Reply to Anonymous Referee #1

We would like to thank Anonymous Referee 1 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 the corresponding clarifications/modifications in the text (marked italic in this document).

Review of "A new method for evaluating the impact of vertical distribution on aerosol radiative forcing in general circulation models" by Vuolo et al., submitted to Atmos. Chem. Phys.

The manuscript studies and quantifies the coupling between the direct radiative forcing exerted by a given aerosol component and the radiative effects of other aerosols and clouds. Non-linearities in aerosol direct radiative effect and forcing caused by clouds and other aerosol species are often mentioned in the literature, but rarely quantified in a consistent framework, which this study does successfully, with a well-chosen focus on black carbon aerosols.

1 Main comments

• I fail to understand why F(B) appears in Equation 1. Shouldn't that be F(clear) in order to define both RF(A|B) and RF(A) with respect to the same reference? Equation 2 looks ok, but it simply says that the radiative forcing exerted by agents A and B is the sum of the radiative forcings exerted by A and B each taken alone, plus a coupling term dRF. This does little to clarify the dRF term. Moreover, the role of those equations in the paper is unclear, since they are never used again.

Note that we have changed the notation in Eqs. 1 and 2 in the final text (A becomes B, B becomes C) to relate better to Fig. 1, but here we use the notation as used in the discussion paper.

The equation is correct: The forcing of A, when B is present as part of the environment (*RF(A|B)*), is equal to the net TOA flux when A and B are present (*F(A+B)*), minus the net TOA flux when only B is kept in the atmosphere (*F(B))*. Both net fluxes contain also other contributions from the atmosphere. Since this part of the equation is not essential and possibly confusing, we have removed the right part of Eq. 1.

For clarification, the two equations and the text read now:

$$dRF=RF(A|B)-RF(A). \quad (1)$$

Since RF(A|B)= F(A+B)–F(B), we can rewrite Eq. (1) in the following way:

$$dRF=[F(A+B)-F(clear)]-[F(B)-F(clear)]-RF(A)=RF(A+B)-RF(A)-RF(B), \quad (2)$$

where we added and subtracted F(clear) to express the coupling term dRF in terms of the total forcing and the single-components forcings.

We have further added after Eq. 2:

This difference dRF corresponds to the nonlinearity of combining forcing (or the coupling term).It is of interest both for the "cloudy sky aerosol forcing" where the radiative interaction between clouds and

aerosols give rise to an important extra forcing term, in the form of Eq. (1), and for the superposition of different aerosol components, where the nonlinearity arises in the form of Eq. (2).

The coupling term *dRF* is important in the results and discussion sections, where it corresponds to:

RF_{CL}- RF_{CS} in Sect. 4.2, pag. 18824 (cloudy-sky minus clear-sky forcing, analogous to Eq. (1))

and to:

$RF_{CS}(AER)$-$\sum RF_{CS}(spec)$ in Sect. 5.2 (now Eq. 4), pag. 18831 (the forcing from the ensemble of the aerosols minus the sum of forcings from individual components, analogous to Eq. (2))

• Section 3.4.2: It took me a while to understand how the experiments are designed once the lowest and highest cloud levels have been identified. What I understand now is that in the abv experiment, aerosols below the highest cloud level are set to zero to keep only those aerosols that are above clouds. For the in experiment, aerosols that are below and above clouds are set to zero. For the blw experiment, experiments above the lowest cloud level are set to zero. Is that correct?

Yes this is correct. A clarifying sentence has been added early on in Sect. 3.4 ("Model simulations"):

The method requires multiple model runs where the radiative TOA forcing due to specific fractions of the aerosol burden is computed. For example aerosol optical depth above clouds are kept as in the reference run, while aerosol optical depth in and below clouds is set to zero.

In addition, the text leaves several questions open:
– What is set to zero, exactly? Mass-mixing ratios or aerosol optical depths?

