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# Contributions of nitrated aromatic compounds to the light absorption of water-soluble and particulate brown carbon in different atmospheric environments in Germany and China

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## S1 Correction procedure for Aethalometer data

The Aethalometer data was corrected against MAAP data (Multi–Angle Absorption Photometer) to account for multiple scattering effects on the filter matrix. The time resolution of MAAP and Aethalometer was set to 1 s. First, the particulate light absorption coefficient $b_{abs}$ was calculated for the MAAP using the following equation:

$$b_{abs,MAAP}[Mm^{-1}] = [BC_{MAAP}] \cdot 6.6\frac{m^2}{g} \cdot 1.05 \qquad \text{(S1)}$$

Where $[BC_{MAAP}]$ is the mass concentration of black carbon (BC) derived from the instrument, 6.6 $m^2\ g^{-1}$ is the mass absorption cross-section (MAC) for ambient aerosol particles determined for the wavelength 670 nm. The factor 1.05 accounts for an adjustment to the correct instruments operating wavelength of 637 nm (Müller et al., 2011).

Accordingly, the particulate light absorption coefficient was calculated for the Aethalometer:

$$b_{abs,Aetha} = [BC_{Aetha}] \cdot \frac{MAC_\lambda}{C} \qquad \text{(S2)}$$

Where $[BC_{Aetha}]$ is the mass concentration of BC derived from the instrument and C is a normalization factor. MAC values used for the Aethalometer are given in Table S2.
Since the MAAP operates at 637 nm $b_{abs,\ Aetha}$ was interpolated to 637 nm using Eq. (S4). The AAE was calculated using the wavelength pair of 590 and 660 nm according to the following equation:

$$AAE = -\frac{\ln\left(\frac{b_{abs,590}}{b_{abs,660}}\right)}{\ln\left(\frac{590}{660}\right)} \qquad \text{(S3)}$$

Now $b_{abs,637}$ can be calculated as follows:

$$b_{abs,637} = b_{abs,590} \cdot \left(\frac{637}{590}\right)^{-AAE} \qquad \text{(S4)}$$

When $b_{abs,637,\ Aetha}$ is plotted against $b_{abs,MAAP}$ the slope of the regression line corresponds to the correction factor C, that is 1.69 for the TROPOS (winter) campaign and 2.06 for the Waldstein (summer) campaign (see Fig. S1 and Fig. S2).

## S2 Figures and Tables

**Table S1.** Mass absorption cross–sections (MAC) for ambient aerosol at a given wavelength λ used to calculate the particulate light absorption coefficient $b_{abs}$ from Aethalometer data (Drinovec et al., 2015).

| λ [nm] | 370 | 470 | 525 | 590 | 660 | 880 | 940 |
|---|---|---|---|---|---|---|---|
| MAC [$m^2\ g^{-1}$] | 18.47 | 14.54 | 13.14 | 11.58 | 9.89 | 7.77 | 7.19 |

**Table S2.** Molar extinction coefficients determined under acidic and alkaline conditions (in $M^{-1}$ $cm^{-1}$).

| Compound | $\varepsilon_{370}$ pH ≈ 2 | $\varepsilon_{370}$ pH ≈ 10 |
|---|---|---|
| 3NSA | 2509 | 2948 |
| 5NSA | 1349 | 2271 |
| 4NP | 1653 | 12243 |
| 2M4NP | 3061 | 7673 |
| 3M4NP | 2105 | 11408 |
| 2,6DM4NP | 1575 | 1916 |
| 2,4DNP | 1239 | 14075 |
| 3,4DNP | 1650 | 8521 |

