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).

p-5296, l-18,19: Nothing personal here, just a general "why has the 'norm' changed to this" statement: Referencing what you know is fine but if previous work had already showed the fact that you are stating, why not refer to the archival study? People like to see that their work is referred to in a fair and substantive manner.

We agree that other references should be included here which refer to the pioneering work that has been performed in the field of nucleation studies. Therefore, the following references were added and cited within the first sentence of the introduction:

The measurement of gaseous sulfuric acid is important since H_2SO_4 is one of the key compounds responsible for atmospheric new particle formation (Eisele and Tanner, 1993; Weber et al., 1995, 1997, 1999; Curtius , 2006; Kulmala and Kerminen, 2008).

Eisele, F. L. and Tanner, D. J.: Measurement of the gas phase concentration of H_2SO_4 and methane sulfonic acid and estimates of H_2SO_4 production and loss in the atmosphere, J. Geophys. Res., 98(D5), 9001–9010, 1993.

Weber, R. J., McMurry, P. H., Eisele, F. L., and Tanner, D. J.: Measurement of expected nucleation precursor species and 3-500-nm diameter particles at Mauna-Loa-Observatory, Hawaii, J. Atmos. Sci., 52, 2242-2257, 1995.

Weber, R. J., Marti, J. J., McMurry, P. H., Eisele, F. L., Tanner, D. J. and Jefferson, A.: Measurements of new particle formation and ultrafine particle growth rates at a clean continental site, J. Geophys. Res., 102(D4), 4375-4385, 1997.

Weber, R. J., McMurry, P. H., Mauldin R. L., Tanner, D. J., Eisele, F. L., Clarke, A. D., and Kapustin, V. N.: New particle formation in the remote troposphere: A comparison of observations at various sites, Geophys. Res. Lett., 26, 307-310, 1999.

p-5298, l-2: '… works, it is stable and reliable,…' >Should include the standard flows for sheath and sample gas.

The sentence has been reformulated and the flow rates have been added.

It is demonstrated that the corona ion source works, it is stable and reliable. Furthermore, it shows a negligible cross-sensitivity to SO_2 for sulfuric acid measurements when using a sample flow rate of 7.5 slm (standard liters per minute) and a sheath gas flow rate of ~22 slm, respectively.

p-5300, l-15: replace very well with highly. This adverb is disputable: see increase in NO₃⁻ with time in Fig. 4. Thus, ' ..reproducible within normal variations of ion count rate.'

We agree with the referee that the statement "very well" might be too strong in this case and therefore replaced it by the suggested formulation.

p-5300, l-17: Were the needles cut off and machined in any way? They look like they are sort of cylindrically 'barbed' in the figure.

It is true that the needles have a barbed shape near the needle tip; however, they were not machined in any way. To clarify this, the following sentences have been added to section 2.2.

p-5300, l-5: *No adjustment needs to be made to a needle; each one can be used as is.*

p-5300, l-19: *The diameter of the hole is just big enough such that the needle can be pressed through the hole. It is then held in place because it is slightly cylindrically barbed.*

p-5302, l-6: 'defined'? what is meant by that?

By "defined" we meant "adjustable". Therefore, the sentence has been re-formulated to read as follows:

The H_2SO_4 has been provided by an external calibration source which provides adjustable and stable concentrations of H_2SO_4.

p-5302, l-17: How do you know linearity above a certain amount of H_2O? This is not shown here yet it is not needed: all that is needed is the comparison between alpha and corona. The results show a good agreement EXCEPT at low H_2O which appears to be a systematic difference between alpha and corona rather than the random difference (+ or - temperature error) alluded to by the authors.

Within the calibration system [H_2O] is photolyzed by illumination with 185 nm light in order to produce OH. The concentration of OH is directly proportional to the water vapor concentration. Due to the reactions with SO_2 and O_2 the OH is rapidly converted to SO_3. If the concentration of H_2O is large, the subsequent reaction which converts SO_3 into H_2SO_4 is not limited by the water vapor concentration. Therefore the resulting H_2SO_4 should be directly proportional to [OH] which depends linearly on [H_2O]. However, since this feature of the calibration system is not of importance in this paper, as it has been pointed out also by the second referee, the whole section between p-5302, l-17 and p-5303, l-2 has been removed. However, more discussion has been included to section 3.1 in order to cover the second part of the referee's point (see below).

