Total aerosol effect: forcing or radiative flux perturbation?

Ulrike Lohmann, Trude Storelvmo1, Andy Jones2, Leon Rotstyn3, Surabi Menon4, Johannes Quaas5, Annica Ekman6, and Dorothy Koch, Reto Ruedy7

1Institute of Atmospheric and Climate Science, ETH Zurich, Switzerland.  
2Met Office Hadley Centre, Exeter, UK.  
3Centre for Australian Weather and Climate Research, CSIRO, Aspendale, Victoria, Australia.  
4Lawrence Berkeley National Laboratory, USA.  
5Max Planck Institute for Meteorology, Hamburg, Germany.  
6Stockholm University, Sweden.  
7NASA GISS, New York, NY, USA.  

Correspondence to: Ulrike Lohmann (ulrike.lohmann@env.ethz.ch)

Abstract. Uncertainties in aerosol forcings, especially those associated with clouds, contribute to a large extent to uncertainties in the total anthropogenic forcing. The interaction of aerosols with clouds and radiation introduces feedbacks which can affect the rate of rain formation. Traditionally these feedbacks were not included in estimates of total aerosol forcing. Here we argue that they should be included because these feedbacks act quickly compared with the time scale of global warming. We show that for different forcing agents (aerosols and greenhouse gases) the radiative forcings as traditionally defined agree rather well with estimates from a method, here referred to as radiative flux perturbations (RFP), that takes these fast feedbacks and interactions into account. Thus we propose replacing the direct and indirect aerosol forcing in the IPCC forcing chart with RFP estimates. This implies that it is better to evaluate the total anthropogenic aerosol effect as a whole.

1 Introduction

Aerosols affect climate directly by scattering and absorption of shortwave and thermal radiation (direct effect). The global-mean net direct effect at the top-of-the-atmosphere (TOA) is a cooling that partly offsets the warming due to greenhouse gases. It is estimated as -0.5 W m^{-2} with a 5 to 95% confidence range of -0.1 to -0.9 W m^{-2} (Forster et al., 2007). In addition, aerosols modify the radiation budget indirectly by acting as cloud condensation nuclei and ice nuclei. The cloud albedo enhancement (first indirect effect, cloud albedo effect or indirect aerosol forcing) of warm stratiform clouds refers to an increase in cloud droplet number concentration due to anthropogenic
aerosols for a constant liquid water content (Twomey, 1977). These more numerous and smaller cloud droplets increase the total surface area and thus cloud albedo. The cloud albedo effect can be calculated as a forcing because of the assumption of a constant liquid water content. Global-mean model estimates of the cloud albedo effect have remained rather constant over time (see figure 1) and amount to roughly -0.9 W m\(^{-2}\). The -0.9 W m\(^{-2}\) estimate that is obtained from the average over all published estimates, treating each of them equal (one paper one vote) is slightly larger than the estimate of the cloud albedo effect in the fourth assessment report of the Intergovernmental Panel on Climate Change (IPCC) where a different weighting procedure was used. There the median value of the indirect aerosol forcing was estimated as -0.7 W m\(^{-2}\) with a 5 to 95% range of -0.3 to -1.8 W m\(^{-2}\) (Forster et al., 2007). The rather large uncertainty in both the direct and indirect (cloud albedo effect) forcing accounts for a large fraction of the uncertainty in the total anthropogenic forcing (Kiehl, 2007).

In addition to the cloud albedo effect, there are multiple other effects such as the cloud lifetime effect, the semi-direct effect and aerosol effects on mixed-phase, convective and cirrus clouds (Lohmann and Feichter, 2005; Denman et al., 2007). These effects need to be evaluated as radiative flux perturbation (\(RFP\)) (Haywood et al., 2009) or climate forcing (Forster and Taylor, 2006) because these effects do not act "instantaneously". This means that the difference in the top-of-the-atmosphere radiation budget between two simulations, one with pre-industrial emissions and one with present-day emissions is evaluated. \(RFP\) estimates thus involve fast feedbacks and interactions in the climate system that induce changes in the meteorology. This does not conform to the usual definition of "radiative forcing" Forster et al. (2007), in which only one thing is changed while leaving everything else constant.

Also, telling the multitude of different effects that refer to different physical processes apart is not easy as different interactions can take place at the same time. Also, if aerosols and/or cloud droplet number concentrations are calculated interactively in the model, the calculation of the aerosol radiative forcing is not straightforward because aerosols will then also influence the precipitation formation and with that cause an additional change in cloud properties. If these interactions and feedbacks are taken into account, then the difference between simulations with different aerosol emissions is a radiative flux perturbation (\(RFP\)). The advantage of the \(RFP\) method over the strictly defined forcing is that it allows the radiative impact of aerosols on both cloud albedo and precipitation efficiency to be evaluated. As shown in figure 1, if estimates of other aerosol-cloud interactions are considered next to the cloud albedo effect, then these estimates are mostly larger than the cloud albedo effect alone. This suggests that most of the model-calculated additional effects do not offset the cloud albedo effect, but rather constitute an additional cooling. Although the total indirect effect shows more scatter than the cloud albedo effect, more recent estimates indicate smaller (less negative) values (see supplement). Some of the smallest estimates result from estimates of the indirect aerosol forcing from satellite data or result from general circulations model (GCM) estimates.
that constrained the indirect aerosol effect by satellite data. Also, some aerosol interactions with mixed-phase clouds can partly offset the forcing due to the cloud albedo effect.