**Table S3**. Chemical parameters for each campaign. The concentrations are given as: minimum-maximum (mean±standard deviation)

| | Waldstein (summer) | Melpitz (summer) | TROPOS (winter) | Melpitz (winter) | Xianghe (summer) | Wangdu (summer) |
|---|---|---|---|---|---|---|
| PM 10 [μg $m^{-3}$] | 11.1-24.4 (17.3±3.92) | 18.7-41.0 (26.3±6.57) | 6.35-85.8 (40.9±21.2) | 23.8-67.6 (40.6±13.5) | 19.7-297 (118±59.7) | 18.8-143 (63.4±27.8) |
| OC [μg $m^{-3}$] | 2.68-6.57 (4.63±0.95) | 4.45-9.71 (6.38±1.58) | 1.56-25.3 (10.6±6.32) | 4.97-23.0 (12.6±6.15) | 5.51-29.6 (13.9±5.39) | 7.09-48.9 (17.8±8.91) |
| EC [μg $m^{-3}$] | 0.15-0.42 (0.24±0.07) | 0.24-0.46 (0.35±0.08) | 0.29-3.31 (1.53±0.69) | 0.60-2.02 (1.30-0.54) | 0.86-7.01 (3.07±1.44) | 1.06-9.37 (3.55±1.73) |
| WSOC [μg $m^{-3}$] | 2.06-4.53 (3.05±0.63) | 2.20-6.09 (3.92±1.26) | 0.80-8.64 (4.39±2.26) | 2.64-7.02 (4.65±1.56) | 2.08-10.03 (5.21±1.84) | 2.53-23.6 (7.80±4.67) |
| Levoglucosan [μg $m^{-3}$] | | | 0.05-1.02 (0.46±0.24) | | 0.01-0.38 (0.12±0.08) | 0.03-2.01 (0.29±0.41) |

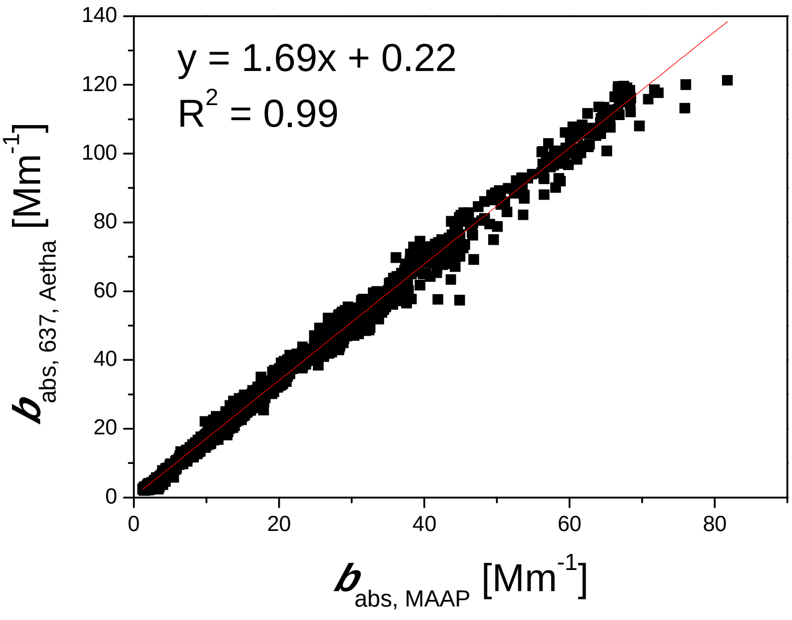


**Figure S1.** Correlation plot of the particulate light absorption coefficient derived from the Aethalometer at 637 nm ($b_{abs,637,Aetha}$) and the particulate light absorption coefficient derived from the MAAP at 637 nm ($b_{abs,MAAP}$) at the TROPOS (winter) campaign.

