Review of "libcloudph++ 1.1: aqueous phase chemistry extension of the Lagrangian cloud microphysics scheme" by Jaruga and Pawlowska (GMD-2018-96)

The manuscript describes and validates the addition of aqueous sulfur chemistry to the *libcloudph++* Lagrangian cloud model (LCM) (Arabas et al., 2015), which are a new class of recently developed highly detailed microphysical models. The presented test cases are an adiabatic parcel simulation based upon the intercomparison by Kreidenweis et al. (2003) and a kinematic two-dimensional stratocumulus study. This manuscript presents a profound description of consideration of aqueous sulfur chemistry in the *libcloudph++* LCM, which will allow much more sophisticated investigations than with earlier models. (However, the manuscript misses the opportunity to demonstrate all possible advantages of this great new development.) All in all, the manuscript fits the scope of Geoscientific Model Development and I recommend its publication once my comments are addressed.

A Slightly Major Comment

P. 8, l. 2 - p. 9, l. 7, Fig. 1: The liquid water content is one of the most important parameters to steer aqueous sulfur chemistry (and probably in cloud physics in general). Therefore, it is necessary that the same liquid water content is reproduced by the presented model to validate it with the intercomparison results of Kreidenweis et al. (2003), in which seven other models are capable of reproducing the same liquid water content. The authors state that the reason for this discrepancy results from the unspecified pressure profile in the study of Kreidenweis et al. (2003). However, there might be another potential source for the deviation: it could be the slightly too high initial temperature in the current study, which is stated as 285.5 K (Tab. 1 of the manuscript) but 285.2 K in Kreidenweis et al. (2003, their Tab. 3). Moreover, it might be helpful to clarify the calculation of the pressure (and density) profile by using equations (the description on p. 8, ll. 21-29 is hard to follow). Finally, why does the axis stating the height above cloud base start with 100 m but the time above cloud base with 0 s (Fig. 1)?

Minor Comments

Introduction: Many previous models (so-called "moving bin" or "moving grid" models) used a Lagrangian framework for the representation of aqueous sulfur chemistry as done in this study. Accordingly, they are based on similar (sometimes identical) model equations as the described in the current manuscript. As the LCM, these "moving bin" models do not suffer from numerical diffusion (e.g., p. 4, ll. 7-8). However, these models are only applicable in simplified parcel or trajectory simulations (e.g., Feingold et al., 1998; Kreidenweis et al., 2003). The authors should make clear that their main advancement is the possibility to use a Lagrangian representation of aqueous sulfur chemistry in multidimensional simulations with a fully coupled dynamics model, which hasn't been possible before.

P. 2, ll. 8-24: Please rearrange/rewrite these two paragraphs. Especially the first paragraph gives no real reasons why "aqueous phase oxidation of sulfur" is an important chemical reaction, it only states it is a very common reaction. Moreover, there are many interesting points in these two paragraphs but it would be nice to connect them more closely with the presented study/model.

P. 2, ll. 28–29: What about the collection of submicron aerosol by droplets (e.g., Ardon-Dryer et al. (2015), Ladino et al. (2011))? These processes are not considered in the current model but also affect the size of aerosols.

P. 3, l. 2: What is meant by an "atmospheric aerosol particle" in this context? There must certainly be a size restriction to the cited statement since very small aerosols are not cloud-active.

P. 3, ll. 5-6: The effect of collision and coalescence on the amount of aerosol inside a cloud droplet (in absence of aqueous sulfur chemistry!) has been considered in an LCM framework before (see Hoffmann, 2017).

P. 4, ll. 11-12: This is a vague statement since it depends on the equations considered and the applied numerical solvers.

P. 5, l. 2: How does the wind speed affect the chemical scheme?

P. 5, ll. 8-13: Does this restriction affect the results? And if yes, how? And what happens to giant and ultra-giant aerosols, which remain less diluted throughout their lifecycle?

P. 5, ll. 20-22: In what aspect is the LCM beneficial? Using a low number of super-droplets, the LCM will suffer from the same problems as a “moving bin” scheme.

P. 5, Eq. (1): For a reaction $ab \rightleftharpoons a + b$ dissociation constant is defined as $\mathbb{K}_{ab} = [a][b]/[ab]$. Accordingly, Eq. (1) is just a relationship to adjust a measured dissociation constant $\mathbb{K}_{ab}$ to a given temperature. Denote the meaning of T_0 and R.

P. 6, Eq. (2): I understand that this equation is based on the neutral charge condition. For clarity, it might be necessary to comment on how the expressions for the individual terms are derived. (Or state an appropriate reference.)

