Our choice was indeed to focus on the effect of aerosol particle hygroscopic growth on the properties which determine the atmospheric particles interaction with light (particle size, and aerosol absorption and scattering coefficients). This is an essential step before the aerosol radiative forcing can be calculated. We show that at a relative humidity of ca. 90% (which is not unusual) the enhancement factors can reach values close to 2 for scattering, and backscattering coefficients, while it remains close to 1 for absorption. The effect of particle hygroscopic growth on certain optical properties is therefore far from marginal. This means that the aerosol hygroscopicity must be known and accounted for at each atmospheric research station where the measurements of the aerosol properties are performed at room RH or in dry conditions before these dozens of data sets acquired across the world can be used for assessing the current direct aerosol forcing based on experimental data. Thus, the scientific value of the results presented in our manuscript is perhaps not outstanding, but it is certainly high enough in the eyes of scientists dealing with the radiative impact of aerosols, and probably higher than if the manuscript was just discussing the variations in the aerosol hygroscopic growth factor we observed at our station. However, we agree that the number of figures could be reduced, even to present and discuss all the results included in the current version of the manuscript. For instance, the current Fig. 8 of the manuscript, which shows the regression between the values of calculated the aerosol absorption and scattering coefficients derived from measurements and Mie calculations, respectively, is just illustrating that there exists for each data point (hourly average) a “reasonable” complex aerosol refractive index “m” from which the Mie theory can reproduce the aerosol properties derived from measurements. This figure could be deleted, as could also be Figures 3 and 14 of our current manuscript, since corresponding information could be given in the text. Dry refractive indices real parts as high as 1.7, as retrieved for Dec. 2008, could perhaps be explained by a mixture of various components, including fullerenes-like species (m = 1.736 – 0.035i; Lee et al., 2011), but we cannot actually exclude that our hypothesis of an internally mixed aerosol was not verified for all days during that month. Daily measurements of the aerosol (PM2.5) chemical composition performed at our site actually support the predominance of carbonaceous aerosol in our area (Fig. 1), and can well “qualitatively” explain the low GF and single scattering albedo values we observe. Gilardoni et al. (2011) recently showed that the main source of carbonaceous aerosol during winter at our site is wood burning. However, the lack of size-segregated aerosol chemical composition data, and the large uncertainties of the refractive index and hygroscopic growth factor of organic carbon would render the reconstruction of the latter variables based on the available chemistry data quite fuzzy.

All corrections needed to derive the aerosol absorption coefficient based on Aethalometer data were of course taken into account according to methods presented in several previous works, as quoted in our manuscript (Weingartner et al., 2003; Schmid et al., 2006). We tested other Aethalometer data inversion schemes (Arnott
et al., 2005; Virkkula et al., 2007), which did not provide significantly different values for the aerosol absorption coefficient at our site. Furthermore, as already mentioned in our manuscript and illustrated on Fig. 2 below, the corrected Aethalometer data (at 660 nm) are in excellent agreement with the Multi Angle Absorption Photometer (at 670 nm) data. As it was recently shown that the latter instrument (MAAP) “compares excellently with the photoacoustic reference” instrument (Mueller et al., 2011), we think there are pieces of evidence that the accuracy of the aerosol absorption coefficients we used is good enough for our purpose (determining a “reasonable” aerosol refractive index, so that we can perform sound Mie calculations in various RH conditions) and adequately reflected by the uncertainty we attributed to these values [-10, 0 %]. We could certainly include some more discussion on this argument in the manuscript, keeping in mind that none of the aerosol single scattering albedo value we compare our data with was calculated from primary measurements of the aerosol absorption, or from measurements performed with an absorption photometer specifically calibrated against a reference photoacoustic instrument.

The question of the accuracy of the dependence of the aerosol growth factor with RH is probably more critical. As suggested by Referee # 1, we fitted “gamma law” curves (GF = (1-RH)^γ) to humidograms of atmospheric aerosols performed in contrasted conditions (before and after a rain event). Both comparisons (Fig. 3) suggest that the “gamma law” can adequately represent the variation of the aerosol hygroscopic growth factor with RH at our site. It is certainly worth mentioning these comparisons in a revised version of our manuscript.

References


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Fig. 1: mean PM$_{2.5}$ composition in Ispra

Figure 2: comparison of aerosol light absorption coefficient data derived from Multi Angle Absorption Photometer and from Aethalometer plus Nephelometer measurements (Ispra, 2008).
Fig. 3: Measured humidograms on 12.06.2008. The black and red circles represent the measurements taken before and after the rain started, respectively.