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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical composition, structures, and light absorption of N-containing aromatic compounds emitted from burning wood and charcoal in household cookstoves" *by* Mingjie Xie et al.

Anonymous Referee #1

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Xie et al. identified and quantified individual nitrogen-containing aromatic compounds (NACs) found in cookstove aerosol produced from water boiling tests. The study focused on two different fuels, charcoal and red oak, and mainly compared and contrasted emissions of NACs from cold start and hot start phases of the WBT. A unique aspect of this study is a focus on filter artifacts by comparing NACs on a quartz fiber filter placed downline of a PTFE membrane. In addition, they quantified the absorption of individual NACs at 365 nm based on their measured concentrations. The authors identified 17 different structures of NACs from their MS-MS spectra. The main con-

clusions of this study are that the back up quartz fiber filter concentrations of NACs were very high, sometimes even larger than on the front PTFE filter highlighting the importance of understanding these sampling artifacts for quantification of semivolatile species better. They also conclude that the NACs in this study make up less than 5% of the extractable absorption at 365 nm on the PTFE filter.

General comments: The results of this paper should be published because this study quantifies particulate emissions of NACs from cookstoves, which is understudied. The results also demonstrate the need to understand sampling artifacts from filters when they are used for quantitative analysis. However, some of the key conclusions of the paper may be misleading for the reader. For example, it is concluded that <5% of the extractable absorption is from NACs and they not significant brown carbon chromophores in cookstove smoke. However, much higher percentages were observed on the back up quartz filter, some of which may be in the particle phase in the atmosphere.

Specific comments:

1. There are some well-documented problems with WBTs, mostly arguing that their combustion efficiencies don't match those in the real world (Johnson et al., 2008, 2010). If the combustion efficiency in real homes is lower, this could result in less NACs due to less flaming and lower NOx. Given this, it would be helpful to have a measure of combustion efficiency, such as modified combustion efficiency, so that it can be compared with field measurements in the future. This may be possible, given the paper mentions gaseous pollutants were measured (Line 140). Even without this, it would be helpful to have more of a description of the cookstoves and WBTs which would help with the interpretation of the results.

1a. The stoves are listed in tables in the supplement, however, they are not really discussed in the experimental section of the main paper. How are they different? Where are they used around the world?

1b. Please include more information about the water boiling tests in the experimental section, as most readers of the journal will not be familiar with it. You should also mention the simmer phase is included for the hot start sample in some tests, if this is correct.

1c. Could use more reasoning as to how red oak and charcoal are different as seen in Figure S1 C and F by relating hot start and cold start phases to the observed types of combustion. For example, hot start is mostly smoldering for charcoal (high OC emissions with very low BC and therefore low NOx to make less NACs).

2. Regarding source apportionment for NAC measurements (Lines 385-402), these fractions of NAC/OM will be very different in the field because OM can come from many sources. The NAC should be ratioed to a combustion product such as CO or EC.

3. It is implied in lines 412-420 that NACs identified in this study are not significant BrC chromophores, however, if the quartz filter (Qb) is included the fraction is likely higher. It may be more appropriate to give an upper limit given that NACs on Qb could partition into the particle phase in the atmosphere. It is difficult to conclude that NACs are not significant BrC chromophores given the measurements on the sampling artifact that other studies have not considered. Also, NACs may be higher for fuel/stove/cooking activity combinations that result in more flaming combustion which produces NOx, an important reactant for NAC formation. Another factor is that the fractional absorption by NACs was not directly measured. Surrogates were used to quantify NAC concentrations and approximate MACs were used to calculate the Abs365.

4. It is assumed that because Abs365,tNAC% at 365 nm is 7-11 times higher on the quartz fiber backup filter, that NACs may be important light absorbers in the gas phase (lines 442, 425-429, 432-434). To claim this in the paper, more discussion and reasoning for should be given.

4a. Those on the backing filter are not necessarily in the gas phase in the natural environment. As you explain in the paper, there are both positive and negative artifacts and there is not likely a straightforward way of calculating what would be in the gas

phase.

4b. The vapor pressures of these molecules are very low, and the fraction in the gas phase is low. However, for some nitroaromatics such as 2-nitropenol the vapor pressure is higher. Are the concentrations for some molecules higher on the back up filter compared to the front filter and do we expect them to have higher vapor pressures?

4c. What are the absorption cross sections for these molecules in the gas phase and their expected gaseous concentrations that would lead us to believe they are significant? Are they long-lived enough in the gas phase to be important? Only solution phase MACs at 365 nm are used to claim that gas phase absorption is significant and this is not sufficient.

5. Line 132: Omit that you did kerosene tests. It is not critical as you do not discuss these results.

References

Johnson, M., Edwards, R., Alatorre Frenk, C. and Masera, O.: In-field greenhouse gas emissions from cookstoves in rural Mexican households, Atmos. Environ., 42(6), 1206–1222, doi:10.1016/j.atmosenv.2007.10.034, 2008.

Johnson, M., Edwards, R., Berrueta, V. and Masera, O.: New Approaches to Performance Testing of Improved Cookstoves, Environ. Sci. Technol., 44(1), 368–374, doi:10.1021/es9013294, 2010.

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