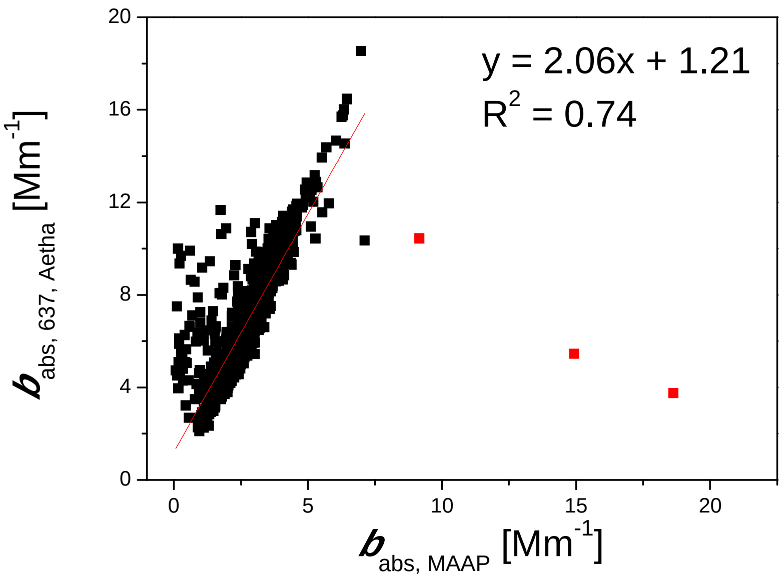


**Figure S2.** Correlation plot of the particulate light absorption coefficient derived from the Aethalometer at 637 nm ($b_{abs,637,Aetha}$) and the particulate light absorption coefficient derived from the MAAP at 637 nm ($b_{abs,MAAP}$) at the Waldstein (summer) campaign. Red data points are excluded from the regression fit.