A complementary approach to estimate either the indirect aerosol effect or the total anthropogenic aerosol effect is to infer it as a residual using the observed temperature record over land, and estimates of the ocean heat uptake and the evolution of greenhouse gas and solar radiative forcing (Anderson et al., 2003; Hegerl et al., 2007). These so-called inverse estimates constrain the total cooling forcing over the 20th century, attributable to anthropogenic aerosols, to a likely range\(^1\) of -1.7 to -0.1 W m\(^{-2}\) (Hegerl et al., 2007). A total anthropogenic aerosol effect that is more negative than -1.7 W m\(^{-2}\) would thus be inconsistent with the observed warming.

The question that remains is how the total aerosol effect that includes fast feedbacks and interactions due to the cloud lifetime effect, semi-direct effect or aerosol interactions with mixed-phase and ice clouds can be compared with the forcings from the well-mixed greenhouse gases (GHG). The difference between the forcing (as strictly defined) and the \(RFP\) (change in TOA net radiation between two GCM simulations with pre-industrial versus present-day aerosol emissions, see also below) due to the aerosol indirect effect was first investigated by Rotstayn and Penner (2001). They found from their atmospheric GCM coupled to a mixed layer ocean model that the differences in the climate sensitivity due to using the \(RFP\) method were smaller than the differences in the climate sensitivity due to different forcings. They hence argued that \(RFP\) estimates from aerosols should be compared to forcing estimates from GHG. Put differently, given that cloud responses to aerosol perturbations are much quicker compared with the timescale of global warming, it makes sense from an energy balance perspective (Murphy et al., 2009) and is more suitable in the conceptual framework of radiative forcing and climate sensitivity (Gregory et al., 2004; Knutti and Hegerl, 2008; Quaas et al., 2009a) to include the radiative impact of fast feedbacks and interactions in estimates of the effects of aerosols.

The issue of how to include fast feedbacks and interactions is not new. One approach suggested by Joshi et al. (2003) and Hansen et al. (2005) is to obtain an efficacy \((E)\) and to display it next to forcing estimates. \(E\) is defined as the ratio of the climate sensitivity parameter for a given forcing agent to the climate sensitivity parameter for \(CO_2\). A comparison of \(E\) for different forcing agents from different models is given in Forster et al. (2007). Instead of introducing \(E\) in addition to forcing estimates, we suggest to replace the total anthropogenic aerosol forcing by its \(RFP\) as detailed below.

2 Radiative forcing versus radiative flux perturbation

In this paper we compare the forcings due to two well-mixed greenhouse gases, the direct aerosol forcing and the cloud albedo effect as described in Table 1 from five atmospheric GCMs with the...

\(^{1}\text{likely refers to a } > 66\% \text{ probability of occurrence}\)
respective $RFP$ that take fast feedbacks and interactions into account. Indirect aerosol effects beyond the cloud albedo effect cannot be compared this way because they comprise fast feedbacks and interactions and thus no forcing calculation can be done for them. The versions of the participating GCMs are: CSIRO in low resolution (Rotstayn et al., 2007; Rotstayn and Liu, 2009), EC-Earth (Storelvmo et al., 2009), ECHAM5 (Lohmann et al., 2008), GISS (Menon et al., 2008), and HadGEM2 (Collins et al., 2008). These models vary in the complexity with which they describe aerosol-cloud interactions and thus provide a reasonable spread in radiative forcing and radiative flux perturbation estimates. All models include anthropogenic emissions of sulfate precursors, organic and black carbon. Therefore the direct aerosol effect accounts for black carbon in all models and the semi-direct effect of black carbon is accounted for in the $RFP$ calculations. However, only in the CSIRO and ECHAM5 GCMs does hydrophilic black carbon also contribute to the number of cloud droplets and thus to the cloud albedo effect. The radiative forcing and $RFP$ calculations are conducted by using prescribed sea-surface temperature and sea ice extent, which is also referred to as the Hansen-style method to estimate forcing (Hansen et al., 2002).

For the forcing calculations using the traditional forcing definition, denoted $F$, the radiation code of the models was called twice keeping the meteorology fixed. The differences between two radiative transfer calculations due to pre-industrial GHG or aerosol concentrations versus their present-day values were extracted at the top-of-the-atmosphere and at the tropopause (or at 100 hPa which some GCMs took as a surrogate for the tropopause). The forcing calculation at the tropopause is necessary to account for the fast stratospheric temperature adjustment as a response to the warming due to molecular absorption by greenhouse gases (Hansen et al., 1997). In the second set of experiments, the simulations were run for 5-10 years each after a spin-up period of several months under conditions appropriate for the present-day climate. As the meteorology is different when varying greenhouse concentrations or aerosols, here the radiative effects of the forcing agents will be evaluated as $RFP$, defined as the difference in the net TOA radiation balance between the pre-industrial and present-day simulations.

