Atmos. Chem. Phys. Discuss., 7, S6028–S6033, 2007
www.atmos-chem-phys-discuss.net/7/S6028/2007/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Vertical profiles of urban aerosol complex refractive index in the frame of ESQUIF airborne measurements" *by* J.-C. Raut and P. Chazette

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Received and published: 16 October 2007

The authors thank the referee for his positive response to the paper and useful comments. We have revised the paper to take these into account:

1/ **Did the authors conduct a sensitivity study of the back trajectories from Hysplit? This can be done by considering a volume of end points and tracing back perhaps up to about a hundred trajectories to demonstrate a lack of sensitivity on the precise location of the end point. Also back trajectories in the PBL are suspect due to the effects of turbulent mixing and large scale eddies.**
As advised by the referee1, we performed a sensitivity study on the back trajectories

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from Hysplit. That study was conducted considering a set of end points surrounding the exact location of Paris (48.85°N, 2.33°E). The corresponding locations belong to the range [48.55°N 8211; 49.05 °N] and [2.23°E 8211; 2.43°E]. This is particularly crucial in the lowest altitudes. In our case (500 m), we did not observed any influence of the multiple trajectories in altitude. It guarantees the lack of sensitivity of the precise location of the end point. One has to note that Hysplit model is working at a large scale that does not cover turbulent mixing processes. The several depositions on the surface along the way of the backtrajectories arriving at 500 m have not any physical significance. But they simply suggest that the air mass at this altitude level may stay in the PBL. This complementary information has been added to the text of the paper.

2/ **How rapid is the convergence in the iterative process to retrieve the ACRI? Is uniqueness guaranteed?**

The retrieval of the complex refractive index does not require any iterative procedure. The determination of the real part is however the result of a minimization of the differences between the measured cross-section and a look-up table. The duration of the computations increases with the smoothness of the grid building the look-up table. (401 * 401) couples of values of real and imaginary parts have been considered in this study. The uniqueness of the real part is guaranteed when the real part can be considered constant in the likely range of values for the imaginary part (Raut and Chazette, 2007). It also triggers off the uniqueness of the imaginary part in the second step of the retrieval, where the unknown parameter (imaginary part) is the only one value enabling to simulate a BER equal to the measurement (synergy lidar 8211; sunphotometer). We have completed our explanations about the ACRI retrieval in the paper.

3/ **Why is it not possible to retrieve the refractive index separately for the**

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accumulation (fine) and coarse modes?

Theoretically, an aerosol size distribution might present different indices of refraction corresponding to each mode in the distribution, whereas the measured scattering coefficient and the BER are global features of the aerosols at a given altitude. The convergence of a Mie code towards those two measured parameters can be seen as the resolution of a system with two equations and two unknown parameters (one real part and one imaginary part). Taking into account the different indices in each mode would assume the introduction of supplementary unknown values without additional constraint. In this case, the methodology would not permit to deduce a unique couple of solutions.

4/ **What errors are introduced by assuming the aerosols are internally mixed?**

We have found this question very interesting and try to answer it properly. We performed a sensitivity study on the mixture state of the aerosols so as to assess the errors introduced when assuming that particles are internally mixed in Sect. 5.3. The homogeneous internal mixture supposes that any portion of the particle would have the same composition as the particle itself. The first error in assuming that all particles are internally mixed is the mixing of both accumulation (fine) and coarse modes, whereas dust particles are generally independent. We have therefore considered the case of an external mixture of dusts and well-mixed fine particles and investigate through a Mie code the equivalent ACRI for the whole size distribution that gives the same scattering coefficient and single-scattering albedo as the external mixture. In comparison with the global homogeneous internal mixing, both real and imaginary parts of the refractive index dramatically increase by 20

The second error concerns the mixing of black carbon within the fine part of size

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distribution. Although black carbon may be internally-mixed with other components in a particle, black carbon cannot be well-mixed since soot is irregularly shaped and solid, containing from 30-2000 graphitic spherules aggregated by collision during combustion (Katrinak et al., 1993). Thus black carbon must be distinct and not a well-mixed component of the particle. We have therefore divided each particle belonging to the fine mode into two smaller particles: one is only constituted of black carbon and the second one is an homogeneous mixture of particulate organic matter and water soluble fraction. It is then possible to assess an ACRI for the fine part providing the same scattering coefficient and single-scattering albedo as calculated through a Mie code. Hence we used as input parameter a size distribution with two modes (one for soot, one for organic matter and water soluble) whose modal radius were given by the mass fractions in the fine part. The same method applied to the external mixture of fine and coarse aerosols finally provides the equivalent ACRI for the entire aerosol size distribution. As expected, the influence of the black carbon amount on ACRI values is lower since soot represents very small particles that do not have a strong effect on optical efficiency. Table 4 summaries the different values of ACRI computed with chemical analyses considering the different states of mixture.

Jacobson (1999) reported that transmission electron microscopy indicates that the coating of elementary carbon is also a plausible physical configuration and his calculations showed that the absorption cross-sections of the core treatment were greater than those of elementary carbon particles externally mixed from water particles and less than those of well mixed elementary carbon 8211; water particles. The actual ACRI values in our study are thus probably between the extrema given in Table 4. This discussion has been added to the manuscript.

Katrinak, K.A., Rez, P., Perkes, R., and Busek, P.R.: Fractal geometry of carbonaceous aggregates from an urban aerosol, Environ. Sci. Technol., 24, 539-547, 1993.

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Jacobson, M.J.: A physically-based treatment of elemental carbon optics: implications for global direct forcing of aerosols, Geophys. Res. Let., 27 (2), 210 8211; 220, 2000.

5/ **In Equation (1) it is assumed that epsilon is constant. Shouldn8217;t epsilon be different for the accumulation and coarse modes?**

The Hänel size growth coefficient used in Eq. (1) has been taken from the results of Randriamiarisoa et al. (2006) thanks to an analysis on the influence of RH on the chemical composition of Paris urban aerosol. This study had shown that the water soluble fraction was belonging to the accumulation mode indicating that the retrieved represents hygroscopicity of the accumulation mode. The coarse mode particles did not appear hydrophilic. This part has been added to the text.

6/ The correction has been done: 8220;Aerosols are not very hygroscopic (Randriamiarisoa, 2006).8221;

7/ **It would be useful to compare the aerosol size distributions from PCASP and AERONET. Can this be included?**

We have included a comparison between aerosol size distributions from PCASP and AERONET in Fig. 8. The first mode of AERONET size distribution centred at 0.1 μm is highly predominant with 99/100 of the total number of particles and is optically efficient. The modal radius is in accordance with the accumulation mode measured with the PCASP and presented on Table 1 with a smaller dispersion. It is now well known that AERONET is not able to manage the detection of very small particles with a

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radius lower than 50 nm, whereas the sampling of coarse aerosols is a limitation of the PCASP. However, these size distributions have only a slight influence on the BER (Raut and Chazette, 2007). In this study, AERONET products, such as complex refractive indices and size distribution, have been used only for the discussion on the BER retrieval.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 10799, 2007.