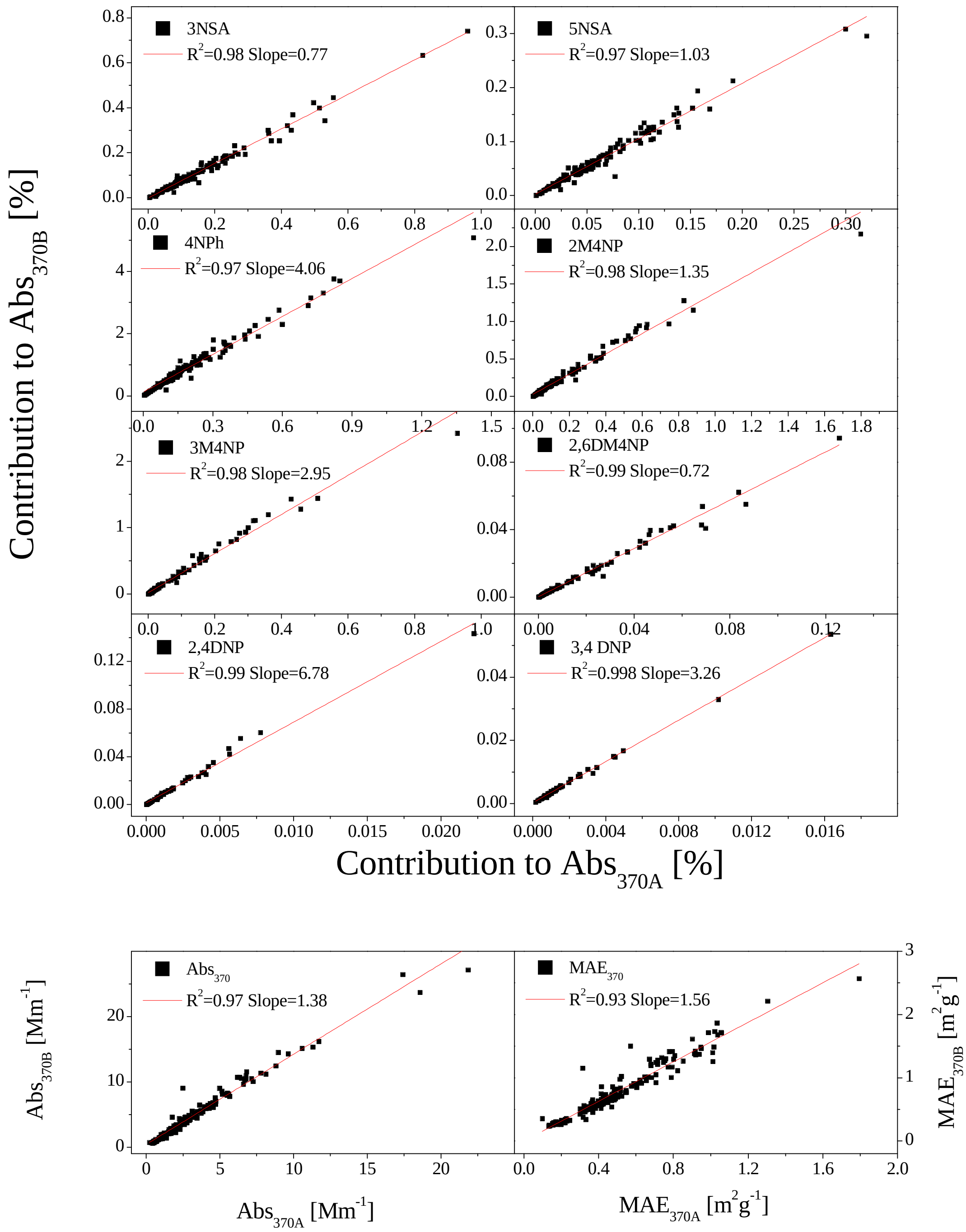


**Figure S3.** (Top) Correlation plot of the contribution to the aqueous light absorption coefficient $Abs_{370}$ for each target compound under alkaline (indicated by the subscript “A”) and acidic (indicated by the subscript “B”) conditions. (Bottom) Correlation plot of the aqueous light absorption coefficient and mass absorption efficiency (MAE) under alkaline conditions with acidic conditions. All data points were measured at 370 nm.