P. 6, ll. 15-16; p. 7, l. 15: How is this “small” timestep determined? Is there a criterion? How does the user know that an appropriate timestep is chosen?

P. 7, ll. 26-28: Why is H_2SO_4 used instead of S^{IV} to denote sulfuric acid? Please clarify.

P. 8, ll. 4-5: What is meant by “initial conditions above supersaturation”?

P. 8, ll. 15-20; p. 12, Fig. 3: How is the aerosol distribution transformed to superdroplets? Is the distribution binned and a superdroplet represents each bin or is a random generator used that follows the given distribution? This is in important information to understand the slight fluctuations in the initial spectrum (Fig. 3).

P. 10, ll. 9-10: How is it possible to have activated particles at a relative humidity of 100 %? To activate a droplet, a *super*saturation (RH>1) is necessary.

P. 13, ll. 3-4: The Hall (1980) collection efficiencies are not very suitable for big droplets since they neglect the effect of a coalescence efficiency of less than unity. How are “big” droplets defined?

P. 16, Fig. 4b: What is meant by “spec.” in the panel‘s title?

P. 18, l. 5: Eq. (5) shows that the oxidation of H_2O_2 is not independent of the acidity ($[H^+]$ in the denominator). (Although the dependence might be very weak.)

P. 18, l. 19-20: “This is because … aerosol particles.” Is this a direct result of Eq. (2)?

Technical Comments

P. 1, ll. 20-21: “clearly defined user interface” is a slightly subjective statement.

P. 1, l. 21: Make clear that “separation of concerns” is a software design principle.

P. 2, l. 17: Define IPCC.

P. 2, ll. 33-34: All these references should be in one pair of parentheses.

P. 3, l. 20-22: As a service to the reader, it would be nice to state how many new attributes are added for the consideration of aqueous sulfur chemistry.

P. 4, l. 15: The citation is "Lee et al. (2014)" not "Junghwa et al. (2014)".

P. 4, ll. 22-23: The pH is a scale used to quantify the acidity of a solution. It is (linguistically) awkward to keep track of a scale and not the underlying physical meaning.

P. 6, Eq. (2): The underbrace needs to include all terms on the right-hand side of the equation.

P. 10, l. 4: These values are stated in Tab. 2 of Kreidenweis et al. (2003).

P. 12, ll. 7-8, 15: Semicolons in an enumeration of references are unusual.

P. 13, l. 21: "the the"

P. 15, l. 27, 29: Is Fig. 4f addressed of Fig. 4c?

References

Arabas, S., Jaruga, A., Pawlowska, H., and Grabowski, W. W. (2015). libcloudph++ 1.0: a single-moment bulk, double-moment bulk, and particle-based warm-rain microphysics library in C++. Geoscientific Model Development, 8(6), 1677.

Ardon-Dryer, K., Huang, Y. W., and Cziczo, D. J. (2015). Laboratory studies of collection efficiency of sub-micrometer aerosol particles by cloud droplets on a single-droplet basis. Atmospheric Chemistry and Physics, 15(16), 9159-9171.

Feingold, G., Kreidenweis, S. M., and Zhang, Y. (1998). Stratocumulus processing of gases and cloud condensation nuclei: 1. Trajectory ensemble model. Journal of Geophysical Research: Atmospheres, 103(D16), 19527-19542.

Hall, W. D. (1980). A detailed microphysical model within a two-dimensional dynamic framework: Model description and preliminary results. Journal of the Atmospheric Sciences, 37(11), 2486-2507.

Hoffmann, F. (2017). On the limits of Köhler activation theory: how do collision and coalescence affect the activation of aerosols?. Atmospheric Chemistry and Physics, 17(13), 8343-8356.

Kreidenweis, S.M., Walcek, C.J., Feingold, G., Gong, W., Jacobson, M.Z., Kim, C.H., Liu, X., Penner, J.E., Nenes, A., and Seinfeld, J.H. (2003). Modification of aerosol mass and size distribution due to aqueous-phase SO2 oxidation in clouds: Comparisons of several models. Journal of Geophysical Research: Atmospheres, 108(D7).

Ladino, L., Stetzer, O., Hattendorf, B., Günther, D., Croft, B., and Lohmann, U. (2011). Experimental study of collection efficiencies between submicron aerosols and cloud droplets. Journal of the Atmospheric Sciences, 68(9), 1853-1864.

Lee, J., Noh, Y., Raasch, S., Riechelmann, T., and Wang, L. P. (2014). Investigation of droplet dynamics in a convective cloud using a Lagrangian cloud model. Meteorology and Atmospheric Physics, 124(1-2), 1-21.