

## **Answer to the reviewers: reviewer 1**

We thank both reviewers and Y. Fang for their helpful comments and suggestions.

Two important concerns of the reviewers can be summarized as follows:

- Does this work contain sufficient new scientific development, it does not present new (model) results and builds on already published work.
- The system that is studied is not representative of the actual atmosphere, aerosol is much more complex.

We will address these first, in the answers to both reviewers. After that we will answer their comments point by point, in the separate documents.

### *General answer*

Many recent studies are devoted to the influence of (changing) aerosol properties on climate and many studies are devoted to the atmospheric hydrological cycle and how it behaves under climate change. However, the link between the hydrological cycle and the aerosol lifetime has not been investigated extensively in the context of climate change. Therefore, we believe our work presents a new perspective on (the causes of) aerosol radiative forcing and in our opinion this merits publication of our paper.

Our study focusses on only one aspect of atmospheric aerosol, i.e., the dependence of its lifetime on the hydrological cycle and how they respond to a temperature change. We focus therefore specifically on highly soluble aerosol that is removed exclusively by wet deposition while other aerosol types and processes relevant for aerosol are neglected. We emphasize strongly that this does not mean that they are considered unimportant, but they are neglected in order to present the matter in a clear and illustrative way. With these constraintment the described system is more or less representative of soluble inorganic aerosol species as sulfate and nitrate that have a strong anthropogenic signature. We have discussed this more clearly in the introduction section.

We note that the work of Pruppache and Jaenicke (1995) only pertains to our section 2.1. They presented expressions for the water vapor and aerosol lifetimes but did not include expressions for temperature sensitivities (our section 2.2). We made that more clear in the text. In the first version of the manuscript the first few equations were relatively simple, following the notation of Pruppacher and Jaenicke. In the present manuscript we use equations that give a better representation of clouds in the real atmosphere and in climate models. The water vapor and aerosol removal rates are now expressed by:

$$L_v = 1/\tau_v = \left( \sum e_{c,i} p_{c,i} c_i U_i / H \right), \text{ and}$$

$$L_{AP} = 1/\tau_{AP} = \left( \sum e_{a,i} p_{a,i} c_i U_i / H \right)^{-1},$$

where subscript  $i$  refers to different cloud types (as in the previous version),  $e_c$  and  $e_a$  are the condensation and aerosol activation efficiencies, and  $p_c$  and  $p_a$  are the fractions of water and aerosol transferred from the cloud to the precipitation phase (replacing the precipitation volume fraction  $f$  used in the previous version).

Of course, many and large uncertainties are currently associated with the climate sensitivity of the hydrological cycle and with the exact magnitude and causes of the aerosol radiative forcing. Our study does not report on new model simulations or observational data and it does not present a quantitative discussion of uncertainties associated with climate sensitivity or aerosol radiative forcing. Our study is therefore limited to a theoretical exploration of the link between water vapor and aerosol lifetimes in the atmosphere and their response to climate change. An important aspect is that climate change itself induces a change in aerosol lifetime, and thus influences the burden and the associated radiative forcing, even when aerosol characteristics (emissions, size distribution, hygroscopicity, etc.) remain the same. This suggests that the link between aerosol lifetime and the hydrological cycle may contribute to intermodel variabilities of the aerosol radiative forcing. This can not be quantified without dedicated model experiments. We have therefore included suggestions for experiments based on the derived equations that can be used to investigate the link between water vapor and aerosol lifetimes and compare the performances of climate models in this respect.

#### *Answers to comments from Reviewer 1*

This manuscript examines simple relations, rather back-of-the-envelope type calculations, attempt to provide a simple characterization of a complex system. Wide range of simulated aerosol lifetime sensitivities to temperature change (with sign differences). Aerosol lifetime sensitivity of 5.3%K<sup>-1</sup>, which is in the upper range. The author uses these relations to show that water vapor and aerosol lifetime are coupled since they depend on the same cloud parameters, and that the ratio of their lifetimes is proportional to the ratio of scavenging and condensation efficiencies.

Scientifically interesting. But certain points that are not clearly presented and certain conclusions that are not well supported. Many of the relations presented are a review of already published work. Section 2.1: Pruppacher and Jaenicke (1995). The figure and table in the manuscript are also a summary of data taken from previous studies. While a review of previous work is useful for the discussion, I was not sure if there was enough new scientific development in this manuscript.

Specifics:

1) fccUc, clearer physical meaning for this term earlier in the text, other than simply calling this the 'cloud response'? (Not best terminology).