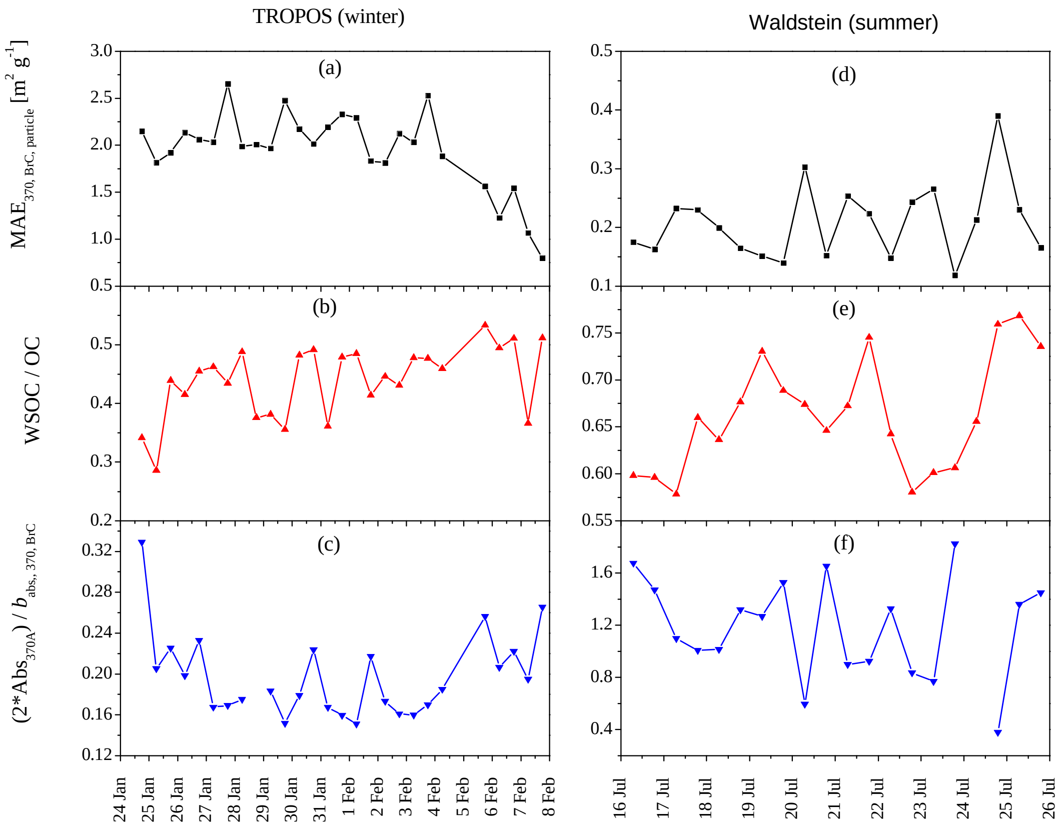


**Figure S4.** Temporal variation of $MAE_{370,\ BrC,\ particle}$ for particulate BrC, the WSOC/OC fraction and the fraction of aqueous extract light absorption to particulate BrC light absorption for the campaigns TROPOS (winter) (a-c) and Waldstein (summer) (d-f). For comparability of aqueous extract 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.

