Reply to Anonymous Referee #2

We would like to thank Anonymous Referee 2 for the useful and constructive comments. These offered us the opportunity to clarify some important points of the manuscript. Please find below the replies to each comment, accompanied by corresponding clarifications/modifications in the text (marked italic in this document).

The authors present a study of the impact of cloud fields and the vertical stratification of aerosols on aerosol direct radiative forcing. Their method is to segment the native aerosol fields in the LMDz model into parts above, in and below clouds and compare these, and to read in external cloud and aerosol fields from the SPRINTARS model. In general the paper is well written and documented, the methods sound and the figures clear and relevant. I recommend that the paper be published in ACP. However, I do have a set of questions for the authors that relate to how the analysis was done and how it is presented. I expect that they can be readily answered by a set of minor revisions to the text and figures.

Main comments:

- In Methods, the authors describe how forcing is calculated in their experiments. However, I miss a thorough discussion of the concept of "RF CL", i.e. cloudy sky radiative forcing. It is not immediately clear how one estimates this in a GCM, and often it is rather calculated by using eq. (3) in this paper with RF CS and RF AS as input. See e.g. the discussion on cloudy sky forcing in Schulz et al 2006 (AeroCom phase 1).

We agree that the concept of RF_{CL} should be explained better.

First, there is terminology ambiguity on all-sky (including clear and cloudy sky calculations) and cloudy-sky forcings as we understand it (forcing just in the sub-grid fraction covered by clouds). We have checked the text and put all-sky forcing where we meant all-sky forcing indeed.

Second, we have used a similar equation as in Schulz et al. (2006) to compute the "cloudy-sky" forcing for the cloudy fraction of the grid cell from all-sky and clear-sky flux perturbations. BUT - in contrast to Schulz et al. (2006), we have computed it at every time step within the model run and outputted daily averages from this instantaneous calculation. This is more accurate than the one in Schulz et al. (2006) which is based on annual values of all-sky, clear-sky forcing and cloud fraction. Note also, we did not weigh for cloud fraction to get to a daily average. In this way, cloudy and clear sky forcings are similar in concept. However, they can not easily be recombined to the actual all-sky forcing.

For clarity we changed the first paragraph in Sect. 3.2 to:

The shortwave radiative fluxes in the model LMDz are computed with the scheme developed in (Fouquart et al., 1980), separately for clear- and all- sky. The reflectivity and transmissivity of a grid cell column - being partly cloudy thus reflecting "all-sky" conditions - are computed using the random cloud overlap assumption (Morcrette et al., 1986). All- and clear-sky fluxes are computed by separate calls to the radiation routine, including or not clouds radiative effects in the computations. This allows computing separately clear- and all-sky aerosol forcings. The shortwave spectrum is divided into two spectral intervals: 0.25-0.68µm and

0.68-4.00µm. The radiation scheme is the one introduced by J.J. Morcrette in the model of the European Centre for Medium-Range Weather Forecasts (ECMWF); for more details we refer to the on-line documentation of the ECMWF research department, available at: http://www.ecmwf.int/research/ifsdocs/pdf_files/Physics.pdf.

And we explain further in Sect. 3.3 now the reverted Eq. 3 to show the computation of RF_{CL}:

Cloudy-sky radiative forcing, RF_{CL}, is calculated at each time step, and for each model cell, as:

$RF_{CL} = RF_{AS} - (1 - CLT)\, RF_{CS}$.   (3)

where CLT is the cloudy fraction in each grid cell. This is similar to the cloudy-sky forcing definition as used by Schulz et al. (2006), but differs in that it is computed at every time step and, when averaged, it is not weighted by the cloud fraction. It is thus similar in concept to the clear sky forcing definition. By using calculations for all time steps we think we obtain a more reproducible forcing number. In Schulz et al. (2006), cloudy-sky forcing was computed only from annual fields of forcings and cloud fraction.

- I also miss some further discussion on the cloud fields and how the aerosol fields look in the abv, in and blw configurations. Global, annual means are given in figure 6, but I think this should be expanded on. This is especially important to be able to compare with other work, e.g. Zarzycki and Bond which performs some of the same analysis that the authors present here for a column model.

One may "look" at the BC abv/in/blw fields in figure 3 and 4 and the cloud fraction in figure 2. We considerably expanded the discussion in Sect. 5.1 of the results presented in Sect. 4.2 (please see text). In particular, we added several elements of comparison with the work of Zarzycki et al. (2010). However, there are important differences between our study and the one of Zarzycki et al. (2010): we split the BC vertical profile in three portions instead of "moving" the same aerosol burden from below to above clouds, so the "BC below/above clouds" of the two studies has not exactly the same meaning. Also, we don't separate forcing computations on the basis of cloud type, as is done in Zarzycki et al. (2010). In spite of these differences, the enhancement of cloudy sky forcing efficiency when passing from below to above clouds is comparable in the two studies: 7.5 in our case against a range between 5 and 13 for low, medium and high clouds in Zarzycki et al. (2010).

