Response to Paul Zieger:

We would like to thank Dr. Zieger for the useful comments. We have responded to each specific comment in detail and will update the manuscript according to his suggestions, which we believe has helped to strengthen and clarify the study. Comments are in blue and italics, and our responses are in black.

Titos et al. present and discuss in their manuscript the results of a measurement campaign at Cape Cod, Massachusetts, where aerosol optical measurements were measured for one year. The focus is the scattering enhancement factor f(RH) which is defined as the aerosol particle scattering coefficient at enhanced relative humidity (RH) divided by its dry value. This parameter was measured by a humidified nephelometer system for approx. 7 months within this year. f(RH) was analysed with regard to air mass origin and compared to other aerosol optical parameters like the single scattering albedo (SSA) and Ångström parameter. A parametrization is being proposed which allows estimation of f(RH) using the SSA as a proxy.

I have read this paper with great interest and would like to share some comments (and questions) to further improve the quality of this manuscript:

• The authors use a two-parameter equation to parametrize the measured humidograms of f(RH), but focus only on the discussion of the γ parameter and ignore the intercept a. How did it vary for the different air masses? I would guess that a > 1 during sea salt periods, or? Looking at Fig. 7, I have the impression that all measured humidograms are slightly biased towards larger values at low RH (where it should, ideally, reach 1). Could this be an effect of the slight disagreement between the two nephelometers at dry conditions?

The "a" parameter was used to recalculate f(RH) values measured at different RH to RH=80% using equation 3 (equation 6 in the revised manuscript). The focus was kept in the γ parameter since it parameterizes the magnitude of the scattering enhancement, which is the main objective of this study. Performing the fitting for RH>40%, a mean value of a = 0.85±0.15 for the whole campaign was obtained, which is in agreement with the value of a = 0.9±0.1 reported by Zieger et al. (2014). The "a" parameter varies with the aerosol transmission efficiency through the humidifier and goodness of the power law fit. Note also that slight differences between the experimental and ideal value of "a" are expected since the "a" value will depend also on aerosol losses in the dry nephelometer and in the humidifier system.

Zieger, P., R. Fierz-Schmidhauser, L. Poulain, T. Müller, W. Birmili, G. Spindler, A. Wiedensohler, U. Baltensperger, and E. Weingartner. 2014. Influence of water uptake on the aerosol particle light scattering coefficients of the Central European aerosol. Tellus B, 66, 22716, http://dx.doi.org/10.3402/tellusb.v66.22716.

• Page 3367, Line 4: Was the difference between the two nephelometers accounted for when calculating f(RH)? Could the authors speculate on why the agreement is much better for PM1 compared to the PM10?

No, the difference was not accounted when calculating f(RH). The difference between both nephelometers might be ascribed to sampling losses in the dry nephelometer and in the humidifier system. Many authors correct the humidified nephelometer measurements by applying an empirical correction factor (the slope of the regression between dry and humidified nephelometers). This correction factor will not affect the gamma parameter which is the main objective of this work but it will modify the "a" parameter. In any case, we would not say that the agreement was much better for PM_1 than for PM_{10}. The slope of the regression was 0.971 ± 0.004 for PM_1 and 1.073 ± 0.001 for PM_{10} but the intercepts differ also for each size fraction. When fitting through zero, the differences between both slopes were lower than 4%. In addition, there are much data dispersion for PM_1 ($R^2 = 0.77$) than for PM_{10} ($R^2=0.99$).

• If I am correct, it should be mentioned in the revised manuscript that the applied humidified nephelometer set-up will only capture the lower branch of the hysteresis curve and will miss the upper branch because no active drying (keeping the humidifier on maximum) is performed before the particles reach the second nephelometer (see Fierz-Schmidhauser et al., 2010).

We agree with Dr. Zieger in that our humidified nephelometer system can only capture the lower branch of the hysteresis curve. We will include this information in the revised manuscript.

• Page 3370, Line 24 and Sect. 4.2: We have made an interesting observation in the Arctic (Zieger et al., 2010) of compensating effects between size and aerosol hygroscopicity. At the beginning of the campaign we had mainly small and less hygroscopic particles compared to the end where large but more hygroscopic particles (mainly sea salt) led to the same magnitude of f(RH). Can this maybe also be seen in your data set, when e.g. comparing size distribution parameters to f(RH)?

