We thank the referee for their constructive comments on our manuscript. We respond to all of them in detail below. Referee comments are repeated in blue, responses are in black and modified text in the manuscript is in *italic* with added text in *green*. Page and line numbers refer to the revised manuscript without annotations.

Referee #1

1) Referee comment: The manuscript by Jaber et al. asks the important question of whether biological (enzymatic) oxidation of phenol and catechol in simulated cloud water is important in the atmosphere. Bacterial (enzymatic) degradation of organic matter in cloud water is an understudied area of atmospheric chemistry that deserves more attention. In the present study, the authors carried out microcosm studies on cloud water surrogates to study biodegradation rates of phenol and catechol by Rhodococcus enclensis, a bacteria strain found to be quite active at oxidizing phenols during a recent (2018) survey of microbes in real cloud water samples. The derived biodegradation rates, along with chemical kinetics data on abiotic degradation of these compounds were combined in a box model to assess the relative importance of chemical and microbial degradation processes in the fate of phenol and catechol in the atmosphere. The manuscript is well organized, concise, and well-written. Not only does the work show that phenol/catechol are consumed by Rhodococcus, but the authors use the derived degradation constants to compare it to abiotic loss processes in the atmosphere. This combination of laboratory and modeling work is a strength. The results conclude that microbial degradation has the potential to be as important as chemical loss processes of the compounds in cloud water, especially in the case of more reactive species such as catechol. This is an important finding that is a valuable contribution to the atmospheric community. I am supportive of publishing this work in ACP after the following questions are addressed.

Authors' response: We thank the referee for their constructive comments. We address all individual comments in detail below.

2) Referee comment: The only major questions I have for study have to do with the applicability of the chosen laboratory experimental conditions to atmospheric conditions. I see that a temperature of 17 degrees Celsius has been chosen as a working temperature for all experiments. I assume this has been chosen to be a typical cloud water temperature?

Authors' response: Yes, the referee is right. As explained in the "Material end Methods" section describing our microcosm mimicking cloud conditions at the puy de Dôme, we mention that (l. 90)

17°C is the average temperature in the summer at this location.

3) Referee comment: I note an absence of any information on solution pH, which is a significant environmental variable that controls both the chemistry (Fe speciation, ROS chemistry) and microbiology (viability of microbes, and enzyme turnover rates). I suggest the authors clarify under which pH conditions all experiments were carried out. Was a buffer used to control pH in these experiments or was solution pH adjusted in any way?

Authors' response: Our experiments were performed in Volvic® mineral water which pH is 7.0. Because of the presence of carbonates, which buffer the system and because no acid is formed as a product of the biotransformation or phototransformation of phenol and catechol, the pH is quite stable during the experiments. As explained in more detail (section 6) bacteria are able to control their intracellular pH and are viable under cloud conditions. In addition, the Fe(EDDS) complex is stable at this pH as specified in the initial manuscript (Li et al., 2010), it is for this reason that we have chosen this iron form to perform our experiments. This information about pH was added in the text (l. 98):

Bacteria pellets were rinsed first with 5 mL of NaCl 0.8% and after with Volvic® mineral water (pH=7.0), previously sterilized by filtration under sterile conditions using a 0.22 µm PES filter.

4) Referee comment: Furthermore, it would be useful to discuss how the pH conditions used in their experiments compare to actual cloud water pH.

Authors' response: The mean pH in cloud water spans a range of ~3 to ~6, as it has been shown in many measurements at different locations (e.g., data compilation in Seinfeld and Pandis (2006)). In the current simulation, the pH was set to a constant value of 4. We note that the pH of the cloud water in the model does not directly affect any chemical reaction in focus here (i.e. radical reactions with phenolic compounds). Only in highly alkaline solutions (pH ~ 10), phenols will significantly dissociate, i.e. in a pH range that is not encountered in cloud water.

We added this information to the text (l. 193):

The pH value of cloud water is assumed to be constant (pH = 4).

5) Referee comment: A discussion of pH should also factor in when discussing the results shown in Figure 2 & 3. Does pH change during these experiments and could that explain trends in the phenol/catechol loss rates over time? Under what pH were the studies listed in Table S-2 carried out under?

Authors' response: Our experiments were performed in Volvic® mineral water which pH is 7.0. Because of the presence of carbonates which buffer the system and because no acid is formed as a product of the biotransformation of phenol and catechol, the pH is quite stable during the experiments. Incubations with *Pseudomonas aeruginosa* (Razika et al. 2010) and with *Pseudomonas putida* EKII (Hinteregger et al. 1992) were performed at pH=7. This information was added in the text:

l. 98: Bacteria pellets were rinsed first with 5 mL of NaCl 0.8% and after with Volvic® mineral water (pH=7.0), previously sterilized by filtration under sterile conditions using a 0.22 µm PES filter.

