Report 1: We thank Referee n°1 for his suggestions and comments which helped improving the manuscript. Comments are addressed point by point below. The manuscript was originally dedicated to GMDD, but got reoriented to ACPD by the editor.

**Comment 1**: My main concern is that the scientific significance of this study has not really been put into context. The section Introduction focuses on explaining the choices for the methods applied in the model framework. It would be more important to explain in Introduction: what are the scientific questions of this study and what is the significance of this topic?

**Reply 1**: As mentioned above, our manuscript was originally addressed to GMDD as it mainly describes the coupling between the chemical mechanism CLEPS 1.0 and a warm cloud microphysical scheme. This explains the technical aspects of the introduction. Following the reviewer's comment, the introduction has been rewritten in order to better highlight the scientific significance of our work:

- The most technical points related to the developments we performed were moved to Sections 2.1 and 2.2;
- Processes related to in-cloud aqueous chemistry are now mentioned, including a more explicit description of those contributing to aqueous concentrations, i.e. nucleation scavenging of particles, exchange of gases between air and droplets through mass transfer and aqueous reactivity. In addition, the use of "transfer" for both particle ("particle-to-cloud transfer") and gas phase ("mass transfer") related processes might be confusing. We have thus changed the terminology throughout the manuscript. The contribution of the particulate phase is now referred to "particle (nucleation) scavenging", while, following the common practice, the exchange between the gas phase and the droplet is still referred to as "mass transfer".
- The importance of aqueous cloud chemistry with respect to global atmospheric chemistry and climate was highlighted.
- The strength of the model regarding its capacity to estimate to contribution of the aforementioned sources to the simulated aqueous concentrations was further stressed, as this information is highly valuable since it cannot be obtained from measurements as stated by Leriche et al. (2007).

**Comment 2**: Page 2, Lines 1-2: "As a first step, CLEPS 1.0 has been integrated in a box model that takes into account neither aerosol particles nor microphysical processes, and thus only allows for the simulation of idealized cloud events" I don't understand what this sentence means? If the box model does not take into account the aerosol particles or microphysical processes, how does it differ from a standalone version of the chemistry model? Has this first step been a part of the work for this manuscript or is there a citation missing?

**Reply 2**: In its first version developed by Mouchel-Vallon et al. (2017), CLEPS 1.0 was limited to the simulation of cloud events with constant microphysics, i.e. constant droplet radius and liquid water content were considered throughout the simulation. In particular, the formation of the cloud through the activation of an aerosol spectrum was not simulated, nor the effect of these particles on cloud chemistry through nucleation scavenging, neither were the microphysical processes determining the evolution of the droplet spectrum. We agree with the reviewer that the abovementioned sentence was confusing, and we thus removed it in the revised version of the manuscript. Instead, we only focus on the strength of the new coupled model: "This paper describes the coupling between the chemistry model based on CLEPS 1.0 and a bulk two-moment warm cloud microphysical scheme allowing for the simulation of cloud events and comparison with long-term observations."

**Comment 3**: Page 2, Line 7: This phrase is very ambiguous "the impact of aerosol particles on the cloud chemistry (by nucleation scavenging)" and it should be clarified, what impact is actually meant.

**Reply 3**: As previously mentioned (Reply 1), the processes contributing to aqueous concentrations are now more explicitly described in the revised version of the introduction. For instance, the sentence mentioned by the reviewer was changed to: "The development of such a coupled model first offers the opportunity to investigate the contribution of the particles serving as cloud condensation nuclei (CCN) to aqueous concentrations of given species by nucleation scavenging, which control the pH of the droplets and further affect oxidation processes (Leriche et al., 2007; Hegg, 2001)."

Hegg, D. A.: The impact of clouds on aerosol populations, IGAC Activ. Newsl., 23, 3-6, 2001.

**Comment 4**: Page 2: It is said that the calculation of chemistry in a sectional cloud parcel model would be computationally too expensive. With current computing resources this sounds strange. What kind of CPU time does CLEPS require for the runs in this study?

**Reply 4**: According to Khain et al. (2000) and Flossmann and Wobrock (2010), several tens of bins are needed in a sectional cloud parcel approach to provide a satisfying description of the cloud. We agree with the reviewer that such an approach would still be fine in the current version of our 0D coupled-model, since the runs performed in the frame of this study on average require a CPU of 49 min (for 2 hours of simulation with a time step of 0.1 second). However, the final goal of our work is to implement our developments in a regional model, for which the sectional approach would definitely not be optimal. This is now clearly stated in the revised version of the manuscript: "They are however computationally too expensive to be used with detailed explicit aqueous-phase chemistry such as that described in CLEPS 1.0 as we aim in the future to include our developments in regional climate models."

Khain, A., Ovtchinnikov, M., Pinsky, M., Pokrovsky, A., and H. Krugliak.: Notes on the state-of-the-art numerical modeling of cloud microphysics, https://doi.org/10.1016/S0169-8095(00)00064-8, 2000.