We observed that when small anthropogenic particles predominated (large SAE values) the particles were less hygroscopic while when coarse particles predominated (small SAE values) the particles were more hygroscopic and the results suggest that the aerosol was dominated by sea salt particles (see Figures 8 and 9). Thus, γ decreased as the SAE increased (the correlation coefficient of γ versus SAE was R = -0.77). This result contrasts with the result of Zieger et al. (2010) that showed a decrease of γ for an increase in the contribution of coarse particles, probably connected with the compensating effects between different varying aerosol properties discussed in their work. This information and the corresponding reference will be included in the revised manuscript.

The interesting observation in our study is that we took advantage of the size segregated f(RH) data and we found that fine sea salt particles were more hygroscopic than coarse sea salt particles (see Figure 3 of the manuscript). This could be due to the large increase in f(RH) of NaCl with decreasing particle size as Zieger et al. (2013) also showed. On the other hand, this behavior might be also due to a shift in the size distribution to a scattering regime with a higher scattering efficiency when the SAE is greater than 1. We have strengthened the discussion of Figure 3 in order to include this information.

In our case, the hysteresis index as introduced in Zieger et al. (2010) did not correlate with the scattering Ångström exponent neither with the sub-micrometer scattering factor, $R_{sp}(\lambda) = \sigma_{sp(Dp < 1\ \mu m)}(\lambda)/\ \sigma_{sp(Dp < 10\ \mu m)}(\lambda)$. Due to the lack of correlation, we did not introduce the hysteresis index and discuss it in the manuscript.

• Sect. 4.4: I think the limitations of the proposed parametrization should be further discussed. It might be true that a simple site-specific proxy can be found to predict f(RH) at Cape Cod, but the same proxy can fail for another (even marine) site. We have done a sensitivity analysis to exactly address this question (see Sect. 6.3 in Zieger et al, 2013) because it was not possible to find one simple parametrization for all different analysed aerosol types. A reliable prediction, especially for climate models, will always need a full determination of the particle number size distribution (fine and coarse mode) and information on the chemical composition or particle hygroscopicity.

We agree with Dr. Zieger in that for a reliable prediction of the aerosol scattering enhancement using Mie theory, the particle number size distribution and the chemical composition are needed. However, that approach also has some disadvantages. For example, for predicting f(RH) using Mie theory, strong assumptions are usually made like spherical and homogenous internally mixed particles, constant refractive index, etc., which led to an overall relative uncertainty in the calculated f(RH) up to 25 % as reported in Zieger et al. (2013). In addition, at least two instruments are needed for measuring the whole size distribution plus instrumentation to measure the chemical composition. This set-up is not as common as it would be desirable, so the global coverage is reduced by using these measurements.

Using the single scattering albedo and/or scattering Angström exponent as estimators for the f(RH) has the advantage that those measurements are widely performed in GAW and ACTRIS networks. However, we agree that the parameterization has limitations. We are aware that this particular study had a strong covariance between SSA and SAE, which allowed a reduction in the γ fit to a single parameter and that this relationship is only valid for the TCAP data. In this particular study, the coarse mode was predominantly dominated by sea salt particles and the presence of other species that typically accumulate in the coarse fraction like dust particles was negligible. Due to the similar characteristics of sea salt and dust particles in terms of SAE and SSA, but strong difference in the hygroscopic behavior, the parameterization proposed in this study would fail under the presence of both types of aerosols as dust does not experience significant hygroscopic growth. Cape Cod may be considered as representative of an aerosol from the Northern Atlantic coast with anthropogenic influence. The same analysis needs to be applied to other regions and aerosol types to catalog exponential fit parameters of γ versus SSA over a variety of aerosol types and atmospheric conditions. In this sense, preliminary analysis to check the validity of this parameterization at different sites (pristine and polluted marine, anthropogenic, rural, desert and forest sites) suggest that the model agrees well with the experimental measurements for

all sites except for the desert site (Titos et al., 2014). The limitations of the proposed parameterization will be further discussed in the revised manuscript.

