Response to Anonymous Referee #1

This paper is investigating the concentrations of nitrated aromatic compounds (NACs) in aerosols collected in Germany and China during winter and summer seasons. The contribution of these compounds to the light absorption of BrC PM and water extracts was determined and discussed. This paper is scientifically very interesting and important for understanding the contribution of individual BrC compounds to the light absorption properties of atmospheric aerosols. The manuscript is well written and I have only a few major and minor comments.

The authors would like to thank the reviewer for recommending the paper and the helpful comments. The revision was carried out carefully according to the reviewer's suggestions.

1. Major comment 1. UV-Vis spectra of WSOC fractions were recorded from 300 to 800 nm, however, only 370 nm wavelength was used in the present study (to compare the data with Aethalometer measurements). Different aromatic compounds can have different spectra (see Pretsch et al. 2008, Samburova et al. 2016) and thus their contribution to the BrC absorptivity can vary based on wavelength. It would be very interesting to see how eight NACs contribute to the light absorption of aqueous filter extracts over the spectrum range between 300 and 800 nm.

Thank you very much for this comment. We agree, that the contribution of NACs to the aqueous extract light absorption over the spectrum range of 300 to 800 nm might be interesting to the reader. Therefore, according to the suggestion of the referee, we added a new diagram to the main script showing the contribution of NACs to Abs over the spectral range of 300 to 500 nm (acidic conditions). This range was chosen, since above 500 nm NACs are only weakly absorbing. A short description of the diagram was added to the text in Section 3.3. Furthermore, a diagram for the contribution of NACs to Abs over 300 to 500 nm under alkaline conditions, as well as the spectral evolution of Abs under acidic and alkaline conditions was added to the supplement.

Detailed changes to the manuscript:
Additional text added to Section 3.3 (lines and pages are given according to the submitted ACPD manuscript)

p. 11, l. 7:
"The focus is on the relative contribution to the aqueous extract light absorption at the wavelength of 370 nm to match the 370 nm channel of the Aethalometer. To give additional information, the relative contribution of NACs to Abs (acidic conditions) over a spectral range of 300 to 500 nm is presented in Fig. 5 as their campaign averages."

p. 11, l. 18:
"The relative contribution of NACs to Abs is wavelength dependent. Under acidic conditions, the contribution of NACs to the aqueous extract light absorption increases towards lower wavelength, reaching a maximum that was found to generally lie in the range of 330 - 350 nm (see Fig. 5). Moreover, it can be seen that maximum values differ for individual compounds. Under alkaline conditions, this maximum shifts towards the range of 400 - 420 nm (see Fig. S6). As stated above, the results for acidic conditions are likely to be more atmospherically relevant. Therefore, a higher influence of NACs towards shorter wavelengths suggests that they can be more important in terms of their influence on atmospheric photochemistry, e.g. O_3 photolysis (Jacobson, 1999)."

removed sentence:
p. 12, l. 10-11:
"This suggests that the influence of NACs is higher towards shorter wavelength, i.e. near their absorption maximum and thus can be more important in terms of their influence on atmospheric photochemistry, e.g. O3 photolysis (Jacobson, 1999)."

Additional diagram in the manuscript:

