Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-187-RC2, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Isotopic partitioning of nitrogen in $PM_{2.5}$ at Beijing and a background site of China" *by* Yan-Li Wang et al.

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Received and published: 6 May 2016

The paper presents a method for determining nitrogen isotope ratios (δ15N) for total nitrogen (TN) in particulate matter (PM). The advantage of this method is that it seems to be simple and very precise. The disadvantage is that TN in PM consists of several components. Therefore the value of δ15N measurements in TN as additional constraint for determining the origin of TN is somewhat limited. The authors try to resolve this by using an isotope model (Iso Source). However, considering the large number of possible sources for TN, the system (described by Eq.1 in the paper,) even with the additional constraint of δ15N measurements in TN, is still underdetermined. Consequently a correct mathematical solution will result in a space of possible solutions, not a single solution and not even a best solution since, without additional assumptions or introducing (often inherent) boundary conditions, all solutions for an underdetermined

system are equally valid. One may argue (see comments of Referee 1) that consequently the scientific value of the information in this paper is very limited. I do not completely agree. A paper thoroughly evaluating possibilities and limitations of an approach to identify the sources for TN in PM based on a simple, precise method for δ15N measurements in TN could be of substantial scientific interest, especially when based on two data sets from locations impacted by very different types of sources for TN. Unfortunately, in its current form, the paper provides little additional insight. The main problems are: i) There is no explanation of the additional constraints, assumptions and boundary conditions that are used to determine best solutions and the various uncertainty ranges given in by Figure 4 and Table 2. ii) There are several factors that can create uncertainty in the solutions: a) The problem of an underdetermined system b) The uncertainty of δ15N values for the various sources of TN in PM c) The variability of chemical composition and δ15N within the two sample sets The potential value of the used approach depends on a). Uncertainty arising from b) can (at least in principle) be reduced by conducting more detailed studies of δ15N in emissions and isotope effects for atmospheric reactions. The variability arising from c) could actually be useful information since it would provide insight into contributions from different source types under different conditions. Unfortunately, the paper does not provide any quantitative insight to which extent a), the fundamental limitation, impacts the reliability of source apportionment based on measurements of δ15N in TN. The qualitative statements and conclusions that isotope ratio measurements and combinations of different types of isotope ratio measurements can provide new insight and constraints are nothing new. iii) In their isotope models for the sample set from a remote location the authors use a fixed offset of 33 ‰ to adjust for isotope fractionation resulting from distribution of NH3/NH4- between gas and PM phase. However, the actual isotope fractionation for N in NH3/NH4- between gas and PM phase will depend on the fraction of total NH3/NH4- over NH4- in PM. The results shown in Figure 3 seem to be consistent with this fundamental principle. Consequently the use of a given fixed value for background samples and no correction for urban samples seems somewhat arbitrary and this may substan-

tially impact the results of source apportionment. iv) The dependence shown in Figure 3 seems interesting. However I do not understand the use of NH4-/(NO3-+1/2 SO42-) as x-axis. Based on stoichiometry it should be NH4-/(NO3-+2 SO42-). I am not sure how this would change the dependence shown Figure 3, but in any case a dependence such as the one in Figure 3 can also result from varying contributions from a source emitting NO2 enriched in δ15N or a source of NH3 depleted in δ15N and so on. These problems need to be addressed quantitatively. After all, the results of source apportionment are numbers and the value of these numbers depends on quantitative understanding of uncertainties. If the authors are able and willing to revise the paper accordingly, a substantially revised manuscript may be considered for publication. In a revised manuscript the authors should pay more attention to avoiding very unusual English phrases and the use of correct grammar as well as providing all necessary details to allow the reader to fully understand what is presented in the tables and figures. In the current version I often had to guess what is shown. I would also like to point out that I find the introduction to be too long, especially since the experimental part also includes several “introductory type” statements. Since a publishable revised version will substantially differ from the current one I will not going into details. In summary, in its current form the paper should not be published in ACP, however a thoroughly revised version could be reconsidered for publication in ACP.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-187, 2016.