*In our paper we used the term "cloud response" for the change in the cloud parameters  $f$ ,  $c$  and  $U$  following a temperature change, i.e.:  $dfcU/dT$ . In the rewritten manuscript the term  $f$  is replaced by  $p_c$  and  $p_a$ , i.e., the fractions of water and aerosol transferred from cloud to precipitation. Now we define the less ambiguous terms "water vapor lifetime temperature response" and "aerosol lifetime temperature response" to describe the sensitivity of these terms for a temperature change.*

1b) SST-cloud cover, but what about  $f$ ? thus the derivative of the term with respect to temperature could be negative while sea surface temperature and cloud cover were positively correlated, if the volume fraction of cloud that produced precipitation decreased with increasing temperature.

*We agree that it is in theory possible that a negative change in cloud cover can be offset by a positive change in  $f$ , or  $p_c$  in the current version. The total only becomes positive if the change in  $p_c$  is relatively, or percentually, larger than that in  $c$ . It implies that a smaller cloud cover is accompanied by a more efficient precipitation formation. This seems contradictory: a smaller cloud cover (i.e., a negative second term on the right hand side of eq. (9)) reflects on average less-saturated conditions, while a higher precipitation formation efficiency (i.e., a positive second term) suggests a higher saturation. We have removed the comparison with SST-cloud cover from the discussion, but added a lay-out of model experiments that can be used for quantification of this aspect.*

2) The calculations in Section 2.2 are based on the work of Held and Soden (2006). So in many respects the first line in Table 1 might reference that publication. Could the author comment on how the 5.3%K-1 sensitivity calculated in Section 2.2 relates to Figure 5b) in Held and Soden, which also shows a roughly 5%K-1 temperature sensitivity related to moisture transport?

*We added the reference to the Table. The 5%/K change in maximum poleward moisture transport (Fig. 5b in the Held and Soden paper) is of the same order as our 5.3%/K cloud processing temperature response. Their equation (4) can be reformulated to*

$$\alpha\delta T = \frac{\delta F_L}{F_L} - \frac{\delta F_S}{F_S} \quad (a)$$

*where  $F_L$  and  $F_S$  are latent and sensible heat fluxes, respectively. Eq. (a) resembles our eq. (7), which when reformulated and neglecting  $e_c$  becomes:*

$$d \ln w_v = d \ln P - d \ln (f c_c U_c) \quad (b)$$

*In climate models  $d \ln w_v$  approximately satisfies the Clausius Clapeyron relation so that  $d \ln w = adT$ . The formation of precipitation water transforms latent into sensible heat, so the second term of the right hand side in eq. (a) is associated with the first right hand side term in eq. (b). The change in latent heat (last term in eq. a) is then associated with the change in the remaining water vapor. This may be a result of a change in cloud properties as expressed by the cloud processing temperature response term in eq. (b). This term appears therefore to be connected in some way with the moisture transport given by Held and Soden (2006), which has about the same magnitude as the cloud temperature response. This however is not very relevant for our study, we have not included this discussion in the text.*

3) Could the author state more clearly what is meant at page 16500, line 4, by the 'tendency of models to maintain relative humidity' and why this is expected?

*We have included references to model studies that show that RH does not change much with temperature. Sherwood et al. (2010) find an absolute change of RH smaller than ~ 2% in the troposphere below ~ 300 hPa. Above that pressure altitude simulated changes become larger, upto 4%. They also find a large variability across models. Wright et al. (2010) show similar findings for ocean slab-GCMs.*

4) The last paragraph of Section 2.2 is a useful discussion about the precipitation sensitivity term and how there is considerable uncertainty related to this term, such that even the sign of the 'cloud response' term is in question as a result. Thus I was not clear why the author chose to present the 5.3%K-1 result in the Table 1 as the only result of this study - a range of values between -0.5 K-1 and 6.0 K-1 results when Eqn. 7 is applied using the precipitation sensitivities given in Section 2.2, paragraph 2 (and under the assumption that the water vapor concentration sensitivity is roughly constant as noted at page 16500, line 4).

