We thank the referee for the insightful comments and suggestions.

Major issues:

1. The title of the paper suggests, probably too boldly, that constraints are being placed on the magnitude of the first aerosol indirect effect. By mixing satellite observations of cloud properties with model aerosol properties in order to determine the slope parameter (dlog(reff)/dlog(SO4)) it is not possible to constrain the effect in a consistent way. I realize that determine the 'true' value of this slope and its error bounds are not objectives of the paper but the title suggests otherwise. I would replace the work 'constraints' by 'studies' or some other term.

We agree that the title may be misleading. We therefore changed the title to 'Constraints on interactions between aerosols and clouds on a global scale from a combination of MODIS-CERES satellite data and climate simulations' to emphasize that the paper very broadly addresses relationships between aerosols and clouds on a global scale without any attempt to attribute these relationships to any specific physical process or situation.

2. Their results (e.g. Figure 3) suggest that the model clouds are less susceptible to aerosols than satellite-determined cloud properties. This is contrary to previous studies that show that GCMs are likely over-estimating the magnitude of the first aerosol indirect effect. What is the explanation for this different result?

The cloud droplet number in the model is empirically related to the sulphate concentration (Eqn. 1) according to the parameterization by Dufresne et al. (2005). As indicated in the text, the parameters in this parameterization were tuned to match a satellite-based relationship between cloud droplet effective radius and aerosol index from POLDER. This leads to a weaker indirect effect for this approach compared to parameterizations in some GCMs which are based on *in situ* observations.

3. The study focuses on clouds with tops below 700 hPa which encompasses many cloud situation. Although the criterion will reduce the likelihood of mixed phase clouds it does not eliminate them. What fraction of the cases do they estimate consist of clouds with some ice present? Also their study includes low cloud with very different dynamical regimes (stratus to stratocumulus to cumulus) and for the entire globe except polar regions. Geographical and dynamical influences in their cloud susceptibilities could yield important insights. Is it possible for the authors to include such an analysis?

We looked at the fraction of liquid water in low clouds relative to liquid + ice in the same clouds, using the ISCCP-like CERES satellite product (http://eosweb.larc.nasa.gov/PRODOCS/ceres/level3_isccp-d2like_table.html). Results shown in Figure below indicate that the fraction is very close to 1 for most regions, especially in the Northern Hemisphere. At high latitudes in the Southern Hemisphere, the fraction is on the order of 0.8 - 0.9.

We also note that the MODIS Science Team effective radius results are just for liquid clouds.

This seems to confirm that the assumption of liquid clouds is rather reasonable in the study.

We did not attempt to investigate geographical and dynamical influences on the relationships between aerosols and clouds. We agree that it would in principle be possible to extend the current study in that regard. This would likely lead to further constraints for models. In any event, an investigation of aerosol/cloud relationships on global scales seems to be a necessary first step in this direction.

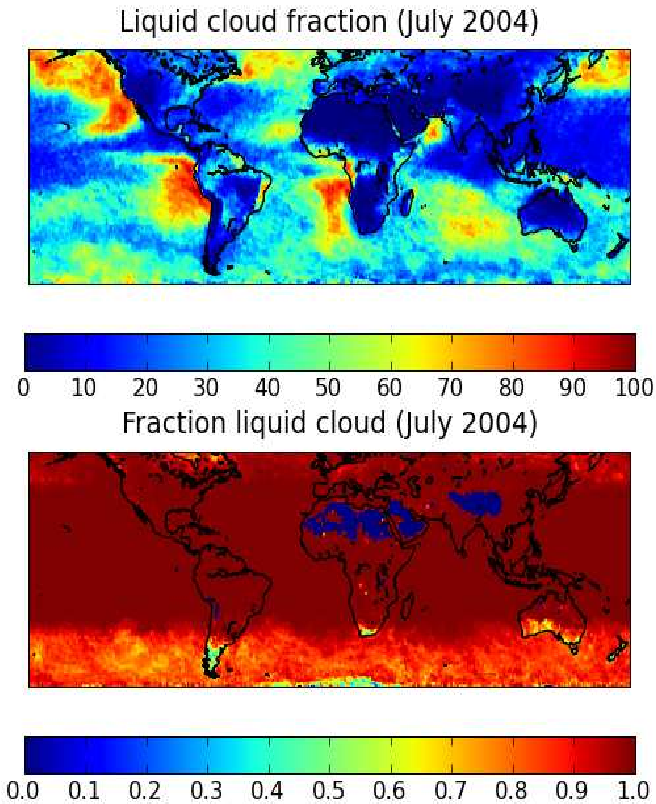

Figure. The monthly mean low cloud fraction (top) and monthly mean fraction of liquid water in low clouds relative to liquid + ice in the same clouds. The data is for July in 2004 and from CERES ISCCP-like cloud product. The dark blue over Africa and Asia indicate there is no data available in these regions.