Titos, G., Jefferson, A., Sheridan, P. J., Andrews, E., Lyamani, H., Ogren, J. A. and Alados-Arboledas, L.: Estimating aerosol light-scattering enhancement from dry aerosol optical properties at different sites, European Geoscience Union Assembly, Vienna, 2014.

• How should a be treated within the proposed parametrization?

The parameter "a" is not included in the parameterization so the equation proposed can be used to estimate the γ parameter but not "a". The "a" parameter would act as a normalization factor but the scattering enhancement characterized by the γ parameter will not be affected.

• Page 3363, Line 17: The authors should also mention recent studies of Zieger et al. (2011, 2012) where humidified nephelometer measurements were explicitly performed to validate or compare remote sensing measurements of the aerosol extinction coefficient with in-situ measurements at ambient conditions.

We will include the aforementioned references.

• Page 3364, Line 11: The longest campaign was actually 4 months long (Cabauw).

We apologize for the mistake; 3 months will be replaced by 4 months in the revised manuscript.

• Fierz-Schmidhauser et al. (2010) also performed PM10 and PM1 measurements of f(RH) (actually together with the humidograph system of DOE/ARM).

We will include the reference in the revised manuscript although the intention here was only to mention a few examples of papers that reported scattering enhancement factors in PM_1 and PM_{10}. The paper of Fierz-Schmidhauser et al. (2010) was not initially listed here because the paper is mainly focused on the comparison of both humidifier systems and not in the characterization of aerosol hygroscopicity in PM_{10} and PM_1 size fractions.

• Page 3366, Line 7: Could you state the mean and standard deviation of temperature and relative humidity within the dry nephelometer?

The mean ± standard deviation of the temperature and relative humidity within the dry nephelometer for the whole measurement campaign were T = 26±4 ºC and RH = 30±13 %. It is important to note that, as stated in Page 3368-line 21, for the calculation of f(RH) and γ parameter only cases when the RH inside the dry nephelometer was below 40% were used. This information will be included in the revised manuscript.

References

Fierz-Schmidhauser R., Zieger P., Wehrle G., Jefferson A., Ogren J., Baltensperger U., and Weingartner E., Measurement of relative humidity dependent light scattering of aerosols, Atmos. Meas. Tech., 3(1), 39–50, doi:10.5194/amt-3-39-2010, 2010.

Zieger P., Fierz-Schmidhauser R., Weingartner E., and Baltensperger U., Effects of relative humidity on aerosol light scattering: results from different European sites, Atmos. Chem. Phys., 13(21), 10609–10631, doi:10.5194/acp-13-10609-2013, 2013.

Zieger P., Kienast-Sjögren E., Starace M., v. Bismarck J., Bukowiecki N., Baltensperger U., Wienhold F., Peter T., Ruhtz T., Collaud Coen M., Vuilleumier L., Maier O., Emili E., Popp C., and Weingartner E., Spatial variation of aerosol optical properties around the high-alpine site Jungfraujoch (3580 m a.s.l.), Atmos. Chem. Phys., 12, 7231–7249, doi: 10.5194/acp-12-7231-2012, 2012.

Zieger P., Weingartner E., Henzing J., Moerman M., de Leeuw G., Mikkilä J., Ehn M., Petäjä T., Clémer K., van Roozendael M., Yilmaz S., Frieß U., Irie H., Wagner T., Shaiganfar R., Beirle S., Apituley A., Wilson K., and Baltensperger U., Comparison of ambient aerosol extinction coefficients obtained from in-situ, MAX-DOAS and LIDAR measurements at Cabauw, Atmos. Chem. Phys., 11(6), 2603–2624, doi:10.5194/acp-11-2603-2011, 2011.

Zieger P., Fierz-Schmidhauser R., Gysel M., Ström J., Henne S., Yttri K., Baltensperger U., and Weingartner E., Effects of relative humidity on aerosol light scattering in the Arctic, Atmos. Chem. Phys., 10(8), 3875–3890, doi:10.5194/acp-10-3875-2010, 2010.