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Aerosol lifetime and climate change

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Abstract

The dominant removal mechanism for atmospheric aerosol is activation of particles to cloud droplets and subsequent wet deposition in precipitation. The atmospheric lifetime of aerosol is thus closely coupled to the atmospheric cycling time of water vapor. Changes of hydrological cycle characteristics resulting from climate change therefore directly affect aerosol lifetime, and thus the radiative forcing exerted by aerosol. This study expresses the coupling between water vapor and aerosol lifetimes and their temperature sensitivities in fundamental equations and in terms of the efficiency of processing of air by precipitating clouds. Based on climate model simulations these temperature sensitivities are estimated to be on the order of $+5.3\,\%\,K^{-1}$, but this may be an overestimation. Generally, shifting spatial and temporal patterns of aerosol (precursor) emissions and precipitation, and changes in aerosol activation efficiency probably influence aerosol lifetimes more than climate change itself, resulting in a wide range of simulated aerosol lifetime sensitivities between aerosol-climate models. It is possible that the climate sensitivity of models plays a role. It can be argued that climate sensitivity is intrinsically coupled with the simulated (temperature sensitivity of the) aerosol lifetime through the distribution of water vapor and aerosol between the lower and upper troposphere. This implies a fundamental relation between various feedback forcings (water vapor, lapse rate, cloud) and the aerosol forcing, illustrating the key role of the hydrological cycle in different aspects of the climate system.

1 Introduction

Aerosol influences the radiative budget by scattering and absorption of solar radiation by the particles themselves (the direct and semi-direct effect), and by influencing cloud reflective properties (the first indirect effect) (Twomey, 1974; Forster et al., 2007; Lohmann and Feichter, 2005) and precipitation formation (the second indirect or lifetime effect) (Albrecht, 1989; Rosenfeld et al., 2008). Estimated direct and indirect

forcings together range between −0.6 and −2.4 W m^{-2}, respectively (Forster et al., 2007) and this uncertainty has not been reduced much in the last decades (Lohmann et al., 2010).

The radiative forcing exterted by a radiatively active atmospheric trace species depends on its amount or atmospheric burden. Under assumption of steady-state the burden is determined by the emission and/or production strength and by the removal efficiency. Wet deposition is the dominant removal mechanism for many aerosol components, and its efficiency is determined essentially by two parameters, i.e. the efficiency of the processing of air by (precipitating) clouds (Pruppacher and Jaenincke, 1995) and the uptake of aerosol in cloud water through activation scavenging. The latter depends on chemical composition and size distribution of the aerosol, the effects of surfactants and of semi-volatile inorganic and organic aerosol components, and on cloud dynamics (Jensen and Charlson, 1984; Jacobson et al., 2000; Dusek et al., 2006; Topping and McFiggans, 2012; Derksen et al., 2009). Comparison of global aerosol model performances yields a large range of aerosol lifetimes and burdens, and a strong anticorrelation between both. For example, in the atmospheric (bulk) sulfur model intercomparison study COSAM (Comparison of large-scale sulfur models; 1999) simulated wet deposition efficiencies for Europe range between 0.1 and 0.5 day^{-1}, and they are associated with sulfate lifetimes between 6 and 1 days, respectively (Roelofs et al., 2001). Differences in the representation of the hydrological cycle (cloud occurrence, precipitation distribution) were mentioned as the main cause of the intermodel variabilities. A more recent intercomparison involving more sophisticated aerosol models again reports global aerosol wet removal efficiencies between 0.1 and 0.5 day^{-1}, with global aerosol lifetimes between 3.5 and 0.5 days (Textor et al., 2006; their Figs. 3a and 4a). On the other hand, when different representations of aerosol processes are implemented in the same climate model simulated lifetimes and burdens are more consistent. For example, global sulfate lifetimes from studies that use the climate model ECHAM (version 4 or 5) with different representations for aerosol and aerosol scavenging range between 3.6–4.4 days (Roelofs et al., 1998, 2006; Stier et al., 2005; Zhang

et al., 2012). This indicates a significant influence of the simulated hydrological cycle characteristics.

