

## ***Interactive comment on “Estimates of aerosol radiative forcing from the MACC re-analysis” by N. Bellouin et al.***

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We thank the two reviewers for their useful comments, which led to significant improvements to the manuscript. Changes were made to the MACC aerosol forcing algorithm, as described below.

- An error was discovered in the calculation of the marine aerosol background. This error had a sizeable impact on estimates of anthropogenic aerosol optical depth (AOD) and radiative forcing (RF). Anthropogenic AOD over the 2003–2010 period is now 0.073, up 50% from its previous value of 0.048. Anthropogenic events previously undetected, such as the North American pollution outflow over the North Atlantic or the biomass-burning plume transported from Southern Africa over the

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Indian Ocean, are now identified. However, suspected misidentifications occur in the Southern Ocean, but are associated with small anthropogenic AODs. All-sky direct and first indirect RF are now stronger at  $-0.7$  and  $-0.6 \text{ Wm}^{-2}$ , respectively, at the top of the atmosphere. Previous values were  $-0.5$  and  $-0.4 \text{ Wm}^{-2}$ , respectively.

- A more realistic cloud droplet effective radius of  $14 \mu\text{m}$  is now used in the calculation of cloud optical depth, down from  $20 \mu\text{m}$  previously. The new value and its uncertainty range (standard deviation of  $2.5 \mu\text{m}$ ) are taken from MODIS and CERES satellite data. This change has also contributed to the stronger first indirect RF mentioned above. The uncertainty bars of the former and revised estimates largely overlap.
- In response to the reviewers' criticisms, the uncertainty analysis has been completely reworked by moving away from the simple “bottom-up” combination of relative uncertainties included in the first draft to a Monte-Carlo analysis where uncertain input distributions and algorithm parameters are randomly varied within their probability density functions (PDFs). Year 2003 is then processed by the MACC algorithm 250 times, each time using different sets of uncertain parameters. The uncertainty analysis produces PDFs of component AODs, radiative effects, and radiative forcings. The Monte-Carlo framework and the characteristics of the PDFs used (shapes, means, and standard deviations) are now documented in a dedicated section of the revised manuscript.

The revised algorithm and the new uncertainty analysis yield a globally-averaged anthropogenic AOD of  $0.073 \pm 0.013$  over the period 2003–2010. Clear-sky anthropogenic direct RF is  $-2.5 \pm 0.5 \text{ Wm}^{-2}$  at the top of the atmosphere,  $-5.5 \pm 1.0 \text{ Wm}^{-2}$  at the surface. Scaled to all-sky conditions assuming no direct effect in cloudy sky, direct RF becomes  $-0.7 \pm 0.2 \text{ Wm}^{-2}$ . Combined with a cloudy-sky direct RF of  $+0.02 \pm 0.2 \text{ Wm}^{-2}$  (Schulz et al., 2006), the best estimate of all-sky direct RF remains the

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same, but its standard deviation increases to  $0.3 \text{ Wm}^{-2}$ . If direct RF is also corrected for differences between present-day natural and pre-industrial aerosols, the best estimate becomes  $-0.4 \text{ Wm}^{-2}$ . The uncertainty on that number is at least  $0.3 \text{ Wm}^{-2}$ , but does not include the potentially large uncertainty in the pre-industrial state. All-sky first indirect RF is  $-0.6 \pm 0.4 \text{ Wm}^{-2}$ , and direct and first indirect RF add up to  $-1.4 \pm 0.5 \text{ Wm}^{-2}$ . PDFs of radiative forcings for the year 2003 are shown in Figure 1. Note that the PDFs for first indirect and total RF are asymmetrical, because the first indirect RF cannot be negative in MACC, in accordance with the physics of the first indirect effect (additional aerosols can increase or leave unchanged the cloud droplet number concentration, but not decrease it).

