

Answer to the reviewers: reviewer 2

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 2

The manuscript tries to highlight the importance of hydrological cycle in the issue of aerosol-climate interaction. While the general message is quite agreeable and the general connection between aerosol-precipitation-climate outlined in the text is reasonable, the manuscript falls short in delivering concrete and substantial new insights in this overarching challenge in my opinion. The simple, bulk 'equations' derived in this text make strong and questionable assumptions about the nature of aerosol and precipitation fields. In the current form, this manuscript may not be ready for publication as an original research article in my opinion, but the decision will be deferred to the editor. Comments on this manuscript are presented in the following, where only major comments are detailed and only a few minor comments are mentioned.

We refer to the General answer above

1. The issue discussed here is certainly scientifically interesting and warrants much more investigation. It covers a wide range of specific subjects that can be explored. However, the

writing and discussion presented in this manuscript are often too general and do not go into much depth. This is also reflected in the amount of original results reported in this manuscript. The whole argument seems to build upon published results with few new contributions. I suggest the author to consider to write a manuscript with a more focused theme and change the abstract substantially because in its current form it is way too general given the amount of supporting materials actually presented in this paper.

In our manuscript we discuss two climatologically highly important atmospheric phenomena that are mostly investigated separately and independently from each other. We believe the novelty of our work comes from connecting these two phenomena in a theoretical framework and discuss the possible relevance for aerosol climate forcing. We do this in a simplified way, using back-of-the-envelope equations that illustrate and clarify this relation rather than quantify it. We see our study as a new perspective on aerosol climate effects that qualitatively explains (part of the) intermodel variabilities in aerosol radiative forcing in terms of simulated characteristics of the hydrological cycle. Given the large uncertainties associated with the hydrological cycle and with aerosol, accurate quantification is not feasible at the moment, although we present a first-order quantification based on global aerosol and climate model results. We also present suggestions for dedicated simulation exercises to investigate intermodel variabilities. We have rewritten the abstract, the introduction and larger parts of the text to present the matter in a more coherent and focussed way.

2. Many of the equations used in Section 2 come from a published work. However, for the specific subject in discussion I have reservation on the applicability of these simple arguments. For example, one outstanding feature of the aerosol and precipitation fields is their heterogeneity. For this simple, but first order reason alone, equation 2 cannot automatically follow from equation 1 because there is no single value of e_c . For example, the tropical deep convection and coastal stratocumulus would have totally different e_c . Similar flaws can be found in subsequent derivations, which makes discussions in sections 2 and 3 questionable. The idea of a single 'aerosol lifetime' parameter is also not very helpful. For example, aerosol types such as sea salt in the tropics can have very short lifetime and participate in hydrological cycle very actively, however they do not exert much forcing to the system. On the other hand, dust from Africa may not interact with major precipitation systems for a long time and therefore have much longer lifetime while they are critical in determining aerosol total forcing.

The reviewer points out correctly that e_c is different for different clouds. Also, aerosol particles of different hygroscopicity activate with different efficiencies, i.e., with different e_a . We can conclude that Pruppacher and Jaenicke (1995) introduced these simplifications because their goal was quantification of the water vapor and aerosol lifetimes while observations of several parameters necessary for their calculations were not available in sufficient detail. In the present manuscript we changed the equations to be more representative for an atmosphere with different clouds as described above. Further, we included more discussion on the heterogeneity of aerosol and why, for our purpose, a

constraintment of the system to soluble aerosol is required.

3. No sound physical ground for expecting simple scaling relationship among aerosols, cloud, precipitation and global averaged temperature is provided except using simple equations that are questionable. This is unlike the relationship between water vapor and temperature, which has a whole body of literature and physical ground to rely on. The author has to provide such convincing conceptual arguments before quantify them with simple models.

The relation between water vapor and temperature is of a direct physical and thermodynamical nature. The Clausius Clapeyron equation shows how the saturation water vapor pressure depends on temperature. Such a physical connection, which would certainly make aerosol modelling less cumbersome, is not active between aerosol and temperature. Instead, aerosol is a highly complex phenomenon in our atmosphere for which an unambiguous relation with temperature can not be expected to exist. However, we believe the equations presented in our study do have a physical ground: the atmosphere contains soluble aerosol, moist air can be involved in cloud formation, and soluble aerosol is involved in cloud formation and is taken up in cloud water. We have included a new Figure 1 to clarify this. The equations describe this, albeit in a simplified way when compared to the actual complexity of aerosol and aerosol removal. We do not agree that they are questionable, they are derived from valid steady state equations and therefore, within the constraints discussed in the manuscript, provide a valid representation of the system under study.

4. Much more concrete results and analyses are needed even one accepts all the derivation and general arguments made in the text. The current title and abstract seem to cover quite a broad subject while the effort and evidence to support conclusions and arguments made in the text fall short.

As remarked above, our study presents the possible existence of a direct relation between hydrological cycle characteristics and the (temperature response of the) aerosol lifetime. In that sense our manuscript presents a new perspective on aerosol forcing and associated uncertainties. We have rewritten parts of the manuscript to make this clearer. We agree with the reviewer that more data are needed to demonstrate and quantify this relation, specifically from aerosol-climate models. We have made suggestions for a model intercomparison based on the presented equations that may be able to provide quantification of the relation aerosol-hydrological cycle.

Minor comments:

1. Before using any term it would help the readers tremendously if a clear definition and their units are provided.

We have mentioned the units and gave clearer definitions where necessary.

2. P16496 L 9: should be Randall et al. not 'Randell'.

The typo has been corrected

3. P16496 L 28: precipitation and aerosol are never 'homogeneous' in any stretch.

The term homogeneous can be misleading here. We meant that mass mixing ratios of the species present in the domain (i.e., water vapor and aerosol) are uniform throughout the domain. We have formulated this more clearly now.

4. Section 2.1: aerosols that do not actively participate in precipitation or survive precipitation can still affect clouds and climate in many ways. Thus, the bulk consideration presented here may not begin to capture the full range of aerosol-climate interactions even with the strong assumptions made here.

We did not intend this study to be representative for all aerosol species. It focusses on aerosol species for which wet deposition is the dominant removal mechanism and, hence, is directly linked to water vapor. We have mentioned this more clearly in the current manuscript.

5. P 16499 L 15-19: low clouds should contribute relatively little to the fCU term while all the references are about low, warm clouds. In addition, SST dependence may not be an entirely valid analog to the sensitivity that the author is after here.

It is still unclear which contribution different type clouds give to aerosol removal. Liu et al. (JGR, 2001) find for ^{210}Pb and ^7Be that 30-35% is removed by wet deposition from stratiform cloud and ~60% by convective clouds. The remainder, i.e., 10%, is removed by dry deposition. For sulfate, simulated contributions to removal are ~4% for dry deposition, 53% for stratiform and 43% for convection (Croft et al., ACP, 2010). This means that, in terms of aerosol removal, both cloud types are comparably important. The SST-cloud cover relation concerns stratiform clouds, and therefore could be relevant for the discussion. However, we removed this from the manuscript as, on itself, it was indeed not a convincing argument.