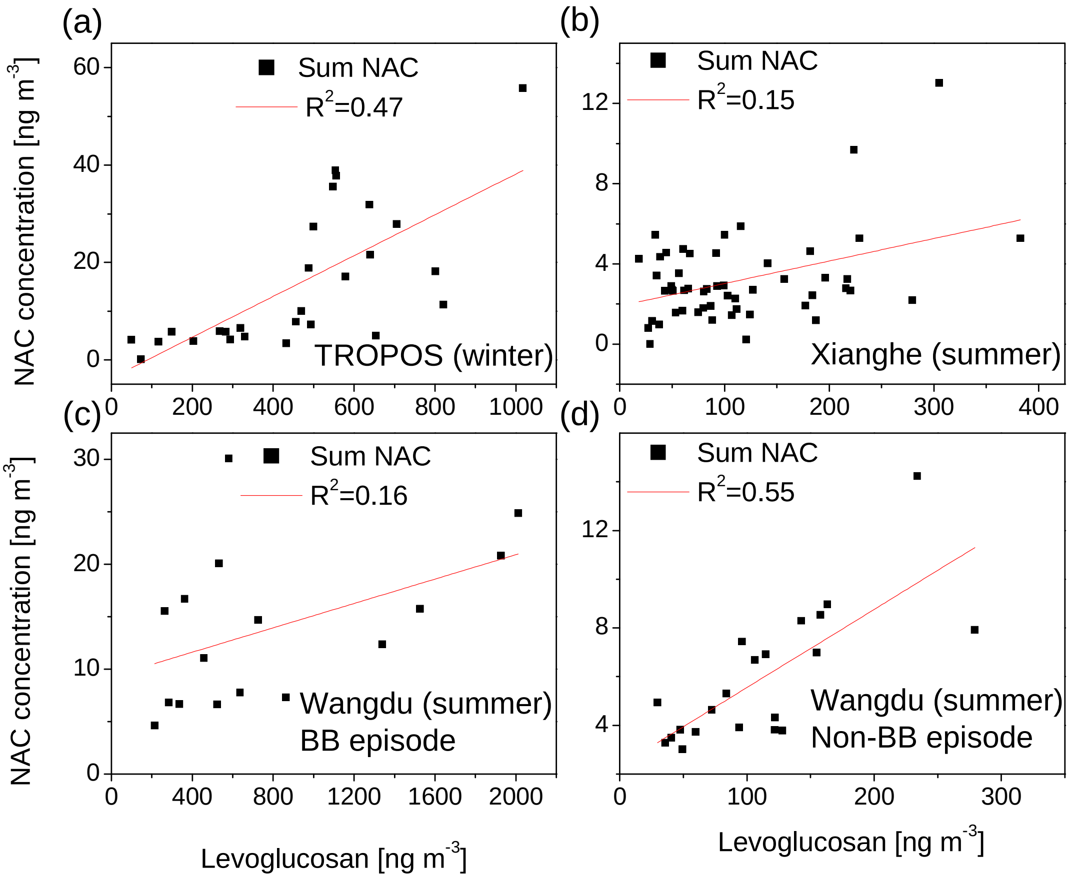


**Figure S5.** Correlation plot of nitrated aromatic compound (NAC) concentration with levoglucosan concentration for the campaigns (a) TROPOS (winter,), (b) Xianghe (summer), (c) Wangdu (summer, BB episode) and (d) Wangdu (summer, non-BB episode). $Abs_{370}$ is given for acidic conditions (indicated by the subscript "A", black squares) and for alkaline conditions (indicated by the subscript "B", red dots).