In cases where GCMs have aerosols that interact with cloud microphysics and where the aerosols are radiatively active at the same time, $RFP$ calculations for individual aerosol effects are more complicated. Here the interaction between aerosols and cloud droplets is artificially deactivated by prescribing a cloud droplet number concentration $N_c$ for the calculation of precipitation formation. Moreover, aerosol concentrations were put to zero for the time evolution of the model. Then the forcings due to the direct aerosol effect and the cloud albedo effect are obtained from the difference of the forcing calculations in a simulation with present-day and one with pre-industrial emissions. Taking the difference between present-day and pre-industrial forcing is necessary as in each simulation the total forcing (present-day minus zero aerosols and pre-industrial minus zero aerosols) is calculated. $RFP$ calculations are performed as for GHGs. For all radiative flux perturbations, the interannual standard deviation was calculated (Snedecor and Cochran, 1989).
The estimates of $RFP$ vs. $F$ at TOA and at the tropopause for the different forcing agents from the five GCMs are shown in Figure 2. The difference between tropopause and TOA forcing is only important for CO$_2$ as an increase in CO$_2$ warms the troposphere but cools the stratosphere. If a stratospheric temperature adjustment would have been allowed in these simulations, then $F$ at TOA would equal $F$ at the tropopause. Therefore for CO$_2$ $RFP$ at TOA needs to be compared to $F$ at the tropopause as shown in the right panel.

For the majority of these different estimates, the $F$ values for the net radiation at the tropopause fall within the $RFP \pm$ their interannual standard deviation. Deviations occur mainly for the larger forcings (carbon dioxide and the first indirect effect) especially for those models with larger forcings for a given species. For individual models explanations can be found that relate to the way the cloud feedback differs in these simulations. The negative $F$ and $RFP$ values for the aerosol effects and their deviations from the one-to-one line are reflected in the shortwave $F$ and $RFP$ values. The positive $F$ and $RFP$ values for the greenhouse gases and their deviations from the one-to-one line are dominated by their longwave signals (Figure 2). The scatter plot of the net radiation tropopause forcing versus $RFP$ also includes a literature estimate of the direct aerosol effect by Hansen et al. (2005).

Deviations between the forcing and $RFP$ estimates are smaller in the clear-sky case where the influence of cloud feedbacks is much smaller (Figure 3). Unfortunately the clear-sky results are only available for the TOA forcing but not for the tropopause forcing. Changes in total cloud cover, liquid and ice water path remain below 1% of their present-day values in all $RFP$ simulations and models (not shown). Thus, the zonal and annual mean pattern of the $RFP$ estimates are a noisy version of the forcing distributions because of the inclusion of fast interactions and feedbacks in the latter but are not fundamentally different (Figures 4, 5, 6).

This is a very powerful result as it shows that $RFP$ estimates are consistent with forcing calculations using the traditional approach for all the species/effects considered here. This implies that in the global mean fast interactions due to aerosol-cloud interactions but also the water vapor, lapse rate and land surface temperature feedbacks are not that important for the investigated species/effects.

3 Conclusions

In this paper we argued that feedbacks and interactions that are fast as compared to the time scale of global warming should be included when estimating the total anthropogenic aerosol effect. Doing so allows the total anthropogenic aerosol effect, which we cannot evaluate as a forcing precisely because it includes fast feedbacks and interactions and needs to be obtained from the $RFP$ method, to be compared to the forcings due to well-mixed greenhouse gases. Thus, it can be included in future IPCC bar charts that compare the different radiative forcing agents. Moreover, replacing the global-mean aerosol forcing by its $RFP$ is warranted because it is the overall aerosol flux perturbation that
is needed for the global energy balance (Murphy et al., 2009).

4 Appendix: References for Figure 1

4.1 Cloud albedo effect:

Kaufman and Chou (1993), Jones et al. (1994), Boucher and Lohmann (1995), Chuang et al. (1997), Feichter et al. (1997), Lohmann and Feichter (1997), Rotstayn (1999), Lohmann et al. (2000), Kiehl et al. (2000), Jones et al. (2001), Williams et al. (2001), Ghan et al. (2001), Rotstayn and Penner (2001), Chuang et al. (2002), Kristjánsson (2002), Rotstayn and Liu (2003), Suzuki et al. (2004), Quaas et al. (2004), Dufresne et al. (2005), Ming et al. (2005), Chen and Penner (2005), Takemura et al. (2005), Quaas and Boucher (2005), Penner et al. (2006), Kvalevag and Myhre (2007), Quaas et al. (2008), Lebsock et al. (2008), Wang and Penner (2009), Storelvmo et al. (2009), Rotstayn and Liu (2009), Haerter et al. (2009)

4.2 Total aerosol indirect effect:

4.2.1 Cloud albedo and cloud lifetime effect:

Lohmann and Feichter (1997), Rotstayn (1999), Lohmann et al. (2000), Jones et al. (2001), Williams et al. (2001), Ghan et al. (2001), Lohmann and Lesins (2002), Menon et al. (2002), Kristjánsson (2002), Peng and Lohmann (2003), Kristjánsson et al. (2005), Ming et al. (2005), Rotstayn and Liu (2005), Takemura et al. (2005), Quaas et al. (2006), Storelvmo et al. (2006), Storelvmo et al. (2008a), Rotstayn and Liu (2009), Hoose et al. (2009)

4.2.2 Cloud albedo, cloud lifetime, direct and semi-direct effect:

Lohmann and Feichter (2001), Penner et al. (2003), Penner et al. (2006), Lohmann et al. (2007), Rotstayn et al. (2007), Posselt and Lohmann (2008), Posselt and Lohmann (2009), Quaas et al. (2009b)

