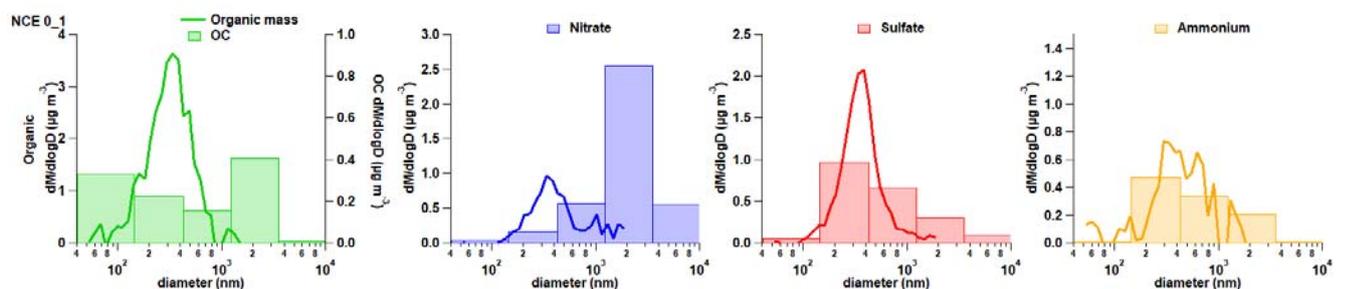


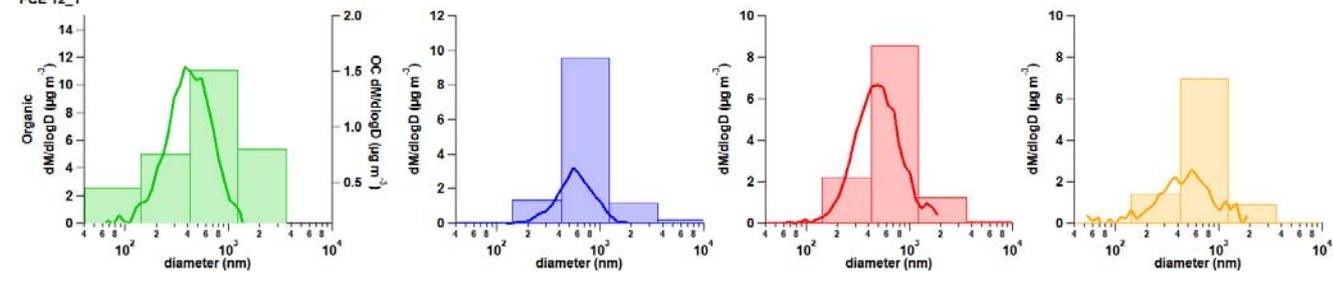
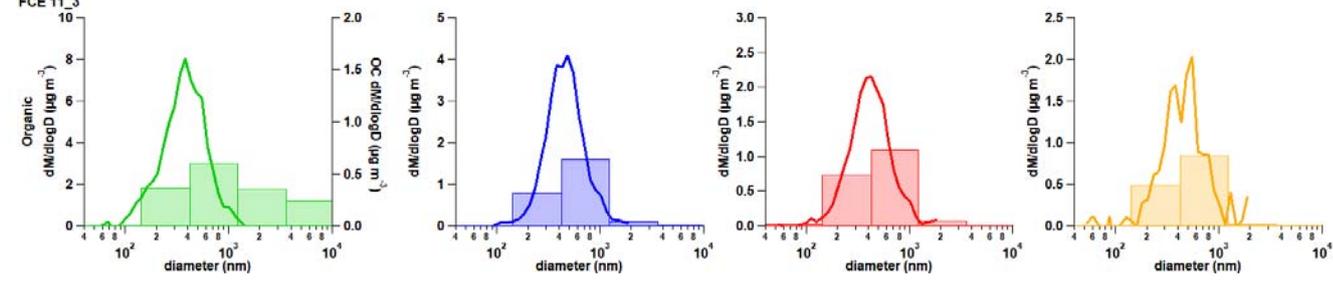
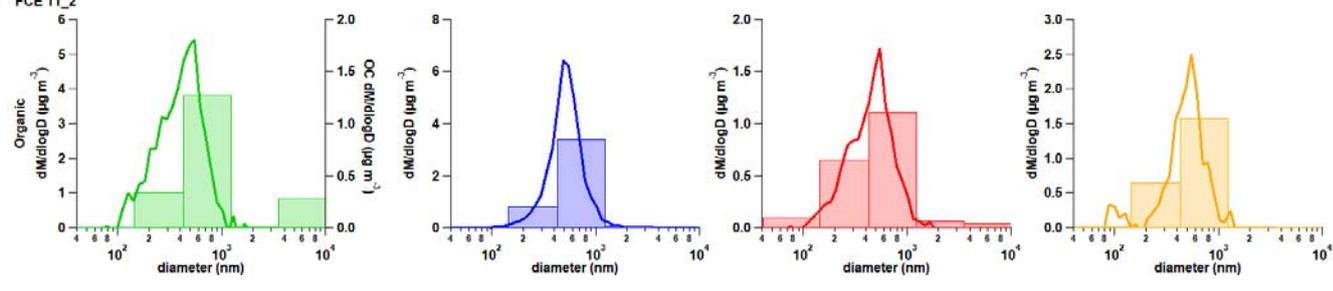
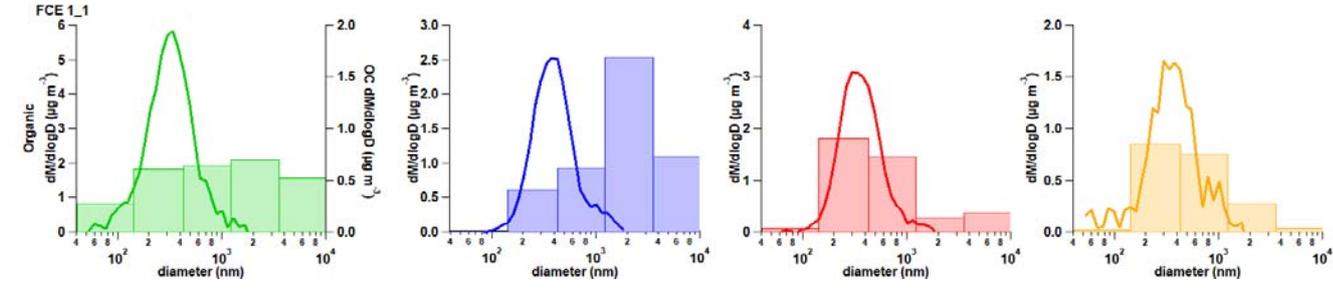
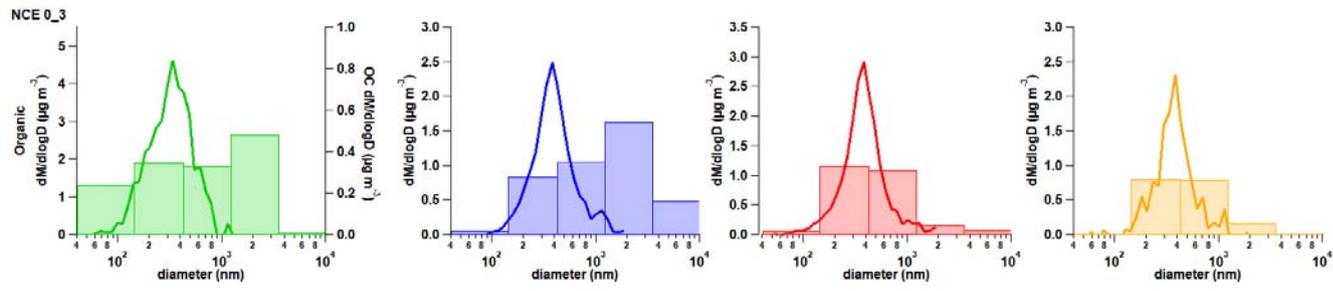
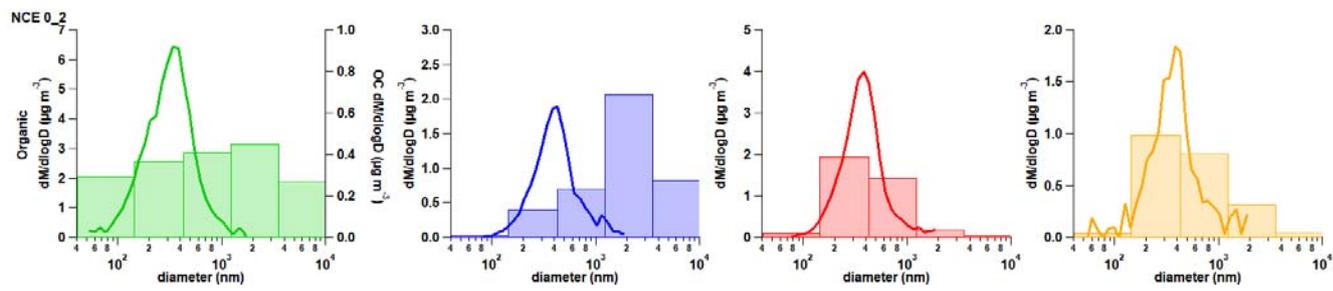
Poulain and coauthors describe a measurement campaign at a forest site in central Germany in 2010 involving the investigation of sources of carbonaceous aerosols using aerosol mass spectrometer (organic and inorganic aerosol) and multi angle absorption photometer (equivalent black carbon) measurements. Particle number-size distributions and light scattering were also measured using a dual mobility particle sizer and a nephelometer, respectively. Although originally chosen as an upwind site for a cloud processing study the dataset is also well