The hydrological cycle also interacts with radiation (Chahine, 1992). Clouds contribute to cooling through reflection of solar radiation and to warming by absorption and emission of longwave radiation, and water vapor acts as a greenhouse gas and contributes to warming. Due to the temperature dependence of the saturation water vapor pressure, the water vapor burden is sensitive for climate change and a positive water vapor-temperature feedback occurs (Dessler and Sherwood, 2009), with other feedbacks associated with atmospheric lapse rates and clouds (Randell et al., 2007). The impact of the feedbacks on surface temperature is expressed in the so-called climate sensitivity. For climate models this is often defined as the change of the simulated surface temperature following a doubling of carbon dioxide (Randell et al., 2007). Model estimates of the climate sensitivity range between 2.1° and 4.4° (Solomon et al., 2007), and this uncertainty range has also not decreased much in recent decades (Schwartz, 2008; Roe and Baker, 2007). The relatively large uncertainties in aerosol forcing and climate sensitivity provide sufficient leeway for individual climate models to simulate a surface temperature increase over the past 150 yr that is consistent with observations, thereby displaying a typical correlation between both parameters (Kiehl, 2007), but they cause large uncertainties in future projections of climate change (Andreae et al., 2005; Knutti and Hegerl, 2008).

The preceding indicates that in order to better understand (differences in) simulated global aerosol distributions and forcing also the coupling between the hydrological cycle and aerosol must be taken into account. The current study investigates this coupling on a basic level, building on a previous study of Pruppacher and Jaenicke (1995). Section 2 presents expressions for the atmospheric lifetime and atmospheric concentration of water vapor and aerosol as well as their temperature sensitivities, based on the the processing efficiency of air in precipitating (warm) clouds and under assumption of steady-state and a homogeneous air mass. In Sect. 3 simulation results from several

aerosol climate model studies and a possible connection with climate sensitivity are discussed. Section 4 presents conclusions of this study.

2 Water vapor and aerosol lifetimes

2.1 First-order removal

First we derive an expression for the first-order removal rate of water vapor, L_v, and its atmospheric lifetime, τ_v, from the steady-state equation for water vapor, $w_v = E / L_v$, with w_v the tropospheric concentration of water vapor and E the evaporation flux. The flux of air passing through all clouds is (Pruppacher and Jaenicke, 1995):

$$F_{\text{air}} = A_E \sum \left(c_{c,i} U_{c,i} \right) \tag{1}$$

with $c_{c,i}$ and $U_{c,i}$ the average fractional cloud cover and updraft velocity for cloud type i, respectively, and A_E is the earth surface area. Introducing e_c for the condensation efficiency, i.e. the ratio of the cloud water mass and the mass of water vapor entering a cloud, the rate at which global water vapor condenses in all clouds becomes:

$$F_{\text{air,c}} = \left(\sum c_{c,i} U_{c,i} \right) A_E w_v e_c \tag{2}$$

If f_i is the volume fraction of clouds of type i that produce precipitation, and assuming that all condensate is removed as precipitation in these clouds, the columnar precipitation flux $F_{\text{air,p}}$ becomes:

$$F_{\text{air,p}} = \left(\sum f_i c_{c,i} U_{c,i} \right) A_E w_v e_c \tag{3a}$$

Simplifying this for clarity gives the precipitation flux P, which equals evaporation E:

$$P = \left(f c_c U_c / H \right) w_v e_c \tag{3b}$$

where H is the height of the troposphere, and P and w_v are in units of column burden ($\text{g m}^{-2}\,\text{s}^{-1}$ and g m^{-2}) or concentration ($\text{g m}^{-3}\,\text{s}^{-1}$ and g m^{-3}). The first-order removal rate L_v (e.g. day^{-1}) and lifetime τ_v (day) for water vapor then become:

$$L_v = 1/\tau_v = \left(f c_c U_c / H \right) e_c \tag{4}$$

Using climatological observations for precipitation, cloud parameters, water vapor content and precipitating fraction, and assuming e_c is 0.3, a typical water vapor lifetime of 9.0 days can be calculated (Pruppacher and Jaenicke, 1995).