4.2.3 Cloud albedo, cloud lifetime, direct effect and aerosol effects on mixed-phase clouds:

Lohmann and Diehl (2006), Jacobson (2006), Storelvmo et al. (2008a), Hoose et al. (2008b), Storelvmo et al. (2008b), Koch et al. (2009), Lohmann and Hoose (2009)

4.2.4 Cloud albedo, cloud lifetime, direct effect and aerosol effects on convective clouds:

Menon and Rotstayn (2006), Lohmann (2008), Unger et al. (2009)
4.2.5 Inverse estimates of the direct and indirect aerosol effects:

Andronova and Schlesinger (2001), Knutti et al. (2002), Gregory et al. (2002), Forest et al. (2002), Knutti et al. (2003), Forest et al. (2006), Stott et al. (2006), Shindell and Faluvegi (2009)

Acknowledgements. We like to thank Yi Ming, Steven Schwartz, William Collins, Sandrine Bony, Leo Donner and the other participants of the FIAS workshop on “Clouds in the perturbed climate system” in March 2008 for useful discussions and Sylvaine Ferrachat for technical help. UL was supported by NCCR Climate and by CSCS. SM was supported by the Office of Science at the U.S. DOE under Contract No. DE- AC02-05CH11231. SM and DK acknowledge support from the NASA MAP program. AJ was supported by the Joint DECC, Defra and MoD Integrated Climate Programme - DECC/Defra (GA01101), MoD (CBC/2B/0417,Annex C5). LR was supported in part by ACCSP.
References

Anderson, T. L., Charlson, R. J., Schwartz, S. E., Knutti, R., Boucher, O., Rodhe, H., and Heintzenberg, J.: Climate forcing by Aerosols - a hazy picture, Science, 300, 1103–1104, 2003.

Andronova, N. G. and Schlesinger, M. E.: Objective estimation of the probability density function for climate sensitivity, J. Geophys. Res., 106, 22605–22611, 2001.

Boucher, O. and Lohmann, U.: The sulfate-CCN-cloud albedo effect: A sensitivity study with two general circulation models, Tellus, Ser. B, 47, 281–300, 1995.

Chen, Y. and Penner, J. E.: Uncertainty analysis for estimates of the first indirect aerosol effect, Atmos. Chem. Phys., 5, 2935–2948, 2005.

Chuang, C. C., Penner, J. E., Taylor, K. E., Grossmann, A. S., and Walton, J. J.: An assessment of the radiative effects of anthropogenic sulfate, J. Geophys. Res., 102, 3761–3778, 1997.

Chuang, C. C., Penner, J. E., Prospero, J. M., Grant, K. E., Rau, G. H., and Kawamoto, K.: Cloud susceptibility and the first aerosol indirect forcing: Sensitivity to black carbon and aerosol concentrations, J. Geophys. Res., 107, doi: 10.1029/2000JD000215, 2002.

Collins, W. J., Bellouin, N., Douttiaux-Boucher, M., Gedney, N., Hinton, T., Jones, C. D., Liddicoat, S., Martin, G., O’Connor, F., Rae, J., Senior, C., Totterdel, I., Woodward, S., Reichler, T., J., and Halloran, P.: Evaluation of the HadGEM2 model, Tech. rep., Hadley Cent. Tech. Note 74, Met Office, Exeter, UK, 2008.

Denman, K., Boucher, G., Chidthaisong, A., Ciais, P., Cox, P., Dickinson, R., Hauglustaine, D., Heinze, C., Holland, E., Jacob, D., Lohmann, U., Ramachandran, S., Silva Dias, P., Wofsy, S., and Zhang, X.: Couplings between changes in the climate system and biogeochemistry, in Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averty, M. Tignor, and H. L. Miller, pp. 499–588, Cambridge Univ. Press, Cambridge, United Kingdom and New York, NY, USA, 2007.

Dufresne, J. L., Quaas, J., Boucher, O., Denvil, S., and Fairhead, L.: Contrasts in the effects on climate of anthropogenic sulfate aerosols between the 20th and the 21st century, Geophys. Res. Lett., 32, 2005.

Feichter, J., Lohmann, U., and Schult, I.: The atmospheric sulfur cycle and its impact on the shortwave radiation, Clim. Dyn., 13, 235–246, 1997.

Forest, C. E., Stone, P. H., Sokolev, A. P., Allen, M. R., and Webster, M. D.: Quantifying uncertainties in climate system properties with the use of recent climate observations, Science, 295, 113–117, 2002.

Forest, C. E., Stone, P. H., and Sokolov, A. P.: Estimated PDFs of climate system properties including natural and anthropogenic forcings, Geophys. Res. Lett., 33, 2006.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., and Van Dorland, R.: Radiative Forcing of Climate Change, in Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averty, M. Tignor, and H. L. Miller, pp. 129–234, Cambridge Univ. Press, Cambridge, United Kingdom and New York, NY, USA, 2007.

Forster, P. M. D. and Taylor, K. E.: Climate forcings and climate sensitivities diagnosed from coupled climate model integrations, J. Climate, 19, 6181–6194, 2006.
Ghan, S. J., Easter, R. C., Hudson, J., and Bréon, F.-M.: Evaluation of aerosol indirect radiative forcing in MIRAGE. J. Geophys. Res., 106, 5317–5334, 2001.

Gregory, J. M., Stouffer, R. J., Raper, S. C. B., Stott, P. A., and Rayner, N. A.: An observationally based estimate of the climate sensitivity. J. Climate, 15, 3117–3121, 2002.

