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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Numerical simulation of flow, H_2SO_4 cycle and new particle formation in the CERN CLOUD chamber" *by* J. Voigtländer et al.

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First of all, we thank the referee for the constructive and helpful comments and suggestions. To address all points, large parts of the manuscript were rewritten. Thereby, the discussion of the (unrealistic, since only theoretical) simple approach of the flat fan was left out. In the revised version, we focus on the arc shaped fan case, which was adjusted to measured flow field data of the CLOUD experiment.
Furthermore, we used nucleation rates published in Kirkby et al. (2011) for the particle dynamics simulation and compared simulated flow field data with a measured velocity profile for the current 2-fan set up. These data were not available when we submitted the first version of the manuscript. With these modifications,

we think the structure and the scientific relevance of the manuscript is significantly improved.

"In the abstract, as well as in the conclusions and at several other locations in the manuscript, it is stated that 'a 1-fan configuration, as used in first CLOUD experiments, may not be sufficient to ensure a homogeneously mixed chamber', and that 'to mix the tank properly, 2 fans are necessary'. This is concluded from the fact that the model results with the 1-fan-flat configuration could be well adjusted to the sulphuric acid data (Figs. 3 and 7), however did only badly represent the measured velocity profile. Therefore, the 1-fan-arc configuration was adjusted to match the velocity profile, but did a much worse job in reproducing the measured sulphuric acid data. "

It was not stated in the manuscript that the 1-fan arc shaped configuration "did a worse job in reproducing the measured sulphuric acid data". It is stated that, within calculated volume weighted standard deviations, the simulations are in agreement with the experimental data. The conclusion (2-fans are necessary to provide a good mixing) evolved because, with respect to simulated pattern of the flow field, H_2SO_4 standard deviations, and surface exchange times, the adjusted model indicate that only 1 half of the CLOUD chamber is influenced for a 1-fan set up. In the revised version, experimental data of the current 2-fan configuration compared to simulation results are included to underlay these results (see above).

"Also the standard deviation of the concentrations from the mean value is much broader for this configuration, from which the authors conclude that the mixing is not sufficient with such a fan configuration. As I think, such an insufficient mixing behaviour of the 1-fan configuration as used in the experiments is already indicated by the divergence of the velocity profile measured above the fan. This divergence could eventually markedly be reduced with an improved fan setup including a flow nozzle or a hood around the fan, as briefly mentioned on page 20016 of the manuscript. Such a modification would presumably approach the 1-fan-flat model configuration and therefore, according to the

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model results shown in the manuscript, results in the most homogeneous conditions."

We agree with the referee that the insufficient mixing of the tanks' contents can be already supposed by the divergent velocity profile of the non-housed 1-fan set up (CLOUD-1). To improve the efficiency of the mixing fans, hoods have been mounted for the current 2-fan set up (CLOUD-5). As shown in Fig. 1 (here) it was found that the effect of the hoods is only small. Measured velocity profiles of the "improved" 2-fan set up (blue dots) are very similar to the older ones (black dots). Numerical simulations suggest that the hoods increase the average velocity of about 30 percent. The low efficiency of the hoods can be explained by the overall small flow velocities in the chamber.
In conclusion the hoods do not result in the supposed improvement. The comparison of both configurations (hood - no hood) has been added to the revised version of the manuscript (chapter 3.2):
"To improve the efficiency of the mixing fans hoods were installed around each of them (current CLOUD-5 set up). However, measured velocity profiles above the mixing fans are quite similar compared to the 1-fan configuration without a hood (CLOUD-1, Fig.4(b)), indicating that the effect of the hoods is only small.
...results in Fig. 5(d) illustrate the simulated flow field for a 2-fan configuration adjusted to the improved CLOUD-5 set up, showing that the mixing fans still produce a divergent flow field. The low efficiency of the hoods is caused by the overall low flow velocities in the chamber."

"Furthermore nothing is said about the fan location close to the chamber bottom. Was this location optimised for achieving homogeneous mixing? "

In the manuscript, we give a short note about the location of the mixing fan(s) (p. 20016, l. 11-13). The mixing fan is located close to the chamber bottom (the fan axis is about 30 cm above the bottom of the tank). Designing the CLOUD chamber, the fan location was not optimised with respect to homogeneous mixing. The fan was positioned according to constructive aspects. The characteri-

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zation of the flow field and the mixing state was aim of the simulations shown in the manuscript.
Nothing was changed in the manuscript.