The steady-state aerosol concentration is given by $w_{AP} = S_{AP} / L_{AP}$, with S_{AP} aerosol emissions (note that aerosol emissions are implicitly assumed to be primary in our approach). By considering the flux of air in clouds (Eq. 1) and a scavenging efficiency for uptake of aerosol in cloud water (e_a), the flux of aerosol removed through precipitation, P_{AP}, is:

$$P_{AP} = \left(f c_c U_c / H \right) w_{AP} e_a \tag{5}$$

where P_{AP} and w_{AP} are in similar units as P and w_v. Note that e_a actually co-depends on updraft velocity (Jensen and Charlson, 1984) and also on entrainment of aerosol particles into the cloud (Derksen et al., 2009), but this is not considered here. The aerosol removal rate L_{AP} and lifetime τ_{AP} are:

$$L_{AP} = 1/\tau_{AP} = \left(f c_c U_c / H \right) e_a \tag{6}$$

For arbitrary values for e_a of 0.5 and 1.0 Pruppacher and Jaenicke (1995) estimate aerosol lifetimes of 5.34 and 2.67 days, respectively. Eq. (6) shows that the aerosol lifetime depends on the same cloud parameters as the water vapor lifetime. Both lifetimes are directly coupled and proportional according to a scavenging-condensation efficiency ratio $\tau_{AP} / \tau_v = e_c / e_a$. This ratio, when applied to aerosol-climate models, can be used to compare the relative cycling speeds of water vapor and aerosol in models, and thus may be a useful tool to analyze intermodel variabilities in simulated aerosol burdens.

2.2 Temperature sensitivity of water vapor lifetime

The temperature sensitivity of the water vapor concentration is derived from the steady-state equation while the removal rate L_v is separated into different parts for cloud properties and for the condensation efficiency:

$$\frac{\mathrm{dln}w_v}{\mathrm{d}\Delta T} = \frac{\mathrm{dln}P}{\mathrm{d}\Delta T} - \frac{\mathrm{dln}(f c_c U_c)}{\mathrm{d}\Delta T} - \frac{\mathrm{dln}e_c}{\mathrm{d}\Delta T} \quad (7)$$

The temperature sensitivity of the water vapor lifetime is:

$$\frac{\mathrm{dln}\tau_v}{\mathrm{d}\Delta T} = -\frac{\mathrm{dln}(f c_c U_c)}{\mathrm{d}\Delta T} - \frac{\mathrm{dln}e_c}{\mathrm{d}\Delta T} \quad (8)$$

Held and Soden (2006) present simulated temperature sensitivities for water vapor and precipitation in response to a LW forcing for several climate models, with average temperature sensitivities of 7.5 % K^{-1} for the atmospheric water vapor concentration and 2.2 % K^{-1} for the precipitation flux. Examination of the Clausius-Clapeyron equation shows that $\mathrm{dln}e_c/\mathrm{d}\Delta T$ is generally smaller than 0.5 % K^{-1}. Without this term the temperature sensitivity of the water vapor lifetime can be attributed entirely to a cloud response involving f, c_c and/or U_c, which then has a magnitude of –5.3 % K^{-1} (Eq. 7). The negative temperature response of $(f c_c U_c)$ is qualitatively consistent with observations from COADS (Comprehensive Ocean Atmosphere Data Set) and ISCCP (International Satellite Cloud Climatology Project) that indicate a negative correlation between local sea surface temperatures and low-level cloud cover (Clement et al., 2009), but it is not supported by other observational studies (Wylie et al., 2005; Evan et al., 2007).

The derived cloud response implies a temperature sensitivity of the water vapor lifetime of +5.3 % K^{-1} (Eq. 8). It must be remarked that the magnitude of the response depends on the nature of the forcing, i.e. shortwave or longwave. This determines the so-called fast response with adjustment of radiative and non-radiative (latent and sensible heat) fluxes (e.g. Allen and Ingram, 2002; Lambert and Faull, 2007; Bala et al.,

2009). For example, Bala et al. (2008) simulate a precipitation temperature sensitivity of 1.5 % K^{-1} for a LW forcing and and 2.4 % K^{-1} for a SW forcing, respectively. However, simulated water vapor sensitivities are similar under both forcings, probably reflecting the tendency of models to maintain relative humidity. The cloud response may therefore be larger for a LW than a SW forcing. Note that a recent estimate of the precipitation temperature sensitivity based on ocean salinity is much larger, 8 % K^{-1} (Durack et al., 2012), which implies a relatively small cloud response (Eq. 7).

