Response to comments of referee #2

General Comments:

Reviewer: This manuscript provides a 5 year long time series of atmospheric aerosol optical properties data in Central Europe. This work could act as a reference for presenting long time series of aerosol optical properties, uncertainties of the measurements, quality control and air mass origin dependence. Therefore it should be published after only few minor revisions. As a general comment I would like to see a discussion about diurnal variations of the parameters discussed in the manuscript.

Response:

Thanks for the comments.

As suggested by the reviewer, a section has been added in the manuscript to discuss the diurnal variations of aerosol optical properties. As shown below:

"3.5. Diurnal cycles of aerosol optical properties

Figure 8 shows the average diurnal cycles of dry aerosol optical properties, the volume fraction and effective radius (r_{eff}) of submicron aerosol in warm season (Apr. to Sep.) and cold season (Oct. to Mar.) measured in Melpitz. Values of percentiles are not shown to facilitate better visualization of the data. Basically, all parameters show some diurnal patterns, with lower contrast compared with their annual cycles.

The σ_{sp} and σ_{ap} shows similar diurnal patterns, with high values during night and low values during daytime. This can be mainly attributed to the diurnal variation of the boundary layer height. The developing of boundary layer during daytime causes a vertical mixing thus dilutes the pollutant near the surface. During night, new emitted aerosol accumulates in the shallow nocturnal boundary layer. σ_{sp} and σ_{ap} therefore remain at a relative high level. This effect is more active in warm season due to surface heating, thus causes a higher contrast between the values in daytime and nighttime than in cold season. The diurnal variation of aerosol emission is also important to explain the diurnal pattern of σ_{sp} and σ_{ap}, especially in cold season. σ_{ap} shows a evident maxima around 21:00 in cold season, might stem from the increase of BC emission from domestic heating at night. The high values of σ_{sp} and σ_{ap} in the morning might also be a reflection of the beginning of anthropogenic activities in the region.

Recalling the definition of ω, the diurnal variation of ω is due to the phase difference between the diurnal cycles of σ_{sp} and σ_{ap}. Corresponding to the two peaks of σ_{ap} in the morning and evening, two evident decrease of ω can be found in the diurnal pattern of both σ_{sp} and σ_{ap}. In warm season, the diurnal pattern of σ_{sp} and σ_{ap} are similar, the ω therefore shows less diurnal variability than in cold season. ω in warm season shows a high level between 09:00 and 18:00, mainly stemming from the enhancement of light-scattering aerosol from secondary aerosol formation and aging processes, which can be also found in the diurnal variation of aerosol number size distribution (fig. 6).

The diurnal variation of b is mainly attributed to the diurnal variation of aerosol number size distribution. In warm season, as shown in fig. 6, the new particle formation begins at around 09:00, thus causes a decrease in the effective radius for submicron aerosol, as shown in fig 8(H). After the nucleation, the new formatted particles grow in size and decrease in number concentration via coagulation and condensation processes, and cause a continually increase of the effective radius for submicron aerosol. According to Collaud Coen et al. (2007), decrease in aerosol mean size may increase b. Therefore, compared with the diurnal variation of the effective radius for submicron aerosol, a reverse pattern is shown for b. In cold season, the secondary aerosol production and aging processes is less active, both b and the effective radius therefore thus show less diurnal variability.

$\alpha_{450\text{-}550nm}$ and $\alpha_{550\text{-}700nm}$ show similar diurnal patterns. Similar as b, α is also determined by the shape of aerosol number size distribution. As mentioned in section 3.4, α smaller than 1 indicates that the size distribution is dominated by coarse mode, and α larger than 2 indicates that fine mode particles dominates the size distribution (Eck et al., 1999; Westphal and Toon, 1991). Schuster et al. (2006) found that α is sensitive to the effective radius of submicron aerosol and the volume fraction of submicron aerosol to total aerosol. In warm season, α decreases with the decrease of volume fraction of submicron aerosol and the increase of effective radius in the early morning. After 09:00, α begins to increase with the decrease of effective radius and remains at a relative stable level in the afternoon. After 18:00, α increases again with the increase of volume fraction of submicron aerosol. In cold season, such pattern can also be seen but with less contrast."