"All in all, it is completely unclear how the authors conclude from the results presented in the manuscript that only a 2-fan configuration provides good mixing in the chamber. "

It is, in principal, possible to achieve good mixing also for a 1-fan configuration as shown by the 1-flat-fan simulation or by the theoretical investigations in Schütze and Stratmann (2008). However, for adjusted fan settings the simulation results suggest that a 1-fan configuration does not provide a good mixing in the chamber, as the upper half of the tank is not influenced by the mixing fan. This holds also for the configuration of a housed fan because of the low flow velocities in the tank (see above).

"Unfortunately, no model results on the sulphuric acid distribution is shown for the 2-fan configuration compared to the 1-fan configuration. Such a comparison is only shown in Figure 12 for the gas temperature change after a wall temperature drop experiment. But again, the more interesting plot of the temperature distribution inside the chamber (lower panel) is only shown for the 2-fan configurations."

Calculated H_2SO_4 data for the 2-fan configuration has been included in the revised version. The comparison of measured and simulated H_2SO_4 lifetime data for a 2-fan configuration show that the volume-to-surface exchange is increased for a 2-fan set up (Fig. 2, here).
Furthermore, the lower panel in Fig. 12 (Fig. 8 in the revised version) shows the distribution for both, the 1-fan and the 2-fan configuration.
The results of the flat fan simulation were left out in the revised version.

"In summary I must state that the paper in its present form is not substantial enough for publication in ACP as a research article, both in terms of the scientific content and

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the balance between conclusions and presented results. I recommend to resubmit the paper, after some major revision, as a technical note paper. Below, some further recommendations and suggestions for revision are included."

For the experiments within this project the investigation of the mixing state in the CLOUD chamber is a fundamental issue. To our opinion, the manuscript should be placed in the special issue for the CLOUD experiments, especially as we think the paper's scientific content has significantly increased.

Specific comments and questions:

"In the introduction it is stated that large uncertainties in understanding the current climate change due to aerosols and clouds "partly result from solar-related contributions, such as the effects of galactic cosmic rays on aerosols and clouds". It is, however, not at all clear up to now whether ionisation through cosmic rays significantly affects the climate system. Therefore this sentence should be modified in a way to state that the role of cosmic ray ionisation for climate change is still unclear and deserves further investigation."

We followed this suggestion and changed the sentence:
"Largest uncertainties in understanding the current climate change are attributed to aerosols and clouds (IPCC2007). These uncertainties partly result from solar-related contributions and require further research. For example, still under discussion are galactic cosmic ray ionization effects on aerosols and clouds (e.g., Carslaw et al., 2002; Enghoff and Svensmark, 2008; Kirkby, 2007; Kulmala et al., 2010; Svensmark and Friis-Christensen, 1997).".

"p. 20015, l. 23: Why should a 1-fan configuration introduce large wall effects? What means large wall effects here and what are then the wall effects of a 2-fan configuration? Be more specific here."

We wanted to state that there is a strong back flow jet at the wall, which may

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influence the measurements. However, this sentence was deleted in the revised version.

"The discussion of the measured sulphuric acid profile on page 20019, last paragraph of Section 4.1.1, is rather unclear. First of all it would be necessary to also measure the measurement uncertainty here. Furthermore, a comparison of concentration profiles measured at different fan speeds would shed more light on the actual mixing situation in the chamber, eventually more than the modelling studies which are done under unrealistic conditions (at least those with the flat plane configuration, see discussion above). "

Concentration profiles measured at different fan speed are, in principal, available (100%, 50% and 25% runs). In the experiments it was found that the wall losses decrease with decreasing fan speed. Qualitatively, such a behavior can be also shown in the numerical simulations. However, for the lower fan speeds it is in our opinion not meaningful to adjust the model to the measured flow field. Therefore, we prefer to not discuss low fan speed simulations in the manuscript. Concerning the experimental uncertainty of the gaseous H_2SO_4 measurements, the following section was added to the manuscript (section 4.1.1):
"The experimental uncertainties of gaseous H_2SO_4 concentration measurements are about a factor of 2. On the other hand, observed short term fluctuations of the H_2SO_4 concentrations, which represent the combination of instrumental noise and local fluctuations in the small sampling volume, were much smaller (less than 20 percent, see Fig. 6). It can be concluded that the experimental uncertainties might influence the (initial) average H_2SO_4 concentration, but did not affect the temporal characteristics of the H_2SO_4 concentrations at the sampling point, as well as the results of the comparison with the modeling data."