2.3 Temperature sensitivity of aerosol lifetimes

The temperature dependence of the aerosol concentration is given by:

$$\frac{\mathrm{dln}w_{AP}}{\mathrm{d}\Delta T} = \frac{\mathrm{dln}S_{AP}}{\mathrm{d}\Delta T} - \frac{\mathrm{dln}(f c_c U_c)}{\mathrm{d}\Delta T} - \frac{\mathrm{dln}e_a}{\mathrm{d}\Delta T} \quad (9)$$

or, as function of time:

$$\frac{\mathrm{dln}w_{AP}}{\mathrm{d}t} = \frac{\mathrm{dln}S_{AP}}{\mathrm{d}t} - \frac{\mathrm{dln}(f c_c U_c)}{\mathrm{d}\Delta T}\frac{\mathrm{d}\Delta T}{\mathrm{d}t} - \frac{\mathrm{dln}e_a}{\mathrm{d}t} \quad (10)$$

where $\mathrm{d}\Delta T/\mathrm{d}t$ reflects the rate of temperature change. Equation (10) illustrates that the temporal evolution of atmospheric aerosol is governed by three factors. The first term on the right hand side describes changing aerosol emissions; these may be associated with anthropogenic activities but also with temperature-dependent processes such as climate-biosphere feedbacks and chemical transformation rates (Carslaw et al., 2010). The second term refers to the response of cloud properties to a change of temperature as discussed above. The third term refers to changes in the aerosol activation or scavenging efficiency. The temperature sensitivity of the aerosol lifetime is given by:

$$\frac{\mathrm{dln}\tau_{AP}}{\mathrm{d}\Delta T} = -\frac{\mathrm{dln}(f c_c U_c)}{\mathrm{d}\Delta T} - \frac{\mathrm{dln}e_a}{\mathrm{d}\Delta T} \quad (11)$$

Including the estimated lifetime temperature sensitivity the aerosol concentration changes according to:

$$w_{\mathrm{AP},\Delta T} = w_{\mathrm{AP},0}\left(\frac{S_{\mathrm{a},\Delta T}}{S_{\mathrm{a,o}}}\right)\left(\frac{e_{\mathrm{a},0}}{e_{\mathrm{a},\Delta T}}\right)\exp(0.053^{*}\Delta T) \quad (12)$$

and the aerosol lifetime to:

$$\tau_{\mathrm{AP},\Delta T} = \tau_{\mathrm{AP},0}\left(\frac{e_{\mathrm{a},0}}{e_{\mathrm{a},\Delta T}}\right)\exp(0.053^{*}\Delta T) \quad (13)$$

Equations (12) and (13) describe effects of changing emissions, scavenging efficiency and temperature on w_{AP}, where the subscript 0 denotes a reference atmosphere. For example, for constant e_{a} the aerosol concentration doubles when emissions are doubled, while the lifetime does not change. Including also a climate warming of 2.7° the concentration increases by ∼130 %, i.e. 30 % more than without climate change, due to an increased lifetime. This invokes a negative feedback: the higher aerosol concentration exerts a negative SW radiative forcing that results in a surface cooling, and according to Eqs. (7) and (9) the cooling induces more efficient processing of air by precipitating clouds and a decrease of the aerosol burden. This negative feedback between atmospheric aerosol and precipitation acts through the surface temperature, and it is therefore fundamentally different from the aerosol second indirect (lifetime) effect where precipitation production is directly affected by the influence of aerosol on cloud microphysics. For the observed global temperature increase of 0.76° between the pre-industrial era and the present-day (Solomon et al., 2007) Eq. (5) implies a relatively modest 4 % increase of the aerosol burden due to climate change. Evidently, the large contribution of anthropogenic emissions to the global aerosol burden in the contemporary atmosphere (Charlson et al., 1992; Dentener et al., 2006) has affected the aerosol burden far more since pre-industrial times than climate change effects. Also, e_{a} may have changed during the last 150 yr, for example because scavenging efficiencies in polluted air are smaller than in cleaner pre-industrial air (e.g. Jensen and Charlson,

1984), and insoluble aerosol such as dust becomes soluble due to interaction with pollution which favors its removal from the atmosphere (Stier et al., 2006).