- Finally, and maybe most crucially, I find the Discussion section underdeveloped. The authors place much emphasis on the core results of their experiments and the resulting fields, but both the "BC vs clouds" and stratification analyses seem rushed. They are both interesting, and I feel that the authors can heighten the impact of their paper by putting greater emphasis here. E.g. in sec. 5.1 a new analysis method is introduced through an equation (which btw is missing an equation number), which may be interesting for others to use for similar analyses. However the results are not really discussed. Some findings are mentioned, but the question in the section title (on the role of clouds and aerosol vertical positions for black carbon forcing) isn't really addressed thoroughly. The same applies for sec. 5.2, which is where the authors apply equations 1 and 2. The nonlinearity calculates is shown in figure 8, but what do the results imply for the analysis, for the model variability of aerosol forcing, and for total aerosol forcing uncertainty?

To answer this concern, we have now reworked Sect. 5.1 with a more detailed discussion about the meaning of the correspondence between BC altitude relative to clouds and the sign of the nonlinearity (please see the revised text). We discuss the implications of our findings in terms of relation between the contribution of BC_{abv} optical depth to the total one with its contribution to the forcing. These results are now compared to the ones from Zarzycki et al. (2010). We also discuss the implications in terms of the potential variability of aerosol forcing due to the vertical distribution (Sect. 5.1, second paragraph). Indeed, the portion of the BC total optical depth above clouds is about 30%, but it is responsible of about 55% of the cloudy-sky forcing: the contribution of the BC above clouds to the forcing is almost twice its contribution to the total optical depth, showing a strong nonlinearity between optical depth and cloudy-sky forcing. This effect is slightly weaker than the one found by Zarzycki et al. (2010) for low clouds, where BC above clouds accounts for about 20% of the global burden and for 50% of the forcing.

A discussion of the meaning and implications of the study on vertical superposition of aerosol components has been added to Sect. 5.2. In particular, we stress the fact that the sign of the nonlinearity is determined by which component (scattering or absorbing) is below and which one is above, and that the error on the forcing due to lacking vertical superposition (aerosols components juxtaposed rather than superposed) can be as high as 100%.

Minor comments:

- The abstract is quite lengthy, and at the end uses model names and technical numbers. I would propose making the abstract shorter and more pointed, to increase interest in the paper.

We shortened the abstract, replacing some cited results with others more pointed towards interpretation. The technical explanations of the method have been reduced, in order to highlight the motivation of the analysis and its key points. The names of the two models LMDz and SPRINTARS have been kept for clarity. It emphasizes the distinction between the first part of the analysis (with the quantification of forcings and “vertical distribution split” for LMDz only) and the second one (study of the impact of the vertical distribution on inter-model differences with SPRINTARS).

- (I note that reviewer 1 had questions about equations 1 and 2. I believe I follow the authors reasoning here, and agree that the calculation is relevant for the discussion section. However, see my comment above.)
- page 18816, line 5: The authors state an assumption about hygroscopic growth. As the forcing of some species is likely highly dependent on this assumption, and it therefore impacts the per-species comparisons made later in the paper, I would like to see this assumption discussed in somewhat further detail. Is the analysis sensitive to it?

We excuse for using a confusing description of the hygroscopic growth. For the optical parameter calculation we use a more simple approach than for the INCA model for computational reasons. The analysis is thus not sensitive to the assumption that the composition influences the hygroscopic growth in a direct way. For example the sulfate mass extinction coefficient in a given grid cell is the same in all experiments and model configurations (even for reading SPRINTARS).

We have changed the description of the hygroscopic growth parameterization to the following:

The model takes into account the observation that two types of particles with different HG factors appear upon hydration of dry particles of a given diameter. This is represented by the two modes: a soluble one and an insoluble one. HG changes the particle diameter, the aerosol composition and particle surface characteristics. Typical wet diameters for each mode had been precalculated in another LMDz-INCA model run and used for computation and tabulation of optical parameters as a function of mode, species and relative humidity. The optical properties of the individual components are computed with this look-up table, using the ambient relative humidity in the model grid cell. The overall optical properties of the global aerosol are then computed by summing the different contributions of the aerosol components to the extinction.

On the other hand, different values of humidity at different altitudes can affect the aerosol optical depth, thus the forcing of the individual species. That is, there is a “differential impact” of hygroscopicity on optical depth according to altitude. In particular, humidity being generally higher close to the surface, this is expected to enhance the aerosol forcings (but a priori not forcing efficiencies per unit optical depth) in the experiments “blw” with respect to “abv”. We have introduced these considerations in Sect. 4.2.

Nevertheless, when computing nonlinearity as $dRF=RF(AER)-\sum(RF_i)$, the differential effect of hygroscopicity with altitude is removed because we keep exactly the same vertical distribution of the single component and humidity when computing its individual forcing or the total forcing.

- page 18824, line 7: "NRF_CS" should be "NRF"?

It is NRF_{CS} because it’s the forcing efficiency evaluated in clear-sky conditions. We added the mention of 'clear-sky' in the sentence to take away any ambiguity.

- figures 4 and 5: Please consider adding a column with the ratio of the two first (or (A-B)/A) for clarity.

We avoided adding figures with this kind of ratios because of the very strong emphasis it pulls towards regions/points where the forcing, taken as reference, is close to zero. Computing these ratios with global averages (Table 3) avoids the inconvenient by smoothing out local differences.