees closely with data obtained in other areas of the world. This data can be employed by researchers to quantify* NH_3 *emissions contributing to ambient atmosphere. This is valuable as many* NH_3 *emission sources are nonpoint sources making them difficult to quantify. Using the nitrogen isotope signatures the authors have estimated urban source contribution before and after a major event in Beijing in which air quality measures were employed during the event. The authors provide isotopic evidence that vehicles are a major contributor to urban* NH_3 *concentrations and the isotopic signature in the city changed to reflect the reduction in vehicle emissions resulting from air quality measures. The estimation of other source contributions is likely confounded by potentially overlapping source signatures and this should be more adequately addressed by the authors. This a novel approach to assessing* NH_3 *source contributions in an urban setting and the authors have presented the data in a clear and concise manner. If the major and minor issues below are addressed, I believe the manuscript could be accepted to ACP.*

Thanks for the recognition of our contribution. Please check our point-by-point responses to the major and minor issues raised.

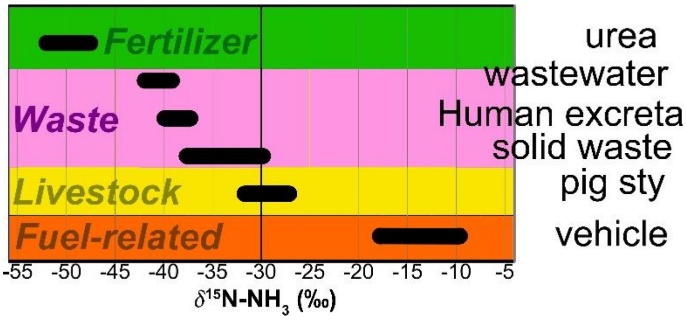
The authors use an isotope mixing model to predict NH_3 *source apportionment. The endmember signatures used in the model are vehicles, fertilizer, livestock waste, and human waste. The authors provide evidence that the vehicle endmember signature is significantly different from the other three endmembers but the other endmembers signatures are similar and with more sampling it is likely that these signatures will overlap. This is because the three 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. I don't think the authors have a viable case for assigning the source signatures they present to each source. I do however believe they could combine the "volatilized* NH_3*" sources as one endmember and vehicles as another endmember. While this would only provide insight to vehicle source apportionment, I think it is a more realistic approach. If the authors keep the mixing model as is, they need to explain the caveats associated with the estimates of the "volatilized* NH_3*" sources and include error analysis. Rather than reporting specific values a range should be reported that represents the deviation and error involved in the calculations.*

(1) We agree with the reviewer's comments that some of the isotopic signatures of volatilized sources could overlap and this problem should be addressed properly.

We think the source classification in our study is reasonable.

Our NH_3 source classification (Figure below) is based on:

a. The difference of isotopic signatures.
In our MS we concluded that "NH_3 emitted from volatilized sources has relatively low $\delta^{15}N$ values, allowing them to be distinctly differentiated from NH_3 emitted from traffic sources that are characterized by relatively high $\delta^{15}N$ value". Obviously, traffic-derived and fertilizer-volatized NH_3 has the highest and the lowest $\delta^{15}N$ values, respectively. However, "three sources are essentially the same volatilized NH_3" does not necessarily signify that all volatilized NH_3 has a similar variation range of $\delta^{15}N-NH_3$ values (data overlap), nor does it mean that these volatilized NH_3 sources should be classified as a single source category. Temperature is an important parameter, but not the only parameter in determining the differences of $\delta^{15}N-NH_3$ values among different NH_3 sources. In other words, even if data overlap may occur within volatilized sources, they can still be further classified as several sub-categories because of their fundamentally different emission mechanisms (e.g., organic N *vs*. inorganic N; see discussion later);



Variation range of isotopic signatures for different NH_3 sources in our study.

b. The results of emission inventories or the practical emission situation of our targeted study area.
If our sampling site is located in rural areas, then we agree with the reviewer that all volatilized sources can be combined as a single source because NH_3 source contributions in rural areas are much simpler: agricultural volatilized sources predominate the NH_3 emission budget. However, in urban areas, except for on-road traffic, there are many other important non-agricultural NH_3 sources (also volatized sources). Urban wastewater treatment plants and solid waste are arguably two of the most important NH_3 sources in urban China. As a case study, we've identified that human excreta stored in septic tanks in Shanghai is a stable and important source of atmospheric NH_3, contributing to over 11% of the total NH_3 emissions in the Shanghai urban areas. Therefore, it is of critical importance to incorporate the isotopic signatures

of waste-related NH_3 sources into the isotope mixing model.