3 Aerosol lifetimes in aerosol-climate models

3.1 Comparison of temperature sensitivities

The foregoing shows that aerosol lifetime, the hydrological cycle, and their responses to climate change are coupled, at least for the relatively simple system and homogeneous air mass considered in our approach. Three-dimensional aerosol climate models display large temporal and spatial variabilities in water vapor and aerosol concentrations, they consider mechanisms for secondary aerosol formation and aerosol is removed from the atmosphere in various ways, and this may decrease the relevance of this coupling to some extent. We compare Eq. (13) with the results of four global modelling studies that report computed temperature sensitivities for precipitation and global (sulfate) aerosol lifetimes, listed in Table 1. The studies apply a LW or SW forcing, or a mixture of both. Simulated temperature sensitivities for precipitation range between 2.0 and 2.8 % K^{-1}, while aerosol lifetime temperature sensitivities range between −6 and 6 % K^{-1}.

The GFDL study (Fang et al., 2011) considers ideal soluble tracers and an aerosol removal efficiency that depends on simulated precipitation rates only. Ideal tracer emissions occur mostly in (sub)tropical regions, concurring with the strongest evaporation fluxes. This study is therefore to some extent comparable to our relatively simple framework, and the computed aerosol lifetime sensitivity is in relatively good agreement with our derived value. Fang et al. (2011) attribute the lifetime increase to a decreased precipitation frequency. We note that the precipitation frequency appears more critical for aerosol scavenging than the precipitation rate (Jacob and Winner, 2009), which is consistent with the use of the precipitation volume fraction in Eq. (6).

Comparison is less straightforward for the other model studies that consider size-resolved aerosol and more realistic aerosol dynamics, microphysics and emission distributions for several aerosol species. From the study with ECHAM-HAM (Kloster et al., 2010) we consider two scenarios: one with changes in greenhouse gas (GHG) emissions only and the other with changes in both GHG and aerosol (precursor) emissions. Different precipitation temperature sensitivities are computed, consistent with the different responses to LW and SW forcings as described above. The computed temperature sensitivities of the sulfate lifetime are positive but smaller than our estimate. The two studies with GISS (Racherla and Adams, 2006; Liao et al., 2006) also use a detailed aerosol scheme but simulate a negative temperature sensitivity for the aerosol lifetime. This is attributed by the authors to the increase in precipitation, but it is more likely that an increased precipitation frequency is responsible. According to the authors changes in simulated seasonalities of precipitation and aerosol concentrations further contribute to the lifetime decrease. A study with HadGEM2-ES (Bellouin et al., 2011; not listed in Table 1) computes a sulfate lifetime that varies from 3.8 days in 1850 to 3 days in 1960 and then to four days at the end of the 21st century. The large variability in lifetime in this study is mainly attributed to the influence of shifting spatial patterns of emissions, clouds and precipitation. In addition, and noting that aerosol activation is co-determined by updraft speed which varies with cloud type, shifting transport patterns may alter the relative contributions by different cloud types to the total aerosol removal, thereby altering the average e_a. This probably also plays a role in the ECHAM-HAM and GISS studies.

3.2 The influence of climate sensitivity

As mentioned in the introduction a significant correlation exists between the climate sensitivity (CS) of aerosol-climate models and the aerosol forcing applied in climate simulations (Kiehl, 2007). On the other hand, several studies report a positive correlation between CS and the simulated cloud radiative feedback at solar wavelengths (Schwartz, 2008; Andrews et al., 2012). A positive cloud radiative feedback

is consistent with reduced cloud cover, especially in the lower troposphere (LT). This is consistent with a negative cloud response (i.e. a negative temperature sensitivity of $f c_c U_c$), and therefore associated with longer water vapor and aerosol lifetimes (Eqs. 8 and 11). Since a longer lifetime also implies a stronger forcing, it appears that CS, lower tropospheric cloud response and aerosol forcing are inherently coupled in aerosol-climate models.