We set to zero aerosol optical depths, as this parameter enters directly in flux/forcing computations in LMDz. We specify now in Sect. 3.4.2 ("The Experiments") in this and analogous form:

Experiment "abv" (above), with aerosol optical depth set to zero except above the highest cloudy model layer.

We have also added the following:

Once the cloud top and bottom are identified, aerosols optical depths are set to zero in different portions of the column for the different experiments, according to points 1) to 4). Setting aerosol optical depth to zero is equivalent to "eliminate" those aerosols, at least in what concerns flux and forcing computations. The fluxes are indeed computed in the model for each layer separately, by use of the layer aerosol transmittance and reflectance, which assume their clear-sky values (1 and 0, respectively) for aerosol optical depth set to zero.

– The method used to isolate aerosol layers with respect to clouds will affect the single-scattering albedo and asymmetry parameter of the total aerosol column, so changes in normalised radiative forcing are not only due to the vertical position of the aerosol. Correct?

Yes, in an implicit way: the aerosol composition and aerosol size of each isolated aerosol layer implies a layer specific effective single scattering albedo and asymmetry parameter, which is different than the rest of the column or other portions of the column. The normalized forcing corresponds exactly to the vertical position of the aerosol layer, but is indeed in a strict sense "not

only due to the vertical position". Isolating this effect would require experiments where the single scattering albedo and asymmetry parameter for each aerosol species is uniform everywhere. Such uniform optical aerosol characterstics are not realistic and we have chosen to characterize the "simple dependence" on vertical distribution, with implicit changes of aerosol properties. A word of caution was added to Sect. 3.4.2:

Note, that our method of eliminating the radiative effect of the aerosol in portions of the column implicitely changes also the effective single scattering albedo and asymmetry parameter of the aerosol, since composition and aerosol size differs in between vertical layers of the atmosphere. It is beyond this study to isolate the impact of vertical position on one hand and vertical variations in intensive aerosol optical properties on the other hand.

Note also: the Normalized Radiative Forcing (*NRF)* is computed as the ratio between the *RF* and the aerosol optical depth. The fluxes themselves and the *RF* are computed in the model for each layer separately, by use of the layer transmittance (*T*) and reflectance (*R*). For aerosol scattering and absorption, T_a and R_a depend indeed on aerosol optical depth (*tau*), single scattering albedo (*ssa*) and asymmetry parameter (*g*), but have the form:

$T_a=1/(1+f(tau,ssa,g))$, with *f(tau,ssa,g)* being a function that goes to zero with tau
and
$R_a= h(tau,ssa,g)$, with *h(tau,ssa,g)* being a function that goes to zero with tau.

This means that, in what concerns flux and forcing computations, the fact of setting aerosol optical depth to zero is equivalent to "eliminate" those aerosols ($T_a=1$, $R_a=0$). The detailed description of LMDz scheme for fluxes computation and their dependency on optical depth, single scattering albedo and asymmetry parameter can be found in the radiation's part of the "ECMWF research department IFS documentation - Part IV: Physical processes":
http://www.ecmwf.int/research/ifsdocs/pdf_files/Physics.pdf

– Why did the authors decide to set aerosols to zero in cloud-free columns? Leaving those untouched would have guaranteed that differences between experiments only originate from the cloudy sky, and removed the complications discussed in Section 4.2.

The choice to set aerosols optical depths to zero in completely clear columns (cc) was made to make the results more explicit in terms of the effect of aerosol-clouds superposition. We considered that the aib, abv, in and blw experiments (without aerosols in cc columns) respond better to our goal.