The Monte-Carlo analysis offers three advantages over a simple combination of uncertainties. First, it is applied within the algorithm, instead of externally on global, multi-annual averages. Second, uncertainties propagate naturally and systematically from the uncertain input distributions and parameters to anthropogenic AOD, then on to direct and indirect RF. Third, the Monte-Carlo method is able to account for uncertainties whose impacts on RF compensate each other, either at the parameter level (e.g. a large amount of absorbing aerosols can exert the same top-of-atmosphere direct RF than a smaller amount of scattering aerosols) or at the global scale (e.g. underestimated anthropogenic AOD in Africa can be compensated by overestimates in North America). The 5 to 95% uncertainty range on the sum of direct and first indirect RF estimated by the Monte-Carlo method is  $-2.2$  to  $-0.6 \text{ Wm}^{-2}$ , smaller than the interval  $-2.9$  to  $-0.1 \text{ Wm}^{-2}$  quoted in the first version of the manuscript and with more robust foundations.

We now respond to individual reviewer comments, which are written in bold.

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## 1 Responses to comments by Reviewer 1

### Major comments:

**1. Section 3 describes how the total AOD is separated into individual components. I would suggest that they do readers' favor by including a flow chart to streamline the procedure, similar to that in Bellouin et al. (2005).**

It is a good idea. A flow chart is now included.

**2. The dust AOD was simply taken from the reanalysis. This represents a large uncertainty. As stated (line 5-26, p20080), "a large fraction" of mineral dust is fine-mode due to assumptions made on the size of emitted dust particles. How large is the fraction? If dust particles have been assumed to be too small in size, they may have been transported too far and global dust AOD may bias high. In the uncertainty assessment (line 18-29, p. 20082), uncertainties associated with dust AOD were not accounted for. I would like to suggest that they evaluate their dust AOD with some observations, such as AERONET observations in desert regions, dust AOD over ocean from Bellouin et al. (2005, 2008), MISR non-spherical AOD, and AIRS/IASI (Peyridieu et al., ACP, 10, 1953-1967, 2010; ACPD, 12, 23093-23133, 2012).**

It is indeed likely that mineral dust particles are transported too far in the model if their size distribution is biased towards smaller sizes. (The FMF of mineral-dust was 0.96 in that version of the MACC re-analysis.) However, if the erroneous transport translates into overestimated optical depths compared to observations, then data assimilation can mitigate the errors where satellite data is available. The forward model of the ECMWF IFS (i.e. without data assimilation) was included in the validation of global dust modelling by Huneeus et al. (2011) and its performance was similar to the other models. Benedetti et al. (2009) and Mangold et al. (2011) have shown that data assimilation improves performance, including where mineral dust dominates the total

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AOD.

Nevertheless, we agree that the simulation of mineral dust AOD represents an important uncertainty in our estimates. Two sources of mineral dust uncertainty are now included in the Monte-Carlo analysis. First, the uncertainty in the global averaged mineral dust AOD is taken from the AeroCom diversity for that variable, which corresponds to a standard deviation of 0.014 (Table 4 of Kinne et al., 2006). Second, the uncertainty in the mineral dust AOD in each grid-box is assumed to be the same as for the total AOD, which is in turn assumed to be that of the MODIS retrieval ( $0.03 \pm 5\%$  over ocean,  $0.05 \pm 15\%$  over land). That second form of uncertainty is applied as a random error in each grid box.

**3. Figure 6: a trend based on 8 years data is not very useful. How did you fit the trend? What is confidence level of the trend? I would suggest the confidence level is marked in the figure.**

The figure showing 8-year linear trends was intended as an illustration of how the MACC aerosol forcing products can be used in the future. In the text, we were cautious not to over-interpret the results. We agree that 8 years is too short a period to achieve statistical significance, and that the figure is potentially misleading. The figure is therefore removed, and the manuscript now states that trend analysis will become possible as more years are analysed by the ECMWF IFS.

**Other comments:**

**1. throughout the paper: “sea-salt aerosol” should be changed to “marine aerosol”, including sea-salt, sulfate transformed from DMS, and maybe organics.**

Agreed, we have changed “sea-salt aerosol” to “marine aerosol” throughout, except where referring to the sea-salt aerosol tracer of the IFS model.

C10911

**2. p.20075, line 11: what do you mean “correct”?**

Agreed, the wording was not good. Sentence now reads “Those distributions are then provided to the radiation scheme where aerosol direct effects are included in the calculation of radiative fluxes.”

**3. p.20075, line 27-28: the statement is not correct because satellite observations of above-cloud AOD are emerging.**

Agreed, sentence rewritten as “aerosol optical depths (AODs) are not yet routinely retrieved in cloudy sky”.