"Additional experiments with stable aerosols added at one location and sampled at some others may also be helpful to get a better idea of the mixing situation in the chamber. "

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Experiments with stable aerosols are a good idea, but currently not available. Therefore, in the scope of this manuscript we concentrate on the volume-to-surface exchange. Internal mixing may be studied in future investigations. Nothing was changed in the text.

"Concerning the sulphuric acid data one could argue, that part of the concentration fluctuation is due to the inhomogeneous UV illumination and therefore an inhomogeneous sulphuric acid production rate. Then, after the UV is switched off, the sulphuric acid should approach a more continuous profile within the internal mixing time scale of the tank. This, however, does not seem to be the case, because the fluctuation pattern does not change much after UV off, both in Figs. 3 and 7. "

The fluctuations can be explained by instrumental noise and local fluctuations in the small sampling volume due to turbulent or diffusional mixing. They are relatively small and the data should be suitable for a comparison with the simulation results. It is beyond the scope of the manuscript to explain these fluctuations in detail.
Nothing was changed in the text.

"It would also be of interest to know here the 1/e reaction time scale of sulphuric acid formation in comparison to the internal transport and mixing time scale."

The 1/e reaction time scale could not be calculated because the H_2SO_4 formation rate is not calculated by means of a full chemistry model. We agree with the referee that a full chemistry model would be a better choice. But because concentrations of the precursor gases are several orders of magnitude larger than the H_2SO_4 concentrations and quasi constant during one experiment, it should be a suitable assumption to apply a constant production rate. Furthermore, it should be kept in mind that a fluid and particle dynamics simulation including full chemistry is computationally extremely expensive with the additional information gain being moderate in our opinion.

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Nothing was changed in the manuscript.

"Concerning the model runs discussed in Section 4.1.2 and later in the manuscript, it is not clear how the sulphuric acid formation was treated. Was it assumed to be formed at constant rate throughout the tank, or at constant rate only in the UV illuminated part of the tank, or proportional to the UV light intensity? "

It was written in the manuscript (p. 20022, l. 25-27) that a constant H_2SO_4 production rate was assumed for the UV illuminated part of the tank (Fig. 6 red and yellow).

"It would certainly be better to run a model with full UV-OH-SO2 chemistry. How realistic is the assumption of a constant formation rate? "

See discussion above.

"Also it is not clear in the discussion e.g. on page 20020, lines 18 to 22, whether the authors refer to the internal mixing time scale or the time scale of volume-to-surface exchange. I think both are different to each other."

We investigated the time scale of volume-to-surface exchange. We corrected this in the text. Internal mixing will be investigated in the future.

"Why were the model runs in Fig. 3 not started at the mean sulphuric acid concentration measured before time zero?"

We included a corrected model run in the revised version of the manuscript.

"p. 20021, l. 8-12: This is just one example of the inconsistency in the manuscript. If I understand right, the result of the 1-fan-flat model configuration, which is an ideal one and also not the one favoured throughout the manuscript, is used to argue, the the actual sampling location is representative for sulphuric acid measurements during CLOUD experiments. This is an odd conclusion, if finally a fan configuration is suggested in the manuscript which was neither used for the measurements nor the model

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runs compared here."

We think, the revised version of the manuscript is much clearer now. Large parts of the manuscript were rewritten. The (unrealistic) flat fan approach was left out of the discussion and the criticized argument was deleted. As also suggested by the referee we now discuss the flow field first.

"p. 20022, l. 1-9: I wonder why the authors did not first discuss the velocity profile, then use the configuration in agreement with the velocity profile to adjust the model to the sulphuric acid measurements, and based on that discuss possible improvements based on other fan configurations. I think the model should first be demonstrated to match all available measurements."

The model was adjusted to reproduce the measured flow field. After that no further adjustments are required and modeled and measured H_2SO_4 concentrations could be directly compared. This part of the manuscript was rewritten (chapter 3.2):
"One of the key parameters in the numerical simulations is a proper description of the mixing fans. As simulations on a 2-D grid do not allow a consideration of the rotating fan blades, the fans are represented by zero thickness pressure jump layers (e.g. Fluent User's guide). The pressure jump across the fan plane is described by a polynomial function dependent on flow speed. To provide an accurate representation of the flow field, pressure jump and shape of the fan planes have to be adjusted to experimental flow field data. For this study this was done by a comparison with a radial velocity profile determined 50 cm above the bottom mixing fan in the CLOUD chamber.
The measured velocity profile compared to the simulated one is shown in Fig. 4. Experimental data for a 1-fan configuration (no hood, CLOUD-1) are represented by the black dots (Fig. 4(a)), data for the current 2-fan configuration (CLOUD-5) are given by the blue curves (Fig. 4(b)). Figure 4(a) shows that 2-D simulations with the simplest approach of a flat disc shaped fan layer are not suitable to re-