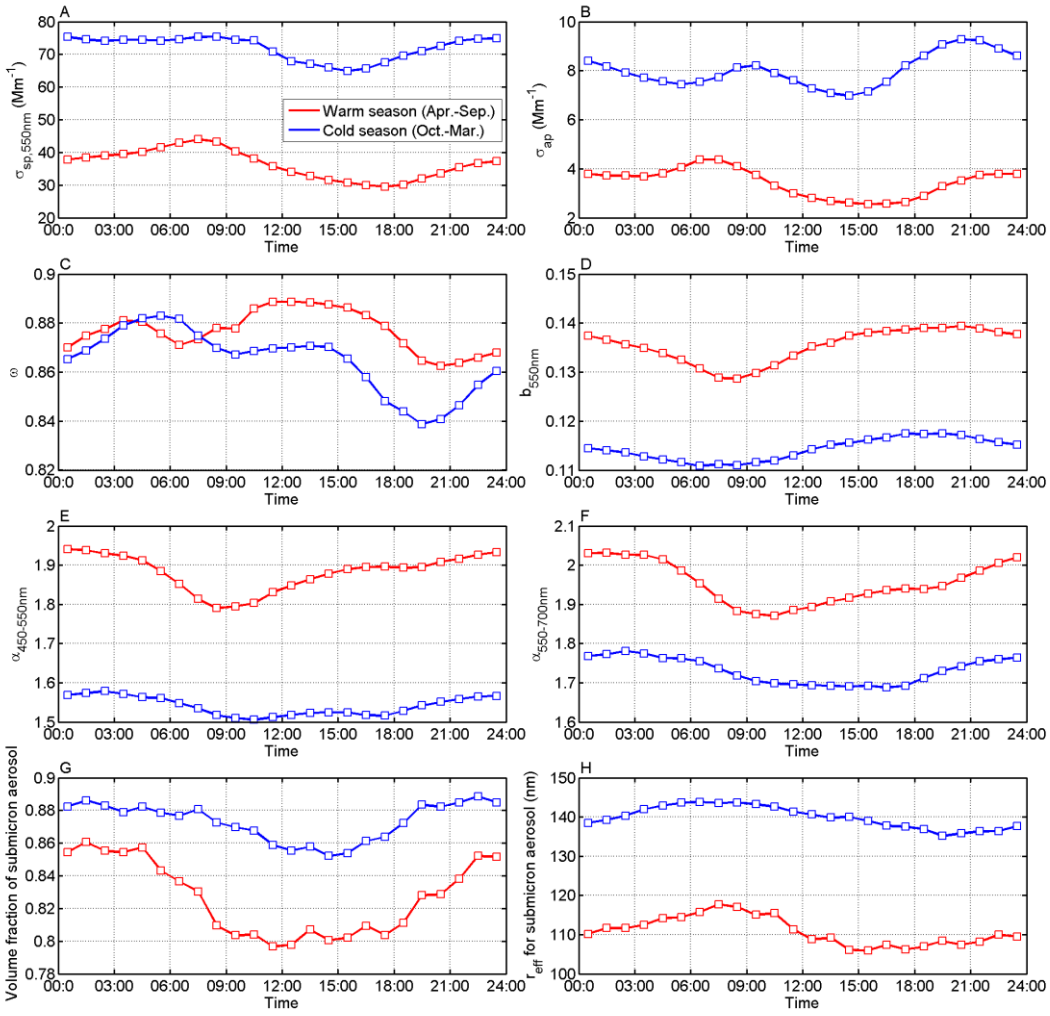

Figure 8. Average diurnal variations of aerosol scattering coefficient (A), absorption coefficient (B), single scattering albedo (C), hemispheric backscattering fraction (D), Ångström exponent at 450-550 nm (E) and at 550-700 nm (F), volume fraction of submicron aerosol (G), and the effective radius of submicron aerosol. For each panel, the red and blue lines represent the results of warm (Apr.-Sep.) and cold season (Oct.-Mar.), respectively.