**4. p.20077, line 14-15: “Simulations by numerical aerosol models are not affected by the limited sampling of satellite retrievals”. What does this sentence really mean? Please clarify.**

Sentence changed to “In contrast to satellite retrievals, numerical aerosol models simulate the full spatial and temporal distributions of aerosols, without data gaps.”

**5. p.20078, line 18-20: This SO<sub>2</sub> to sulfate transformation scheme appears too simple to me. Is it justified?**

It is a simple representation of the sulphur-cycle chemistry, justified by its low cost, calibration against the LMD-LOA model, and reasonable simulation of the sulphate aerosol distribution. However, Mangold et al. (2011) have shown on a case study that sulphate mass concentrations can be overestimated by the model, and this bias is only partially removed by the assimilation. The forward aerosol is currently undergoing improvements, which include a more sophisticated representation of aerosol chemistry. In this study, we only use modelled speciation indirectly, mitigating the impacts of this simple representation.

**6. p.20078, line 27: please clarify how the model trajectory is updated during the**

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### **assimilation process.**

The word “trajectory” was indeed ambiguous, since aerosol assimilation does not yet affect the simulated temperatures, pressures, or specific humidities. The manuscript now reads “Aerosol data assimilation consists in the minimisation of a complicated cost function and updates the modelled aerosol masses in order to match observations more closely.”

### **7. p.20079, line 20-23: “interestingly”. Why is this comparison interesting?**

We agree that the statement is merely noteworthy. “Interestingly” has been deleted.

### **8. p.20081: how does the model define FMF? Is the definition consistent with MODIS or some in-situ observations?**

Fine mode particles are those with radii smaller than  $0.5 \mu\text{m}$ , following MODIS. The manuscript now includes the definition of non-dust FMF, which is

$$f' = \frac{\tau_{\text{sulphate}} + \tau_{\text{black-carbon}} + \tau_{\text{organic-matter}} + \tau_{\text{seasalt}} \cdot f_{\text{seasalt}}}{\tau_{\text{sulphate}} + \tau_{\text{black-carbon}} + \tau_{\text{organic-matter}} + \tau_{\text{seasalt}}} \quad (1)$$

where  $f_{\text{seasalt}}$  is the FMF of sea-salt, assumed to be 0.3. (This parameter is part of the Monte-Carlo uncertainty analysis, where its standard deviation is 0.2.) The equation above is consistent with the size distributions assumed in the model, where sulphate, organic matter, and black carbon AODs are fully in the fine mode, and 30% of globally-averaged sea-salt AOD is due to the two bins below  $0.5 \mu\text{m}$ .

### **9. p.20081, line 24: “using emissions of natural aerosols only”. Please specify what kinds of natural sources are considered here. Do you consider biomass burning aerosol to be completely natural?**

The list of natural aerosol sources used in HadGEM to derive anthropogenic fractions over land is now given in the manuscript: mineral dust, sea-salt, ocean- and land-based  
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dimethyl sulphide, sulphur dioxide from degassing volcanoes, and a climatology of biogenic aerosols. Biomass-burning aerosol is considered completely anthropogenic, as there is little information about its origins in the emission datasets, and the distinction is virtually impossible from observations alone.

### **10. p.20082, line 1: please specify if you considered the seasonal and spatial (on a scale of model horizontal resolution) variations of anthropogenic fraction?**

The manuscript now specifies that anthropogenic fraction has no seasonality, and does not vary within the regions specified in Table 1. This will be improved upon in the next version of the MACC aerosol forcing products.

### **11. p.20082, line 14-15: “Sea-salt AOD is maximum over mid-latitude oceans where near-surface wind speeds are large” It is interesting that maximum sea-salt AOD is not in the “roaring forties”.**

Maxima of marine AOD seen in the Arabian Sea in JJA have now been removed by the correction of an error in the calculation of the marine background. Maxima are now located within 40 to 50 degrees of latitude.

### **12. p.20082, line 24-29: please give some details about the Monte-Carlo estimate of uncertainty. Also the uncertainty associated with dust AOD should be included.**

The Monte-Carlo estimate of uncertainty is now fully detailed in a dedicated section, and includes uncertainty in dust AOD.