More importantly, even in natural/rural areas, we still need to take account of emission inventory information. If we combined all volatilized sources as a single source, the underlying logic is that every volatilized source contributes equally to ambient NH_3 concentrations. However, wastewater and solid waste, generally, have an insignificant contribution to the rural NH_3 budget. Besides, **if we simply separate all NH_3 sources into two categories (volatilized source and combustion source) without the consideration of the emission inventory, then we are highly likely to get into a trouble: some $\delta^{15}N$ values of ambient NH_3 samples may be beyond the endmember, or the isotopic signatures of NH_3 sources.**

To facilitate our explanation, we would like to give a counterexample. In a recent paper entitled "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.* (2016) presented the isotopic measurements of size-resolved aerosols in Beijing, summarizing that fossil fuel-related ammonia emissions (including traffic, coal combustion and power plants NH_3 slip) have overtaken agricultural activities as the dominant source of atmospheric NH_3 during the hazy days of 2013.

There are three NH_3 sources, i.e., agricultural NH_3 volatilization, fossil fuel combustion, and power plant NH_3 slip, considered in Pan *et al.* (2016), and the average $\delta^{15}N$ values (specific values) of these sources were used as isotope signatures ($\delta^{15}N$-NH_3) to estimate their relative contributions to the ambient NH_3 in Beijing. We fully understand the author's consideration in terms of the classification of NH_3 sources: agricultural ammonia is emitted at environmental temperature, the process of fossil fuel combustion can directly emit ammonia at high temperature, and ammonia slipped from power plant is the residue 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.

However, we have to point out that temperature-only-based NH_3 source classification has a few severe problems, which may lead to wrong conclusions and therefore could potentially mislead China's policy on future NH_3 emissions reduction.

It is well accepted that agricultural activities-fertilizer application merged with livestock production-are the largest contributors of NH_3 emissions at a regional or global scale. However, recent works reveal that NH_3 volatilized from fertilizer application and livestock waste have distinct $\delta^{15}N$ values, which can also be reflected by the large variation range of $\delta^{15}N$ values for agricultural source in Pan *et al.* (2016) (see Figure 3). This is because 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. In Pan *et al*. (2016), fertilizer application and livestock waste were combined as a single source (volatilized source), which could inevitably underestimate the contribution of agricultural activities to the ambient NH_3 in Beijing.