produce the experimental data. The simulated jet above the fan (Fig.4(a), green line) was not observed in the experiments. In fact, the measured profile suggests a much more divergent velocity field. To match the measured velocity profile, the shape of the fan had to be changed to arc (as illustrated in Fig. 2, velocity profile shown in Fig.4(a), red line). To improve the efficiency of the mixing fans hoods were installed around each of them (current CLOUD-5 set up). However, measured velocity profiles above the mixing fans are quite similar compared to the 1-fan configuration without a hood (CLOUD-1, Fig.4(b)), indicating that the effect of the hoods is only small.

In Fig. 5 cross sectional profiles of the velocity magnitude are presented. The data visualize the jet above the flat fan (Fig. 5(a)) and the much more divergent flow field of the adjusted arc-fan simulation (Fig. 5(b)). Furthermore, it is shown that the upper half of the tank is almost not influenced by the arc shaped fan, as the velocity is almost zero in this part of the chamber. The turbulent intensity around the fan is much larger than for the flat fan approach, but turbulent mixing is limited to the region next to the fan (not shown here). It can be concluded that to mix the tank properly under realistic conditions (arc fan) there is a need for a second fan. In agreement, the simulated flow field for a 2-fan configuration indicates that the whole tank is mixed by such a set up (Fig. 5(c)). Finally, results in Fig. 5(d) illustrate the simulated flow field for a 2-fan configuration adjusted to the improved CLOUD-5 set up (with hoods), showing that the mixing fans still produce a divergent flow field. As already stated above, the low efficiency of the hoods is caused by the overall low flow velocities in the chamber."

"Figures should be included in the same order as first mentioned in the text. I did not find Fig. 10 mentioned in the text body."

Fig. 10 was left out of the revised version.

Minor points and technical corrections:

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"p. 20014, l. 10: . . . mixing state of the tanks content largely . . ." "p. 20014, l. 25: Do you mean attributed instead of contributed? "

We corrected these sentences.

"p. 20016, l. 9-12: Should be rephrased " "p. 20016, l. 13: Suggest to refer here to the thin lines in Fig. 2 to better identify the fan location."

We followed the suggestion of the referee and changed these lines to (chapter 2):
"The cylindrical CLOUD chamber, located at CERN, is an electro-polished stainless steel tank with a diameter of 3.0 m, a height of approx. 4.0 m and a corresponding volume of 26.1 m^3. The tank was designed, after a pilot experiment (Duplissy et al., 2010), with respect to achieve highest standards of cleanliness and temperature stability (Kirkby et al., 2011). A schematic diagram of the chamber is shown in Fig. 1. Different inlets and outlets at the chamber wall can be used to connect sampling probes, to introduce trace gases into the chamber, and to evacuate the chamber. To continuously mix the tank's contents, two fans can be installed next to the flanges at the top and the bottom (thin black lines in Fig. 2). ..."

"p. 20017, l. 5: I guess you mean the samples, not the sampling probes, to be representative for the whole tank volume."

We corrected the sentence.

"p. 20018, l. 3: How thick is the laminar boundary layer of the tank at certain fan speeds? Would be good to mention that somewhere in the manuscript."

The sentence was corrected to (chapter 3.1):
"Utilizing a near wall approach means that the laminar sublayer with a thickness of about 10^0 mm has to be resolved by the numerical grid. Accordingly, a grid with about 20000 grid cells (Fig. 2) was generated and applied for the simulations

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shown here."
The total thickness of the boundary layer is about 10-20 cm centimeters (Fig. 3, here). In the experiments it was ensured that the samples were taken outside of this layer. We added to the manuscript (section 4.1.1):
"... The subsequent decrease of H_2SO_4 due to the transport to the wall was recorded outside of the boundary layer (thickness about 10 to 20 cm) at one sampling line of the tank. ..."

"p. 20018, l. 10-15: Is the size and type of the fan somewhere mentioned in the manuscript? What is the diameter of the model fan planes? Was the pressure drop assumed to be constant throughout the fan plates."