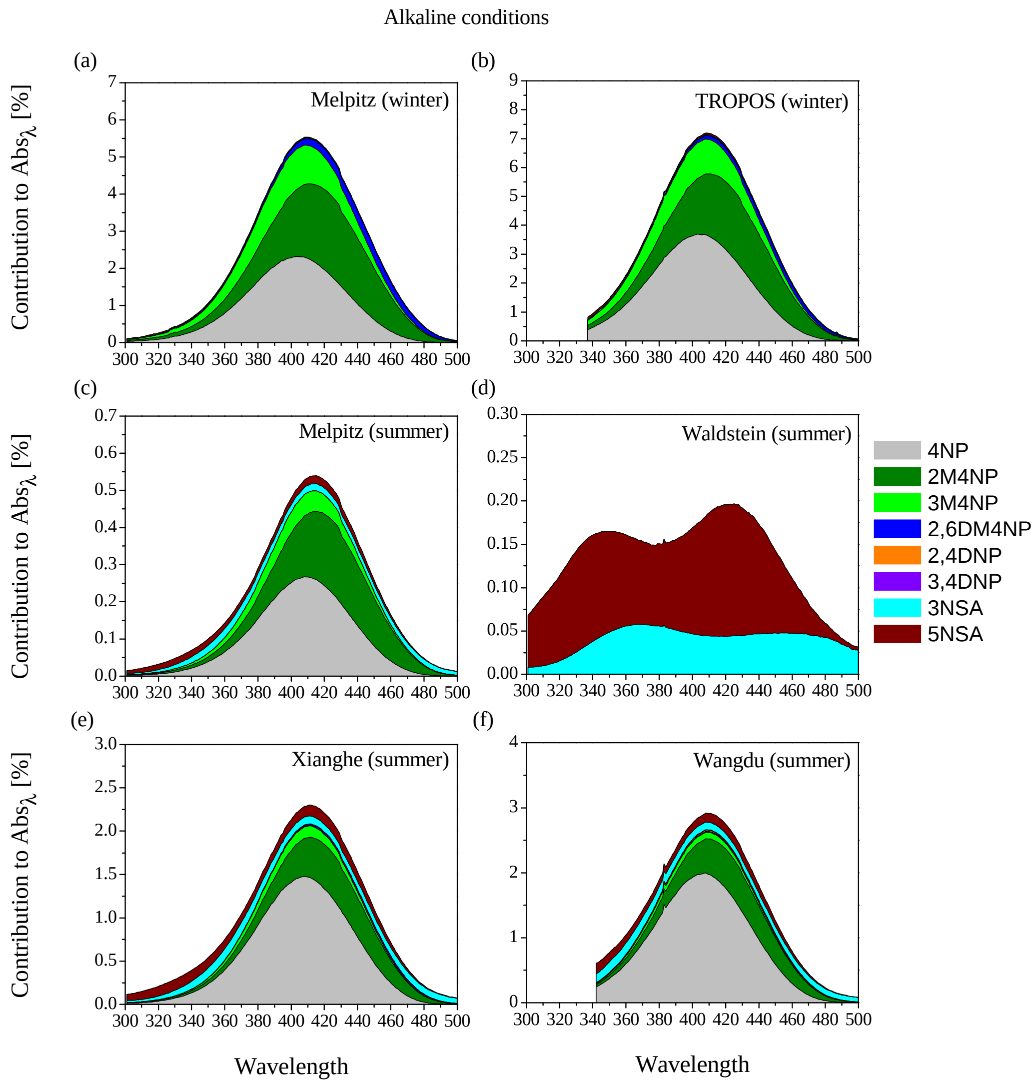


**Figure S6.** Relative contribution of NACs to $Abs_{\lambda}$ over the spectral range of 300 to 500 nm for each measurement campaign (a-f) under alkaline conditions. The data is presented as campaign averages. Due to instrumental issues, data for lower wavelengths is not always available.