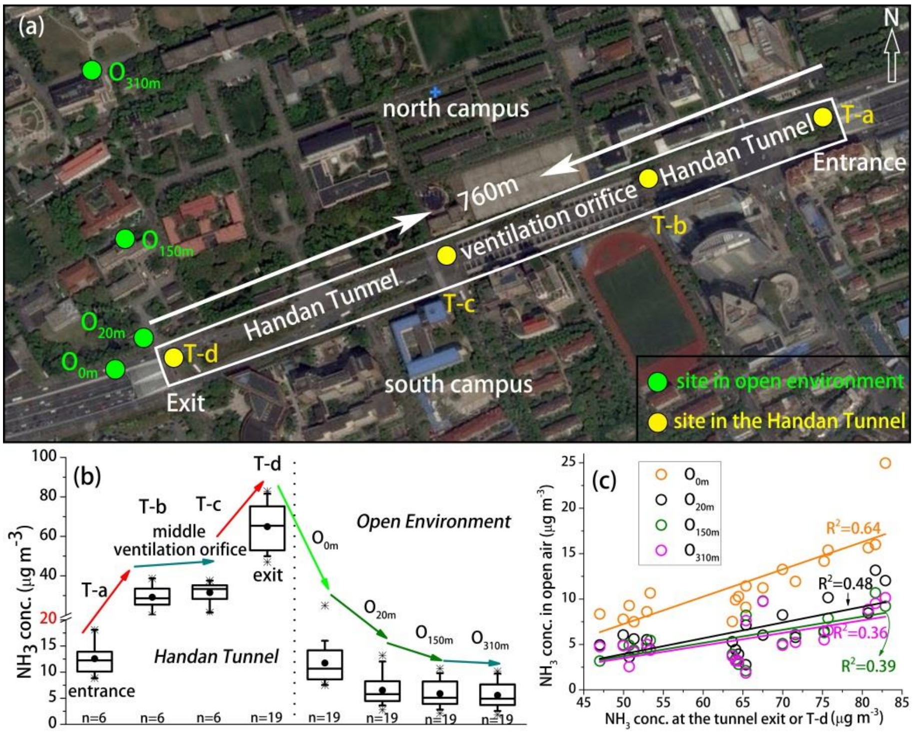
If temperature is a decisive factor, then power plant NH_3 slip considered as a major NH_3 source in Pan *et al*. (2016) should be reasonable. However, Pan *et al*. (2016) claimed that during haze periods, **49%** of NH_3 in the ambient atmosphere of Beijing was derived from power plant NH_3 slip, which 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. In 2016, all CFPP in Beijing will be shuttled and replaced with gas-fired power plants to cut pollution. The replacement by the four gas-fired power plants will help cut emissions by 10000 t of sulfur dioxide and 19000 t of nitric oxide annually. 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 two or three orders of magnitude smaller than that of NO_x. Moreover, it is necessary to consider that although there are many CFPPs surrounding Beijing in the North China Plain, most of these are co-located with intensive agricultural production areas.

(2) We also fully agree with the reviewer that the range of isotopic values, instead of a specific value of a given NH_3 source should be served as input into isotopic mixing model. Unfortunately, the isotopic mixing model-IsoSource-used in the current study only allows a fixed isotopic value input for each NH_3 source. The good news is that the isotopic signatures of each source in our study have very narrow variation range, and there is only a few data overlap between the source signatures of waste and livestock (see figure above). These could significantly reduce the uncertainty of source apportionment by IsoSource. Besides, the uncertainty of model simulation also reported in Figure 3 in the MS. Nevertheless, we think future studies should include the adoption of more sophisticated Bayesian mixing models.

Vehicle sampling Unlike the US, a major urban NH_3 source in China is human waste which has been found to have a $\delta^{15}N$-NH_3 value of ~ -41 to -30 per mil in this study. The vehicle exhaust in this study was sampled in a tunnel not directly from tailpipe. Most highly trafficked tunnels have ventilation systems that flush the tunnel with ambient air constantly. If the urban ambient air NH_3 is mainly from waste and the tunnels are flushed with ambient air, this mixing would lower the $\delta^{15}N$ value of the NH_3 sampled at the tunnel. Do these tunnels have ventilation systems? Could mixing with ambient air confound the $\delta^{15}N$ vehicle signal?

The tunnel we chose does have ventilation system and the air in the tunnel is absolutely the mixture of vehicle exhaust and ambient air (even without the ventilation system). We think the mixing with ambient air (including NH_3 emissions from human excreta) in the tunnel is not an important problem. Two reasons are as follows:

(1) The NH_3 conc. in the Tunnel (T-d in figure below; samples for isotopic analysis were also collected at) is nearly **11** times than that in ambient air (O_{310m} and O_{150m} in figure below) (Chang *et al*., 2016). Therefore, it is safe to conclude that the NH_3 emissions in the Tunnel are dominated by vehicles instead of ambient air.



(**a**) Location of the eight sampling points in (labeled in yellow; inside the tunnel from the entrance to the exit) and out (labeled in green; varying in distance from the tunnel) of the Handan tunnel. The campus of Fudan University was separated into north and south parts by the tunnel. (**b**) Box-whisker plots of the NH_3 concentration sampled at each site, setting 20 as the breaking point of y axis. The box boundaries represent the 25th and 75th percentile, the horizontal line is the median, and the whiskers mark the 10th and 90th percentiles. (**c**) Relationship between the NH_3concentration at T-d (the exit of the Handan tunnel) and the other four sites varying in distance from the Handan road in the open environment.