suited to source apportionment of local and transported carbonaceous aerosols, which is the focus of this work. Good mass closure was obtained for AMS+MAAP data relative to expected mass concentrations derived using size distribution data and composition-dependent density values. AMS mass concentrations were observed to agree well with supporting on-line measurements (Monitor for AeRosol and Gases in Ambient Air, MARGA) and off-line size-resolved filter analysis (ion chromatography/UV-Vis and OC/EC analysis). Organic aerosol as measured by the AMS was apportioned using PMF/ME-2 constrained with reference mass spectra for hydrocarbon-like organic aerosol (HOA) and biomass burning organic aerosol (BBOA). Five factors were derived: HOA, BBOA and three oxidized organic aerosol factors: semivolatile (SV-OOA), less oxidized (LO-OOA) and more oxidized (MO-OOA). eBC was found to be predominantly associated with longrange transport through multiple linear regression analysis, which is somewhat unexpected, and this observation is supported by the size-dependence of eBC during different air mass periods. Under marine air masses, locally emitted carbonaceous aerosol sources become more important in terms of fractional composition, however continental air masses from the East result in the worst local air quality at the site. Overall, I find the manuscript to be well written and the quantification and apportionment procedures are rigorous and comprehensive. The dependence of aerosol composition on air mass origin is established well and the findings reported here are a useful reference point for central European background sites. I only have a few minor comments.