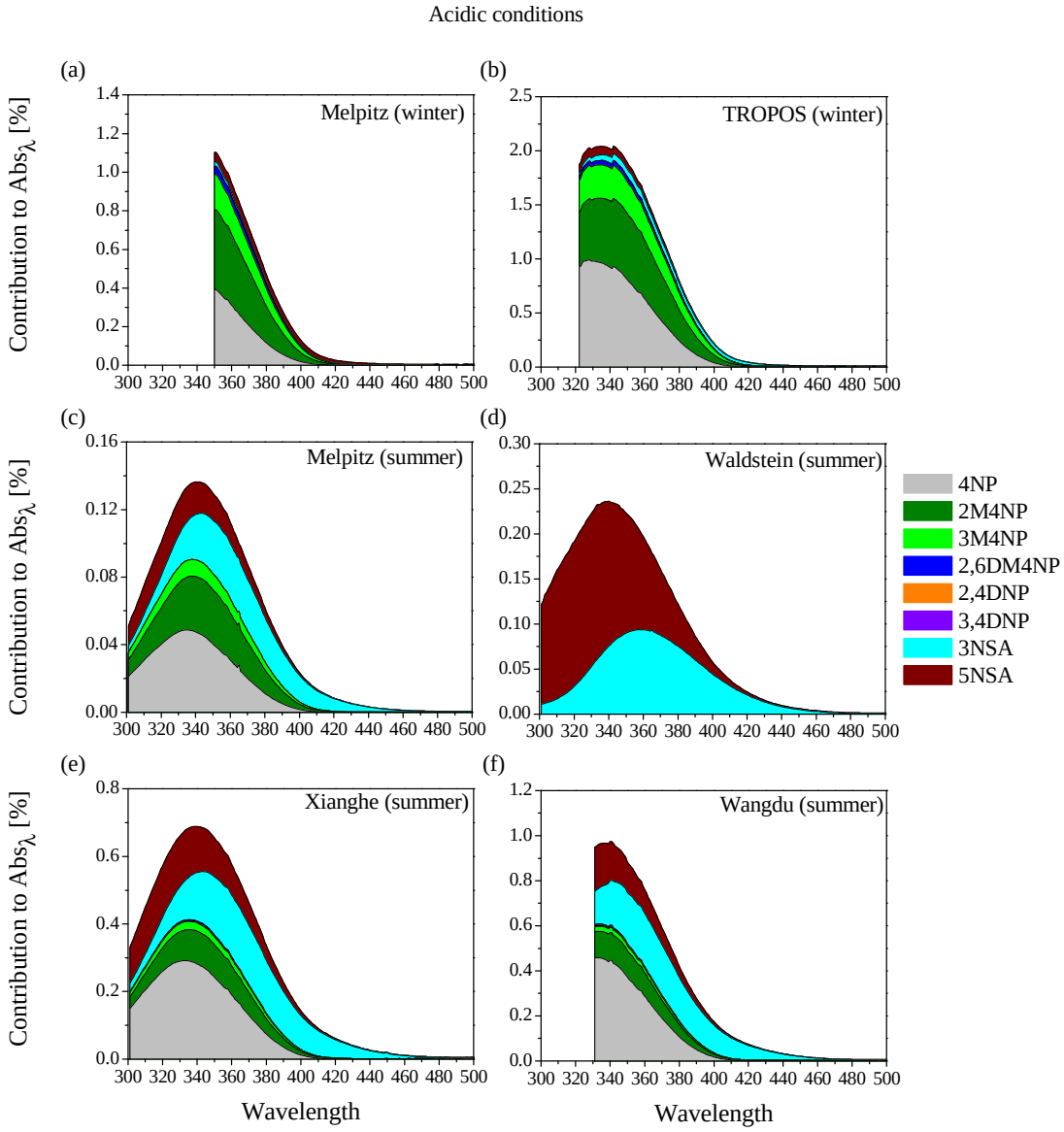

Figure 5. *Relative contribution of NACs to Abs$_\lambda$ over the spectral range of 300 to 500 nm for each measurement campaign (a-f) under acidic conditions. The data is presented as campaign averages. Due to instrumental issues, data for lower wavelengths is not always available.*

Figure numbers of diagram appearing later in the text were changed accordingly.

Additional diagrams in the supplement:

