Anonymous Referee #3
Received and published: 29 September 2016

General comments
The authors present quantitative data on concentrations and light absorption contributions of eight nitrated aromatic compounds (NACs) measured in atmospheric particles at five different locations in Germany and China during two different seasons. Light absorption by brown carbon is an important topic for the overall assessment of the direct aerosol effect; many open questions remain related to the extent and organic compounds involved. The diversity of measurement sites and the comparison of two different methods for light absorption assessment make this study very interesting. The manuscript is well written. I thus recommend publication after the comments below have been addressed.

We would like to thank the referee for the recommendation and the helpful suggestions

Specific comments
1. The focus is on WSOC and water-soluble BrC. What I am missing is an assessment/estimate of the fractions WSOC/OC and water-soluble BrC/BrC, to get an idea of comparability and validity of methods. I am also missing a direct comparison of babs, the light absorption coefficient of particles, and Abs, and as well the calculated MAE (why was MAE not calculated for the Aethalometer data, based on babs and total PM mass/total PM organic mass, if available?) At least a comparison plot of the relative temporal evolution of these parameters
for the Waldstein (summer) and TROPOS (winter) campaigns is highly interesting from both a scientific and methodological point of view and should be added to the paper (could also be in the SI).

We agree, that this information might be interesting to the reader. Therefore, as suggested by the referee, a diagram was added to the supplement containing the temporal variation of MAE_{370} for particulate BrC, the fraction WSOC/OC and the fraction of aqueous extract light absorption to particulate BrC light absorption for the campaigns TROPOS (winter) and Waldstein (summer). Abs and b_{abs} are not directly comparable. According to the method by Liu et al. 2013 (Atmos. Chem. Phys) a conversion factor of 2 was applied to Abs. A brief comparison of Abs and b_{abs} is given in Section 3.1.3.

Additional text:
p. 9, l. 2:
"Normalizing the determined babs for particulate BrC by the according OC content gives the mass absorption efficiency for BrC in the particle (MAE370, BrC, particle). The temporal variation of MAE370, BrC, particle is displayed in Fig. S4."

p. 9, l. 8
"and an average $MAE_{370,\ BrC,\ particle}$ of 1.95 $m^2\ g^{-1}$ was calculated"

p. 9., l. 19:
", a $MAE_{370,\ BrC,\ particle}$ of 0.21 $m^2\ g^{-1}$" and "The temporal variation of the fraction of the converted Abs370 to the particulate BrC light absorption is given in Fig. S4."

Additional diagrams in the supplement:

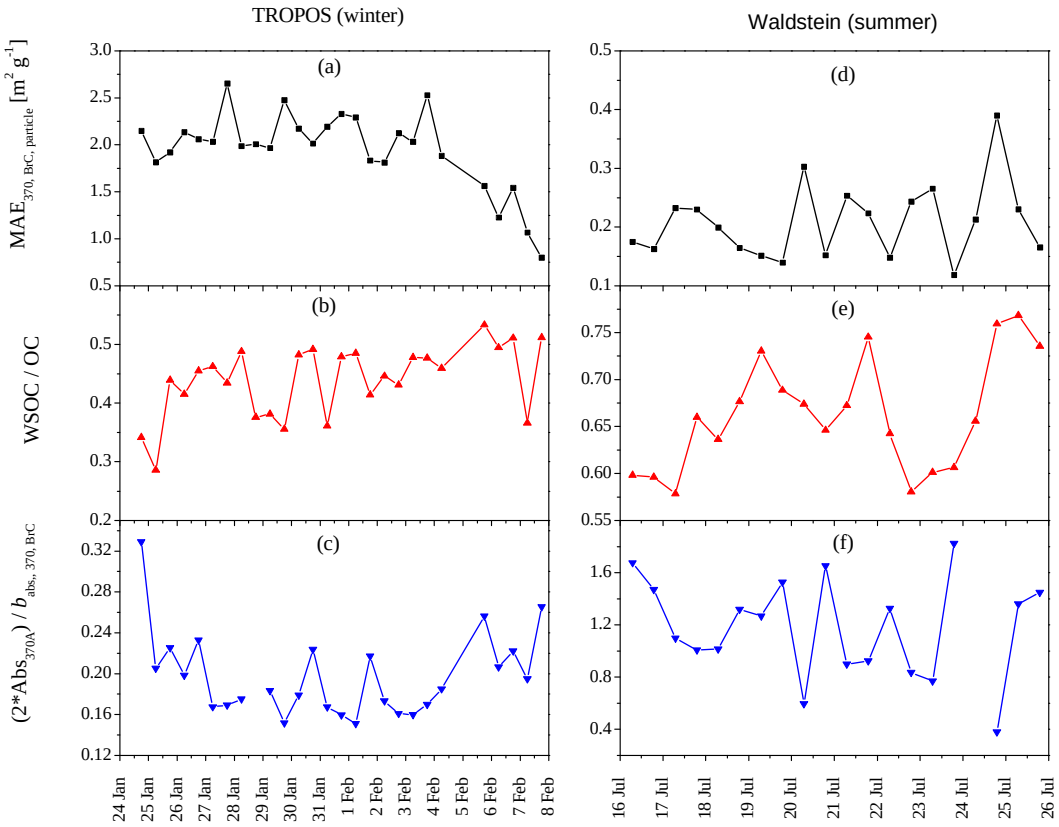

Figure S4. *Temporal variation of $MAE_{370,\ BrC,\ particle}$ for particulate BrC, the WSOC/OC fraction and the fraction of aqueous extract light absorption to particulate BrC light absorption for the campaigns TROPOS (winter) (a-c) and Waldstein (summer) (d-f). For comparability of aqueous extract light absorption and the particulate BrC light absorption, Abs_{370A} (acidic conditions) was multiplied by a factor of 2, according to the method mentioned in the main text. $MAE_{370,\ BrC,\ particle}$ was determined by normalizing $b_{abs,\ 370,\ BrC}$ by the according OC content of the sample.*