We would like to thank the referee for his/her constructive comments and suggestions made to improve and clarify our manuscript. Our responses are given below. For clarity, comments from the referee are in black, our responses in blue, and change on the text of the manuscript in **bold blue**.

R1: How do the size distribution shapes for the impactor off-line analysis (Figure 2) agree with the AMS size distributions for nitrate/sulfate/ammonium/OC(or OA)?

A1: Comparison between AMS and Berner impactor size distribution was added to the supplementary information for each FCE and NCE as followed:





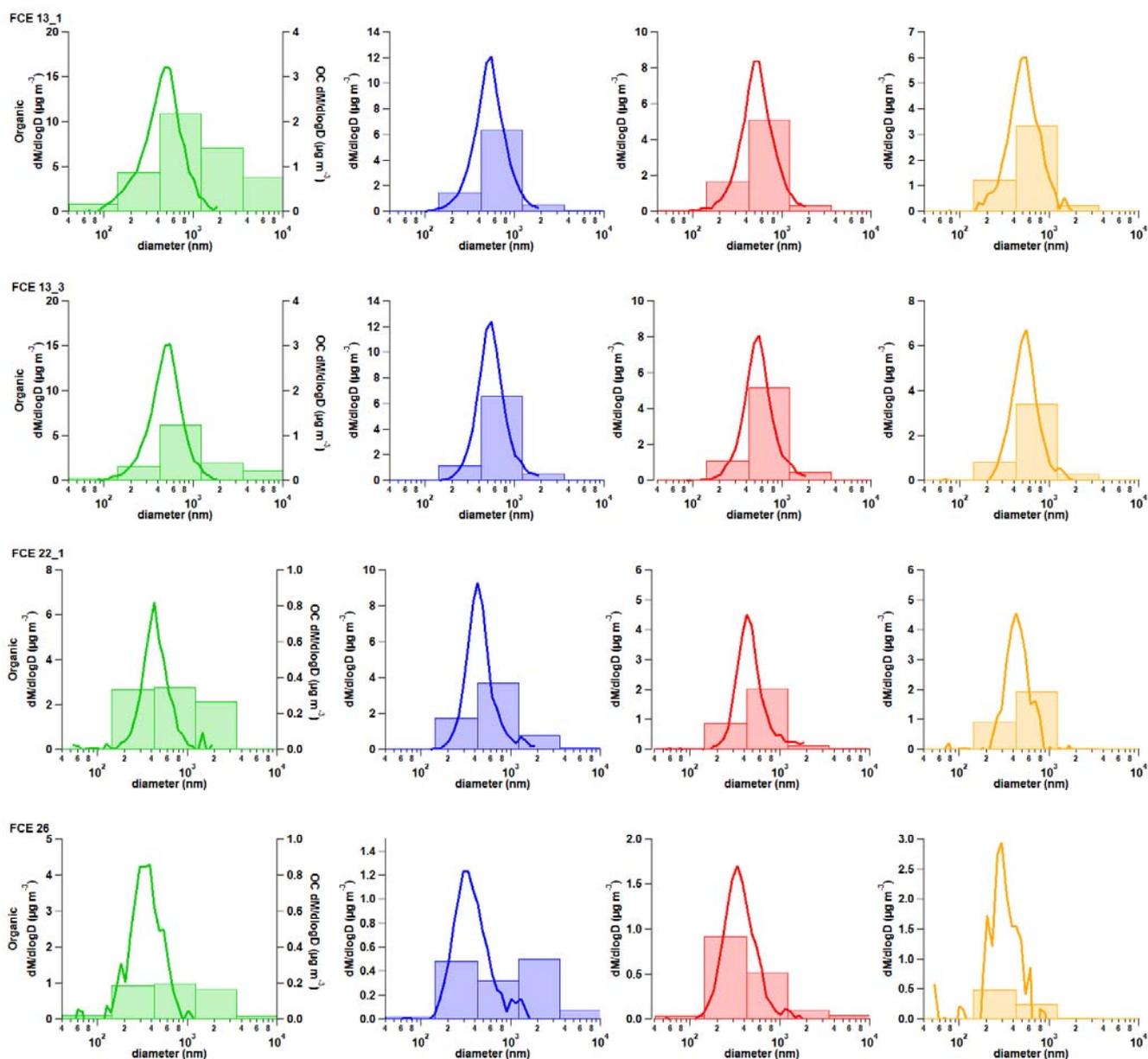


Figure SI-5: Comparison between the AMS and Berner impactor size distribution for organic (organic mass for the AMS and OC for the Berner impactor), nitrate, sulfate, and ammonium for the different FCE and NCE events.

