

Interactive comment on "Four-year long-path monitoring of ambient aerosol extinction at a central European urban site: dependence on relative humidity" by A. Skupin et al.

Anonymous Referee #2

Received and published: 4 June 2015

This study summarizes measurements of aerosol extinction coefficient using an openpath spectral technique at the urban Leipzig, Germany site. Analysis takes advantage of significant daily diurnal variability in ambient relative humidity (RH), by calculating hygroscopic enhancement factors at high and low RH. This technique is advantageous in that it is independent of inlet artifacts and the uncertainty of sample conditioning, but has a major implicit assumption that the dry aerosol loading (i.e., extinction coefficient) is constant throughout the day. The ramifications of photochemical production and dynamical variability on this technique require further assessment. Results suggest that optical properties did not vary with different transport paths to the site, implying

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that local sources are most important. Overall, the paper could provide an interesting assessment of sub-saturated particle hygroscopicity using a unique technique, but requires major revisions to address this major assumption.

Major Critique

1. As I can see, the calculation of RH enhancement factors relies on the assumption that dry extinction is constant throughout the day. Many factors could force this assumption to fail, including secondary aerosol production, advection to the site of aerosol emitted elsewhere or an airmass with lower concentrations, temperature-driven partitioning of semi-volatile material, boundary-layer dilution, and wet depositional loss. Without explicitly showing that these mechanisms are not altering the dry extinction, your calculation of RH enhancement factors may be significantly inaccurate or at least highly uncertain. Since this is the major focus of the paper, the assumption needs to be addressed for publication. External data sources are almost certainly necessary to provide this evidence, or to provide additional constraint on selecting appropriate cases. Another path forward may be to analyze the approximately 1300 days that were not used in the analysis to assess the typical diurnal trend for aerosol extinction at the site for different meteorological patterns. Regardless, I feel that this assumption must be addressed quantitatively before publication.

Minor Comments:

Page – Line

12585 - 8: remove "occurring"

12589 – 14: I am a bit confused by the statement regarding a factor of 2 difference from the upper and lower PBL. If this layer is truly mixed, there should be no significant gradient. Please comment, or at least remove the word "obviously".

12589 – 20: Please comment on the remaining 1317 days that did not exhibit a 'pronounced' diurnal cycle. Do these days still fall on a typical hygroscopic curve even with the smaller dynamic RH range? Is the extinction coefficient constant during days with no RH change?

12589 – 20: Please note the RH range considered to be a 'pronounced' diurnal cycle.

12590 – 1: Please comment on, or add, the trajectory heights to Figure 5. The periods are only similar if their heights are also similar.

12590 - 20: How do you know the aerosols are 'aged'?

12590 – 23: Advection is explicitly used here in a description of this case, which directly contradicts the implicit assumption of diurnal, aerosol loading consistency.

12596 - 11: Please provide wavelength-corrected comparison for extinction. This is not a complex correction, and simply stating the presumed 10-15% offset is not acceptable.

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 12583, 2015.