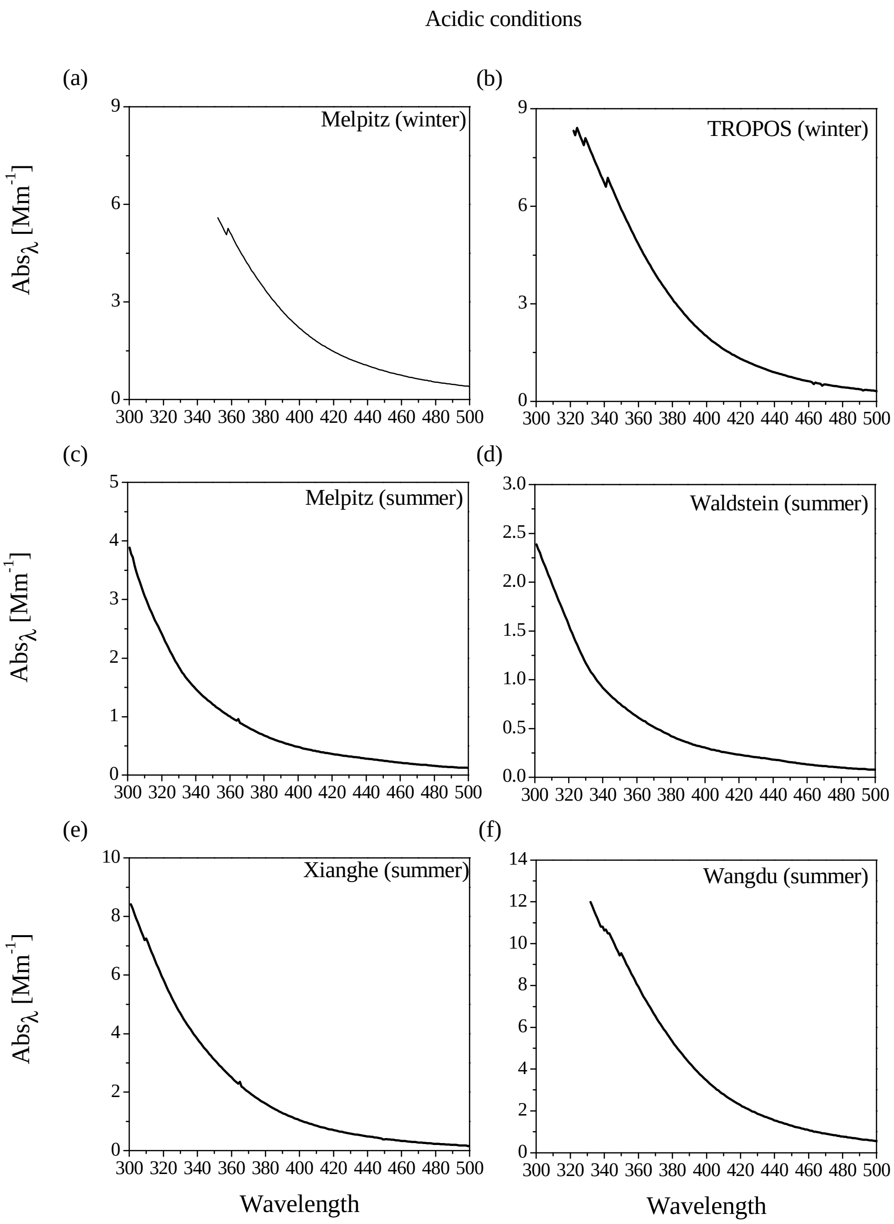


**Figure S7.** Abs$_\lambda$ over the spectral range of 300 to 500 nm for each measurement campaign (a-f) under acidic conditions. The data is presented as campaign averages. Due to instrumental issues, data for lower wavelengths is not always available.

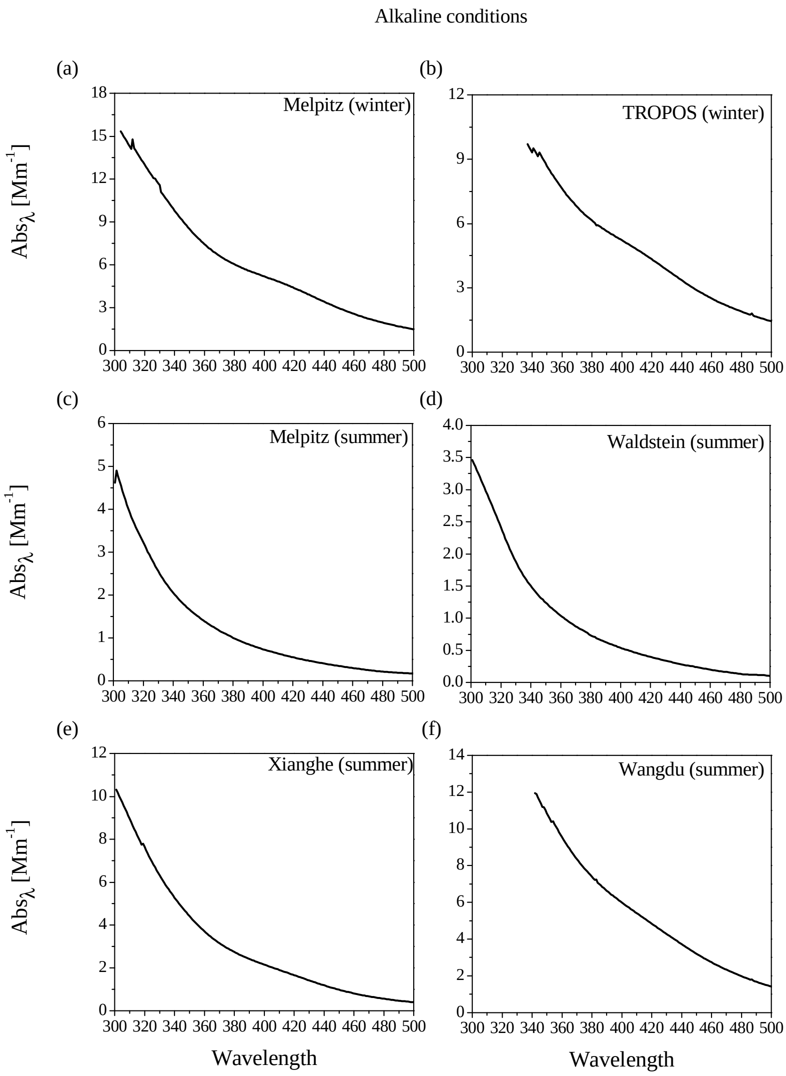


**Figure S8.** $Abs_\lambda$ over the spectral range of 300 to 500 nm for each measurement campaign (a-f) under alkaline conditions. The data is presented as campaign averages. Due to instrumental issues, data for lower wavelengths is not always available.

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