We would like to thank both reviewers for their constructive comments on our manuscript, and appreciate pointing out parts needing improvement. Below are our point–by-point answers to the comments.

Anonymous Referee #1

Brus et al. measured the RH and temperature dependence of the diffusion coefficients of gaseous H2SO4, using a flow tube, and the performance of the experimental set-up is supported by CFD simulations. A much stronger effect of temperature, compared to theories, has been reported, and cluster kinetics modeling has been used to explain and interpret their experimental data. Diffusion coefficients of H2SO4 are of great importance in atmospheric chemistry, and the results reported by this work are interesting. Nevertheless, I do have a few major concerns which the authors should address before I can recommend this manuscript for final publication:

1) Line 384-386: I do not believe that the CFD simulation can confirm the assumption that the wall of the flow tube is an infinite sink. Actually this is a critical assumption already made in the model (line 166). The modeling result that [H2SO4] on the flow tube wall is less than 6% of [H2SO4]0 is determined by the spatial resolution used in the model; if a higher resolution is used, [H2SO4] on the flow tube wall will be closer to 0. A proper way to confirm this assumption is to measure the diffusion coefficients as a function of pressure in the flow tube, as described in previous work (Fickert et al., 1999; Liu et al., 2009). Though I believe that wall used by Brus et al. should be an infinite sink for H2SO4, these incorrect statements need to be changed and the proper way to confirm this assumption should be mentioned.

Ad 1) This is probably a misunderstanding and we will reformulate the text to clarify the issue. The mentioned 6 % of initial H_2SO_4 concentration on the wall is not a result of the simulations, but a boundary condition for the simulations in which we aimed to assess "how big mistake we could make in determining the diffusion coefficient if the wall is actually *not* acting as an infinite sink". Lines 193-201: we set as a boundary condition for the CFD model the H_2SO_4 concentration on the wall to be 0-100% of the initial H_2SO_4 concentration. We then investigated the change in the slope (ln([H_2SO_4]) vs. distance), and determined the change in the diffusion coefficient accordingly. We found that if [H_2SO_4] on the wall is up to 6 % of [H_2SO_4]$_0$ we are able to see only 10% change in the determined diffusion coefficient, which is also our experimental uncertainty. In other words, when the wall is emitting up to 6% of [H_2SO_4]$_0$, we are not able to recognize it in our experiment.

The method of diffusion coefficient measurements as a function of pressure is not suitable for our study. Generally, in systems including easily nucleating substances like H_2SO_4, a small change in the pressure can initiate strong new particle formation, i.e. secondary losses in the system. However, we will also mention other methods for testing the assumption of an infinite wall loss sink in flow tube experiments, as suggested by the reviewer.

2) I disagree with their proposed temperature dependence of H2SO4 diffusion coefficients (D). Examination of the experimental D at different T and RH shown in Figure 3 reveals that for the same RH, within the experimental uncertainties there is no significant difference between D measured at 278 K and at these measured at 288 K. The very strong dependence of D on T suggested by the authors are based on three data points at i) 298 K and 4% RH, ii) 288 K and 8% RH, and iii) 278 K and 26% RH (line 290-296). From i) to iii), T decreases and RH increases, both very likely leading to the decrease in D; in addition, Figure 3 shows that for RH below 15%, RH has a strong effect. One may conclude that instead of temperature, the change of RH can play the major role; therefore, the strong temperature dependence suggested by Brus et al. is not convincing if not wrong.

Ad 2) The suggested temperature dependency is not based on the three data points mentioned by the reviewer, but as stated e.g. in Table 1, as “unweighted averages over RH”, which is clarified also on lines 292-294: “The temperature dependency of the experimental diffusion coefficients was found to be a power of 5.4 for the whole dataset and temperature range. “

However, since one might find it incorrect or strange to include averages of the whole dataset, which indeed covers a different RH range for each temperature, we provide new values for the power dependency only for the range of RH that is covered at all three temperatures (15-70%) (please see Table I, last column and last row). The values in Table I in the manuscript will change as follows: the power dependency for the temperature range 278-288 K will change from 2.18 to 1.9, the value for 288-298 K will change from 8.7 to 9.4, and for the whole dataset and temperature range (278-298 K), the change is from 5.35 to 5.56. The manuscript text will change accordingly.

3) Even if their proposed temperature dependence is correct and can be justified, I feel this study is not complete or does not provide much insight with broad implications. A strong T dependence was found and can be explained by clustering with amines. However, the possible presence of amines is an experimental artifact. This manuscript has not yet answered the key question, i.e. the true dependence of D(H2SO4) on T, and thus currently may not be suitable for publication by ACP which requires studies with general implications for atmospheric science.

Ad 3) This manuscript had no ambition to answer the question of the “true dependence of D(H_2SO_4) on T”, i.e. the T dependence for the single H_2SO_4 molecule not bound to any other molecules, mentioned by the reviewer. We understand that the title might be misleading, and will thus change it to be as follows: Temperature-dependent diffusion of H_2SO_4 in air at atmospherically relevant conditions: laboratory measurements using laminar flow technique

First, it is very hard to find an environment in the atmosphere where pure H_2SO_4 would exist; even at high altitudes it is likely to be hydrated. As we state on page 3, lines 85-88 : “Such base impurities are unavoidably present also in our experiment, and most probably they originate from the humidification of the carrier gas (e.g. Benson et al., 2011; Kirkby et al., 2011; Neitola et al. 2015 already cited in the manuscript).”

Within the boundary layer, impurities like ammonia or amines are always present (e.g. Ge et al. 2011). The ranges of H_2SO_4 concentrations, RH, and temperature used in this study, as well as the concentration of impurities, represent typical ambient values and are atmospherically relevant. Thus we consider our study to be suitable for general implications in atmospheric science. We will also add the reference to Ge et al. (2011), and will add some further discussion on the evidence that in components that bound to sulfuric acid are likely present in any natural environments (e.g. Petäjä et al. (2011) already cited in the manuscript).

References

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