Gregory, J. M., Ingram, W. J., Palmer, M. A., Jones, G. S., Stott, P. A., Thorpe, R. B., Lowe, J. A., Johns, T. C., and Williams, K. D.: A new method for diagnosing radiative forcing and climate sensitivity. Geophys. Res. Lett., 31, doi: 10.1029/2003GL018747, 2004.

Haerter, J. O., Roeckner, E., Tomassini, L., and von Storch, J. S.: Parametric uncertainty effects on aerosol radiative forcing. Geophys. Res. Lett., 36, doi: 10.1029/2009GL039050, 2009.

Hansen, J., Sato, M., and Ruedy, R.: Radiative forcing and climate response. J. Geophys. Res., 102, 6831–6864, 1997.

Hansen, J., Sato, M., Nazarenko, L., Ruedy, R., Lacin, A., Koch, D., Tegen, I., Hall, T., Shindell, D., Santer, B., Stone, P., Novakov, T., Thomason, L., Wang, R., Wang, Y., Jacob, D., Hollandsworth, S., Bishop, L., Logan, J., Thompson, A., Stolarski, R., Lean, J., Willson, R., Levitus, S., Antonov, J., Rayner, N., Parker, D., and Christy, J.: Climate forcings in Goddard Institute for Space Studies SI2000 simulations. J. Geophys. Res., 107, doi:10.1029/2001JD001143, 2002.

Hansen, J., Sato, M., Ruedy, R., Nazarenko, L., Lacin, A., Schmidt, G. A., Russell, G., Aleinov, I., Bauer, M., Bauer, S., Bell, N., Cairns, B., Canuto, V., Chandler, M., Cheng, Y., Del Genio, A., Faluvecchi, G., Fleming, E., Friend, A., Hall, T., Jackman, C., Kelley, M., Kiang, N., Koch, D., Lean, J., Lerner, J., Lo, K., Menon, S., Miller, R., Minnis, P., Novakov, T., Oinas, V., Perlwitz, J., Perlwitz, J., Rind, D., Romanou, A., Shindell, D., Stone, P., Sun, S., Tausnev, N., Thresher, D., Wielicki, B., Wong, T., Yao, M., and Zhang, S.: Efficacy of climate forcings. J. Geophys. Res., 110, doi:10.1029/2005JD005776, d18104, 2005.

Haywood, J. M., Donner, L. J., Jones, A., and Golaz, J.-C.: Global indirect radiative forcing caused by aerosols: IPCC (2007) and beyond, in Clouds in the Perturbed Climate System, edited by J. Heintzenberg and R. J. Charlson, pp. 451–467, MIT Press, Cambridge, 2009.

Hegerl, G. C., Zwiers, F. W., Bracancon, P., Gillett, N. P., Luo, Y., Orsini, J. A. M., Nicholls, N., Penner, J. E., and Stott, P. A.: Understanding and attributing climate change, in Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor, and H. L. Miller, pp. 663–746, Cambridge Univ. Press, Cambridge, United Kingdom and New York, NY, USA, 2007.

Hoose, C., Lohmann, U., Erdin, R., and Tegen, I.: Global influence of dust mineralogical composition on heterogeneous ice nucleation in mixed-phase clouds. Environ. Res. Lett., 3, doi:10.1088/1748–9326/3/2/025 003, 2008b.

Hoose, C., Kristjánsson, J. E., Iversen, T., Kirevag, A., Seland, Ø., and Gettelman, A.: Constraining cloud droplet number concentration in GCMs suppresses the aerosol indirect effect. Geophys. Res. Lett., 36, doi:10.1029/2009GL038568, 2009.

Jacobson, M. Z.: Effects of Externally-Through-Internally-Mixed Soot Inclusions within Clouds and Precipitation on Global Climate. J. Phys. Chem., 110, 6860–6873, 2006.

Jones, A., Roberts, D. L., and Slingo, A.: A climate model study of indirect radiative forcing by anthropogenic
sulphate aerosols, Nature, 370, 450–453, 1994.

Jones, A., Roberts, D. L., Woodage, M. J., and Johnson, C. E.: Indirect sulphate aerosol forcing in a climate model with an interactive sulphur cycle, J. Geophys. Res., 106, 20,293–30,310, 2001.

Joshi, M., Shine, K., Ponater, M., Stubler, N., Sausen, R., and Li, L.: A comparison of climate response to different radiative forcings in three general circulation models: towards an improved metric of climate change, Clim. Dyn., 20, 843–854, 2003.

Kaufman, Y. J. and Chou, M. D.: Model Simulations of the Competing Climatic Effects of So2 and Co2, J. Climate, 6, 1241–1252, 1993.

Kiehl, J. T.: Twentieth century climate model response and climate sensitivity, Geophys. Res. Lett., 34, 2007.

Kiehl, J. T., Schneider, T. L., Rasch, P. J., Barth, M. C., and Wong, J.: Radiative forcing due to sulfate aerosols from simulations with the National Center for Atmospheric Research Community Climate Model, Version 3, J. Geophys. Res., 105, 2000.

Knutti, R. and Hegerl, G. C.: The equilibrium sensitivity of the Earth’s temperature to radiation changes, Nature Geosci., 1, 735–743, 2008.

