

Interactive comment on “A chemical analyzer for charged ultrafine particles” by S. G. Gonser and A. Held

Anonymous Referee #1

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The manuscript entitled “A chemical analyzer for charged ultrafine particles” by Gonser and Held describes a newly developed collection and evaporation unit for aerosol particles with diameters below 30 nm.

The particles can either be charged by exposure to bipolar ions from a radioactive source or unipolar ions from corona discharge. The charged particles are size-selected by a radial differential mobility analyzer (rDMA) before they enter the collection and evaporation unit. Within this device the aerosols are electrostatically precipitated onto a Nichrome filament. After a certain collection period the filament is moved close to the entrance region of a mass spectrometer. The application of a current to the filament results in the evaporation of the non-refractory aerosol compounds. These are transferred into the ion source region of the mass spectrometer by means of a capil-

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lary. Electron ionization and subsequent analysis of the ions in a time-of-flight mass spectrometer yields information about the particle composition.

The chemical analysis of nanometer-sized aerosol particles is a big challenge due to the extremely small mass concentrations of these particles. Therefore, only very few instruments exist which can be used to obtain insights into the composition of these small aerosols. However, knowledge about their composition is required in order to understand how freshly nucleated particles can grow to sizes where they can act as CCN (cloud condensation nuclei). In this respect the development by Gonser and Held is an important and innovative approach to gain further knowledge in the underlying mechanisms of this process.

The manuscript is well structured and contains information about the set-up and the characterization of the instrument. However, some aspects should be described in more detail and additional information which is important to fully understand the method is missing.

Overall, I recommend the publication of the manuscript after the points listed below have been considered.

Points which should be addressed:

page 3864, line 1 (and in the following): Did the authors consider the production of OH from the corona discharge? This could also lead to the oxidation of gas phase substances and their subsequent partitioning into the aerosol phase. A short discussion should be added.

page 3864, line 28: How does the “pre-separator” work? It seems that this is a crucial aspect of the design when using the unipolar corona charger. Therefore, the operational principle and the design of the “pre-separator” should be mentioned.

page 3865, line 12: The way the sentence is formulated it is not clear if the rDMA is an exact copy of the design by Zhang et al. (1995) or whether it has further been

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optimized. If it has been optimized then the modifications should be described.

page 3866, line 1 (or end of section 2.2): Has the performance of the rDMA been validated against theoretical transmission curves? Stolzenburg and McMurry (2008, Aerosol Sci. Technol.) report the theory for calculating the expected transmission efficiency in a tandem DMA set-up as function of the DMA geometries, applied flow rates and voltages. In order to demonstrate that the newly set-up DMA selects particles accurately with respect to their sizes; these theoretical response curves could be added to Fig. 3. In any case, a discussion should be included whether the SMPS which is used as a reference yields the peak concentration at the same diameters as the rDMA.

page 3866, line 20: How can it be assured that both the sample flow and the purge flow have the same velocity at the upper edge of the collection tube? The geometry shown in Fig. 4 is quite complex and later in the manuscript (page 3873, line 16) it is indicated that the flow could be turbulent in this region. This inconsistency should be resolved.

page 3866, line 28: I have the impression that the word “sealed” is not correct here. If the PEEK piston would indeed align with the conical cavity then it would not be possible to flush the filament with nitrogen (from inlet 8) while the aerosol material is being desorbed and transported into the mass spectrometer.

page 3867, line 2: How is the temperature controlled? Probably this is done by varying the current through the filament. However, is the temperature of the filament also measured? Values for the filament temperatures are given throughout the manuscript without describing how these were actually determined. This procedure needs to be described.

page 3868, line 28 (and in the following): How is it known that the flow still contains a considerable amount of gas-phase organics and that ~90% of these could be removed with a charcoal scrubber? Was this measured with the same mass spectrometer in “gas-phase mode”?