The type of the fan is not given in the manuscript as different fan types were used for the different campaigns (4-blade, 8-blade). On the other hand, the type of the mixing fan is meaningless for the simulations because in the model only a parametrization of a zero thickness pressure jump layer is used, which has to be adjusted to experimental flow field data. As mentioned in the manuscript, the pressure drop was not assumed to be constant throughout the fan plates. In fact, a polynomial fit function dependent on the velocity magnitude was used. We changed the respective sentence in the text (chapter 3.2):
"The pressure jump across the fan plane is described by a polynomial function dependent on flow speed."

"p. 20019, l. 3: What is the relation between the pressure drop and the fan speed? Was the velocity profile be measured for different fan speeds, and would the 1-fan-arc configuration represent different actual fan speeds, and match the actual velocity profiles, just by adjusting the δp?"

The relation between calculated flow velocity and pressure drop at the fan layer is a polynomial function. Considering a 2-D simulation both, the fan shape and the pressure jump have to be adjusted to corresponding experimental data (see

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above).
Nothing was changed in the text.

"p. 20019, l. 17: . . . time axis was set to . . ."

We modified this sentence (section 4.1.1):
"Thereby, the time axis was shifted so that the H_2SO_4 decrease starts at the zero line."

"p. 20021, l. 19/20: Measurements, if accurate enough, always should reflect the real picture."

"p. 20024, l. 11: It is unclear here what the back flow jet actually is and haw it acts on the wall exchange."

These sentences were deleted in the revised version.

"p. 20024, l. 25 - 28: This paragraph needs to be re-written."

This part of the manuscript was completely rewritten (chapter 3.2, see above)

"p. 20026, l. 9: Would be good to also show the temperature deviations for the flat fan configuration."

The detailed flat fan discussion was deleted.

"p. 20026, l. 21: Again, do you mean the internal mixing time or the wall exchange time? I guess the internal mixing time scale is somewhat shorter but is not shown here. The exchange time between the mixed volume and the walls can already be taken from the sulphuric acid decrease in Fig. 3. The transport time for sulphuric acid should be somewhat longer because of the larger molecular weight and thereby the smaller diffusion coefficient compared to heat transport."

We investigated wall exchange times (see above).

"p. 20028, l. 15: Is it a realistic assumption to neglect the Kelvin term here?"

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We are aware that the growth law applied for the simulations is a simplified one, as a kinetic description with respect to water is neglected, the concentration/vapour pressure of H_2SO_4 is assumed to be zero and the ratio of wet to dry particle diameter is calculated neglecting the Kelvin term. However, the growth law was already used in this form elsewhere (Herrmann et al., 2010). To us, it is an appropriate tool for the investigations of the mixing state shown in the manuscript. But we agree with the referee that for future investigations the description of particle growth can be improved.
Nothing was changed in the text.

"p. 20029, l. 1 - 17: It is nice to see the model can somehow describe particle formation and growth. But I do not see the results to provide additional information on the mixing state of the tank without additional particle measurements. It seems obvious that the sulphuric acid particles should at least be as well mixed as the precursor gases."

To increase the scientific value of the revised version, we included nucleation rates recently determined in the CLOUD experiments (Kirkby et al., 2011; see above) into the particle dynamics simulation.
To us it is not so obvious that such simulations do not provide additional information, as the dependence between precursor concentration and particle number distribution is highly non-linear. Furthermore, the developed model and the presented results represent a valuable contribution to ongoing and future data analysis within the CLOUD project. We did not follow the suggestion of the referee to delete the particle simulation.

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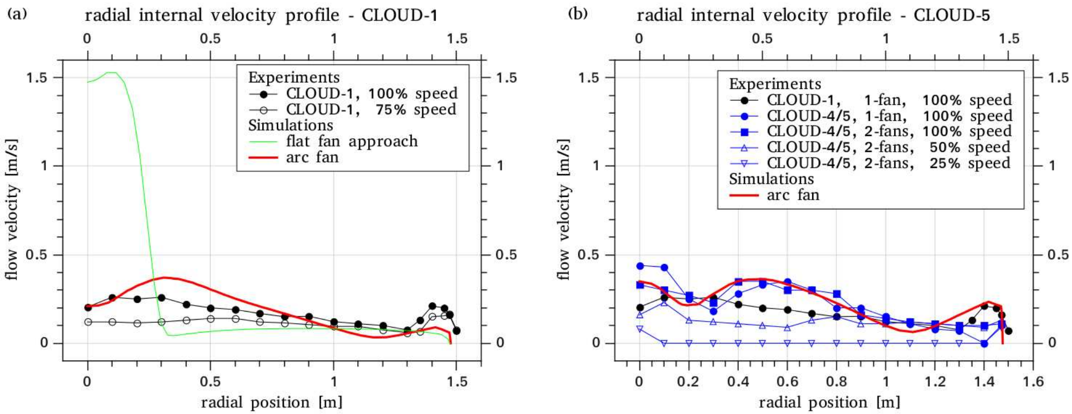

Fig. 1. Measured internal velocity profile 50 cm above the fan measured for the current CLOUD-5 set up.