Supplement, S-1.1:

l. 2/3: To calculate the biodegradation rate of phenol and catechol by Pseudomonas putida EKII, based on experiments performed at pH=7.0, we used the following data from Hinteregger et al. (1992):

l. 12/13: To calculate the biodegradation rate of phenol and catechol by Pseusomonas aeriginosa, based on experiments performed at pH=7.0, we used the following data from Razika et al. (2010):

6) Referee comment: In the absence of the authors own data on pH effects, does the literature provide any insights into the effect of pH on phenol monooxygenases/hydroxylases and/or the activity of Pseudomonas and Rhodococcus strains?

Authors' response: Actually, the experiments are performed with bacteria and not purified enzymes. In that case, phenol monooxygenases/hydroxylases' activity takes place inside the cell and are not impacted by the external pH. Bacteria are able to regulate their internal pH (which is usually ~6.5 -7) when exposed to external pHs within a very broad range, except at extreme conditions, e.g., pH < 2 or pH > 10. In that case only acidophilic or alcalinophilic bacteria can adapt to such extreme pH values. In our case, cloud water collected at the puy de Dôme have moderate pHs ranging from 3.8 to 7.6 (Deguillaume et al., 2014).

In our previous biodegradation experiments with 17 different cloud bacteria using artificial cloud water at pH =5.0 (continental) and pH= 6.5 (marine), respectively (Vaïtilingom et al., 2011), no influence of the pH was observed on the measured biodegradation rates. Also Razika et al., (2010) showed that biodegradation rates of phenol by *Pseudomonas aeruginosa* (used in this paper) were very similar when incubated at pH=5.8, 7.0 and 8.0, respectively.

We added the following text at the beginning of Section 3.1, l. 245-249:

The transformation rates described in this work were measured at pH=7.0 which is within the range of pHs encountered in real clouds as observed at the Puy de Dome (3.8 < pH < 7.6, Deguillaume et al, 2014). However, bacteria are able to control their intracellular pH under such conditions, and it has been shown that pH variation has a low impact on their biodegradation ability (Vaitilingom et al. 2011; Razika et al. 2010).

7) Referee comment: Under what pH is the modeling carried out under?

Authors' response: The pH in the model is set to a constant value of 4 which is representative of moderately polluted to polluted air masses, such as downwind of urban areas. As none of the reactions (OH, NO_3 + phenolic compounds) is directly dependent on the pH within the ranges typically found in clouds (~ 2 < pH < ~6), this value does not have a large impact on the model results.

We added this information to the text (l. 193):

The pH value of cloud water is assumed to be constant (pH = 4).

8) Referee comment: Although not done in this communication, future work should be focused on characterizing these rates as a function of T and pH.

Authors' response: We thank the referee for this suggestion. However, as explained above, the intracellular pH is regulated by the bacterial cells under cloud pH conditions. Therefore, we do not think that it is necessary to perform experiments (which are highly time consuming) at different pHs.

9) Referee comment: On line 354-355, the authors state, "...we caution that these results likely represent an upper estimate that might not correspond to the moderate pH values encountered in clouds." Please elaborate on this in light of the abovementioned questions. What do the authors mean by "moderate" and why wouldn't their experiments be applicable to the moderate pH values encountered in clouds? I feel that the lack of attention to pH is a major issue that needs to be addressed in the final manuscript.

Authors' response: As outlined above, the pH value does not have any significant impact on the chemical reactions of the aromatics in the model. The last sentence in Section 3.3. ("*However, we caution that these results likely represent an upper estimate that might not correspond to the moderate pH values as encountered in clouds.*") only refers to the sensitivity model study. In Section S-4 of the supplement, we discussed that the rate constants applied in the original model study by Hoffmann et al. (2018) and also adopted in our study were determined in the original literature by Gurol and Nekouinaini (1984) at pH = 1.5. The rate constant shows a decreasing trend with increasing pH; however, the exact pH dependence is not known. Thus, we concluded that the predicted contribution by the ozone reaction is likely an overestimate. In order to make it clear, we modified the text as follows (l. 359-364):

However, we caution that these results of the model sensitivity study including the ozone and HO_2/O_2^- reactions likely represent an upper estimate. The rate constant used in the model was determined at pH = 1.5. In the original study, a decreasing trend with increasing pH was suggested; however, the exact pH dependence was not given. Thus, the prediction shown in Figure S-3 ~~that~~ might not correspond to the moderate pH values as encountered in clouds and thus might be an overestimate of the role of the ozone reaction.

10) Referee comment: Lastly, for the modeling work, simulations are performed on monodisperse cloud droplets with a diameter of 20 microns, with specific drop number concentration and liquid water content. Please indicate how these were chosen and whether they are representative of typical cloud water.

Authors' response: The drop diameter of 20 microm is a typical value that is often assumed in large scale models as being representative for clouds in clean or moderately polluted regions (cf, for example, the overview by Ervens (2015)). The cloud liquid water content is also typical for stratocumulus or cumulus clouds (~ 0.1 – 1 g m^{-3}, e.g. Pruppacher and Klett (2003)). In several recent studies, the dependence of the OH(aq) concentration on cloud drop sizes has been discussed, e.g. (Chakraborty et al., 2016; Ervens et al., 2014).

We show below a figure from additional model simulations using another monodisperse droplet size distribution (drop diameter 10 μm, Simulation I) and a polydisperse one with droplet sizes between 1 and 30 μm (Simulation III). Simulation II corresponds to the results shown in the manuscript.