### **13. p.200823, line 5-6: Why is the impact of the FMF uncertainty reduced when the method only compares the FMF to a threshold?**

When the FMF is used in a mathematical expression, its uncertainty propagates directly to the value being computed. When the FMF is compared to a threshold, its un-

certainty only propagates when it is large enough to bring the FMF below the threshold, which does not happen all the time, thus lowering the impact of the uncertainty. The corresponding statement in the paper has been made clearer.

**14. p.20084, line 15-16: “Anthropogenic aerosols contribute most to the absorption” Can you give a percentage?**

Global, multi-annual averaged component absorption aerosol optical depths are now included in Table 2. Anthropogenic aerosols represent 88% of total aerosol absorption.

**15. p.20090, line 12: “Given the available data”, what data?**

The wording is improved by now saying “Given current knowledge”.

**16. p.20094: what are optimal finger printing techniques?**

They are statistical techniques (similar to least-square regressions) used to increase the signal-to-noise ratio in detection and attribution studies. The paper now refers the interested reader to appendix 9A of the IPCC AR4.

**17. p.20094, line 25-27: need a reference here to back up the statement that calibration issues result in spurious trends of AOD.**

Added reference to Zhang and Reid, Atmos. Chem. Phys., 2010.

## **2 Responses to comments by Reviewer 2**

**This paper is an attempt to provide another forcing estimate based on assimilated aerosol data for AOD as well as anthropogenic fraction of AOD over land from a model. Over oceans, a method is used based solely on observations, but that also includes a number of unverified assumptions. My problem with**

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**the paper is that most of the justification of the assumptions in deriving these estimates is not given, and that the “uncertainty” analysis is not spelled out in detail and does not include the range of uncertainties from the literature that are known to affect such estimates. The authors need to correct the paper to address the following concerns (the majority of which and the most important of which concern the uncertainty analyses).**

The uncertainty analysis has been much improved in response to the reviewer’s comments. We now use a Monte-Carlo uncertainty analysis, where uncertain input distributions and parameters are varied within their probability density functions. That method is more mathematically sound and, we hope, more convincing.

**Abstract, line 13: “being bounded” should be “are bounded”**

In the sentence, “results” was used as a verb, so “being bounded” was grammatically correct. The sentence has however been rewritten to avoid confusion.

**Page 20075, line 17: actually the susceptibility of precipitation formation has been shown to be too high in climate models likely leading to incorrect responses to aerosol change (Wang et al., GRL, 2012)**

This is indeed a convincing result, and in line with previous results from a model intercomparison study. We have added the following to the revised manuscript: “[...] is debated (Stevens and Feingold, 2009), with evidence of climate models overpredicting the strength of the second indirect effect (Quaas et al., 2009; Wang et al., 2012).”

**Line 23: specify from an observational-only view**

The sentence now reads “From a purely observational point of view”.

**20076: define “DRF efficiencies” so that this jargon is avoided.**

DRF efficiencies are now defined as DRF per unit optical depth.

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**Line 23-24: “concludes with the TOA . . .” needs to be reworded. i.e. “concluded that the best estimate of TOA . . .”**

The sentence has been reworded as suggested.

**Page 20077: line 5: wording: change to “ spread in model results”**

The sentence has been reworded as suggested.

**11-13: I think the referencing here does not reflect the literature. I would suggest that the Penner et al. reference does not suggest that model estimates are necessarily correct. Only that the use of the satellite method with a model leads to a similar discrepancy, since the satellite method does not properly retrieve the pre-industrial forcing. The Quaas et al. paper does not demonstrate that model estimates of pre-industrial forcing are in error.**

In light of the reviewer’s remark, we feel that this statement is more confusing than helpful, and we remove it from the revised manuscript, i.e. delete from “It is debated” to “(Quaas et al., 2011).”

**Page 20078: line 21-22. Diagnosis of AOD from aerosol mass needs to also specify the aerosol size distribution as well as refractive indices. In addition, one needs to know whether the separate types are internally or externally mixed.**

The reader is now referred to Section 4.2 of Morcrette et al. [2009] for details of the aerosol optical properties used in the ECMWF aerosol model. In addition, the paper now states that external mixture is assumed for AOD calculations.