We note that most aerosol resides in the LT and its atmospheric lifetime is dominated by the LT hydrological cycle. It is therefore useful to consider water vapor burdens and lifetimes (Eqs. 7 and 8) in the LT separately from those in the upper troposphere (UT). Simulated temperature sensitivities for water vapor in the UT and LT are, on average, $14\,\%\,K^{-1}$ and $6\,\%\,K^{-1}$ (supplementary material in Soden et al., 2002; John and Soden, 2007), but model results display considerable variability around these values. The vertical transport efficiency in the troposphere may be a key parameter, in such a way that models with relatively large CS deposit more water vapor in the UT in response to a warming and less water vapor in the LT, and vice versa. This is supported by the fact that simulated values for the water vapor and lapse rate feedbacks are smaller or more negative for models with low CS than high CS (between 1.5 and $2.4\,W\,m^{-2}$ and -0.4 and $-1.27\,W\,m^{-2}$, respectively). Also, the combined water vapor and lapse rate feedbacks correlate relatively well with the cloud radiative feedback (Soden and Held, 2006). The latter ranges between $0.15\,W\,m^{-2}$ and $1.15\,W\,m^{-2}$ for models with small and large CS, respectively (see Fig. 1, based on data reported in Soden and Held, 2006). If the amount of additional water vapor in the LT is relatively small (i.e. $d\ln P/d\Delta T$ in the LT is significantly less than ~ 0.06) saturation can only be restored if the cloud response leads to an increase of the water vapor lifetime, i.e. $d\ln(f c_c U_c)/d\Delta T$ is negative (Eq. 8), consistent with a positive cloud radiative feedback, and with longer aerosol lifetimes (Eq. 11). Models with a relatively small CS simulate a larger increase of water vapor in the LT which is consistent with a smaller or negative cloud radiative feedback, and consequently these models may simulate only a small increase or a decrease of aerosol lifetime upon a warming. The signs of the

simulated temperature sensitivities of aerosol lifetime listed in Table 1 are consistent with this. We note that intermodel differences in vertical transport between the LT and UT also affect the vertical distribution of aerosol (precursors). Since aerosol lifetimes are generally larger in the UT, model transport characteristics probably lead to stronger increases in aerosol lifetime in models with relatively large CS.

However, observations and reanalyses do not corroborate the simulated water vapor trends in the upper troposphere (Trenberth et al., 2005; Paltridge et al., 2009; Dessler and Davis, 2010). Further, the boundary layer in models is generally too dry (up to 25 %) and the upper troposphere too moist (by 75 % on average) (John and Soden, 2007), probably due to too strong upward transports (Lindzen et al., 1996). This is in accordance with several studies that suggest that the actual climate sensitivity of the Earth may be at the lower end of the current uncertainty range (Harvey and Kaufmann, 2002; Gillett et al., 2012). For our analysis this again implies that the temperature sensitivity of water vapor and aerosol lifetimes may be considerably smaller than $+5.3\,\%\,K^{-1}$.

4 Summary and conclusions

With a few fundamental equations, adequate for steady-state conditions and a well-mixed atmosphere, we explored the connection between aerosol and water vapor lifetimes. The removal of water vapor and wet deposition of aerosol are governed by the processing efficiency of air by precipitating clouds, expressed in terms of precipitating fraction, cloud cover and updraft speed. Both lifetimes are proportional through the efficiencies of condensation, e_c, and of aerosol scavenging, e_a.

The temperature sensitivity of the aerosol lifetime is expressed in terms of a cloud response involving the parameters mentioned above, and of the aerosol scavenging efficiency. Based on simulated temperature sensitivities for water vapor content and precipitation we derive a negative cloud response, i.e. a combined decrease in cloud cover, precipitating volume and updraft velocity, of $-5.3\,\%\,K^{-1}$. The estimated temperature sensitivity of the water vapor and aerosol lifetimes then becomes $+5.3\,\%\,K^{-1}$,

consistent with global model simulations that apply idealized soluble tracers and primary emissions. Observations of cloud cover and recent estimates of the precipitation temperature sensitivity, that are much larger than predicted by climate models, indicate that the actual magnitude of the cloud response is probably smaller. The aerosol-climate models considered in our study display a wide range of temperature sensitivities. This is partly due to processes not represented in our study, for example, atmospheric transport of aerosol between emission and removal (Bellouin et al., 2011; Zhang et al., 2007; Carslaw et al., 2010; Zhou et al., 2011), gaseous and in-cloud production of secondary aerosol matter, and compensating effects between aerosol wet removal and other removal processes, i.e. below-cloud scavenging and dry deposition. We also neglected any size-dependence of aerosol-cloud interactions, the influence of aerosol scavenging on precipitation formation (the "lifetime" effect) and other climate-chemistry feedbacks (Raes et al., 2010) that affect the removal rate.