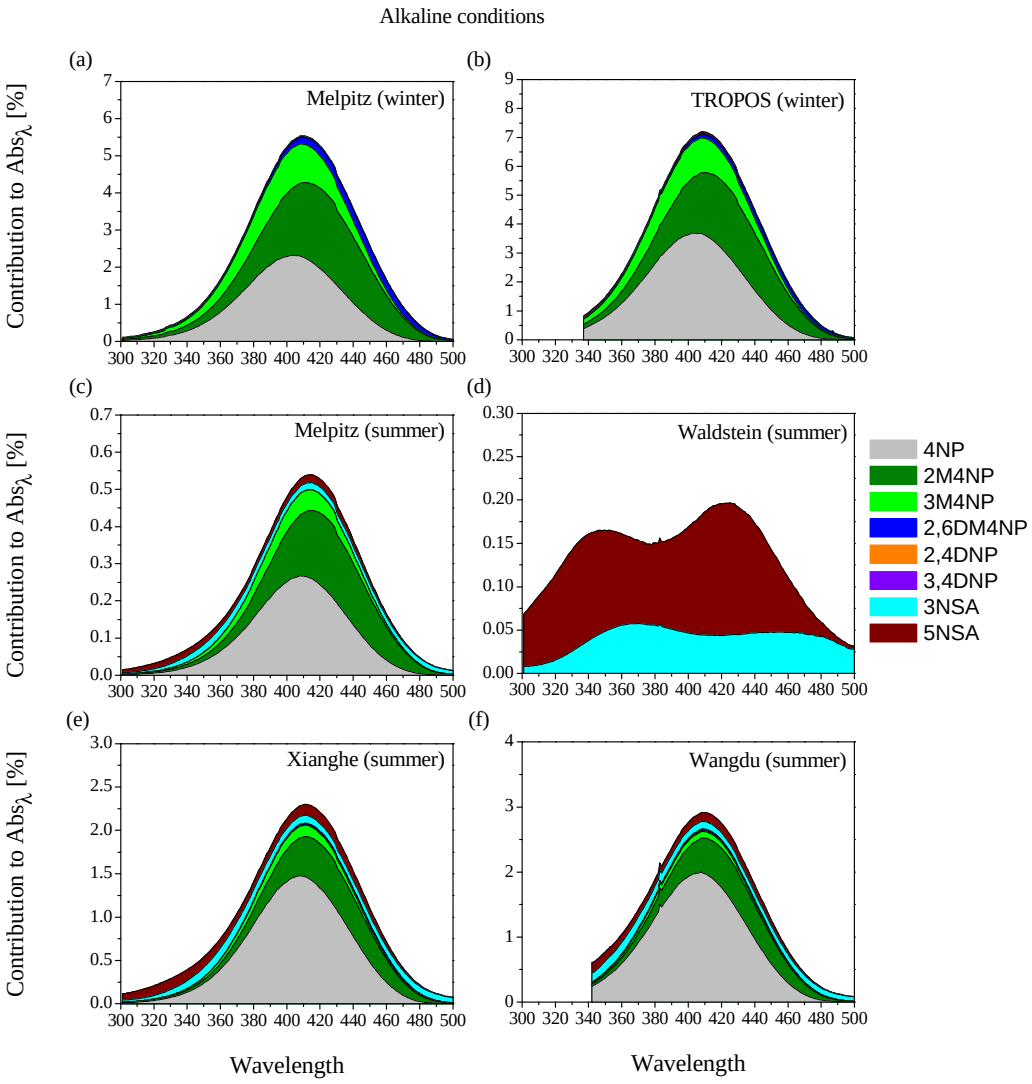

Figure S6. *Relative contribution of NACs to Absλ over the spectral range of 300 to 500 nm for each measurement campaign (a-f) under alkaline conditions. The data is presented as campaign averages. Due to instrumental issues, data for lower wavelengths is not always available.*

