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 #2

This manuscript describes lab measurements of the ability of two bacteria species present in cloudwater to react with phenol and catechol molecules. The authors then run simple day and nighttime box model simulations to apportion the reactivity of these molecules to three bins: gas phase, aqueous phase chemical and aqueous phase biological reactivity. They find that bacterial transformation of catechol is an important loss process during the day, comprising 17% of the total losses in the daytime model. Daytime biotransformation of phenol, and nighttime biotransformation of either species, are minor loss pathways. This work will be of interest to those interested in SOA formation and cloud processing, and is publishable after minor revision.

Authors' response: We thank the referee for their constructive comments and address all of them individually below.

Specific Comments

1) Referee comment: Table 2: How realistic is it to model bacterial degradation rates as the fastest measured in Figure 2? Some discussion on this point could strengthen the conclusions.

Authors' response: As explained in the initial text: *"A lag time of about 2.5 hours is observed, during which phenol is degraded extremely slowly. This is a well-known phenomenon under lab conditions corresponding to the induction period of the gene expression (Al-Khalid and El-Naas, 2012)."*

Given that the bacteria are present in the atmosphere for extended periods of time, it can be implied that this lag time is not of importance in cloud droplets. Therefore, we think that it is reasonable to use the rates of biodegradation, which correspond to the highest slopes in Figure 2 as it represents the real phase of biodegradation.

2) Referee comment: Line 283: The text states that at 10ˆ9 cell concentrations catechol biodegradation "was too fast to be detected within the time resolution of the experiments (Figure 3)." However, the 10ˆ8 data is identical to the 10ˆ9 data, and should be included in this statement.

Authors' response: You are right; we have changed the text as follows (l. 285):

When the cell concentration was 10^8 or 10^9 cell mL^{-1}, the catechol biodegradation was too fast to be detected within the time resolution of the experiments (Figure 3).

We performed various experiments with reduced cell concentrations, from 10^7 cell mL^{-1} to 10^6 cell mL^{-1} (Figure 3).

3) Referee comment: Figure 3 is not very relevant to the aims of the paper and could be moved to the SI section.

Authors' response: We prefer to keep this figure in the main text for the following reasons:

1) To our knowledge, LC-MS has not been used in any previous studies to measure catechol biodegradation rates. Thus, the presented data are original.
2) These experiments are essential for the measurement of the biodegradation rates that are finally used in the model studies.

4) Referee comment: Figure S-3 is much more relevant to the aims of the paper, even though it doesn't necessarily strengthen the conclusions that biotransformation of catechol is significant during daytime. I urge the authors to move Figure S-3 into the manuscript, along with appropriate discussion.

Authors' response: We respectfully disagree with the suggestion to move Figure S-3 to the main text of the manuscript. Given the uncertainties in the rate constants for the HO_2/O_2^- and ozone reactions for relevant cloud conditions (pH), it likely shows a biased picture on the importance of ozone reactions with phenolic compounds. As discussed in the supplement, the rate constant was determined at pH = 1.5 and shows a decreasing trend with increasing pH. Since this trend has not been quantified in the original literature, we want to caution to draw false conclusions based on Figure S-3. To make these concerns this clearer, we extended the last paragraph of Section 3.3 (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 of the role of the ozone reaction. 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.

5) Referee comment: Line 315: The statement about catechol degradation rates "Values are only available for Ps. putida EKII" is confusing, given that values are listed for a second strain listed in Table 2 (from Razika 2010). Only by reading the supplemental information section can the reader ascertain that the Ps. aeriginosa catechol degradation rate listed in the table in the row marked "Razika 2010" was not measured by Razika, but is actually the phenol rate times the ratio 12 (measured for another Pseudomonas strain. This is unintentionally misleading. I suggest that the table entry be "ND" and the phenol rate x 12 be given in the table caption, or in some other way that makes it clear that it is not a measurement of Razika et al.

Authors' response: We agree with the referee that the wording was misleading. We extended the text and table as follows (l. 322-325):

As in the case of phenol, we also calculated catechol biodegradation rates with Pseudomonas strains based on literature data (Table 2). Values are only available for Pseudomonas putida EKII (Hinteregger et al., 1992) and show a biodegradation rate that is twelve times higher compared to that of phenol biodegradation. This confirms that catechol dioxygenases are much more active than phenol hydroxylases as observed for Rhodococcus enclensis. Similar to phenol, catechol biodegradation rates for Pseudomonas strains are somewhat higher than those for Rhodococcus, but within the same order of magnitude. The same ratio (~12) as for the Pseudomonas putida was applied to estimate the biodegradation rate of

catechol by Pseudomonas aeruginosa, for which only the rate for phenol was experimentally determined by Razika et al. (2010).