Specific Comments:

1. Reviewer:
Page 27813, Line 4: Update reference to IPCC 2013 (-0.27 and -0.55 Wm-2)
Response:
Thanks for the correction. The corresponding sentence has been revised as:
"The effective radiative forcing caused by these two effects is estimated as -0.45 Wm-2 and -0.45 Wm-2, respectively, with their uncertainties being the largest among all climate forcing factors (IPCC, 2013)."

2. Reviewer:
Page 27815, Line 5: In Table 1 PM10 values are reported and the corresponding measuring methodology should be described.
Response:
The description of corresponding measuring methodology of PM10 has been added in the last paragraph of section 2.3, as follows:
"As a reference, the average PM_{10} mass concentrations at 53 rural background sites in Germany is also shown in table 1. PM_{10} mass concentrations in Germany are recorded on a continuous basis by federal and regional government air quality monitoring networks Graphical and tabular versions can be assessed on a website of Germany's Federal Environment Agency UBA (http://www.umweltbundesamt.de/). Depending on each individual regional environment agency PM_{10} might be measured by a different method, including the gravimetric reference method, TEOMs, beta-gauges, and optical particle counters. Final versions of the data, however, are always corrected to the levels of the gravimetric reference method (CEN, 1998) involving 24 h filter sampling."

3. Reviewer:
Page 27815, Line 26: Why have these numbers been chosen? An explanation or at least a reference would be useful
Response:
The sentence "The particle densities for fine and coarse mode were assumed to be 1.6 gcm−3 and 2.5 gcm−3, and the shape factors as 1.0 and 1.26, respectively." has been revised as:
"According to McMurry et al. (2002), the fine mode particles were assumed as spherical (shape factor =1), with a density of 1.6 g cm^{-3}. The coarse mode particles were assumed to be mainly mineral dust. According to Kaaden et al. (2009), the shape factor and density were assumed to be 1.26 and 2.5 g cm^{-3}, respectively."

4. Reviewer:
Page 27818, Line 25: During night the stability of the atmosphere is quite different than the day. The approach the authors follow is representative for the day but not for the night, as stated in the following sentence there is usually a development in the boundary layer. Some sensitivity test would be useful or at least stating it in the text.
Response:
It is true that our used classification scheme (BCLM) refers to mixed layer conditions during day-time (12:00 UTC = 13:00 local time). It is certainly a limitation of this method that classification is only available for every 24 h. Nevertheless, we refrained from using a higher time resolution because this would inevitably compare data sets recorded at different times of the day.

To account for your suggestion, we plotted additional diurnal cycles of aerosol optical parameters, embedded in an entirely new section (3.5) of text. Here, the night-time values will almost always correspond to situations of stable stratification, while the day-time values will correspond to the air mass type indicated in the BCLM scheme.

5. Reviewer:

Page 27825, Line 18: There is a more recent study for Finokalia station with longer time series, Kalivitis et al., Atmospheric Research, 2011.

Response:

Following reviewer#1's comment 10, the comparison between aerosol scattering coefficient measured at Melpitz and Finokalia has been removed.

6. Reviewer:

Page 27827, Line 5: As already stated in the previous paragraph during winter the emissions from combustion sources are higher and this should be reflected in ω in the region and therefore this must be the reason for the ω drop.

Response:

Thanks for this comment. To clear this point, sentences below have been added in the corresponding paragraph:

"In winter, the local emission of black carbon is higher due to a larger amount of fossil fuel combustion. It was also found that the residential wood burning during the cold seasons has a significant contribution on the elemental carbon in Europe (Genberg et al., 2013). The higher emission of BC and less activity of aerosol aging and secondary aerosol formation causes a lower ω in winter."

7. Reviewer:

Page 27826, paragraph 3.4: Accumulated precipitation is higher in the summer months rather than the winter ones in the region under study and the rain/wash out processes should also be taken into account to interpret the data series.