4. In the middle of Section 3 (top of page 13950) it is argued that long time averaging is desirable because it reduces the weather-related variations compared to climate features, and hence they use seasonal means of the MODIS retrievals of cloud properties. However it has been pointed out (e.g. Stevens and Feingold, 2009 and others) that the multitude of microphysical and dynamical processes can lead to a 'buffering' of the indirect effect and I would claim that doing long time averages will contribute to this problem. More discussion and recognition about this issue is needed.

We agree that the buffering of the indirect effect that is associated with a range of different processes is an important aspect for aerosol indirect effects in the atmosphere and we therefore included a brief discussion in the introduction and made modifications elsewhere in order to make clearer that this study is very broad in its scope. Specifically, the study does not address any specific mechanisms underlying aerosol indirect effects.

5. I am somewhat surprised that the cloud droplet activation is being parameterized simply by the old-fashioned aerosol mass concentration instead of having a prognostic equation for aerosol number. Why is not practical to have a more advanced treatment of activation with a prognostic equation for aerosol number? What evidence is there that the aerosol mass concentration parameterization works sufficiently well to predict the cloud droplet number? Obviously all dynamical effects are being ignored with this parameterization. I can understand using Eqn 1 for long-term averaged quantities but it is my understanding that Eqn. 1 is being used at the time and space step of the GCM run.

The focus of this study is on the development and testing of a new diagnostic approach for aerosol/cloud interactions. The approach is sufficiently general so that it can in principle be applied to any model and parameterization. We selected the parameterization by Dufresne et al (2005) as an example for this study because it was easy to implement in our model and it can be regarded as a rather generic approach. The parameterization employs the same basic functional relationship between sulphate and cloud droplet concentrations that is used in several other global models (e.g. models that are based on the approach by Boucher and Lohmann, 1995) and observation-based studies. As has been shown in numerous studies, including ours, observed dependencies of cloud microphysics on aerosols can be broadly captured by this simple approach (i.e. Fig. 3).

The parameterization in this study has been "state of the art" in climate research for many years as is evident from the 4th climate assessment report by the IPCC and other publications. Despite its extraordinary simplicity, there seems to be no clear evidence yet that this particular type of parameterization produces biases or uncertainties in globally mean simulated aerosol indirect effects that are *per se* much worse than those from more complex modelling approaches. The more complex schemes that currently exist are relatively new. They may not always be more accurate than simple schemes given the need to constrain a greater number of processes and their interactions with other processes, which are not always well known.

A microphyics schemes for prognostic simulations of aerosols has recently been introduced in our GCM. Results from this approach will also be compared to satellite observations in the near future. However, this approach is still under development so we were not yet in a position to include any results in the paper.

We modified the text accordingly.

6. One of the main results of this paper is that organic aerosols are contributing to the indirect effect. Does the organic contribution include both the hydrophilic and hydrophobic components? if it is just the hydrophilic component then it seems to be an obvious result. Why should this be considered a new finding? The higher sensitivity to sulphate when using Eqns 2 and 3 is also obvious since the coefficient in front of the log(SO4) term is 0.50 instead of 0.20. Why didn't the authors develop their own improved parameterization for cloud droplet number with a better balance between sulphate and organic effects on clouds? Also the parameterization should probably use the sum of the sulphate and organic masses instead of a logarithmic product to avoid their problem when the organic mass goes to zero.

Organic carbon (OC) in this paper refers to the hydrophilic component (see text on p. 13950). Although this component is believed to be an important contributor to the indirect effect, effects on organic carbon are not taken into account in many models. Based on satellite data, we determined that effects of OC on cloud droplets are similar in efficiency than effects of sulphate (Figs. 4, 5, 8 and 9). Furthermore, based on the new diagnostic approach, we concluded that the coefficient in front of the log(SO4) term in Eqns. 2 and 3 is too large and that the corresponding coefficient in Eq. 1 is more reasonable (Fig. 8). We think that this is a new finding and that this example clearly demonstrates that our method can be used to put constraints on relationships between OC and cloud droplets from models. We are not aware of any similar constraints from other methods.

We did not attempt to develop our own empirical parameterization to capture relationships between OC and cloud droplets. It seems unlikely to us that a simple parameterization can be developed that would be physically meaningful and be able to capture joint effects of sulphate and OC on cloud droplets. We believe that a combination of prognostic aerosol and cloud microphysics schemes will eventually enable realistic simulations of effects of OC on clouds in GCMs.

7. In Figure 3 the aerosol dependence is given as a column burden. However the cloud droplet parameterization (Eqn.1) is based on aerosol concentration at the level where the cloud forms. This inconsistency can lead to biases in the results. How have they accounted or correlated for this problem?

We considered the dependency of the results on the depths and location of the layers and did not find any notable differences in mean dependencies when shallower layers were used for results below 700 hPa. As pointed out in the description of results in Fig. 4 in Section 5, the diagnosed mean relationships between cloud droplet radius and sulphate burden in the layer between the surface and 700 hPa are similar to the theoretically expected relationship from Eq. 1.