If we had kept the aerosols in cc columns, the aerosol optical depth of the "aib+cc" experiment wouldn't be the sum of the optical depths from abv+cc, in+cc and blw+cc experiments, because these last experiments would also contain the contribution from cc columns. We would then have:

tau(aib+cc)= tau(abv+cc) + tau(in+cc) + tau(blw+cc) – 2 tau(cc)*

and would find:

RF(aib+cc) ≠ RF (abv+cc) + RF (in+cc) + RF (blw+cc),

this difference arising both from the nonlinearity of the forcing and from the difference in optical depth (*tau(aib+cc) ≠ tau(abv+cc) + tau(in+cc) + tau(blw+cc)*)

By setting *tau(cc)=0*, we have instead:

tau(aib)= tau(abv) + tau(in) + tau(blw),

and still :

RF(aib) ≠ RF (abv) + RF (in) + RF (blw).

For this set-up the last non-equality reflects directly the nonlinearity of the forcing, which is desirable for our goal.

We have added in Sect. 3.4.2.:

For these 4 experiments the optical depth is also set to zero in completely clear columns, so that the aib result can be directly compared to the sum of the abv+in+blw experiments.

• *The authors derive the aerosol radiative forcing with respect to an atmosphere containing no aerosols. This definition is different to the IPCC definition, which takes pre-industrial aerosols as a reference (see below). That difference is not a problem in itself, but it raises an intriguing question: since pre-industrial aerosol radiative forcing will also suffer from non-linearities due to coupling between the different aerosol species and clouds, how does the pre-industrial coupling term compare to the present-day coupling term?*

Indeed, we chose to take as reference an atmosphere without aerosols (see below) to limit the number of experiments needed to interpret the differences in forcings obtained with different experiments/models and to add information of radiative effects of dust and seasalt. The question of the comparison of the pre-industrial coupling term with the present-day one would imply performing additional experiments to estimate beyond this study. We have added a sentence to the conclusions:

We have not yet characterized the nonlinear coupling terms for aerosol forcing neither for pre-industrial nor anthropogenic aerosols. It is difficult to predict even the sign of such coupling because it depends on co-variations of aerosol species and clouds as well as their respective vertical position. However, we suggest that total aerosol forcing should be systematically compared to the sum of the forcing from its components to understand any non-linearities better.

Note also the response to reviewer 2 with respect to differences to the corresponding findings from Myhre et al. (2013) on anthropogenic aerosols.

2 Other comments

• *Throughout the paper, clearly indicate that radiative forcings are diagnosed at the top of the atmosphere.*

We thank the reviewer for pointing that our. We added the clarification throughout the text.

• Page 18811, line 7: "(190 compared to 179)". What is the quantity being mentioned here, and its units?

The quantity is a forcing efficiency, and the units Wm^{-2} per unit optical depth. We should have placed this expression in parentheses after the sentence "increasing its cloudy-sky forcing efficiency with respect to LMDz". Following the advice of Referee 2 concerning the abstract, these numbers have been removed from the text.

• Page 18811, line 12: The dependency of direct radiative effect in aerosol amount (equivalent to aerosol optical depth, if the aerosol size distribution, chemical composition and environmental factors such as surface albedo remain the same) is indeed non-linear, but not strongly so. See Figure 3 of Boucher et al. (1998).

Boucher et al. (1998) considered an idealized experiment to compute forcing, where forcing efficiencies decreased by 10% when sulfate AOD increased from 0.1 to 0.5. We do come to a similar conclusion when considering global average. The nonlinear effect, comparing total aerosol forcing with the sum of the components forcing is small (14%), but when taking into account the regional patterns, it can be higher than 100%. In Boucher et al. (1998) the forcing comes only from sulfate (scattering) aerosols, while we consider 5 aerosol species: sulfate, sea salt, particulate organic matter, desert dust and black carbon. We presume that, in our case, absorbing black carbon plays an important role on the local nonlinearity of total forcing.
We have added:

although Boucher et al. (1998) suggested a small non-linearity for a range of global sulfate burdens corresponding to 0.1-0.5 AOD

• Page 18812, line 3: The presentation of assumptions in Bellouin et al. (2008) and Evan et al. (2009) is misleading. The text should note that Bellouin et al. (2008) acknowledge the importance of direct aerosol forcing in cloudy-sky, and that Evan et al. (2009) calculate the forcing at the surface, where the cloud "masking" assumption is arguably more acceptable than at the top of the atmosphere.

