Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer based source apportionment using radiocarbon measurements of ambient aerosol

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Black carbon (BC) measured by a multi-wavelength Aethalometer can be apportioned to traffic exhaust and wood burning. The method is based on the differences in the aerosol absorption wavelength dependence originating from these two sources (Sandradewi et al. 2008). This dependence is typically parameterized by the absorption Ångström exponent (α). While the spectral dependence (α values) of the traffic-related BC light absorption is low, wood smoke particles are characterized by enhanced light absorption in the blue and near ultraviolet. Source apportionment results using this methodology (the Aethalometer model) are hence strongly dependent on the α-values assumed for the pure emissions from these two sources (traffic α_TR and wood burning α_WB). Most studies use a single α_TR and α_WB pair in the Aethalometer model, derived from previous work. However, an accurate determination of the source specific α-values is currently lacking, and in some recent publications the applicability of the Aethalometer model was questioned.

We present an indirect methodology for the determination of α_WB and α_TR by comparing BC source apportionment using the Aethalometer model with radiocarbon ^14C measurements of the EC fraction conducted on filters from several locations and campaigns across Switzerland during 2005-2012. The data obtained at eight stations with different source characteristics also enabled the evaluation of the performance and the uncertainties of the Aethalometer model in different environments. The best combination of α_TR and α_WB (0.9 and 1.68, respectively) was obtained by fitting the Aethalometer model outputs against the fossil fraction of EC (EC_F/EC) derived from ^14C measurements. Aethalometer and ^14C source apportionment results are well correlated (r = 0.81) and the fitting residuals exhibit only a minor positive bias of 1.6% and an average precision of 9.3%. This indicates that the Aethalometer model reproduces reasonably well the ^14C results for all stations investigated in this study using our best estimate of a single α_WB and α_TR pair. We also show that α_WB values previously used in literature (~2) result in significant residuals and biases. Therefore we recommend to use the wavelength pair 470 nm and 950 nm and the best α combination as obtained here (α_TR = 0.9 and α_WB = 1.68) in future studies when no or only limited additional information are available.

Figure 1: (a) Comparison between BC_TR/BC and EC_F/EC and (b) residuals of BC_TR/BC compared to EC_F/EC (ΔBC_TR/BC).