Atmos. Chem. Phys. Discuss., 11, C14656–C14662, 2012
www.atmos-chem-phys-discuss.net/11/C14656/2012/

Atmospheric Chemistry and Physics Discussions

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.

J. Voigtländer et al.

jensv@tropos.de

Received and published: 25 January 2012

First of all we would like to thank referee 1 for his constructive comments and suggestions. In the following the comments will be addressed and discussed.

General comments:

"The authors present CFD-FPM simulations results of mixing state together with particle nucleation and growth in 26 m^3 CLOUD-09 chamber situated at CERN (Switzerland). The manuscript is structured and also reads well. However, it is not very convincing about its scientific significancy in its current form. "

To our opinion, the developed model and the presented results represent a valuable contribution to ongoing and future data analysis within the CLOUD project.

However, to address all comments and suggestions of the referees, large parts of the manuscript were rewritten. With the modifications, we think the structure and the scientific value of the manuscript is significantly increased.

"The authors found that one-fan configuration is not ideal, the flat shaped fan copies well the experimental data obtained from H_2SO_4 loss experiment but does not reproduce the velocity profile experiment. The arc shaped fan does not reproduce well H_2SO_4 loss experiment (either temperature jump sims) but can reproduce velocity profile measurements. Finally, they concluded that two-fan (flat or arc) configuration is favourable to obtain well mixed tank. Since the author list shares names with current publication of Kirkby et al., (2011) one would expect direct comparison to data published in there, it is pity this would significantly increase scientific value of the manuscript. Of course it is understandable that each manuscript has its own history."

The mentioned paper (Kirkby et al., 2011) was not published when we submitted our manuscript. However, we agree with the referee that actual data should be included and performed additional particle simulations applying nucleation rates presented in Kirkby et al., (2011) for the revised version of our manuscript. Furthermore, we now also present results concerning the flow field and H_2SO_4 cycle of the actual set up with 2 mixing fans, and we omitted the discussion of the unrealistic (since only theoretical) approach of the flat fan.

Specific comments:

"1. 4.1.2 Simulation results, page 20020, lines 6-17, concerning diffusion coefficient. This part is bit confusing, can authors make this part clear and be bit more specific, what value of diffusion coefficient and what method of estimation they have actually used for the experimental conditions (T=291.65 K, RH=38%)? How their estimate of DC compares to others, for example Fuller method?"

As the reviewer suggested, we included DC values derived with different methods. The paragraph (section 4.1.2) was rewritten to:

"Furthermore, diffusion coefficients of 0.09 cm^{2}s^{-1} (H_2SO_4 in air) and 0.06 cm^{2}s^{-1} (H_2SO_4 in H_2O) were applied in the simulations (Marti et al., 1997; Hanson and Eisele, 2000; Brus et al., 2010). These values are comparable to to values determined using the methods of Fuller (FSG, Fuller et al., 1966), FSG-LaBas (Lymann, 1993) or Wilcke and Lee (WL, Wilke and Lee, 1955), which yield 0.11 cm^{2}s^{-1}, 0.093 cm^{2}s^{-1} and 0.1 cm^{2}s^{-1}, respectively."

"2. Just from curiosity, did the authors try another approach to obtain DC? For example, to keep DC as a free parameter and by iteration to find the right value of DC that fits experimental observations?"

It is correct that the DC determines the wall losses. Varying DC will change the temporal decrease of H_2SO_4 obtained from the simulations (Fig. 1). The larger the DC, the larger the wall losses of H_2SO_4. However, the differences are not very large for values in the range of literature data (0.09 cm^{2}s^{-1} - 0.11 cm^{2}s^{-1}, see above and Fig. 1). Due to this low sensitivity, fitting DC does not seem meaningful. Consequently, we applied DC from literature for our simulations.
Nothing was changed in the text.

"3. Since authors consider only first-order loss to wall, what particle number concentration would be critical for their system to find secondary loss of H_2SO_4 to particles significant? What is the actual measured and simulated wall loss factor (WLF)? How the obtain WLF compares to other studies concentrating H_2SO_4 - H_2O nucleation measurements?"

Experiments measuring sulfuric acid lifetime have been done at comparably low H_2SO_4 concentrations (typically lower than about 10^7 cm^{-3}). Comparing with data published in Kirkby et al., (2011) nucleation rates are below 10^{-4} cm^{-3}s^{-1} to 10^{-5} cm^{-3}s^{-1}, while the time scale of the sulfuric acid lifetime experiments was in the order of 10^3 s. Under such conditions, considering only first order H_2SO_4 loss to the wall is a reasonable assumption. For the particle dynamics simulations at higher H_2SO_4 concentrations, H_2SO_4 depletion due to condensational growth of the particles is considered in the CLOUD-DPM model. Addressing the referee's concerns we slightly modified the corresponding section in chapter 3.1:
"Both models together form the so-called CLOUD-FPM, a model being capable of handling the coupled fluid and particle dynamical processes taking place inside the CLOUD chamber. In CLOUD-FPM, all relevant properties like velocity, temperature, pressure, turbulence parameters, composition of gas/particle phase, and nucleation/growth of ultra-fine aerosol particles are treated explicitly. The coupling of the models also includes the consideration of mass transfer between gas and particle phase due to particle nucleation and condensational growth."
and included the following sentence into chapter 4.2.:
"Assuming a H_2SO_4 production rate of 2.5$\times$10^6 cm^{-3}s^{-1}, the resulting maximum H_2SO_4 concentration was about 5$\times$10^8 cm^{-3} (see Fig. 9(a)). After reaching its maximum, the average gaseous H_2SO_4 concentration decreases again due to the increasing amount of H_2SO_4 transferred to the particle phase by particle nucleation and condensational growth."