(2) For NH_3 emission from human excreta, we recently quantified that that the population of ~21 million people living in the urban areas of Shanghai annually emitted approximately 1386 t NH_3, which corresponds to over 11.4% of the total NH_3 emissions in the Shanghai urban areas (Chang *et al*., 2015). Therefore, we don't think human excreta is a major (but still important) urban NH_3 source in Chinese megacities like Beijing.

Line 119: All three filters from a sampling event were combined for single analysis. Why?

Was there not enough N for analysis? This doesn't allow for reporting of the deviation, if any among samplers. Did the authors evaluate deviation among samplers? If so this should be included.

Yes, ensuring enough N for isotopic analysis was our top priority. Efficient NH_3 PSD for short-term sampling and for N isotope analysis is still missing. In our previous work, two Ogawa filter samples collected monthly in Shanghai often cannot absorb enough NH_3-N for isotopic analysis. Although the ALPHA PSDs used in Beijing had larger filters to absorb more NH_3, the sampling time in our study was shortened. Therefore, we combined three ALPHA filters for a single analysis. Although we didn't have isotopic deviation among samplers, we had noticed the potential deviation of NH_3 concentrations among different filters. We had two co-located ALPHA PSDs simultaneously operating at our sampling site as part of our monitoring campaign. We didn't find any significant difference between the combined samples (for isotopic analysis) and these single filter samples.

Line 166: When sampling exhaust from septic tanks the authors state "However, the δ^{15}N-NH_3 values of daily samples varied widely (±10 per mil), suggesting that the isotope fractionation may occur during the process of sampling/storage. After many tests by trial and error, we found that a sampling period of 2 hours could provide sufficient N-NH_3 as well as avoid potential fractionation" It seems the daily samples would be more representative of the source and the fractionation that is occurring is representative of the source. The magnitude of fractionation is going to change under varying environmental conditions but this is a symptom of the source type and should be evaluated as the range in source signature.

Compared with previous work, we have every confidence that we had optimized our sampling to provide sufficient N-NH_3 and avoid potential fractionation. As to the sampling period we chosen, please note that the NH_3 conc. in the ceiling ducts were very high. A whole day sampling was unpractical because the sampling filters could be overloaded. Besides, NH_3 conc. in the ceiling ducts only came from septic tanks without the interference of ambient air.

We cannot agree more that "the magnitude of fractionation is going to change under varying environmental conditions". If the fluctuation of daily environmental conditions could alter the δ^{15}N-NH_3 values, then we can expect a much larger difference between different seasons. However, our results show that even sampling in different seasons, the δ^{15}N-NH_3 values of the samples collected from septic tanks didn't show significant difference.

Line 400: The authors state "However, as a direct product of NO reduction on the catalyst surface of TWCs ($2NO+5H_2 \rightarrow 2NH_3+2H_2O$ and/or $2NO+2CO+3H_2 \rightarrow 2NH_3+2CO_2$), NH_3 emitted from light-duty vehicle exhausts can be expected to have similar δ^{15}N-values to vehicle-emitted NO_x." This reasoning is not *sound. NO produced may have a different original* $\delta^{15}N$ *value then the* NH_3 *being used in the TWC and the fractionation factor of the two different compounds caused by the TWC process could be very different. There is not valid evidence to state that the* $\delta^{15}N$ *of vehicle* NO_x *and* NH_3 *would be the same.*

To avoid misunderstanding, we decided to delete this sentence in our revised MS (line 390-405).

Minor comments
Line 28: APEC should not be abbreviated Line 51: delete "extensive" Line 160: "don't" should be replaced with "doesn't" Line 198: "A" should be deleted Lin329: The wording "far ahead" is not the appropriate descriptor here and should be changed.

Revised accordingly. "far ahead" has been replaced by "much more than" in the revised MS.

Figure 3: The x-axis labeling isn't sufficient. Are the boxes in order of sampling period? If so, provide the timeframe on the axis.

Timeframe has been added on the axis.