The following text discussing the comparison was included in the supplementary information section SI-3:

The MARGA (PM₁₀, Fig. SI-2 & SI-3) and Berner impactor PM_{1.2} (sum of the three first stages, Fig. SI-4) mass concentration of ammonium, nitrate, and sulfate also present an excellent correlation with the AMS measurements regarding individual instrumental limitations. This includes the limited number of samples and the reduced sampling time of the Berner impactor, as well as the specific upper size cut-off (near-PM₁ for the AMS, PM_{1.2} for the Berner impactor, and PM₁₀ for MARGA). **The size distribution measured by the Berner-impactor and the AMS was compared to each other for organic (Organic mass for the AMS, OC for the Berner impactor), nitrate, sulfate, and ammonium (Fig. SI-5). Similar size distributions were obtained for the inorganic species, except for FCE 12.1 which shows higher nitrate and ammonium mass concentrations on stage 3 compared to the AMS size distribution. A**

comparison between the two organics measurement methods is more difficult since the AMS directly measures the organic mass concentration, while the OC from the Berner impactor was obtained according to the two-step thermographic method (VDI standard). Moreover, the presence of different nitrate salts can also explain the discrepancy between AMS and MARGA (see discussion on main text).

R2:Figure 2 caption define NCE and FCE here also Figure 2: different colour for sodium needed here

A2: The definition of NCE and FCE was added to the figure caption and the color code of the different chemical species (including sodium) was changed.