Minor issues:

1. The age of the cloud is likely an important factor in altering the apparent correlation between aerosols and cloud effective radius. The authors should point this out and explain why they are unable to address the issue.

We are not sure that we fully understand this comment. The satellite retrievals for cloud droplet effective radius used in the study implicitly account for cloud lifetime effects. The satellite results were not filtered to exclude any particular process.

However, the GCM does not include a parameterization for cloud lifetime effects (i.e. there is no dependency of the precipitation formation rates on aerosol concentrations in this particular version of the model). We agree that cloud droplet radii should in principle depend on cloud lifetime effects. Consequently, one would not expect the model to successfully reproduce features of the satellite retrievals that are related to cloud lifetime effects. However, it seems possible that the cloud lifetime effect has a somewhat subtle effect on global mean relationships between aerosol and cloud droplet sizes. This may not be easy to detect based on the diagnostic approach in this study, which was designed to address very broad features of aerosol/cloud interactions. It seems likely that larger discrepancies between model results and satellite retrievals may be found if relationships are analyzed at smaller spatial and temporal scales than in this study.

Generally, a realistic representation of relationships between aerosols and cloud droplet sizes appears to be a necessary criterion for an accurate representation of aerosol indirect effects in models. But it is likely that this is not a sufficient criterion.

We included a brief discussions of this aspect in various parts of the revised manuscript.

2. What years were used in compiling the MODIS observations?

From 2001 to 2005. The text was changed accordingly.

3. In the second paragraph in Section 4 (bottom of page 13950)

they state there are large cloud droplets over oceans where aerosol concentrations are low and cloud liquid water contents are high. Isn't the cloud liquid water content more a function of the average temperature (i.e. latitude) rather than land versus ocean? Also shouldn't the average cloud updraft speed, which is typically lower over the oceans, be included as a factor? Strong updrafts will activate more aerosols leading to smaller cloud droplet even for fixed total aerosol number.

Thanks for bringing this to our attention. Clouds over the ocean have larger droplets than cloud over land mainly because there are fewer aerosol particles over the ocean. We changed the text accordingly.

Regarding effects of updraft speeds: We agree. In the conclusions, we point out the lack of a dependency of model results on updraft speed as a possible explanation for model biases. Unfortunately, there is not enough information available from the satellite or GCM to look further into this.

4. Since GCM aerosols are being used why wasn't the anthropogenic component of the indirect effect computed?

The diagnostic approach in this study does not provide any information about the anthropogenic component of the indirect effect because satellite retrievals are only available for present-day conditions. Although it may be possible to filter the available satellite retrievals in order to obtain some information about anthropogenic contributions, this would be beyond the scope of this study.

In Section 3 it is stated that simply using the cloud droplet radius from the top cloud layer in the model is too simple and they

use a more sophisticated approach by Klein and Jacob (1999). It is not shown whether the more sophisticated approach is any better than the simpler approach. This needs to be discussed.

The cloud top effective radius was approximated for each subcolumn by averaging from liquid cloud top, determined by the ISCCP simulator, to an optical depth of 3, by $\overline{r_{eff}} = \sum LWP_i/(\sum LWP_i/r_{eff,i})$, where LWP_i and $r_{eff,i}$ are the model layer liquid water path and effective radius. This was done instead of using the effective radius from the uppermost cloud layer (Quaas et. al., 2004). The study of Platnick (2000) suggests that cloud effective radii retrieved at visible and near-infrared wavelengths contain information from the first few optical thickness below cloud top. In this version of CanAM4, it was found that the upper liquid cloud layer often had a cloud optical depth greater than 3 (not shown) and therefore the diagnosed cloud effective radius was often taken from the uppermost cloudy layer. However, by using the mean over a prescribed optical thickness our definition is independent of changes to model configuration, e.g., vertical resolution decreasing the layer optical thickness.

Systematic comparisons of this approach with others has not been performed.

6. Why isn't there an estimate of the radiative forcing due to the indirect effect based on their results?

The main focus of the study is on a new method for the diagnosis of relationships between cloud droplet sizes and aerosol concentrations, based on results from satellite and GCM simulations. In an attempt to demonstrate the usefulness of this method, we compared the diagnosed relationships to results from CanAM4. In principle, any other model could have been used.

There is no information available about radiative forcings from this method.

We would consider an analysis of radiative forcings in CanAM4 to be part of a larger model evaluation study, which would require the application of additional diagnostic methods. We think that this would be beyond the scope of this study.

We modified the text to better emphasize the objectives of this study.

7. In Figures 4,5,8 and 9 I recommend that the y-axis only to 30 um in order to stretch the plots vertically. They are very small and hard to see. Also please add the number of points plotted on each sub-figure (perhaps under the slope information).

The figures were changed as suggested.

8. Last paragraph in Section 2, the word 'microphysics' is misspelled twice.

The text was corrected accordingly.