*An uncertainty range better reflects our current knowledge than a single value, and if the Durack (2012) estimate is included the range is indeed very large: from -0.5 to 6 %/K. However, the Durack estimate is based on surface observations that may or may not reflect the full hydrological cycle so we did not include it in our estimate. In the present manuscript we have listed the range  $5.3 \pm 2.0$  %/K, which is estimated from the spread in Figs. 2c and 2d of Held and Soden (2006). The Durack (2012) estimate illustrates that actual uncertainties may be much larger than the model spread indicates.*

5) The last paragraph in Section 2.3 discusses a potential feedback mechanism under the assumption of constant scavenging efficiency (ea). Acknowledge more clearly here that this assumption of constant ea with temperature change is highly uncertain and perhaps not very likely. This makes the conclusion about the relative importance of emissions versus climate change on the aerosol burdens a highly uncertain result. Thus I am not sure how meaningful this feedback discussion is here, given this uncertainty. Also, could the author

provide a clearer list of the assumptions underlying this analysis? This paragraph also references Eq. (5), which is not the correct equation number.

*The described feedback mechanism does not depend on  $e_a$ , but the reviewer is correct that a change in  $e_a$  constitutes an additional feedback possibility. However, since this was less relevant for the main message of the study we removed this part. We have added more discussion on the possible changes in  $e_a$  and corrected the equation numbering.*

6) Section 3 first sentence, perhaps 'water vapor lifetime' could be used to be more specific as opposed to water vapor lifetime.

*We did not quite understand this comment, the sentence mentions the aerosol lifetime but not 'water vapor lifetime'. We reformulated this sentence for clarity.*

7) Table 1 presents quite a limited selection of previous studies (only 4) – is this enough to draw the conclusions about the sign of the simulated sensitivity of aerosol lifetimes as related to the climate sensitivity of models? The ideas examined by this study would be suitable for a model intercomparison project involving a greater number of global models.

*During the time of writing our manuscript these were the only four studies that listed the information necessary to compile Table 2, i.e., changes in temperature, precipitation, water vapor lifetime and aerosol lifetime. We agree with the suggestion for the intercomparison and included a possible series of experiments based on our analysis.*

8) Page 16503, line 12 states that 'more likely that increased precipitation frequency is responsible' for the negative temperature sensitivity of aerosol lifetime – could not other factors also contribute to this result? Can the author better justify this conclusion?

*This comment was based on discussion with one of the authors of the paper. Their paper mention different reasons: more precipitation, and different seasonalities of precipitation and aerosol concentrations. Analogous to the study of Fang et al (2011) and our analysis it appears more likely that a change in precipitation frequency, rather than a change in total precipitation, results in an actual aerosol lifetime change. However, because of the speculative nature we removed this from the text.*

9) Figure 1 is a plot of data from Soden and Held (2006). Could the author provide a less cursory discussion of the figure to make the paper more accessible to a reader that is not as familiar with this previous study? Perhaps there could be a brief discussion of what is meant for cloud radiative, water vapor and lapse rate feedbacks and why the author has chosen to present the feedbacks in this way. Also perhaps a clearer statement of the main conclusion that the figure supports.

*We have thoroughly rewritten section 3. As it was no longer necessary as basis for our argumentation we removed the original Fig. 1. We included more detail in our discussion of the possible link between aerosol lifetime and climate sensitivity, better explaining the*

*nature of the different feedbacks.*

10) Page 16504, line 25 states that when the temperature response of the term  $f_c U_c$  is negative, this is consistent with a positive cloud radiative feedback. Could the author provide a clearer discussion about how cloud radiative feedback relates to the term  $f_c U_c$ ?

*We describe more clearly the possibility that changes in cloud properties leading to cloud radiative feedback may influence aerosol lifetimes, and vice versa. For example a positive CRF can be the result of a smaller cloud cover and/or a shallower cloud depth (governed by updraft velocity). The same changes are consistent with a reduced processing efficiency of air by clouds, thus affecting the water vapor and aerosol lifetimes. Dedicated model experiments are required to demonstrate that such a relation between CRF and both lifetimes exists, and we suggest model experiments for this purpose that are based on the equations presented in our study.*

11) Page 16505, lines 22-23 – could the author be more specific about what is meant by ‘cloud response’? Also in that paragraph, could the author be clear about the assumptions that underlie the derived ‘cloud response’

*With the term "cloud response" we referred to the temperature dependence of  $f_c U_c$ , or the relative speed with which a column of air is processed by clouds that actually precipitate. In the current version we have used the less ambiguous water vapor lifetime temperature response and aerosol lifetime temperature response.*

12) Page 16506, line 2, I am not sure that cloud cover observations alone can provide an indication about the estimate of the ‘cloud response’ term, given that this term also includes  $f$ , the fraction of clouds that produce precipitation. Also in this paragraph, I think that presenting the range of aerosol lifetime temperature sensitivities would be helpful.

