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Interactive Comment

Interactive comment on "Size-segregated aerosol mass closure and chemical composition in Monte Cimone (I) during MINATROC" *by* J.-P. Putaud et al.

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This paper describes an experimental work aiming at a detailed mass closure of surface aerosols at the high altitude station (2165 m) of Monte Cimone, based on results from a summer 1-month campaign. Several techniques (chemistry, gravimetry, optics) are used in parallel and results are carefully evaluated and compared. It is always difficult to reconcile aerosol measurements from different techniques and I find the approach and the paper very interesting. I have a few remarks, and I especially wish to discuss the approach for the large 1-10 μm size fraction. I believe that the scientific community still has to work on reconciling optical and chemical measurements of the super-micronic aerosol size fraction.

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Interactive Discussion

Discussion Paper

The super-μm particle volume calculations critically depend on the accuracy of measurements of relatively few particles in the largest size range (5-10 μm) by the Grimm optical particle counter (OPC). Section 2.3.1 Measurements is presently not giving me total confidence on these results. I have questions on the derivation of the supermicron aerosol volume:

- Was the OPC recently calibrated against particles of known size?

- What was the OPC individual measurement time? Do you really measure OPC over the whole period of the corresponding impactor sample? A short time contamination by locally produced large particles (car, gust wind, ...) might be missed by the OPC while significantly contributing to total mass. How do the size distribution and the total concentration change during an 11h impactor period? It seems to me that just averaging the number size distribution over the impactor period is uncertain to derive the total volume over a long period if there are variations in the size distribution and the total mass along the period. I would think of using a PM10 monitor in parallel and weighing the size distribution by the total mass.

- How significant were the numbers of particles in the bigger channels of the OPC? I believe that the 2% counting reproducibility reported here from the manufacturer only applies when one counts enough particles in a channel. What is applicable in industrial or polluted environment may not be in free troposphere air. I find that we miss here some statistics of the countings, and figures on size distributions.

- Can you show that there is no cut-off in particle size when you sub-sample the PM10 flow compared to direct sampling with the OPC? Looking at figure 4b, I suspect that you could reconcile your volume with gravimetric measurements for the 9 outliers showing underestimated volume by adding a small number of particles in the bigger Grimm channel, which measurements are possibly doubtful due to both a small number of particles and a possible cut-off caused by the sub-sampling system.

- It would be helpful to compare the Grimm-derived size distribution with other independant optical measurements or to refer to such comparison if they cannot be made with Minatroc data. I find that the volume size distributions should be also illustrated et compared for instance to results from ACE-2. During the dust events of ACE-2, the volume distribution was found to peak over 4-5 μm (see e.g. Formenti et al., 2000 and Collins et al., 2000). Do you reproduce such a distribution with the Grimm?

- By reference to fig.2, it looks that the Saharan period is excluded from the comparisons in figs. 4 and 5. Can you comment?

The cut-off diameter of impactor stage 4 which is used to segregate between the sub-μm and the super-μm fractions is in fact equal to 1.2 μm. To my knowledge, Grimm classes are $>$1 μm and $>$1.6 μm and there is no Grimm size channel $>$1.2μm. It is not specified how you cope with this difference in cut-off between the two instruments.

The use made of cut-off diameters of the Berner impactor also gives me some concern. Collection efficiencies of the impaction stages are considered perfect here. In fact they are not step-like functions and the collection efficiency curves as a function of particle size should be known. Actually, excluding particles bounce-off problems, a certain percentage ($<$50% and decreasing with increasing particle size) of large particles penetrates stage 4 and below, and conversely a certain percentage ($<$50% and decreasing with decreasing partricle size) of particles smaller than 1 μm is captured by preceeding stages with a larger cut-off. The excess fraction of large particles on a given stage should make a somewhat bigger volume than the missing fraction of small particles. This is likely to induce some bias in the retrieved mass fraction from a stage if it is attributed to a size range defined by cut-off sizes in particle number. This bias is not evaluated. It seems to me that the balance in mass between size fractions from an impactor sample requests to take the collection efficiency of the impactor stages into account and to reconstruct the size distribution (see e.g. Gomes et al., 1991). Analytical deconvolution of the impactor data would allow to derive analytically the mass below 1 μm and in the range 1-10 μm, thus a more reliable comparison with OPC measurements (i.e. exactly within the same size range).

In section 2.3.2 Data processing, you state that the volume of particles is calculated based on the diameter measured by the instruments, but the instrument do not really measure the size, they just classify it in a given range, which is pretty large for the bigger Grimm channels (5-7.5 and 7.5-10 μm). You should therefore specify your hypothesis for the reference size of every channel (median size of channels?).

References:

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