***Table 1**: Biodegradation rates [mol cell^{-1} h^{-1}] of catechol and phenol of Rhodococcus and Pseudomonas strains normalized to the exact number of cells present in the incubations. The calculation of biodegradation rates for the Pseudomonas strains are detailed in S-1.*

Bacterial strain (experimental condition)	Biodegradation rate of phenol (10^{-16} mol cell^{-1} h^{-1})	Biodegradation rate of catechol (10^{-16} mol cell^{-1} h^{-1})	References
Rhodococcus enclensis PDD-23b-28 (dark)	1.8 ± 0.5	15.0 ± 0.5	This work
Rhodococcus enclensis PDD-23b-28 (light)	1.2 ± 0.5	ND[1)]	This work
Rhodococcus enclensis PDD-23b-28 (light+Fe(EDDS))	1.0 ± 0.3	ND[1)]	This work
Pseudomonas putida EKII (dark)	0.2	2.4	(Hinteregger et al., 1992)
Pseudomonas aeruginosa (dark)	5.9	70.7 [2)]	Phenol experiments (Razika et al., 2010)
Pseudomonas (average)	**Average: 3.0**	**Average: 36.6**	

[1)] *Not determined;* [2)] *This rate was estimated based on the value for phenol (Razika et al., 2010) and the ratio (~ 12) for phenol/catechol biodegradation rates as determined for Pseudomonas putida by Hinteregger et al. (1992) (cf also Section 1-1 in the supplement)*

6) Referee comment: Line 318: The claim that biodegradation rates of phenol or catechol are generally higher for Pseudomonas than for Rhodococcus has no statistical validity and cannot be made, especially in light of my previous comment. The variability between Pseudomonas strains is larger than the difference between the two species.

Authors' response: We agree with the referee. We have changed the text as follows (l. 322):

Similar to phenol, catechol biodegradation rates for *Pseudomonas* strains *are within the same order of magnitude as those for Rhodococcus.*

7) Referee comment: Figures 4cd and S-3cd: Some of the statements made in the text discussing Figure 4 appear to be quantitatively incorrect when looking at Figure S-3. For example, line 344 "The total microbial activity in the aqueous exceeds that of the chemical reactions (Figure 4c) and contributes up to 17% to the total loss of catechol in the multiphase system." According to Figure S-3c, this statement is likely true when reaction with dissolved OH is the only chemical reaction considered. The statement should be modified to reflect the information shown in both figures.

Authors' response: As pointed out above, the contributions of the HO_2/O_2^- and ozone reactions to the total chemical loss of phenol and catechol are highly uncertain. We modified the text as follows to (i) reflect that other oxidation reactions in addition to OH might take place but (ii) their contributions are very uncertain due to uncertainties in their rate constants.

We added (l. 349):

*During daytime, the loss by aqueous phase processes (chemical and microbial) is >30% for catechol (**Figure 4c**), with contributions by OH(aq), Pseudomonas and Rhodococcus of 14%, 10% and 7%, respectively, when OH as the only oxidant for the phenols in the aqueous phase is considered.*

8) Referee comment: Line 356: These sentences correspond with measurements in Figure 2, but do not correspond with the results shown in Figure 4, where the different processes are compared under the same conditions. It seems that with catechol (not phenol), photo- and biotransformations are of the same order or magnitude, and with phenol reactions with dissolved OH are significantly more important.

Authors' response: We agree with the referee that these sentences were misleading. We modified them as follows (l. 366-368):

Both experimental and modelling approaches show that, in the water phase of clouds suggest that phenol and catechol degradation by microbial and chemical OH(aq) processes may be within one order of magnitude. ~~phenol bio- and photo-transformations are within the same order of magnitude, while catechol biotransformation seems more efficient than OH(aq) chemistry under identical experimental and atmospheric conditions, respectively.~~ When the complete multiphase system is taken into account, phenol chemical transformation is largely dominant in the gas phase whereas the ~~might~~ more water-soluble catechol is more efficiently biodegraded in the aqueous phase.

9) Referee comment: Line 379: This conclusion needs more support. It is clear from this work that microbial processes must be included to give a complete representation of cloudwater chemistry. Whether this complete representation is necessary to improve air quality or climate predictions has not been established.

Authors' response: We agree with the referee that our statement was somewhat pretentious. However, we would like to point out that the implementation of microbial processes will not only help to complete the understanding of the chemical composition of cloud water but also of the atmospheric multiphase system. The fact that, for example, microbial processes may contribute ~10% to the total loss of catechol shows that for some pollutants these processes are an important multiphase sink.

We removed the last part of the sentence:

Thus, atmospheric models may be incomplete in describing the loss of some organic compounds and should be complemented by microbial processes in order to give a complete representation of the atmospheric multiphase system. ~~to eventually allow comprehensive air quality and climate predictions.~~

Technical corrections

10) Referee comment: Line 176: "turned" should be "tuned"

Authors' response: We corrected the typo.

11) Referee comment: Figure 2 caption should specify the Rhodococcus cell concentration.

Authors' response: Figure 2 caption was modified as follows:

Rhodococcus enclensis cell concentration was 10^9 cells mL^{-1}.

12) Referee comment: Figure S1: the figure legend does not match the description in the caption. Is the blue line the lamp spectrum or the absorption spectrum of phenol?

Authors' response: We corrected the legend and it reads now:

Comparison of the actinic fluxes of the lamps used and the emission of the solar spectrum measured in-cloud at the puy de Dôme station. The ~~green~~ blue line represents the actinic flux of the lamp; the brown line corresponds to the actinic flux of the solar emission spectrum in cloud. The ~~red~~ pink line represents the molar absorption coefficient of the Fe-EDDS complex. The ~~blue~~ red line represents the molar absorption coefficient of phenol.

13) Referee comment: Line 360: "might" should be "slightly"?

Authors' response: We removed 'might'.