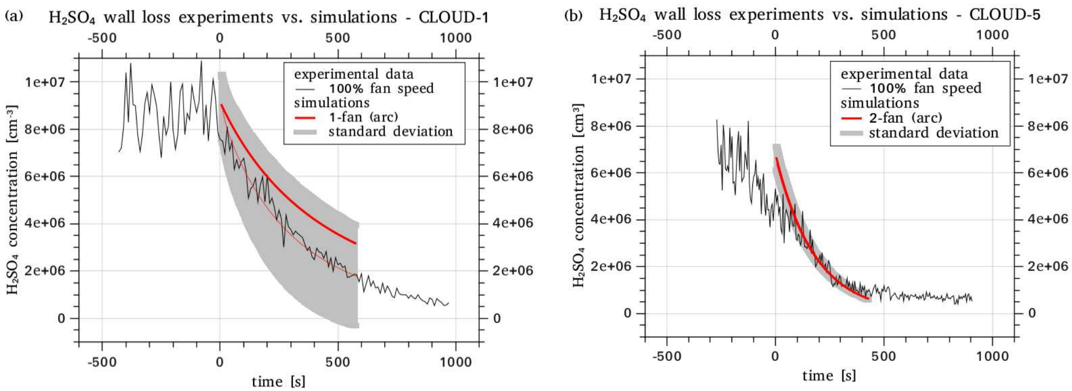

Fig. 2. Sulfuric acid lifetime experiments compared to numerical simulations

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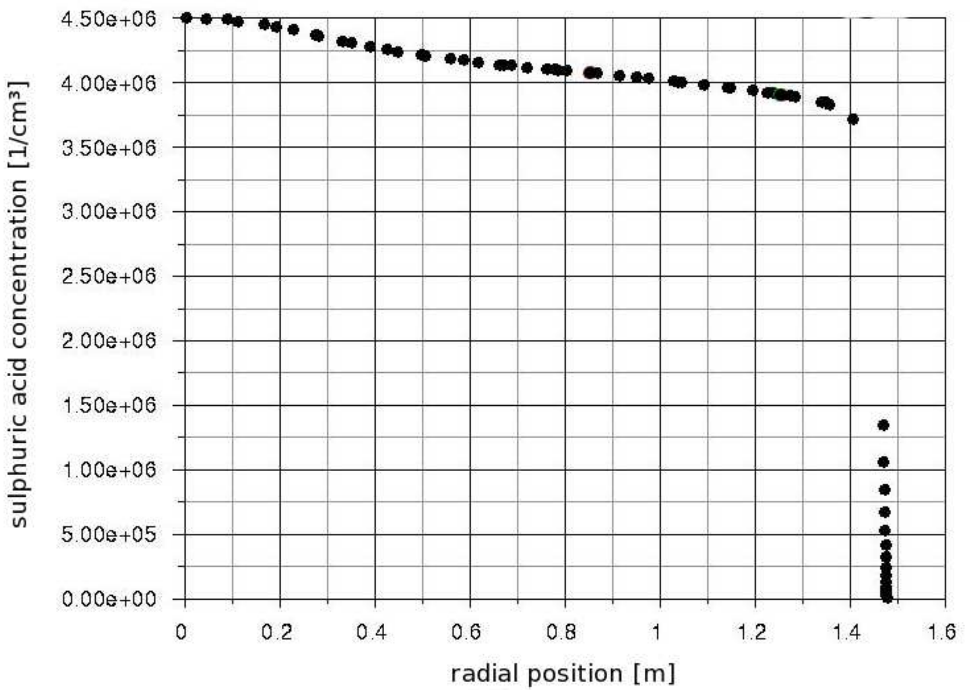

Fig. 3. Example of the calculated sulfuric acid concentration in radial direction (at mid height of the tank). Measurements have to be done outside of the boundary layer (thickness about 20 cm).

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