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page 3869, line 14: If the signal from silicone (m/z 207) shows a constant intensity throughout all the measurements, why is it then used to normalize the other signals? This would mean that the other signal intensities are divided by a constant number. Therefore, the normalization would be useless.

page 3870, line 2: A temperature of 800 °C can be realized at the filament. Again, it should be pointed out how this was determined. In addition, at this high temperature the PEEK material should melt (melting temperature ~340°C). How is this prevented?

page 3870, line 3: How is it known that some of the signals arise from the gas phase? Was the HV turned on and off and then the residual signal was attributed to the gas phase? page 3870, line 12: Again, the normalization with the silicon signal doesn't seem to be useful if it is constant throughout all measurements.

page 3871, line 3: This sentence should be reformulated. It is not clear why there is no restriction on the collection times. This is clearly not the case because for very short times the signals will be below the detection limit.

page 3871, line 23: The method for estimating the collected aerosol mass should be described in more detail. It seems that one requirement is that the particle number concentration coming from the rDMA is constant over time. Another assumption seems to be that the number of particles which are collected on the wire can be determined from the difference in particle concentration between HV on and HV off periods, i.e. losses at the exit of the collection unit as well as losses close to the collection tube are neglected. In this respect it would be interesting to know what the transmission of particles is when the HV is off and when the particle concentration is measured simultaneously at the inlet and the outlet of the collection unit.

page 3871, line 27: A short discussion could be added about the detection limit for different substances. The detection limit of 7 ng is determined for a rather complex system where many different peaks occur in the mass spectrum. For a pure compound with just a few peaks present, the detection limit could be substantially better.

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page 3872, line 27: To my knowledge no impactors are available for these small sizes which work at pressures around 1 bar. Only when the pressure is substantially lowered these low cut-off sizes can be realized with impactors. Please clarify how these small cut-off sizes can be realized with the current set-up.

page 3881 (Fig. 5) and page 3882 (Fig. 6): Fit curves could be added to these figures to indicate the linearity.

page 3882, Fig. 6: In panel d the x-axis should range from 0 to >2 h, currently it starts somewhere around 15 min. Although the signal clearly increases for longer collection time, there seems to be a deviation from linearity here. Please add some discussion why this could be the case.

Additional remark:

I did not understand how the signals in Fig. 5 and Fig. 6 d were exactly derived. Clearly the signal at each mass was integrated over a certain period. However, when heating the wire the aerosol material won't come off all at once. Depending on the temperature the signal will probably rise and then decay after a while. Therefore, could you please add an additional figure which shows the time dependency for one representative m/z value. In this figure the wire temperature should also be shown. One very important aspect is also after what time the signal reaches background level although the heating still persists. In this respect it would also be interesting to discuss potential memory effects. E.g. if the aerosol sample is desorbed and the heating current is turned off and then turned on again after a while, do you see any rise in the signals? In other words how good does the cleaning procedure of the wire work and how exactly is this cleaning performed?

Minor comments:

page 3860, line 3 (also line 24): "the Earth's"

page 3860, line 9: "subsequently" instead of "consequently"?

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page 3860, line 13: maybe better to use the word "defined" instead of "known"

page 3860, line 24: maybe better to use the word "significant" instead of "eminent"

page 3861, line 4: instead of "after sunrise" please reformulate to "in the morning hours"

page 3861, line 27: "ion trap", two words; the word "trapped" could be replaced by "captured"

page 3862, line 17: replace the word "and" by "while"

page 3867, line 4: please delete the word "to" after "heated"

page 3871, line 7: Please replace "with growth rates close to natural rates" by "with similar growth rates that can be observed in the atmosphere"

page 3872, line 17: Using one defined size of 20 nm is not appropriate to report an aerosol concentration; instead a size interval should be used.

page 3873, line 6: Please delete the word "By"

page 3873, line 9: Please delete the word "of"

page 3880, Fig. 4: adding a scale to this figure would be useful

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