

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 1:**

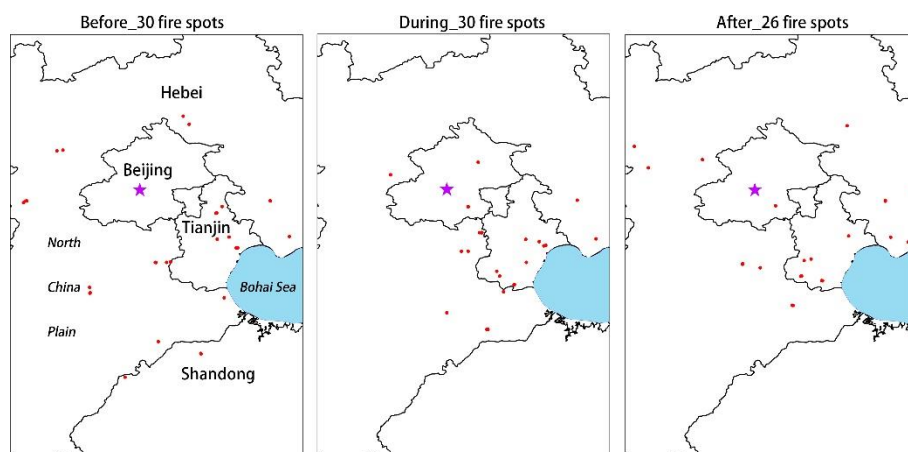
*The manuscript by Chang et al. studied the sources of atmospheric ammonia before, during, and after the 2014 APEC summit in Beijing using stable nitrogen isotope signatures coupled with an isotope mixing model. The source contributions of traffic, waste, livestock, and fertilizer were quantified and compared. The results showed a substantial decrease (58.7%) of traffic emission during the APEC period with strict emission controls. The results also showed that non-agricultural sources (traffic and waste) of NH<sub>3</sub> play an important role in particle pollution in the megacity of Beijing, which has important implications for future air pollution mitigating strategies. This is a pioneer study by applying isotopic measurements into source apportionment of NH<sub>3</sub>. Such an approach could be very powerful in future source apportionment studies particularly if combining with more collocated measurements. I recommend it for publication after addressing the following two comments.*

Thanks for the encouraging comments, and also useful suggestions, which will certainly be taken into account in our field measurements in the future.

*One of the assumptions of this study is that the contribution of biomass burning is considered minimal. The authors need to address the uncertainties of this assumption because Xu et al. (2015) showed that biomass burning contributed 12-19% to total organic aerosols before and during APEC in Beijing.*

Firstly, biomass burning is well-known as a major source of ambient organic aerosols (OA). The factors/sources of OA in non-refractory submicron aerosols identified by aerosol mass spectrometer typically (including Xu et al. (2015)) include HOA, COA, biomass-burning (BBOA), coal combustion (CCOA), semi-volatile OOA (SV-OOA), and low volatility OOA (LV-OOA). Although biomass burning (e.g., crop residues, wild fires) also contribute to NH<sub>3</sub> emissions, their emission factors are much less than that of OA and its large group of precursors (Akagi et al., 2011; Stockwell et al., 2014).

Secondly, we illustrated the locations (red dots) and number of fire spots (<https://firms.modaps.eosdis.nasa.gov/>) in Beijing and its neighboring region from 18<sup>th</sup> October to 29<sup>th</sup> November 2014. The figure below clearly showed that fire spots in each period were sparse, indicating the magnitude of open burning activities was very limited. Such a result can be expected because all crops grown in open-field (e.g. maize and cotton) in Northern China had been harvested before the APEC summit.



Thirdly, there is a considerable amounts of wood burning for heating in EU and US (Clark *et al.*, 2013; Saffari *et al.*, 2013). While this is not the case in Northern China where coal-based heating is overwhelmingly popular (Chen *et al.*, 2013). Besides, wood burning for cooking only exists in some rural areas of China that are far away from our sampling site in Beijing. Moreover, regions having wood burning for cooking also having intensive agricultural activities (e.g., livestock production), which makes  $\text{NH}_3$  emissions from wood burning relatively insignificant. In addition, the atmospheric behavior of  $\text{NH}_3$  is characterized by a short lifetime (1-5 days or less (Warneck, 1999)), low transport height, and relatively high dry deposition velocity (Asman and van Jaarsveld, 1992), high rural  $\text{NH}_3$  emissions do not generally influence urban areas strongly in the gaseous phase unless reacting with acidic gases locally to form particulate  $\text{NH}_4^+$  (Flechard *et al.*, 2013).

In conclusion, we are confident that the contribution of biomass burning to ambient  $\text{NH}_3$  concentrations was minimal during our sampling period in Beijing.

#### Reference:

- Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crouse, J. D., and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, *Atmos. Chem. Phys.*, 11, 4039-4072, doi:10.5194/acp-11-4039-2011, 2011.
- Asman, W. A., and van Jaarsveld, H. A.: A variable-resolution transport model applied for  $\text{NH}_x$  in Europe, *Atmos. Environ.*, 26, 445-464, doi: 10.1016/0960-1686(92)90329-J, 1992.
- Chen, Y., Ebenstein, A., Greenstone, M., and Li, H.: Evidence on the impact of sustained exposure to air pollution on life expectancy from China's Huai River policy, *Proc. Natl. Acad. Sci.*, 6, 110, 12936-12941, doi: 10.1073/pnas.1300018110, 2013.
- Clark, M. L., Peel, J. L., Balakrishnan K, Breyse, P. N., Chillrud, S. N., Naeher, L. P., Rodes, C. E., Vette, A. F., and Balbus, J. M.: Health and household air pollution from solid fuel use: the need for improved exposure assessment, *Environ. Health Perspect.*, 121, 1120-1128, doi: org/10.1289/ehp.1206429, 2013.