Climate sensitivity is predominantly associated with water vapor in the upper troposphere, and aerosol lifetime with the water vapor cycling efficiency in the lower troposphere. Therefore climate sensitivity and aerosol are linked through the vertical distribution of water vapor between the lower and upper troposphere. Although the relation between aerosol concentration and the associated radiative forcing is not straightforward we tentatively conclude that the correlation between climate sensitivity and aerosol forcing found in aerosol-climate models is not (only) an inadvertant result of adjusting the simulated temperature evolution to observations (Kiehl, 2007; Kerr, 2007). Instead it reflects the strong influence of the hydrological cycle, especially the water vapor cycling time, on different aspects and components of the climate system, thereby linking the positive and negative radiative forcings exerted by water vapor, clouds and aerosol. Since only few global model studies of water vapor and aerosol lifetimes were available during our analysis, studies from more aerosol-climate models are needed to evaluate the robustness of this link. In general it can be concluded that a reduction of present uncertainties in aerosol radiative forcing and a better assessment of climate model performance requires an approach directed at the coupling

between the hydrological cycle and aerosol, involving a more accurate quantification of the processing efficiency of air in different types of (precipitating) clouds, of the aerosol scavenging efficiency, and of the global and/or regional changes in these parameters during the last 150 yr.

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Table 1. Simulated temperature sensitivities of precipitation and sulfate lifetime.

	Model	CS[a]	ΔF (W m^{-2})	$\Delta P / \Delta T$ (% K^{-1})	$\Delta \tau_{AP} / \Delta T$ (% K^{-1})
this work	"average"		LW	+2.2	+5.3
Fang et al (2011)[b]	GFDL	3.4	LW	+2.2	+6.1
Kloster et al. (2010)[c]	ECHAM	3.4	LW	+2.0	+2.4
Kloster et al. (2010)[d]	ECHAM	3.4	LW, SW	+2.4	+3.9
Liao et al. (2006)[e]	GISS-II	2.7	LW	+2.0	−3.1
Racherla/Adams (2006)[f]	GISS-II	2.7	SW	+2.8	−6.2

[a] Climate sensitivity (Randall et al., 2007).
[b] Idealized soluble tracer, climate change by 2 × CO_2.
[c] Emission changes for greenhouse gases, yr 2000 vs. 2030.
[d] Emission changes for greenhouse gases and aerosol, yr 2000 vs. 2030.
[e] CO_2 increase from 368 to 836 ppb, unchanged emissions, yr 2000 vs. 2100.
[f] Forcing of surface temperature from changes in ocean boundary conditions, unchanged emissions, yr 1990 vs. 2050.

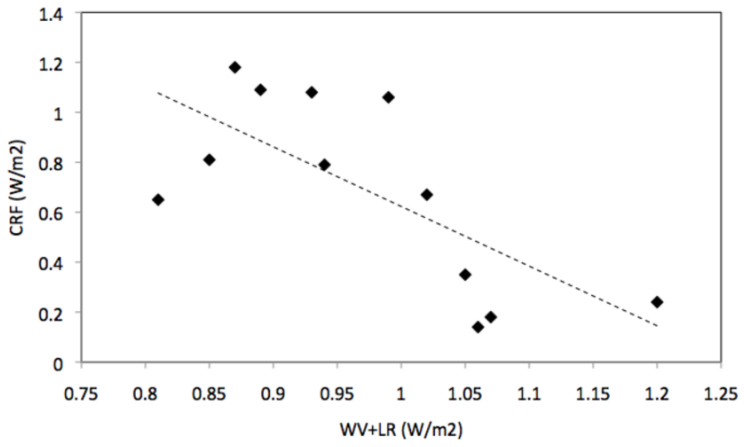

Fig. 1. Sum of water vapor (WV) and lapse rate (LR) feedbacks vs. cloud radiative feedback (CRF) (R^2 = 0.50). Data from Soden and Held (2006), their Table 1.