Table R-1: Microphysical characteristics assumed in model sensitivity studies; Simulation II is the case presented in the manuscript

Simulation	Drop diameter [μm]	N_{dr} [cm^{-3}]	LWC [g m^{-3}]
I	10	550	0.29
II	20	220	0.92
III	1 – 30	293	0.3

It can be seen in Figure R-1 that smaller droplet sizes (Simulation I) tend to cause a higher importance for OH(aq)-initiated processes, in agreement with the conclusions by Ervens et al. (2014). In the latter study, it was discussed that smaller droplets (larger surface-to-volume ratio) allow more OH to be transported into the droplets and, thus, lead to higher rates of OH(aq) reactions. Simulation III represents a case in-between Simulation I and II in terms of the droplet interface and shows corresponding results in terms of the importance of microbial and chemical processes, respectively. Since these dependencies on drop surface and drop surface-to-volume ratio have been discussed in detail in previous publications (Ervens et al., 2014; McVay and Ervens, 2017) and are not focus of the current study, we refrained from adding these model sensitivity studies to the current manuscript.

We extended the following to the text (l. 380/381)

The relative importance of radical chemistry compared to biodegradation will also depend on the radical concentrations in both phases which, in turn, are a function of numerous factors such as air mass characteristics, pollution levels that affect OH concentrations and microphysical cloud properties (e.g., drop diameters, liquid water content) (Ervens et al., 2014).

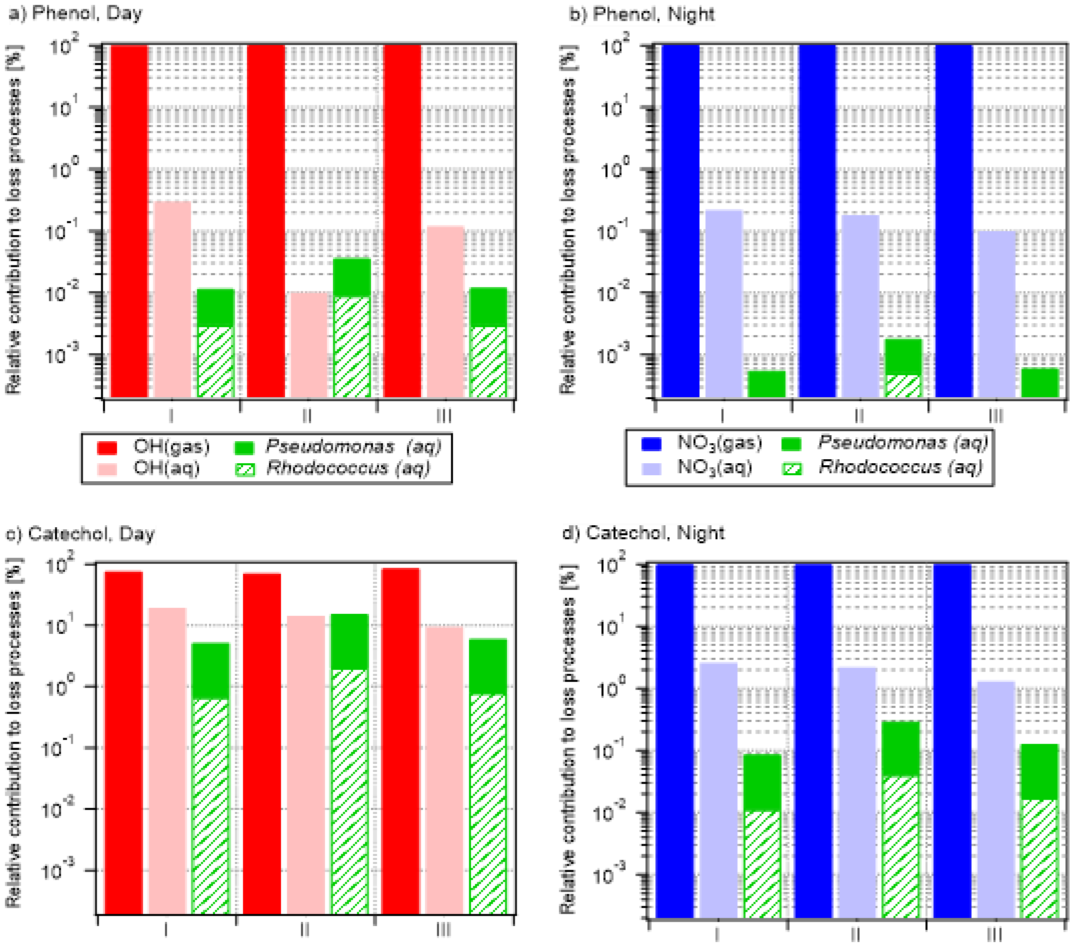

Figure R-1: Relative contributions to loss processes for phenol (a, b) and catechol (c, d) due to radical processes in the gas and aqueous phases and microbial activity by Pseudomonas and Rhodococcus in the aqueous phase for three model simulations (I– III, Table R-1)