Knutti, R., Stocker, T. F., Joos, F., and Plattner, G.-K.: Constraints on radiative forcing and future climate change from observations and climate model ensembles, Nature, 416, 719–723, 2002.

Knutti, R., Stocker, T. F., Joos, F., and Plattner, G. K.: Probabilistic climate change projections using neural networks, Clim. Dyn., 21, 257–272, 2003.

Koch, D., Menon, S., Del Genio, A., Ruedy, R., Aleinov, I., and Schmidt, G. A.: Distinguishing aerosol impacts on climate over the past century, J. Climate, 22, 2659–2677, 2009.

Kristjánsson, J. E.: Studies of the aerosol indirect effect from sulfate and black carbon aerosols, J. Geophys. Res., 107, doi: 10.1029/2001JD000887, 2002.

Kristjánsson, J. E., Iversen, T., Kirkevåg, A., Seland, Ø., and Debernard, J.: Response of the climate system to aerosol direct and indirect forcing: Role of cloud feedbacks, J. Geophys. Res., 110, 2005.

Kvalevag, M. M. and Myhre, G.: Human impact on direct and diffuse solar radiation during the industrial era, J. Climate, 20, 4874–4883, 2007.

Lebsock, M. D., Stephens, G. L., and Kummerow, C.: Multisensor satellite observations of aerosol effects on warm clouds, J. Geophys. Res., 113, 2008.

Lohmann, U.: Global anthropogenic aerosol effects on convective clouds in ECHAM5-HAM, Atmos. Chem. Phys., 8, 2115–2131, 2008.

Lohmann, U. and Diehl, K.: Sensitivity studies of the importance of dust ice nuclei for the indirect aerosol effect on stratiform mixed-phase clouds, J. Atmos. Sci, 63, 968–982, 2006.

Lohmann, U. and Feichter, J.: Impact of sulfate aerosols on albedo and lifetime of clouds: A sensitivity study with the ECHAM GCM, J. Geophys. Res., 102, 13,685–13,700, 1997.

Lohmann, U. and Feichter, J.: Can the direct and semi-direct aerosol effect compete with the indirect effect on a global scale?, Geophys. Res. Lett., 28, 159–161, 2001.

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: A review, Atmos. Chem. Phys., 5, 715–737, 2005.

Lohmann, U. and Hoos, C.: Sensitivity studies of different aerosol indirect effects in mixed-phase clouds, Atmos. Chem. Phys. Discuss., 9, 15045–15081, 2009.
Lohmann, U. and Lesins, G.: Stronger constraints on the anthropogenic indirect aerosol effect, Science, 298, 1012–1016, 2002.
Lohmann, U., Feichter, J., Penner, J. E., and Leaitch, W. R.: Indirect effect of sulfate and carbonaceous aerosols: A mechanistic treatment, J. Geophys. Res., 105, 12,193–12,206, 2000.
Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, Atmos. Chem. Phys., 7, 3425–3446, 2007.
Lohmann, U., Spichtinger, P., Jess, S., Peter, T., and Smit, H.: Cirrus cloud formation and ice supersaturated regions in a global climate model, Env. Res. Lett., 3, doi:10.1088/1748–9326/3/4/045 022, 2008.
Menon, S. and Rotstayn, L.: The radiative influence of aerosol effects on liquid-phase cumulus and stratiform clouds based on sensitivity studies with two climate models, Climate Dyn., 27, 345–356, 2006.
Menon, S., DelGenio, A. D., Koch, D., and Tselioudis, G.: GCM Simulations of the Aerosol Indirect Effect: Sensitivity to Cloud Parameterization and Aerosol Burden, J. Atmos. Sci., 59, 692–713, 2002.
Menon, S., Del Genio, A. D., Kaufman, Y., Bennartz, R., Koch, D., Loeb, N., and Orlikowski, D.: Analyzing signatures of aerosol-cloud interactions from satellite retrievals and the GISS GCM to constrain the aerosol indirect effect, J. Geophys. Res., 113, 2008.
Ming, Y., Ramaswamy, V., Ginoux, P. A., Horowitz, L. W., and Russell, L. M.: Geophysical Fluid Dynamics Laboratory general circulation model investigation of the indirect radiative effects of anthropogenic sulfate aerosol, J. Geophys. Res., 110, 2005.
Murphy, D. M., Solomon, S., Portmann, R. W., Rosenlof, K. H., Forster, P. M. d. F., and Wong, T.: An observationally based energy balance for the Earth since 1950, J. Geophys. Res., 114, doi:10.1029/2009JD012 105, 2009.
Peng, Y. and Lohmann, U.: Sensitivity study of the spectral dispersion of the cloud droplet size distribution on the indirect aerosol effect, Geophys. Res. Lett., 30, doi: 10.1029/2003GL017 192, 2003.
Penner, J. E., Zhang, S. Y., and Chuang, C. C.: Soot and smoke aerosol may not warm climate, J. Geophys. Res., 108, doi: 10.1029/2003JD003 409, 2003.
Penner, J. E., Quaas, J., Storelvmo, T., Takemura, T., Boucher, O., Guo, H., Kirkevág, A., Kristjánsson, J. E., and Seland, Ø.: Model intercomparison of indirect aerosol effects, Atmos. Chem. Phys., 6, 3391–3405, 2006.
Posselt, R. and Lohmann, U.: Influence of giant CCN on warm rain processes in the ECHAM5 GCM, Atmos. Chem. Phys., 8, 3769–3788, 2008.
Posselt, R. and Lohmann, U.: Sensitivity of the total anthropogenic aerosol effect to the treatment of rain in a global climate model, Geophys. Res. Lett., 36, 2009.
Quaas, J. and Boucher, O.: Constraining the first aerosol indirect radiative forcing in the LMDZ GCM using POLDER and MODIS satellite data, Geophys. Res. Lett., 32, doi: 10.1029/2005GL023 850, 2005.
Quaas, J., Boucher, O., and Bréon, F.-M.: Aerosol indirect effects in POLDER satellite data and the Laboratoire de Météorologie Dynamique-Zoom (LMDZ) general circulation model, J. Geophys. Res., 109, doi: 10.1029/2004JD004 317, 2004.
Quaas, J., Boucher, O., and Lohmann, U.: A new estimate of the aerosol indirect radiative forcing by constraints of global climate models using satellite datasets, Atmos. Chem. Phys., 6, 947–955, 2006.
Quaas, J., Boucher, O., Bellouin, N., and Kinne, S.: Satellite-based estimate of the direct and indirect aerosol
climate forcing, J. Geophys. Res., 113, 2008.