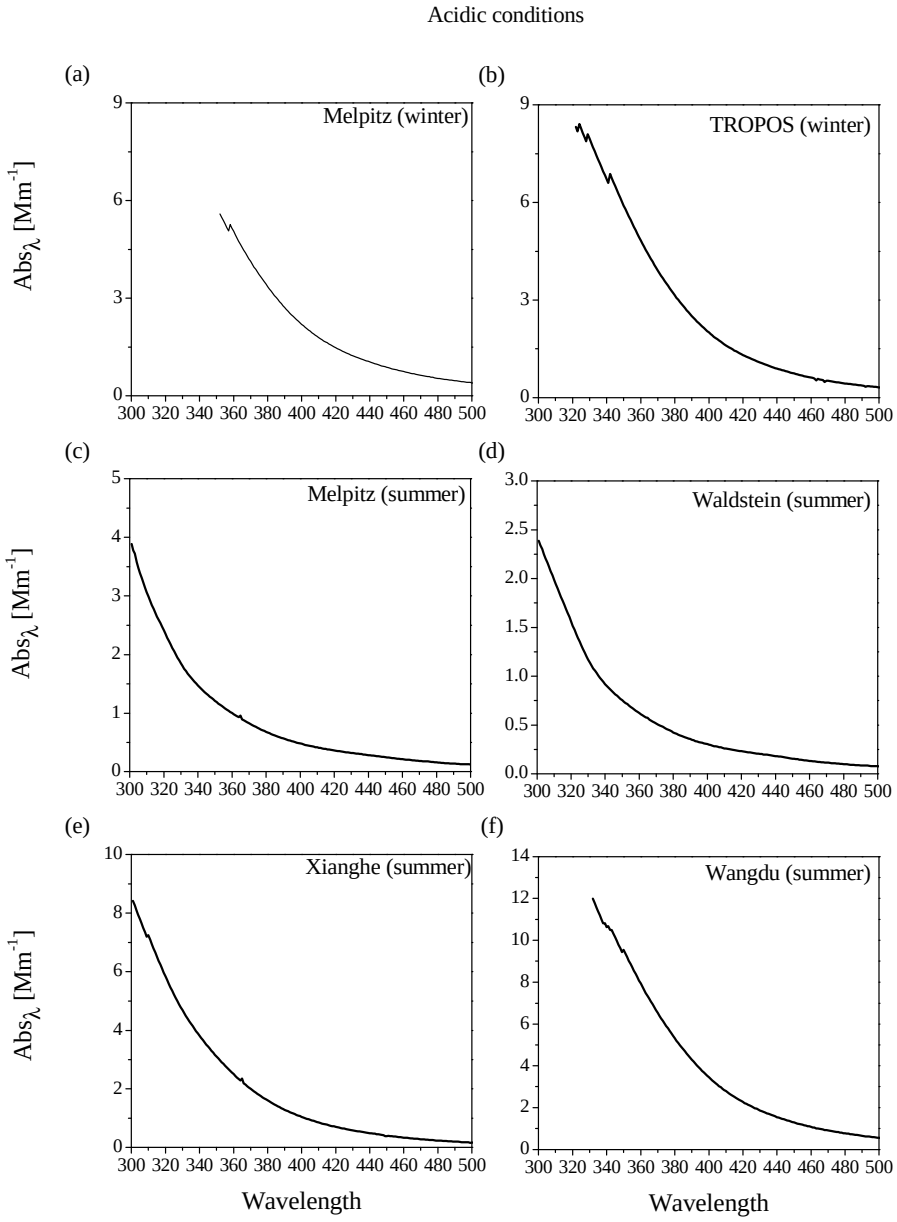

Figure S7. *Abs$_\lambda$ over the spectral range of 300 to 500 nm for each measurement campaign (a-f) under acidic conditions. The data is presented as campaign averages. Due to instrumental issues, data for lower wavelengths is not always available.*

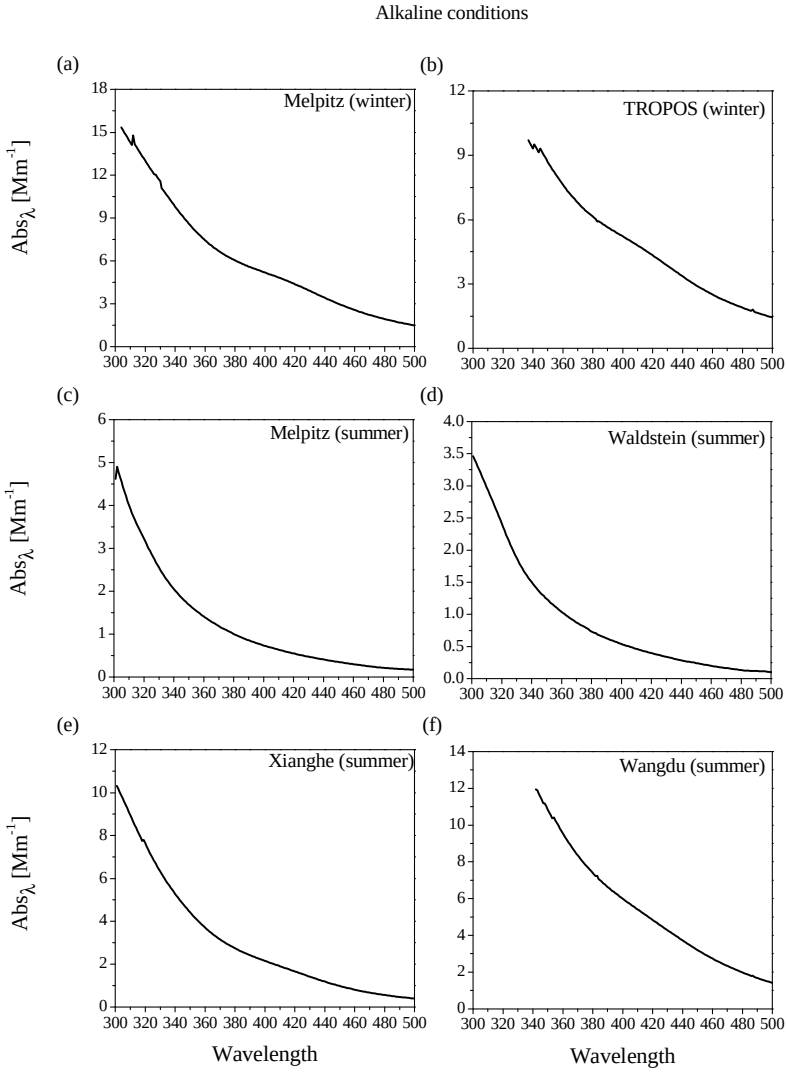

Figure S8. *Abs$_\lambda$ over the spectral range of 300 to 500 nm for each measurement campaign (a-f) under alkaline conditions. The data is presented as campaign averages. Due to instrumental issues, data for lower wavelengths is not always available.*

2. Major comment 2. Did the author compare the absorption properties of PM water extracts at "natural" pH with artificially acidified and alkylated samples?