- Flechar, C., Massad, R. S., Loubet, B., Personne, E., Simpson, D., Bash, J., Cooter, E., Nemitz, E., and Sutton, M. A.: Advances in understanding, models and parameterizations of biosphere-atmosphere ammonia exchange, *Biogeosci.*, 10, 5183-5225, doi: 10.5194/bg-10-5183-2013, 2013.
- Saffari, A., Daher, N., Samara, C, et al.: Increased biomass burning due to the economic crisis in Greece and its adverse impact on wintertime air quality in Thessaloniki, *Environ. Sci. Technol.*, 47, 13313-13320, doi: 10.1021/es403847h, 2013.
- Stockwell, C. E., Yokelson, R. J., Kreidenweis, S. M., Robinson, A. L., DeMott, P. J., Sullivan, R. C., Reardon, J., Ryan, K. C., Griffith, D. W. T., and Stevens, L.: Trace gas emissions from combustion of peat, crop residue, domestic biofuels, grasses, and other fuels: configuration and Fourier transform infrared (FTIR) component of the fourth Fire Lab at Missoula Experiment (FLAME-4), *Atmos. Chem. Phys.*, 14, 9727-9754, doi:10.5194/acp-14-9727-2014, 2014.
- Warneck, P.: *Chemistry of the Natural Atmosphere*, pp. 426-441, Academic Press, San Diego, Calif., 1999.
- Xu, W. Q., Sun, Y. L., Chen, C., Du, W., Han, T. T., Wang, Q. Q., Fu, P. Q., Wang, Z. F., Zhao, X. J., Zhou, L. B., Ji, D. S., Wang, P. C., and Worsnop, D. R.: Aerosol composition, oxidation properties, and sources in Beijing: results from the 2014 Asia-Pacific Economic Cooperation summit study, *Atmos. Chem. Phys.*, 15, 13681-13698, doi:10.5194/acp-15-13681-2015, 2015.

*Coal combustion is the dominant source of aerosol particles during the heating season in Beijing. Unfortunately, such a source is missed in this study, which will affect the source apportionment results.*

We appreciate the constructive criticism. We fully agree with the reviewer that coal combustion is the dominant source of aerosol particles during heating season in Beijing. However, we also think coal combustion-derived NH<sub>3</sub> emissions could be largely neglected **in this study**.

First of all, the emission control measures taken by the Chinese government were really comprehensive and aggressive. For example, Beijing took half of the cars off local roads, closed more than 1000 heavy industrial plants within a 120-mile radius of the city. More importantly, although the convention was held during a heating period, coal-fired power plants operation and urban coal-based heating services in Beijing and Tianjin were suspended until the summit was closed (Zhang, 2014).

Besides, coal combustion is not necessary an important source of gaseous NH<sub>3</sub>. In fact, we have collected NH<sub>3</sub> emissions from coal combustion in a combustion chamber by a glass-fritted bubbler system, and the initial results do not support the view that coal combustion is an important source of NH<sub>3</sub> emissions (unpublished). Li *et al.* (2016) recently provided more robust evidence, suggesting that the average NH<sub>3</sub> emission factors for burning 13 kinds of coal in a traditional heating stove was 1.01 mg g<sup>-1</sup>, and the advanced heating stove with a highly modified combustion efficiency had a much

lower average  $\text{NH}_3$  EF of  $0.13 \text{ mg g}^{-1}$ . Supposing an amount of 19 Mt coal consumption in 2014, the annual  $\text{NH}_3$  emissions from coal combustion in Beijing was only 247-1919 t, which cannot be comparable with any other major  $\text{NH}_3$  sources (e.g., N fertilizer and animal manure/urine emissions).

Lastly, some may argue that coal-fired power plant (CFPP)  $\text{NH}_3$  slip is a major  $\text{NH}_3$  source in Beijing. But we do not think this can be true. In September 2013, a five-year plan was introduced in Beijing to slash coal consumption, and there were only four CFPPs operating near the city's urban areas during wintertime (China Daily, 2015). In 2016, all CFPP in Beijing will be shut down 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 (China Daily, 2015). Although  $\text{NH}_3$  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  $3\text{-}5 \text{ mg NH}_3 \text{ m}^{-3}$ ) in flue gases is two or three orders of magnitude smaller than that of  $\text{NO}_x$  (MOE of China, 2014). Moreover, although there are many CFPPs surrounded Beijing in the North China Plain, most of which are co-located with intensive agricultural production areas.

#### Reference:

- China Daily: Beijing closes coal power plant to cut pollution, [http://www.chinadaily.com.cn/china/2015-03/20/content\\_19869722.htm](http://www.chinadaily.com.cn/china/2015-03/20/content_19869722.htm). (accessible August 13, 2016)
- Li, Q., Jiang, J., Cai, S., Zhou, W., Wang, S., Duan, L., and Hao, J.: Gaseous ammonia emissions from coal and biomass combustion in household stoves with different combustion efficiencies, *Environ. Sci. Technol. Lett.*, 3, 98-103, doi: 10.1021/acs.estlett.6b00013, 2016.
- MOE of China, Emission standard of air pollutants for thermal power plants (GB13223-2003), 2014.
- Zhang, S. R.: Beijing smog: The day after 'APEC Blue', 2014, <http://thediplomat.com/2014/11/beijing-smog-the-day-after-apec-blue/>. (accessible August 13, 2016)