

***Interactive comment on* “Numerical simulation of flow, H₂SO₄ cycle and new particle formation in the CERN CLOUD chamber” by J. Voigtländer et al.**

Anonymous Referee #3

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This paper comprises fluid dynamic modelling studies of the mixing situation in the novel CERN CLOUD chamber. The model results are adjusted to or compared with two sets of measurements, namely the flow velocity profile above the mixing fan inside the chamber and the time series of the sulfuric acid concentration measured at the sampling port location as specified in the manuscript. In the modelling studies, different fan configurations are compared to each other: a 1-fan configuration with just one fan close to the bottom of the chamber driven in the model with either a flat (1-fan-flat) or an arc-shaped (1-fan-arc) pressure drop plane, and a 2-fan configuration with a second fan close to the top of the chamber, both fans also driven either with a flat or an arc-shaped pressure drop plane.

In the abstract, as well as in the conclusions and at several other locations in the

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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. 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 model results shown in the manuscript, results in the most homogeneous conditions. Furthermore nothing is said about the fan location close to the chamber bottom. Was this location optimised for achieving homogeneous mixing? 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. 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.

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 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

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recommendations and suggestions for revision are included.

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.

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.

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). 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. 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 sulfuric 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. It would also be of interest to know here the $1/e$ reaction time scale of sulphuric acid formation

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in comparison to the internal transport and mixing time scale.

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

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

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 favored 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 runs compared here.

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.

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.

Minor points and technical corrections

p. 20014, l. 10: ... mixing state of the tanks content largely ...

p. 20014, l. 25: Do you mean attributed instead of contributed?

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.

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

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.

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.

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 ?

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

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 how it acts on the wall exchange.

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

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

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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 is should be somewhat longer because of the larger molecular weight and thereby the smaller diffusion coefficient compared to heat transport.

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

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 20013, 2011.

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