The pH of the pure water extracts depends on the amount of water used in the extraction process. Furthermore, it would contain a mixture of protonated and deprotonated NACs. To determine the relative contribution of NACs to Abs one would need to know the amount of protonated and deprotonated species in the solution. Under the chosen acidic conditions the NACs are fully protonated and under alkaline conditions fully deprotonated. Thus we determined the lower and upper limits for the contribution of NACs to Abs. Due to the limited amount of aqueous filter exracts (and filter material) we chose to analyse acidified and alkalized filter extracts only.

3. Minor comments: Abstract, Line 13. Use (NACs: nitrophenols and nitrated salicylic acid)
P. 2, Line 26. “in the UV” means in the “UV-Vis range of the spectrum”?

Here, we do refer to the “UV range of the spectrum” and not to the “UV-Vis range of the spectrum, since the observed reduction of photolysis rates and atmospheric oxidents was reported to be in the UV and not UV-Vis. However, to clarify this sentence “range of the spectrum” was added.

4. P.2, Line 35. Use “BC” instead of “black carbon”, since this abbreviation was introduced above (line 18)
P. 10, Line 23. Use abbreviation “NPs” P. 10, Line 40. Use abbreviation “NACs”

Changes were made according to the referee’s suggestion throughout the manuscript.

5. P. 3, Lines 6-7. References are needed after “. . .absorption at 365 nm is used to characterize water-soluble BrC.”

References added “(e.g., Bosch et al. 2014; Cheng et al. 2011; Hecobian et al. 2010)” (These references were already listed in the reference list in the manuscript)

6. P. 3, Line 33. Use “biomass-burning aerosols”

done

7. P. 3, Lines 41-43. This long sentence is a little confusing.

To clarify this part, the sentences was divided into two sentences: “The present study aims to expand the understanding of BrC and water-soluble BrC by investigating its spatial and temporal variation in different, very diverse environments (urban, rural, biogenic, high BB influence). Furthermore, the contributions of individual light-absorbing organic compounds to the BrC light absorption is included.”

8. P4, Line 9. Add the period (not “until 31 January”) for “the first half of the campaign”.

replaced with “(24 to 31 January)”

9. P4, Line 41. It sounds a little better, but optional: “Analysis of WSOC, levoglucosan, NACs and UV/Vis spectrophotometry measurements were carried out using aqueous filter extracts of different portions of filter in ultrapure water.”

This sentence was adopted to the manuscript according to the referee’s suggestion

10. P. 10, paragraph 1. Why significant BB absorption was observed at the German sites during winter campaigns (because there are more domestic biomass-burning events)?

As stated in Section 2.1, we concluded that BB was playing a major role at the German winter sites because of high observed levoglucosan concentrations. It is also known that BB due to domestic heating can play a large role in this area.

11. P.10, Lines 35-39. If BB was not the major contributor to the analyzed BrC particles, what was a possible source of BrC compounds? Suggestions?

A similar question was asked by the anonymous Referee#2. Based on the comment by the anonymous Referee#2 and anonymous Referee#1 a possible source for NACs was added to Section 3.2.

Added text:
p.10, l. 39:
"*It was found that NACs can be a product of the photochemical processing of anthropogenic volatile organic compounds (Jaoui et al. 2008), which might be a possible source for NACs at the Chinese sites besides BB." was added to the text.*

Additional reference:
"Jaoui, M., Edney, E. O., Kleindienst, T. E., Lewandowski, M., Offenberg, J. H., Surratt, J. D., and Seinfeld, J. H.: Formation of secondary organic aerosol from irradiated α-pinene/toluene/NOx mixtures and the effect of isoprene and sulfur dioxide, J. Geophys. Res., 113, D09303, doi: 10.1029/2007JD009426, 2008."

12. P.11, Line 41. Delete space between 4 and % Summary: I recommend this manuscript for publication after minor revisions.

Already published papers in ACP do use a space between the number and %. Therefore, we chose to also use a space between the number and % and the corrections were made accordingly throughout the text.