We removed the reference to Evan et al. (2009) which was indeed not pertinent for TOA forcing, and wrt to the reference to Bellouin et al. (2008) we rectified the sentence as follows:

Some studies on radiative forcing used or had to use (when based on satellite data) the assumption that aerosol effects are negligible in cloudy regions as they are "masked" by clouds (Bellouin et al. 2005). This was shown to be an important source of discrepancy between observation based and model based forcing estimates (Bellouin et al., 2008) .

• Page 18812, line 5: I guess the authors mean "single-column model".

Yes, this is what was meant. We now added it in the text.

• Page 18813, line 16: The definition of radiative forcing is, if indeed taken from the IPCC assessment reports, incomplete. First, the perturbation to the forcing agent is specifically a change since pre-industrial conditions. Second, the tropospheric state remains fixed to unperturbed values.

Therefore, the authors do not compute a radiative forcing in the IPCC sense of the term, but a radiative effect with respect to an atmosphere with no aerosols (as stated in Page 18816, lines 24–26).

We hopefully clarified this way in Sect. 2:

The radiative forcing according to IPCC is the change in net radiative flux at the tropopause due to an anthropogenic perturbation introduced by an atmospheric agent, measured against a preindustrial reference, allowing adjustment of stratospheric temperatures but with surface and tropospheric temperatures and state held fixed at the unperturbed values. The definition of forcing we adopt here deviates from that and considers as reference a state with no aerosols rather than pre-industrial aerosols, and uses TOA flux changes. In IPCC terms it would be named radiative effect. Our approach simplified the experiments and allowed to study also dust and sea salt radiative effects.

• Page 18814, lines 1–2: The text refers to agents A and B, but Figure 1 uses B and C. It would be useful to harmonise the notations.

We had two different notations in the manuscript. One was used at page 18814 and the other one in another Sect. 2. We have now harmonized the notation by using B and C in Sect. 2 as well.

• Page 18814, Figure 1, and Appendix: Note that the method of resolution of the equation of radiative transfer illustrated here is commonly called the "adding doubling" method.

We now mention the "adding doubling method" in the Appendix.

• Page 18815, line 8: AeroCom 1 or 2?

The sentence is corrected:

Aerosol fields are read as input into the model from monthly averages. In the reference experiment (see Sect. 2.4) these aerosols fields correspond to the year 2006 and are from a version of the LMDz-INCA model used for the recent forcing study in the AeroCom phase II (Myhre et al., 2013).

• Page 18815, line 16: What is meant by "(rather hygroscopic)"?

Since this is explained later in the text, we omitted it here.

• Page 18816, lines 5–7: Is hygroscopic growth applied separately to each aerosol component, or for the mixture? The text seems to suggest the latter, but is unclear because the words "our first assumption" on line 3 imply that a second assumption is made.

As far as the wet diameter of the aerosol is concerned, the hygroscopic growth is applied to the mixture of aerosol. We have though shortened the description of the hygroscopic growth factor impact on AOD, since we believe it is not important in this study. It reads now:

HG changes the particle diameter, the aerosol composition and particle surface characteristics. The optical properties of the individual components are computed with a look-up table, using the ambient relative humidity in the model grid cell.

• Page 18817, line 2: But those differences in meteorology are removed after a few timesteps when the model is nudged towards the ECMWF re-analysis, correct?

Differences are non-existing in our simulations, since we use nudging and exclude radiative effects from aerosols on meteorology. We wanted to point to the default GCM functionality. The last sentence reads now:

This explains why different aerosol and cloud fields in LMDz may give rise to different meteorological fields if such effect is permitted by experiment set-up. This is avoided in our experiments.