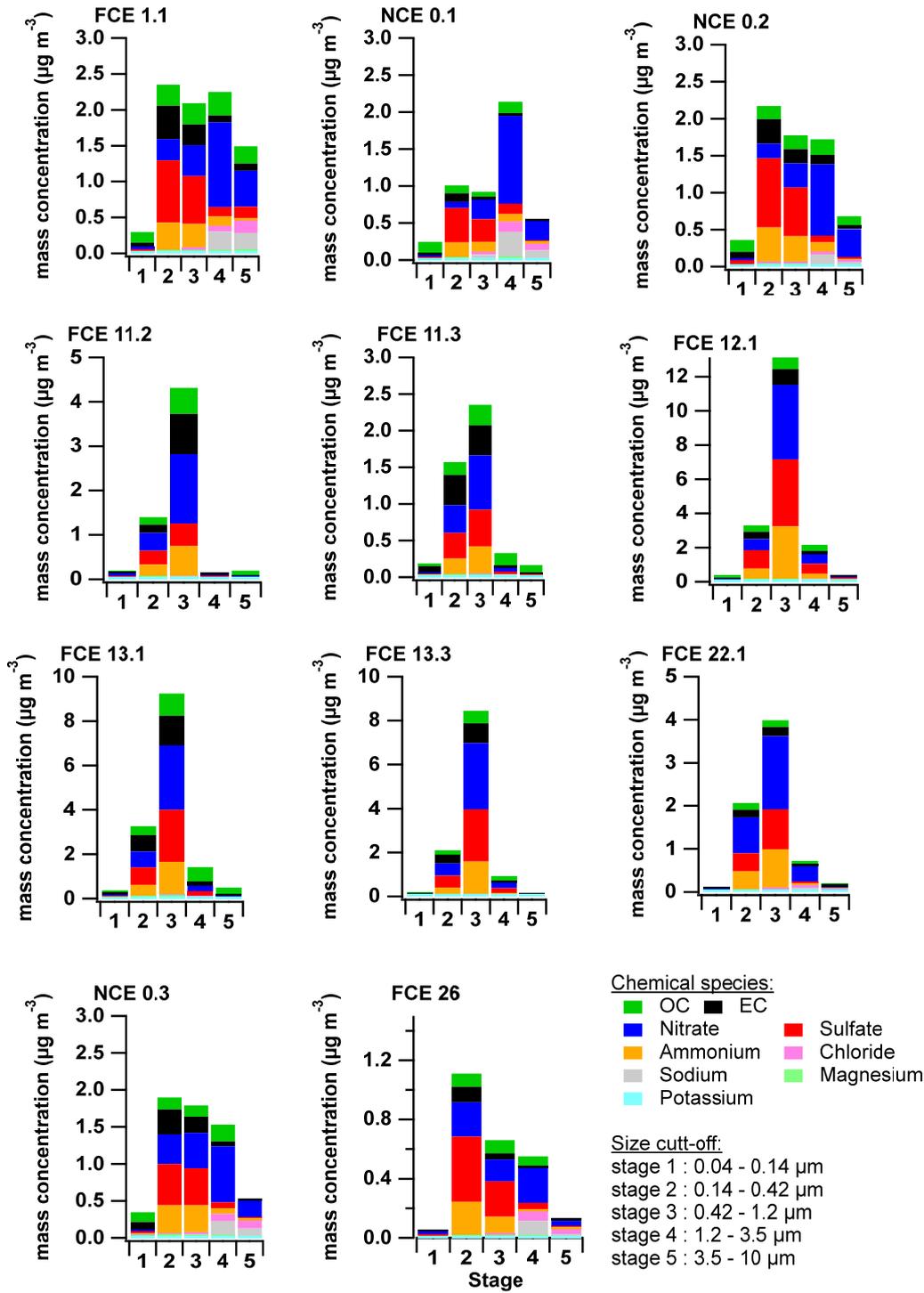


Figure 2: Size distribution of OC, EC, and major water-soluble ions from Berner impactor measurements for the different full cloud events (FCE) and non cloud event (NCE)

R3: Replace size cutting with size cut-off throughout the text

A3: We replaced it in the main text, figures, and supplementary information.

R4 Line 230: The temperature dependence could also be related to less evaporation or oxidation of locally produced vehicle exhaust HOA. Is there more local wind stagnation at lower temperatures? This could also boost the contribution of local vehicle emissions.

A4: Thanks for pointing out this aspect. This is true that during low temperature periods, slower evaporation of the vehicle exhaust, as well as their oxidation, might be expected. Moreover, during the cold period, the wind speed was most of the time below 1 m s^{-1} , which could also contribute to lead higher local emission mass concentration by reducing the dilution effect. Moreover, during the entire campaign, relatively low wind speed was reported ranging up to 5 m s^{-1} .

The text was modified as follows:

This temperature dependency indicates that HOA should be mostly associated with local residential house heating rather than **a significant increase in car emissions. Moreover, the low temperature period can also lead to an artificial increase of the HOA concentration by slowing down the evaporation process of the emitted particles as well as their oxidation processes. Finally, the cold period was also associated with low wind speed and stable stratification (Fig. 1) (Tilgner et al., 2014), which might also contribute to higher concentrations of locally emitted aerosol by reducing both mixing and transport processes.**

R5: Figure 8: add the regression statistics

A5: regression slope and statistics are now included in the figure. The figure caption was modified accordingly.