Quaas, J., Bony, S., Collins, W. D., Donner, L., Illingworth, A., Jones, A., Lohmann, U., Satoh, M., Schwartz, S. E., Tao, W.-K., and Wood, R.: Quantification of Clouds in the Changing Climate System and Strategies for Reducing Critical Uncertainties, in Clouds in the perturbed climate system, edited by J. Heintzenberg and R. J. Charlson, pp. 557–573, MIT press, 2009a.

Quaas, J., Ming, Y., Menon, S., Takemura, T., Wang, M., Penner, J. E., Gettelman, A., Lohmann, U., Bellouin, N., Boucher, O., Sayer, A. M., Thomas, G. E., McComiskey, A., Feingold, G., Hooge, C., Kristjansson, J. E., Liu, Z., Balkanski, Y., Donner, L. J., Ginoux, P. A., Stier, P., Feichter, J., Sednev, I., Bauer, S. E., Koch, D., Grainger, R. G., Kirkevag, A., Iversen, T., Seland, Ø., Easter, R., Ghan, S. J., Rasch, P. J., Morrison, H., Lamarque, J.-F., Iacono, M. J., Kinne, S., and Schulz, M.: Aerosol indirect effects general circulation model intercomparison and evaluation with satellite data, Atmos. Chem. Phys. Discuss., 9, 12731–12779, 2009b.

Rotstayn, L. D.: Indirect forcing by anthropogenic aerosols: A global climate model calculation of the effective radius and cloud lifetime effects, J. Geophys. Res., 104, 9369–9380, 1999.

Rotstayn, L. D. and Liu, Y.: Sensitivity of the first indirect aerosol effect to an increase of cloud droplet spectral dispersion with droplet number concentration, J. Climate, 16, 3476–3481, 2003.

Rotstayn, L. D. and Liu, Y.: A smaller global estimate of the second indirect aerosol effect, Geophys. Res. Lett., 32, doi:10.1029/2004GL021922, 2005.

Rotstayn, L. D. and Liu, Y.: Cloud droplet spectral dispersion and the indirect aerosol effect: Comparison of two treatments in a GCM, Geophys. Res. Lett., 36, doi:10.1029/2009GL038216, 2009.

Rotstayn, L. D. and Penner, J. E.: Indirect aerosol forcing, quasi-forcing, and climate response, J. Climate, 14, 2960–2975, 2001.

Rotstayn, L. D., Cai, W. J., Dix, M. R., Farquhar, G. D., Feng, Y., Ginoux, P., Herzog, M., Ito, A., Penner, J. E., Roderick, M. L., and Wang, M. H.: Have Australian rainfall and cloudiness increased due to the remote effects of Asian anthropogenic aerosols?, J. Geophys. Res., 112, 2007.

Shindell, D. and Faluvegi, G.: Climate response to regional radiative forcing during the twentieth century, Nature Geosci., 2, 294–300, 2009.

Snedecor, G. W. and Cochran, W. G.: Statistical methods, Blackwell Publishing, Iowa, USA, 8th edn., 1989.

Storelvmo, T., Kristjánsson, J. E., Ghan, S. J., Kirkevåg, A., Seland, Ø., and Iversen, T.: Predicting cloud droplet number concentration in Community Atmosphere Model (CAM)-Oslo, J. Geophys. Res., 111, doi:10.1029/2005JD006300, 2006.

Storelvmo, T., Kristjánsson, J.-E., and Lohmann, U.: Aerosol influence on mixed-phase clouds in CAM-Oslo, J. Atmos. Sci., 65, 3214–3230, 2008a.

Storelvmo, T., Kristjánsson, J. E., Lohmann, U., Iversen, T., Kirkevåg, A., and Seland, Ø.: Modeling of the Wegener-Bergeron-Findeisen process - implications for aerosol indirect effects, Env. Res. Lett., 3, doi:10.1088/1748-9326/3/4/045001, 2008b.

Storelvmo, T., Lohmann, U., and Bennartz, R.: What governs the spread in shortwave forcings in the transient IPCC AR4 models?, Geophys. Res. Lett., 36, 2009.