• Pages 18818–18819, sections 3.4.1 and 3.4.2: A Table summarising the configurations and experiments would be helpful.

A table has been added.

• Page 18819, line 24: Are cloud levels determined in each model column, that is the aerosol vertical profile in the abv, in, and blw experiments differ from one column to the next?

Yes absolutely. The test described pp. 18819-18820 is performed at each time step and for each model grid cell. See clarification for Sect. 3.4.2 above.

• Page 18821, line 6: Aerosol direct radiative effects will also cause feedbacks on modelled meteorology (since they modify the radiative budget), so should be listed here.

This has been added in the text.

• Page 18821: It may be clearer to merge the explanation for points 1 and 3 together, since both configurations are in fact double calls to the radiation scheme, with the non-advancing call using aerosols and climatological clouds, and the advancing call using no aerosols and interactive clouds.

We merged the two points emphasizing the fact that, for each “configuration”, we are indeed calling twice the radiation scheme with different cloud fields.

• Page 18831, Table 1, and Figure 8: Results suggest that the radiative forcing by total aerosols is stronger (more negative) than the sum of the component radiative forcings. Myhre et al. (2013) find the opposite result (see their Figure 16), which I find easier to understand: when all aerosols are considered together, there is more scattering and absorption in the atmosphere, thus component aerosols experience smaller radiative fluxes than when they are considered independently, thus their radiative forcing is weaker. Can the authors explain their result?

There are two elements in our answer, which suggest we can "explain our result":

1) Looking closer into Fig. 16 does not convince us about a contradictory finding by Myhre et al. (2013). What should be really compared in that figure are the red (sum of forcings) and the dotted yellow-black pdfs (total aerosol forcing). Only those pdfs contain all aerosol components including SOA and nitrate. The median of these two pdfs are not really different. Also - the result by Myhre et al. is for anthropogenic aerosols and we are investigating a mixture of all aerosols including

dust, sea salt and biomass burning aerosols. Finally, the curves do not reveal the model spread and it would be interesting to indeed see what other models would produce as coupling term.

2) Secondly we believe that aerosol composition and vertical positioning is needed to interpret the difference between total aerosol forcing and the sum of the component forcings. Our point is that the sign of the nonlinearity of the forcing (that is, the difference dRF=RF(AER)-∑RF(components)) strongly depends on the vertical superposition of absorbing and scattering aerosols. The prevailing effect, when putting several aerosols in the same column, is the selection of "which aerosol will intercept incoming radiation first", so, which one will be more effective. Indeed, in our case, the fact that the total aerosol forcing is more negative than the sum of the forcings (dRF<0), seems to be explained by BC being more present in lower layers than scattering aerosols. The fact that, in a similar situation, it is possible to have a negative dRF, can be illustrated with an example: consider, for simplicity, two ideal aerosols B situated below C, the first completely absorbing, the second completely reflecting, and a surface with albedo of 0.4. If the incoming radiation is 100, the net flux in the absence of aerosols (that is, the reference to compute forcings) will be:

Fnet(0)=100-40=60

As the two ideal aerosols have zero transmissivity, the individual forcings will be:

RF(B)= Fnet(B) – Fnet(0) = 100-60=40

RF(C)=0-60= -60

That implies *RF(B)+ RF(C)= -20*

Consider the forcing of B+C, when B is *below* C:

RF(B+C|B below)= 0 - 60= - 60

That is, the total *RF* from the two aerosols (-60) is more negative than the sum of the *RF* from B and C separately (-20). In the general case (transmissivity different from zero) the total forcing is influenced by the fact that the effectiveness of the aerosol that is below is reduced, while the one of the aerosol that is above is enhanced. In the case we considered here, putting B above C would imply a total forcing:

RF(B+C|B above) = 100 – 60 = 40

Thus, the absorbing behavior of the ensemble "B+C" would dominate over the scattering behavior.