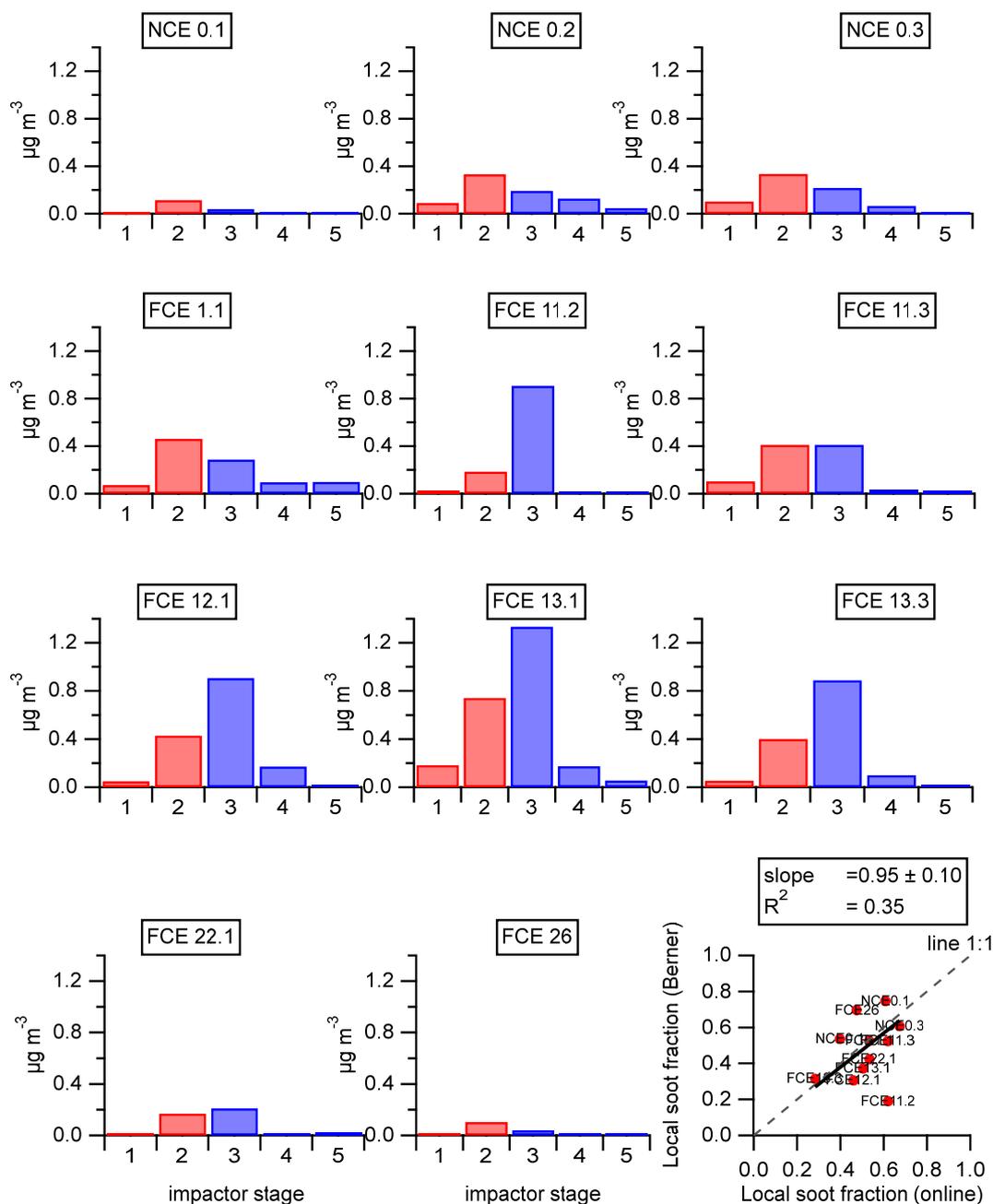


Figure 8: Overview of the EC size distribution measured by the 5-stages Berner impactor. Color corresponds to the following EC classification: red = local and blue = regional/transport. The scatter plot on the bottom right shows the comparison between the local soot fractions estimated using the two different approaches: Berner impactor (y-axis) and on-line multilinear regression (x-axis). **Regression (black line) was made using the least orthogonal distance fit method.**

Reference

Tilgner, A., Schone, L., Brauer, P., van Pinxteren, D., Hoffmann, E., Spindler, G., Styler, S. A., Mertes, S., Birmili, W., Otto, R., Merkel, M., Weinhold, K., Wiedensohler, A., Deneke, H., Schrodner, R., Wolke, R., Schneider, J., Haunold, W., Engel, A., Weber, A., and Herrmann, H.: Comprehensive assessment of meteorological conditions and airflow connectivity during HCCT-2010, *Atmos. Chem. Phys.*, 14, 9105-9128, 10.5194/acp-14-9105-2014, 2014.