"4. 4.5 Simulation of particle nucleation and grow, page 20028, lines 20 and further. The authors speculate about unidentified condensable vapours and insufficient growth rates at very low H_2SO_4 concentrations, why?? Again if they would directly use and compare to data from Kirkby et al (2011), where H_2SO_4 concentrations are for "'pure"' experiment about 5x10^8 cm^{-3} and nucleation rate 1/ccm/s, they could avoid any speculation."

As mentioned above, data from Kirkby et al. (2011) were not published when we submitted our manuscript. For the revised version we performed additional particle dynamics simulations with nucleation rates published in Kirkby et al., (2011), included the results into the manuscript and changed the text accordingly (chapter 4.3):
"For simulations shown in this study, A and K were adjusted to data pub-

lished in Kirkby et al. (2011) (neutral case) resulting in values A = 3 and K = 0.8 $\times$ 10^{-26} cm^{3}s^{-1}.
...
According to Kirkby et al. (2011), particle nucleation rates were up to about 1 cm^{-3}s^{-1} (Fig. 9(b)). Volume average particle diameters plotted in Fig 9(c) show that particle growth rates up to 25-30 nm/h were calculated."

Technical corrections:

"Abstract, page 20014, line 7, the abbreviation "'FPM"' is not explained in (CLOUD-FPM), the first appearance of explanation is on page 20017, line 8."

The name of the model was deleted in the abstract of the revised version.

"Introduction, page 20015, line 22, "but smaller (12 m^2)", probably should be"(12 m^3)"."

We corrected this mistake.

"Introduction, page 20016, line 2, the abbreviation "FPM" is not explained in (CLOUD-FPM)."

We slightly modified this phrase to:
"Simulations were carried out using a coupled computational fluid dynamics (CFD) - particle model called CLOUD-FPM"

"4.2. Cross section profiles, page 20024, lines 16 and 17, "Cross section profiles for the arc shaped fan,. . ., in Fig. 8 (velocity) and Fig. 9 (turbulent intensity)", should be "Cross section profiles for the arc shaped fan,. . ., in Fig. 10 (velocity) and Fig. 11 (turbulent intensity)"."

The Figs. were modified in the revised version.

"page 20029, line 4, ". . .about 6 cm^{-3}s^{-3}. . .", should be ". . .about 6 cm^{-3}s^{-1}. . .""

We corrected this mistake.

"References: Kirkby et al: Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation, Nature 476, 429-433, doi:10.1038/nature10343, 2011."

We included this reference and applied nucleation rates published in Kirkby et al., (2011) in our particle simulations.

References

Fuller, E.N., P.D. Schettler and J.C. Giddings, A new method for prediction of binary gas-phase diffusion coefficients , *Ind. Eng. Chem.*, *58 (5)*, 18–27, 1966.

Kirkby, J., et al., Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation., *Nature*, *476*, 429–433, doi:10.1038/nature10343, 2011.

Lyman, W.J., W.F. Reehl and D.H. Rosenblatt, *Handbook of chemical property estimation methods*, American Chemical Society: Washington, DC (USA), 1990.

Wilke, C.R. and C.Y. Lee, Estimation of Diffusion Coefficients for Gases and Vapors , *Ind. Eng. Chem.*, *47 (6)*, 1253–1257, 1955.

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

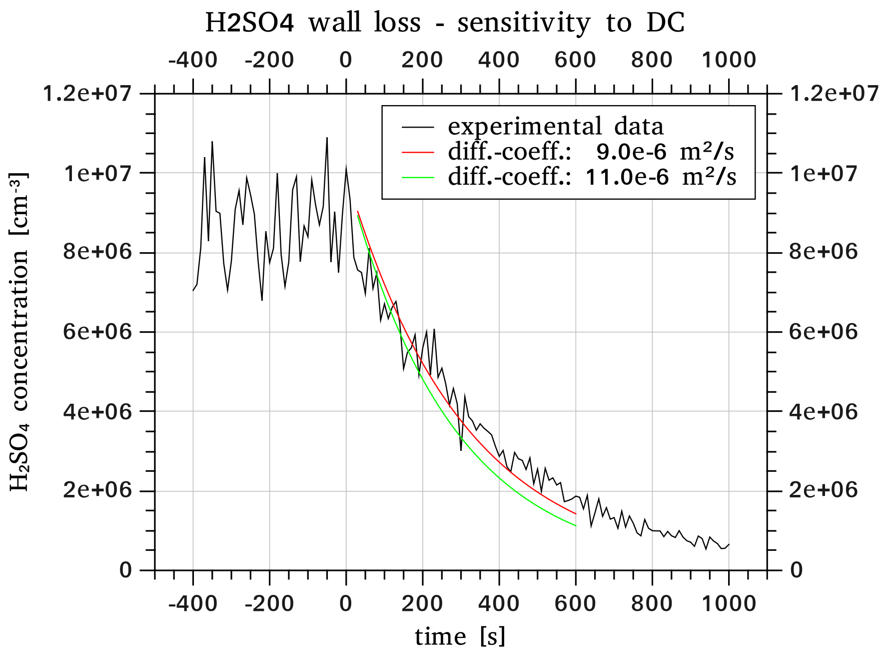

Fig. 1. Sensitivity of the sulfuric acid wall loss to different sulfuric acid diffusion coefficients.