Stott, P. A., Mitchell, J. F. B., Allen, M. R., Delworth, T. L., Gregory, J. M., Meehl, G. A., and Santer, B. D.: Observational constraints on past attributable warming and predictions of future global warming, J. Climate, 19, 3055–3069, 2006.
Suzuki, K., Nakajima, T., Numaguti, A., Takemura, T., Kawamoto, K., and Higurashi, A.: A study of the aerosol effect on a cloud field with simultaneous use of GCM modeling and satellite observations, J. Atmos. Sci., 61, 179–194, 2004.

Takemura, T., Nozawa, T., Emori, S., Nakajima, T. Y., and Nakajima, T.: Simulation of climate response to aerosol direct and indirect effects with aerosol transport-radiation model, J. Geophys. Res., 110, doi:10.1029/2004JD00502, 2005.

Twomey, S. A.: The influence of pollution on the shortwave albedo of clouds, J. Atmos. Sci., 34, 1149–1152, 1977.

Unger, N., Menon, S., Shindell, D. T., and Koch, D. M.: Impacts of aerosol indirect effect on past and future changes in tropospheric composition, Atmos. Chem. Phys. Discuss., 9, 4691–4725, 2009.

Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, Atmos. Chem. Phys., 9, 239–260, 2009.

Williams, K. D., Jones, A., Roberts, D. L., Senior, C. A., and Woodage, M. J.: The response of the climate system to the indirect effects of anthropogenic sulfate aerosols, Clim. Dyn., 17, 845–856, 2001.
### Tables

**Table 1.** Experimental set-up

| Forcing agent | pre-industrial concentration | present-day concentration |
|---------------|-----------------------------|---------------------------|
| CO₂           | 280 ppm                     | 379 ppm                   |
| CH₄           | 0.715 ppm                   | 1.774 ppm                 |
| direct aerosol effect | pre-industrial emissions (1750 or 1860) | present-day (year 2000) emissions |
| cloud albedo effect | pre-industrial emissions (1750 or 1860) | present-day (year 2000) emissions |
Figure Captions

Fig. 1. Model, satellite and inverse estimates of the aerosol indirect effects over the last two decades. Per method or effects considered, each symbol represents one published estimate (one paper one vote). Blue represents estimates of the cloud albedo effect from GCMs (circles), GCMs combined with satellite measurements (squares) and satellite only (triangles). Red represents estimates of both the cloud albedo and cloud lifetime effect from GCMs (circles) and GCMs combined with satellite estimates (squares). The yellow circle represents an estimate of the cloud albedo, lifetime, direct and semi-direct effects. Black circles represent the aerosol effects on stratiform and convective clouds and green circles represent estimates of aerosol effects on liquid and mixed-phase clouds. The black stippled area refers to inverse estimates. In case of multiple estimates per paper, the vertical bars denote the standard deviation. See supplement for the individual papers, from which the estimates are obtained.

Fig. 2. Net, shortwave and longwave radiative flux perturbation versus TOA and tropopause forcing, respectively, from five GCMs. Vertical bars denote the interannual standard deviation in the radiative flux perturbation calculations.

Fig. 3. As figure 2, but for the clear-sky net, shortwave and longwave radiative flux perturbation versus TOA forcing from four GCMs.

Fig. 4. Annual zonal means of $F$ vs. $RFP$ [W m$^{-2}$] for the different forcing agents from the HadGEM2 and ECHAM5 GCMs

Fig. 5. Annual zonal means of $F$ vs. $RFP$ [W m$^{-2}$] for the different forcing agents from the EC-Earth and GISS GCMs

Fig. 6. Annual zonal means of $F$ vs. $RFP$ [W m$^{-2}$] for the different forcing agents from the CSIRO GCM
Published estimates of the aerosol indirect effect

Anthropogenic changes in net radiation at the TOA

Fig. 1. Model, satellite and inverse estimates of the aerosol indirect effects over the last two decades. Per method or effects considered, each symbol represents one published estimate (one paper one vote). Blue represents estimates of the cloud albedo effect from GCMs (circles), GCMs combined with satellite measurements (squares) and satellite only (triangles). Red represents estimates of both the cloud albedo and cloud lifetime effect from GCMs (circles) and GCMs combined with satellite estimates (squares). The yellow circle represents an estimate of the cloud albedo, lifetime, direct and semi-direct effects. Black circles represent the aerosol effects on stratiform and convective clouds and green circles represent estimates of aerosol effects on liquid and mixed-phase clouds. The black stippled area refers to inverse estimates. In case of multiple estimates per paper, the vertical bars denote the standard deviation. See supplement for the individual papers, from which the estimates are obtained.
Fig. 2. Net, shortwave and longwave radiative flux perturbation versus TOA and tropopause forcing, respectively, from five GCMs. Vertical bars denote the interannual standard deviation in the radiative flux perturbation calculations.
Fig. 3. As figure 2, but for the clear-sky net, shortwave and longwave radiative flux perturbation versus TOA forcing from four GCMs.
Fig. 4. Annual zonal means of $F$ vs. $RFP$ [W m$^{-2}$] for the different forcing agents from the HadGEM2 and ECHAM5 GCMs
Fig. 5. Annual zonal means of $F$ vs. $RFP$ [W m$^{-2}$] for the different forcing agents from the EC-Earth and GISS GCMs.
Fig. 6. Annual zonal means of $F$ vs. $RFP$ [W m$^{-2}$] for the different forcing agents from the CSIRO GCM.