The referee also mentions that there appears to be a systematic deviation between the corona and the americium ion source (Fig. 3). We think that the falloff in the ratio (H2SO4_corona / H2SO4_americium) at lower [H_2O] is actually not related to the ion source but rather related to the calibration system. An additional error, besides the temperature measurement, is the adjustment of the exact H_2O concentration which is currently not very well reproducible at the very low [H_2O]. However, this uncertainty is also included in the error calculation for the ratio. For larger moistures, where the adjustment of a stable [H_2O] is much better reproducible the agreement between corona and americium source is nearly perfect.

Another reason, why we think the accuracy of both sources is identical, is the following observation. One source of error one might think about is that the corona ion source could produce spurious amounts of H_2SO_4 by the reaction of OH (produced in the source) and SO_2 in the sample gas as has been tested in section 3.3. If this was the case the concentration measured by the corona source should be higher than the one measured with the americium. This is, however, not the case. In addition, there is no evidence that the contribution from the OH and SO_2 reaction in the corona source has a substantial contribution for [H_2SO_4] > ~2e+05 cm-3/s (section 3.3). Taken this together, it is much more likely that the seemingly systematic deviation for low [H_2O] is rather caused by the calibration system than by the different ion sources. To cover this question the following passage has been added to the manuscript (instead of the section between p-5302, l-17 and p-5303, l-2 as mentioned above):

While the points for the two highest [H_2O] are perfectly corresponding to a 1:1 ratio, the points for the lower [H_2O] seem to move progressively further away from this ratio. For a [H_2SO_4] of ~7e+06 molecule cm^{-3} the ratio reaches a value of 0.75. However, we don't think this behavior is caused by any of the two different ion sources but rather reflects a feature of the calibration system in its current realization. The accuracy for adjusting the humidities becomes lower for dryer conditions and therefore the ratio of the [H_2SO_4] measured by the two sources reflects this uncertainty. As shown in section 3.3 the corona source shows only a negligible cross-sensitivity to SO_2 in the sample flow (from the reaction with OH produced by the discharge). Therefore, this contribution is quite small and if it was responsible for the observed deviation, the ratio should become larger than 1 because additional H_2SO_4 would be produced by the corona ion source. Since this is not the case, the cross-sensitivity to SO_2 is unlikely responsible for the falloff in the ratio. Together, with the very good agreement for the higher concentrations, where the [H_2O] could be adjusted much more accurately, the slight systematic discrepancy does not seem to be caused by any difference between the two ion sources. Especially when taking into account that the points shown in Figure 3 are all corresponding to a ratio of 1 within the range of errors.

As an aside, it is unlikely that the sum of SO_3 and H_2SO_4 will change with H_2O content of the calibration source. Did the authors detect any SO_3^-? Is it possible that it is converted to HSO_4^- by reaction with water ligands on the ions?

The [SO_3] depends on the [OH] in the calibration system. Since the [OH] depends linearly on the [H_2O], the concentration of SO_3 is not constant. In addition, we think it would be challenging to perform quantitative measurements of [SO_3] with the current configuration of our CIMS. The H_2SO_4 is detected through the reaction with NO_3^- whereas SO_3 probably doesn't react with NO3- to form SO_3^- ions - at least not at the same

rate as H_2SO_4. Therefore, an additional calibration would be required for quantitative measurement of SO_3 which is not available to us. Thus we think it is beyond the scope of this paper discussing this question.

p-5304, l-10: 'can vary by 50%.' ? What is then meant by the previous claims of stability and reliability? Perhaps a plot of representative alpha data should be included so these relative words can be substantiated.

In our opinion slow drifts in the primary ion count rate do not influence the accuracy of the CIMS measurement and do have a small effect on its lower detection limits only. As stated, the count rate was more stable for the americium ion source where virtually no drift was observable over the timescales the instrument was used. Therefore, we don't think it is necessary to show a plot of the primary ion count rates for the americium source. However, we do agree that the detection limit of the americium source should be given as well.In addition, following the suggestions of the second referee we have also included some discussion about the possible reasons for the different detection limits as well as information how the mass spectrometer was operated. This has been done by adding the following text passage to the end of section 3.3:

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 ~4.5 s. An evaluation of the detection limit for the americium ion source yielded a value of 1×10^4 cm^{-3} when no SO_2 was added to the sample gas and 2.8×10^4 cm^{-3} if SO_2 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 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 ±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.