Flossmann, A. I., Wobrock, W. :A review of our understanding of the aerosol-cloud interaction from the perspective of a bin resolved cloud scale modelling, Atmos. Res., 97, 4, 478-497, DOI 10.1016/j.atmosres.2010.05.008 (Elsevier), 2010.

**Comment 5**: Page 3, Line 34: Henry's law effective→effective Henry's law

**Reply 5**: Correction was made.

**Comment 6**: Section 2.2.1: The way the activation of droplets is calculated is unclear (even going back to papers Caro et al., (2004) and Leriche et al., (2007). Is it so that the activation of droplets is calculated on each time step, not only at the base of the cloud? The Abdul-Razzak Ghan parameterization calculates the number of activated droplets at the cloud base for an air parcel that is rising adiabatically. This means that the parameterization would not work for the droplet activation in-cloud because the already activated droplets would significantly affect the parcel supersaturation. This effect is not taken into account in the parameterization. What was the updraft speed used for the activation parameterization? Are there estimates for updrafts at puy de Dôme?

**Reply 6**: The number of activated droplets is calculated at each time step by:

$$N_{activated} = \left| N_{available\ for\ activation} - N_{already\ activated} \right|$$

However, activation is mainly efficient during the first time steps of the cloud occurrence.

The updraft at puy de Dôme is 0.4m/s, estimated as a mean value of the time profile described in Leriche et al. (2007), the air parcel rises adiabatically.

**Comment 7**: Page 7, Line 2: What are the "inputs related to the dissolution of the particulate matter in the cloud droplet"?

**Reply 7**: The sentence was changed to: "The term $T_{ap}$ has been introduced in Eq. (10) to take into account the contribution of the soluble fraction of particulate phase to aqueous concentration of species $i$ via particle nucleation scavenging."

**Comment 8**: Page 10: The motivation for the simulation without dissolution is unclear to me. The result from this exercise is that there is less uptake when the particle dissolution is neglected. This seems to me quite obvious since the effective Henry's law constant is (by its definition) always higher than the Henry's law constant.

**Reply 8**: The expression "particle dissolution" was probably not appropriate and has been replaced by "particle scavenging" (by nucleation). The simulation without particle scavenging is of great interest as it clearly demonstrates the fact that accounting for nucleation scavenging is crucial to retrieve reliable concentrations of some species, including for instance nitrate as well as di-carboxylic acids (see Table 4). This last aspect is further illustrated in Fig. 9, which clearly shows that particle scavenging has to be taken into account as it is the main (or even unique) source for some of the chemical species described in the model, such as tartric, succinic and malic acids.

The use of "transfer" for both particle ("particle-to-cloud transfer") and gas phase ("mass transfer") contributions was confusing. The terminology has been thus changed throughout the manuscript to clarify the text. The contribution of the particulate phase is now referred to as "particle scavenging" and/or "nucleation scavenging", while the exchanges between the gas phase and the droplet is still referred to as "mass transfer", following Schwartz (1986).

Schwartz, S.: Mass-transport considerations pertinent to aqueous phase reactions of gases in liquid-water clouds, Chemistry of Multiphase Atmospheric Systems, NATO ASI Ser., vol. G6, Springer-Verlag, New York, 1986.

**Comment 9**: Page 10, Line 29: What would be considered a significant change in the cloud microphysical properties and which properties are meant here? The motivation for the comparison between mass transfer and particle dissolution is also unclear. If I am not mistaken, in Figure 9, it is actually the effective Henry's law coefficient that determines the ratio between "particle dissolution" and "mass transfer" (at least for compounds that are not produced by aqueous phase chemistry). This would mean that mass transfer has little to do with this ratio. Would Figure 9 change at all if, for example, the mass transfer coefficient was doubled?

**Reply 9**: Regarding the first comment the microphysical properties refer to cloud liquid water content and droplet radius, and should have been explicitly mentioned. Regarding the magnitude of this change, the use of the expression "not significant" was also not appropriate, as the difference observed between the two simulations (Run 1 = reference and Run 3 = increased organics loading in the particle phase) is negligible. In fact, concerning the liquid water content ($LWC$), it turns out that the average of the ratio $\frac{LWC(Run\ 3)-LWC(Run\ 1)}{LWC(Run\ 1)}$ is $\sim 3 \times 10^{-6}$. Applying similar reasoning to droplet radius leads to similar conclusions. The abovementioned sentence was thus changed to: "The increased amount of

organic matter in the particulate phase did not affect the cloud microphysical properties, namely the cloud liquid water content and droplet radius, which were rather determined by the dominant inorganic fraction representative of marine aerosols."

The second part of the comment has already been addressed in Reply 8. The results shown in Fig. 9 clearly illustrate one of the major strengths of the model, which is capable of estimating the sources of the compounds that are found in the cloud droplet. Comparing mass transfer from gas phase to aqueous phase and particle scavenging by nucleation thus makes sense, as it indicates which one of these two processes is the major contributor to aqueous concentration of a given species.