

We would like to thank the referee for carefully reading our manuscript and for giving such constructive comments which substantially helped improving the quality of the paper. In the revised version of the manuscript we have tried to consider all the points that were raised. The points that relate to the text and grammar are not discussed here. These have been adopted for the revised version according to the referee's suggestions. The points which needed further discussion are listed below. Therefore, the points raised by the referee are repeated (in bold letters) followed by a detailed reply and discussion by the authors. Additionally, if necessary, text passages that have been added to the revised manuscript are given (written in italics).

While only indirectly relevant to the main point of the manuscript, some discussion of the absolute calibration of the H₂SO₄ calibration source is needed. I.e. how do you know what the humidity dependence of the calibration is without the absolute calibration of the output as a function of H₂O.

A similar point has also been raised by the first referee. However, the absolute calibration is not relevant in this case, because only a relative comparison between the two sources has been performed. The section between page 5302, l. 17 and page 5303, l. 2 has been removed therefore. However, another passage covering the different [H₂SO₄] provided by the calibration source has been included instead (see reply to your remark about Figure 3 below). The performance of the calibration system, also with respect to providing absolute concentrations of H₂SO₄, will be the subject of a forthcoming paper.

A comparison of the S/N for identical [H₂SO₄] and integration periods, as well as the nominal H₂SO₄ background levels between the two sources should be included. What was the primary ion count rate for the americium source? What is the duty cycle and integration time for the scanning of the ion masses by the quadrupole. What is the timescale and level of the 'higher noise' from the corona source (i.e. does normalization by the primary ion cancel this effect or does it impact the precision of the final measurement as compared with the americium source).

In order to discuss the differences between americium and corona source further the below text passage has been added to the end of section 3:

The integration time for m/z 64 and m/z 97 were 0.25 s and 1 s, respectively. One full cycle (which included acquiring data also for six additional m/z values) was adjusted to ~5 s. An evaluation of the detection limit for the americium ion source yielded a value of $1 \times 10^4 \text{ cm}^{-3}$ when no SO₂ was added to the sample gas and $2.8 \times 10^4 \text{ cm}^{-3}$ if SO₂ was added at a similar concentration as for the experiment with the corona ion source, respectively. The count rate for m/z 64 reached values up to $20,000 \text{ s}^{-1}$ for the americium source. The primary ion count rate of the corona ion source is therefore approximately 25 times lower. This effect should therefore lead to a factor $25^{0.5} \approx 5$ lower detection limit while the actual detection limit is about a factor of 6.5 lower. This means that normalization by the primary ion count rate can explain the difference in the detection limits almost completely. The slight drifts in the primary ion count should therefore only lead to a small contribution. These drifts occur on the timescale of several hours and can reach maximum fluctuations of $\pm 50\%$. It is also

worth mentioning that the relative change in sensitivity due to the addition of SO_2 is quite similar for both sources.

Has it been verified that $[H_2O]$ mixing ratio in the discharge does not impact the H_2SO_4 determinations, in terms of background stability, and SO_2 interference?

No systematic study of this influence has been performed. However, the CIMS with its current configuration proved to work reliably over a period of >4 weeks during the CLOUD-10 campaign at CERN. While there were no extreme changes in the $[H_2O]$ concentrations of the sheath gas, the temperature in the hall where the experiments were performed changed between 12°C and 19°C and therefore affected the $[H_2O]$ as well. This means, at least for laboratory measurements where the $[H_2O]$ is not subject to extreme changes no substantial interferences should be expected.

Pg 5299 ln 24: Dimensions are mentioned here in the text, but are not shown in Figures 1 or 2 for reference. It would be nice if dimensions or at a minimum a scale bar were added to Figures 1 and 2.

It is a good idea to include dimensions in the figure. Therefore, these have been added to Fig.1 and Fig.2, respectively.

Pg 5300 ln 1-2: This is not clear. The needle has been moved backwards relative to what?

Instead of having the needle in the center between the two QF40 connectors it has been moved further upstream in order to increase the distance between the needle and the edge of the inner cylinder with respect to the direction of flows. This way the electric field from the HV at the needle should have a smaller effect on the trajectories of the ions which should mix with the sample gas. To make this clearer, the sentence in section 2.2 has been modified:

Therefore, the needle has been moved slightly upstream in order to shield the ion/sample gas mixing zone from the electric field the high voltage creates (see Fig. 2).

Pg 5301 P1: It would be good to describe where the discharge occurs? Is it between the needle and the outer cylinder? Is the actual discharge path well-determined, meaning does the arc migrating from the needle to various points locations on the edge of the hole surrounding the needle? If so, would any increase in stability be gained if the exact arc path was determined by destroying the symmetry of the needle with respect to the closest approach of the outer cylinder?

This is something that has not been examined yet in detail. However, there is actually no arc existing in this type of discharge since the emission current from the needle is too small (several micro-amperes). Therefore, the emission of electrons from the needle tip should resemble rather a spray which covers a relatively large volume of air in contrast to an arc which would represent a narrow and more direct connection between the tip

and the cylinder. Thus, the idea of a migrating arc can probably not explain the fluctuations in the primary ion count rate.

Pg 5302 ln 17-20: Related to major point 1. It has not been demonstrated here that the output of the calibration source is linear with [H₂O] in the calibration source. Without this, I find this sentence lacking support.

We agree with this comment and therefore have removed this passage from the manuscript. Further discussion about the calibration source has however been added to section 3.1 (see below).

Figure 1 and 2: Add dimensions, as mentioned above. Perhaps highlight the region in Figure 1, which is replaced by the apparatus shown in Figure 2.

We have followed these suggestions and modified the figures accordingly.

Figure 3: If this data is shown on a log-log scale would more information be visible? Is the reason for the seeming falloff in sensitivity ratio between the sources at low H₂SO₄ (low [H₂O]) understood? Further discussion regarding this is warranted, as the H₂SO₄ calibration levels are substantially above relevant ambient levels.

The data point for the lowest [H₂O] corresponds to a concentration of $\sim 7 \times 10^6$ cm⁻³ of H₂SO₄. Here, the ratio suggests an average deviation of $\sim 25\%$ between the two sources. This deviation is still lower than the accuracy for most H₂SO₄ measurements. Also, already at the higher end of observed atmospheric concentrations, a value of [H₂SO₄] = 7×10^6 cm⁻³ can be observed frequently during the day time. However, we do agree that there might be a systematic deviation between the measurements for the corona and the americium source. This potential issue has been brought up by the first referee as well. We are arguing that the discrepancy between the two sources is rather related to an uncertainty in the [H₂O] than in the measurement of the [H₂SO₄]. To address this question, more discussion has been added to section 3.1 and we also refer to the reply given to the first referee for further clarification. Additionally, the figure has been modified to show the [H₂SO₄] and [H₂O] on a log-log-scale.

Figure 4: 1-minute average curves do not appear 'by eye' to go through the center of 5s data for the m/z 97 and [H₂SO₄] traces. I suppose this is an artifact of the log scale not showing zero points. This should probably be mentioned in the caption unless there is a better way to display this.

This is right; there are quite a lot of data points showing a zero count rate for the m/z 97 signal. Therefore, following your suggestion, an explanatory sentence has been added to the caption of Figure 4:

Please note that the data for m/z 97 and H₂SO₄ includes many points showing values of zero, which are not visible on the logarithmic scale used here.