Response:

Thanks for this comment. As pointed out by the reviewer, based on the auto weather station data, it was found that the accumulated precipitation in summer is approximately two times as that in winter. The precipitation scavenging of aerosol might also be a reason for the diurnal variation of σ_{sp} and σ_{ap}. Therefore, we added some discussion in the corresponding paragraph, as shown below:

Precipitation scavenging is an important way to clean aerosol particles out of the atmosphere (Buat-Ménard and Duce, 1986; Jaffrezo and Colin, 1988). The precipitation measured near Melpitz shows that the accumulated precipitation in summer is approximately two times as that in winter. The stronger precipitation scavenging of aerosol particles may also be an important reason of the low level of σ_{sp} and σ_{ap} in summer.

<u>Specific Comments:</u>

Reviewer:

Page 27816, Line 1: "Estimatie", change to "estimate".

Page 27816, Line 12: "the the", change to "the".

Page 27824, Line 9: "in Fig.2", missing bracket.

Page 27824, Line 10: “in Fig.2”, missing bracket.
Page 27831, Line 15: “Results shows”, change to show.
Page 27832, Line 12: “showeda”, change to showed a.

Response:

Thanks for these corrections. These typos have been corrected.

Reference

Buat-Ménard, P. and Duce, R. A.: Precipitation scavenging of aerosol particles over remote marine regions, Nature, 321, 508-510, 1986.

CEN (1998): EN 12341, Air quality - Determination of the PM 10 fraction of suspended particulate matter - Reference method and field test procedure to demonstrate reference equivalence of measurement methods. CEN/TC 264 - Air quality, 1998.

Collaud Coen, M., Weingartner, E., Nyeki, S., Cozic, J., Henning, S., Verheggen, B., Gehrig, R., Baltensperger, U.: Long-term trend analysis of aerosol variables at the high-alpine site Jungfraujoch, J. Geophys. Res., 112, D13213, doi:10.1029/2006JD007995, 2007.

Eck, T., Holben, B. N., Reid, J., Dubovik, O., Smirnov, A., O’Neill, N., Slutsker, I., Kinne, S.: Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols, J. Geophys. Res., 104(D24), 31333– 31349, 1999.

Genberg, J., Denier van der Gon, H. A. C., Simpson, D., Swietlicki, E., Areskoug, H., Beddows, D., Ceburnis, D., Fiebig, M., Hansson, H. C., Harrison, R. M., Jennings, S. G., Saarikoski, S., Spindler, G., Visschedijk, A. J. H., Wiedensohler, A., Yttri, K. E., and Bergström, R.: Light-absorbing carbon in Europe – measurement and modelling, with a focus on residential wood combustion emissions, Atmos. Chem. Phys., 13, 8719-8738, doi:10.5194/acp-13-8719-2013, 2013.

IPCC 2013: Myhre, G., D. Shindell, F.-M. Bréon, W. Collins, J. Fuglestvedt, J. Huang, D. Koch, J.-F. Lamarque, D. Lee, B. Mendoza, T. Nakajima, A. Robock, G. Stephens, T. Takemura and H. Zhang, 2013: Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Jaffrezo, J. L. and Colin, J. L.: Rain-aerosol coupling in urban area: scavenging ratio measurement and identification of some transfer processes, Atmos. Environ., 22(5), 929-935, 1988.

Kaaden, N., Massling, A., Schladitz, A., Müller, T., Kandler, K., Schütz, L., Weinzierl, B., Petzold, A., Tesche, M., Leinert, S., Deutscher, C., Ebert, M., Weinbruch, S., Wiedensohler A.: State of mixing, shape factor, number size distribution, and hygroscopic growth of the Saharan anthropogenic and mineral dust aerosol at Tinfou, Morocco, Tellus B, 61(1), 51-63, 2009.

McMurry, P. H., Wang, X., Park, K., Ehara, K.: The relationship between mass and mobility for atmospheric particles: A new technique for measuring particle density, Aerosol Sci. Tech., 36(2), 227-238, 2002.

Schuster, G. L., Dubovik, O., Holben, B. N.: Ångström exponent and bimodal aerosol size distributions, J. Geophys. Res., 111(D07207), 2006.

Westphal, D., and Toon, O.: Simulations of microphysical, radiative, and dynamical processes in a continental-scale forest fire smoke plume, J. Geophys. Res., 96(D12), 22379–22400, 1991.