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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A simplified empirical method for determination of aerosol hygroscopicity and composition" *by* C. H. Chan et al.

Anonymous Referee #2

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Comments of Reviewer #2

The authors present a simple method for determining hygroscopicity and source/type of aerosol based on RH, PM2.5, PM10 and visibility measurements conducted in Macao, China during the period January 2006 – January 2009. The correlations between aerosol extinction coefficients (extracted from visibility data) and aerosol mass concentrations are presented. These comparisons involve also extinction coefficients that are "corrected" by accounting the aerosol water uptake with a theoretical model. Also, the authors present a method for inferring the aerosol type/source based on the conducted measurements. In particular, relative contributions of marine and continental

aerosol sources at the sampling site are considered.

The contents of the study are relevant for Atmospheric Chemistry and Physics. However, as detailed below, the manuscript is so flawed in several respects that I cannot recommend the manuscript for publication. At the very least, the authors need to employ additional measurements to verify their approach and to improve to the usage of English language in the manuscript substantially.

Major Comments

1. Relationship between visibility and extinction. The authors estimate the extinction coefficient of aerosols at ambient conditions, σ(RH), based on the visibility measurements by using the Koschmieder formula. However, the equation is not necessarily valid under atmospheric conditions (see e.g. Horvath, 1971) and the applicability of the formula needs thus to be evaluated.

2. Estimation of the extinction coefficient at dry conditions, σ0, based on σ(RH). The authors employ a previously developed parameterization, taken from the literature, to calculate the ratio γ= σ(RH)/σ0 (Equation 2). The applicability of the parameterization for the conditions during the sampling period remains to be evaluated, however. This is needed because the particle hygroscopicity and hence also the ratio γ depends strongly on the particle size distribution, chemical composition and mixing state. On the other hand, the parameterization is based on certain assumptions on these properties and these assumptions may not correspond to the reality. The validity of the applied parameterization should be verified by measuring σ0 by e.g. with a nephelometer or, alternatively, measuring physical size distribution of aerosols along with their chemical properties and performing calculations with a more detailed model (see Cheng et al., 2006, for example).

3. Estimating the aerosol origin based on the PM and optical measurements and numerical algorithm (Section 3, equations 3 and 4). The algorithm employs another parameterization for calculating γ. Here the algorithm shares the same problems as the

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first method of calculating γ (see above). Another problem is the underlying assumption that aerosols with a certain origin contribute γ in a fashion that can be parameterized using a single relationship (Eqs. 3 and 4, Table 1). Even among particles of a single type, urban aerosol, for example, the physico-chemical properties of aerosols vary strongly (Swietlicki et al., 2008; Wehner and Wiedensohler, 2003). Further problem is that these contributions are assumed to be additive so that the contribution of each aerosol type to can be found by solving a multi-dimensional optimization problem (Section 3). Because of the variability of the aerosol properties in the atmosphere, it is questionable if each aerosol type has its characteristic hygroscopic growth or optical properties that can be distinguished from other aerosol types (for aerosol hygroscopicity, see e.g. Swietlicki et al., 2008). At the very least, the authors should investigate if the solution to the problem is truly a global minima (or maxima).

What the authors present, basically, is a very simple source-apportionment method. The method requires methodological evaluation (as outlined above) and experimental verification. The latter in turn would include measurements of aerosol size distribution, their chemical composition and trajectory analyses for the air masses sampled at the site.

4. Usage of English. The manuscript contains a number of grammatically incorrect and vague expressions and sentences, leading to poor readability. The manuscript should be proof-read by a native English user with suitable scientific background, if possible.

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