*We removed the comparison with observed cloud cover. Due to the difficulty of observing atmospheric trends in humidity, cloud cover, etc, this single comparison was not a convincing argument.*

13) Page 16506, line 2 states that atmospheric transport is not included – would this not be implicit in term 2 if the results are based on estimates for the other terms in Eq. 7, which come from global models that include transport processes (taken from the Held and Soden (2006) compilation of models)? Likewise for some of the remaining list of not included processes.

*The values from Held and Soden refer to hydrological cycle studies and therefore pertain to water vapor only, including its transport through the atmosphere. For wet deposition of aerosol the combined transports of aerosol and water vapor must be considered. The removal efficiency of aerosol depends on the removal of water vapor, and any changes in the transport of one of these species relative to the other directly affects the aerosol lifetime.*

14) Page 16506, line 14-16, could the author add a reference here?

*In the current manuscript the hypothesized link between climate sensitivity and aerosol is presented in a different way now, and this particular sentence has been removed.*

15) Could the author provide a clearer reference to the evidence from this study that supports the tentative conclusion of page 16506, line 19?

*In our discussion we postulate that a change in cloud characteristics leads to cloud radiative forcing as well as a temperature response of the aerosol lifetime, and thus links climate sensitivity and aerosol forcing. We can not unambiguously show this is true, so we suggest dedicated climate model experiments that may quantify the relation.*

16) Table 1 – it is not clear what the term ‘average’ means, and also add units for CS.

*We removed the term average, CS is in degrees K as mentioned now.*

17) Abstract – I think the abstract should mention that even the sign of the aerosol and water vapor lifetime temperature sensitivity is not certain. Also perhaps mention that nature of the forcing (LW) for the derived value of 5.3%K-1 and be clear that these were not model simulations conducted in this study.

*We have rewritten the abstract and made it clearer that the estimate of the aerosol lifetime temperature response was derived from previous model studies of the temperature sensitivity of the hydrological cycle and was not calculated in our study. We specifically included an estimated uncertainty of 2.0 %/K, directly based on these model studies, and mentioned that aerosol-climate models simulate lifetime changes that span a much wider range than this.*

18) Abstract – I find that the argument about the distribution of water vapor and aerosol between the lower and upper troposphere as related to climate sensitivity and temperature sensitivity of aerosol lifetime was not clearly developed in Section 3.2 and had frequent use of the words ‘may’ and ‘probably’ - so I was not sure how much confidence to place in this analysis. The last 2 sentences of the abstract seem to be rather general and not a strong conclusion based on evidence provided by this study.

*We have rewritten section 3.2, starting from the premise that the same processes that lead to CRF also are responsible for the temperature response of the aerosol lifetime, which should be the case for internally consistent aerosol-climate models. It follows that climate sensitivity and aerosol lifetime temperature response are connected. The discussion around the vertical exchange is included as a possible explanation for this connection, relating water vapor transports to the climate sensitivity feedbacks. We have suggested model experiments that can be carried out to investigate this relation further. We have also rewritten the Abstract to present the results more clearly.*

19) Introduction page 16495, line 10, wet deposition efficiency is noted to depend on the efficiency of air processing by precipitating clouds and uptake of aerosol in cloud water – should this not also depend on rates of conversion of cloud water to precipitation (autoconversion, aggregation, accretion)?

*The actual precipitation formation rate and the exact timing of precipitation are not relevant because the steady-state approach covers a relatively long period, from months to years. However, in the equations we added the efficiencies for the transfer of water and aerosol from the cloud to the precipitation phase, which describes the actual atmosphere and clouds in climate models more realistically than the previous manuscript.*

20) Introduction page 16495, line 15-16, lifetime and burden should be positively correlated – do you mean removal efficiency and burden are anticorrelated?

*We corrected this, what we meant is the anti-correlation between lifetime and removal efficiency.*

21) Page 16495, line 25-30, are these studies for changes to stratiform scavenging only? Could the lifetime changes be greater for changes to convective scavenging processes? Is this evidence enough to indicate the significant influence of the simulated hydrological cycle alone?

*The large differences in the representation of clouds in climate models (e.g., Bony et al., 2006) can lead to significant intermodel differences in simulated aerosol lifetime. The fact that aerosol lifetimes are much more in line in simulations with the same model than between different models seems to corroborate this, as we stated in the previous manuscript. However, based on only one model this was rather speculative and we removed this from the text. The equations included in the present manuscript specifically allow for changes in cloud characteristics for different cloud types. We included suggestions for a possible model intercomparison that may help to quantify the role of different cloud types.*