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Atmospheric
Chemistry
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Discussions

Interactive comment on “Online mass spectrometric aerosol measurements during the MINOS campaign (Crete, August 2001)” *by* J. Schneider et al.

Anonymous Referee #2

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General Comment

The authors describe aerosol measurements in the field. They applied several particle measurement techniques. The goal of their paper is to establish new mass spectroscopic techniques with focus on the AMS (Aerodyne) by comparison to approved techniques. The paper is interesting but needs some reorganisation in order to remove ambiguities and inherent contradictions. I suggest the paper should be published in ACP after a few minor and three major revisions along the lines given below.

Major Comments

Major Comment 1

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Section 3.1

4th § + Figure 5 : I agree about the correlation between sulfate and ammonium on 22.8. However, on all other days shown in Figure 3 the ammonium concentration (mostly around 1 microgram/m3) seems to be pretty much detached from the sulfate amounts (with the exception of late 19.8). Figure 5 is misleading, because the sulfate axis is compressed compared to the ammonium axis. The datalook more like an almost horizontal "line" of ammonium data centered around 1 ?g/m3 along a wide range of sulfate data of 1-10 ?g/m3. I suggest a day by day analysis. Moreover, if the authors cast doubt on their ammonium data compared to the MOUDI - which is not justified (see below)- what is worth of discussing ammonium/sulfate ratios? What are the errors on the ratios from AMS and Moudi data, are they significantly different? What kind of regression was performed? Since both quantities, ammonium and sulfate, are afflicted with errors, a simple regression may lead to misleading results. The errors of sulfate and ammonium must be considered in the regression. What are the errors of slope and intercept ?

Major Comment 2

Section 3.2

2nd § + Figure 9 : What is meant here by AMS and Moudi agree well? - The authors should give numbers instead of qualitative statements. Moreover, the following discussions (4th §, see major comment 3) on shifting the size distributions, rises the questions, if AMS and MOUDI are plotted on the correct diameter axis. MOUDI measures aerodynamic diameters, while AMS measures vacuum aerodynamic diameters. Were the data corrected for that fact ? What would be the effect of such a correction on the intended comparison ?! From Figure 9 it seems that the AMS underestimates sulfate more than ammonium compared to the MOUDI. In Table 1 the deviation of ammonium in AMS and MOUDI are close to the deviation of sulfate. How does this relate to the lower ammonium/sulfate ratios found by AMS and the statement "A possible rea-

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son for the lower slopes measured by the AMS may be underestimation of ammonium due to uncertainties in the absolute calibration for this species." (p7 3.1 §3). Figure 9 and Table 1 need coherent discussion considering all findings.

The MOUDI channel 200-350 nm seems to be significantly underestimated in sulfate and ammonium by the AMS (or overestimated by MOUDI), while all other channels are within the errors and within in 20%. Is there any reason for that ?

In the case of nitrate: nitrate appears mainly in the coarse mode and only traces are found in the fine mode. All findings are within the (large) errors bars. So, there is no mismatch between MOUDI and AMS as falsely stated.

Major comment 3

Section 3.2

4th § + Figure10 : What is compared in this figure, daily averages ? The shift of the axis' is absolutely misleading. The authors must show what they have measured, recalculate their data to the suited diameter e.g. the mobility diameter and then compare the data and clearly indicate the size of the introduced shift.The shift by a factor of 2 to match DPS and AMS data could just be justified, although non-neutralized ammonium sulfates tend to be deliquescent down to 40-50% RH. Therfore the proposed density of 1.7 g/cm3 could be already to large. The required shift to match OPC and AMS data is not reasonable. Lets for the moment accept the factor of 2 between DPS and AMS. Further, lets refer in the discussion to the mobility diameter as measured by the DPS. Then a factor of 3.5 has to be applied to bring the OPC measurement in agreement with the large-particle tail of the AMS. Stier and Quinten (J.Aerosol Sci., 29 (1998), 223-225) demonstrated, that a deviation of +/-0.1 in the (real part of the complex) refractive index of the particle material compared to that of Latex induces a shift by +/-1 channels of the observed size. The PCS2000 has 16 channels per decade, thus shifting the first 330 nm channel by a factor of 3.5 to 1155 nm, translates into a shift by 8 to 9 channels. An underestimation of size by 8-9 channels would require a refractive

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index smaller than 1 (1.59 - 8ů0.1) for the particle material. The shifting of the size distributions at once questions also the results in Figure 7 and Figure 9. In Figure 7 it was concluded that DPS and OPC match quite well (shown also for the data of 20.8 and 21.8.).

The whole topic of comparing different aerosol measurements needs a clear cut treatment, i.e. a physical reasonable recalculation of all measured data on a common diameter axis is required. Figure7,9 and 10 must then be coherently discussed. Agreement or deviations between the several aerosol techniques should be clearly pointed out and thoroughly discussed.

Minor comments:

Introduction

4th § : The description of the AMS is misleading and needs extension: What is meant by bulk aerosol ? - integrated over several particle diameters ? What means thermally evaporated ? Alternatively: a clear, compact description of the AMS is put into section 2.1 and 2.1 is then referenced here.

2 Experimental

2nd §+ 3rd § : Please, state clearly, the instruments situated in the Met-Station: MOUDI, Quartz filter sampler (?), TSI 3040 diffusion battery and the instruments housed in the MPI/FZJ container AMS; SPLAT, PCS 2000, PCS2010, TSI CPC3025 (?) The 4th § is redundant as soon as 2nd § and 3rd § are put in a reasonable order.

2.1 The AMS

Already in the previous paragraph it was mentioned that the upper limit of the AMS is 1.5 micrometer (diameter or radius ?). How do the cut off curves look like for diameters between 50 nm and 80 nm and -more important- between 600 nm and 1500 nm where particles are transmitted through the lens with less than 100% efficiency? Is there anything quantitative known about it? How will uncertainties here translate into

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measurement errors and affect the comparison e.g. with MOUDI data?

The authors should introduce and explain in this section the "nitrate equivalent" calibration concept.

3. Results and discussion

The concept of the "nitrate equivalent" needs to be explained in the experimental section.

Figure 1 : the authors must comment on the different colors / species detected under the same mass, at least the basic idea behind it.

3.1 Time series measurements

2nd § + Figure 2: Why do the UCPC's deviate at 21.8 to 22.8

5th § : Can the back trajectory analysis explain any of the authors findings in Figure 3 and 5? If, yes they should describe it; if not they should skip the back trajectories.

3.2 Size distributions

1st § : DPS was named diffusion battery in the previous paragraphs

Figure 7 : What kind of size distributions are given ? The best, typical single measurements or daily averages ? How close was the air plane? Are singular fly bys a good test of the sizing instruments at the ground? Is there a rational, why size distributions measured by the air plane should be the same as those measured on ground, or is this just a lucky case? In the 4th § it is claimed that the optical diameters must be corrected, because the refractive index of the particle material is different from that of the Latex calibration aerosol (see major comment 3). Of course, that would also affect the results shown in this figure, there will then be a quite substantial gap in the data.

4th § DPS was named diffusion battery in the previous paragraphs

3.3 Ion series analysis

2nd § : It is more the processing of the gas-phase VOCs than the direct oxidation of particles

3rd § : I see two cases, may be three, in Figure 11: the 18th, 20th, 23rd, with 21st as most distinct, and 17th, 19th, 22nd of August, is there any rationale in the trajectories or meteorological conditions, that could explain this behavior.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3907, 2003.

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