2. P. 2, l. 14 – 15: With the attention brown carbon is getting in recent years (and in the rest of your introduction) this statement seems too strong here. Add "in global climate models" for specification.

Additions were made according to the referee's suggestions.

Additional text:
p. 2., l. 14:
"in global climate models"

3. P. 3, l. 3: To my knowledge, Sandradewi et al. (Environ. Sci. Technol., 2008, 42 (9), pp 3316–3323, DOI: 10.1021/es702253m) were among the first to introduce the "Aethalometer model" for the separation of BC and BrC (then traffic vs wood burning contributions). Please

cite.

Additions were made according to the referee's suggestions.

Additional text:
p. 3, l. 3
"Sandradewi et al. 2008"

Additional reference:
"Sandradewi, S., Prevot, A. S. H., Szidat, S., Perron, N., Alfarra, M. A. Lanz, V. A., Weingartner, E., and Baltensperger, U.: Using Aerosol Light Absorption Measurements for the Quantitative Determination of Wood Burning and Traffic Emission Contributions to Particulate Matter, Environ. Sci. Technol., 42, 3316-3323, doi: 10.1021/es702253m, 2008."

4. P. 4, l. 9 (compare comment for Table 1 and p. 4, l. 38 - 40): Give time interval for 12h mean.

see reply to 10.

5. P. 4, l. 33: How were they determined, based on what criteria? Please add this information.

The determination of NACs was explained later in the text. To avoid confusion, specific NAC compounds are now mentioned later in the text

p. 4, l. 33 - 36 were moved to p. 5, l. 13.

6. P. 10, l. 27 - 30: Photolysis can be sink of NP as well.

"or photolysis processes" added to p. 10, l. 28.

Technical comments:

7. The abstract is relatively long and dense. It would profit from a bit of streamlining. Consider moving the sentence on p. 1, l. 34 – 24, to l. 34.

As suggested by the referee, to improve the readability, the abstract was shortened.

Additional text:
p.1, l. 25
"with larger values at higher pH"

Removed sentences:

p. 1, l. 25 - 26:
": at pH 10, the aqueous light absorption coefficient Abs370 and the mass absorption efficiency (MAE370) at 370 nm were a factor of 1.6 and 1.4 larger than at pH 2, respectively"

p. 1. l. 29 - 31:
"Furthermore, it was found that the MAE_{370} values in 30 winter in Germany exceeded those of the Chinese summer background stations (average of 0.85±0.24 m^2 g^{-1} compared to 0.47±0.15 m^2 g^{-1})"

p. 1. l. 36 - p. 2. l. 1:
"The absorption Ångström exponent of the ambient aerosol during the campaigns at TROPOS (winter) and Waldstein (summer) was found to be 1.5±0.1 and 1.2±0.3, respectively."

p. 2, l. 8 - 9:
"A correlation of NAC concentrations with Abs_{370} was observed for the BB-influenced campaigns at TROPOS (winter) and Melpitz (winter)."

8. P. 2, l. 14: [. . .] are usually treated [. . .]

corrected

9. P. 5, l. 7: What do you mean by "distributed sources"? Not clear.

Here, we meant it in the sense of "various" or "different". For clarity the word "distributed" was replaced by "various"

10. Table 1: The alignment of some of the columns is off. Please correct for better readability. Also add the sampling times (now give in Table S2) to Table 1.

Table 1 was corrected and the sampling times were added. Table S1, that originally contained the sampling times, was removed from the supplement.

11. Table 2: I suggest highlighting the highest and lowest values in each column/category. The light absorption contribution (in %) is given for NP and NSA individually, but there is not further mentioning of this. I assume NP is the sum of the 6 NP and NSA the sum of the 2 NSA you mention on p. 4, l. 33 -35. This kind of differentiation/grouping is only done in Table 2 – I suggest making that consistent throughout the manuscript. Do A) and B) refer to acidic and alkaline conditions? Please clarify and add this information in the table caption.

"Acidic conditions are indicated by the letter "A" and alkaline conditions are indicated by the letter "B"" was added to the table caption.
As mentioned by the referee, the differentiation for NPs and NSAs was made in Table 2 only. To be consistent with the text, we changed the information in Table 2 to values for NACs instead NPs + NSAs. Furthermore the highest and lowest mean values for each category were marked in bold, as suggested by the referee.

Literature

Liu, J., Bergin, M., Guo, H., King, L., Kotra, N., Edgerton, E., and Weber, R. J.: Size-resolved measurements of brown carbon in water and methanol extracts and estimates of their contribution to ambient fine-particle light absorption, Atmos. Chem. Phys., 13, 12389-12404, doi: 10.5194/acp-13-12389-2013, 2013.