**Page 20079: line 9-10: correcting the model in proportion to the original contribution to total mass would be incorrect, if sulfate, for example, would tend to be in smaller sizes (since it is subject to nucleation, which increases aerosol number but tends to produce smaller sizes) What are the assumptions you are**

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**building into this assimilation product? This and other assumptions need to be spelled out.**

It is true that component AODs do not scale in the same way as component masses. The statement was an over-simplification of the data assimilation scheme, which assimilates total AOD, but uses total mass-mixing ratio as control variable. The actual explanation is complex, but well described in section 2.5 of Benedetti et al. (2009). The manuscript now reads: “During data assimilation, each aerosol component is corrected in proportion of its original contribution to the total aerosol mass. This mass increment is then converted to an AOD and compared against the assimilated MODIS retrieval, until convergence is achieved within observational and model errors.”

**Line 13-14: it corrects for left-out species, but does not correct for the fraction of nitrate that is anthropogenic (without further assumptions). This needs to be spelled out.**

Does the reviewer mean that assimilation only affects total AOD, not speciation, and thus will not impact the anthropogenic identification applied later? If so, the reviewer is correct, but the sentence does already make clear that only total AOD can be corrected by data assimilation.

**Line 20: “interestingly” should be “not surprisingly” since the AOD is assimilated to match the satellite retrieval.**

“interestingly” has been deleted.

**Page 20080: “Unfortunately, it is not possible to completely avoid using modelled fields that are not affected by the data assimilation. For example, although the aerosol forcing estimation does not rely on aerosol speciation, it does use an associated variable, the FMF of the total AOD.” So you are not using the information about refractive index (which requires aerosol speciation) to estimate forcing?**

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We are using speciated optical properties indirectly, since they are used in the calculation of component AODs and the FMF. But we are not using those optical properties when computing radiative effects from the component AODs, see response to comments below.

**Page 20081, line 6: Why aren't advected natural aerosols in addition to advected anthropogenic aerosols part of what is seen over oceans?**

It is indeed likely that advected natural aerosols form a fourth aerosol type over ocean, alongside mineral dust, marine, and anthropogenic aerosols. The manuscript now acknowledges this limitation. The problem is that distinguishing anthropogenic and fine-mode natural aerosols is difficult to do from AOD and FMF alone. Over land, we use model-derived anthropogenic fractions, but this was not extended to oceans. We shall investigate such an extension for future versions of the MACC aerosol forcing products.

**Eq. 1, 2: The sea salt source functions are highly variable. According to Jaegle et al., ACP 2011 and Lewis and Schwartz 2004 (see Jaegle for reference), sea salt is one of the most poorly constrained aerosols. What is the uncertainty associated with this estimate? How does this uncertainty affect your conclusions? For example, in situ observations give a range of mass extinction coefficients from 3.1 to 6.6 m<sup>2</sup>/g. Thus, there is at least a factor of 2 uncertainty in this estimate, which should be factored into your analysis and conclusions.**

The new Monte-Carlo uncertainty analysis yields a standard deviation for the marine AOD of 0.022 over oceans, 28% of the global average of 0.078. Uncertain input distributions that affect the marine AOD estimate are the total AOD, mineral dust AOD, and non-dust FMF. Uncertain algorithm parameters that affect the marine AOD estimate are the parameters used in the first guess of the marine AOD, the prescribed fine-mode fraction of marine aerosols, and the threshold on the non-dust fine-mode fraction.

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Jaeglé et al. (2011) use MODIS and AERONET to test and rank various empirical source functions for sea-salt. So, in effect, they use AOD retrievals to constrain their model – a constraint we get explicitly from assimilating MODIS AODs. As for the uncertainty in specific coefficients, it is unlikely that it propagates directly and in full to the uncertainty in the column-integrated total AOD retrieved by MODIS or simulated by the ECMWF IFS.

**Note: equ 3 seems odd: fine mode fraction  $f$  is difference in  $f \cdot \tau$  for non-dust and seasalt divided by difference of  $\tau$  for non-dust minus seasalt. But it is FMF of  $\tau$ , right?**

The FMF computed by equation 3 is the FMF of non-dust, non-first-guess-marine aerosols. In other words, it is the FMF that non-dust aerosols would have if the first guess of marine AOD was true. Testing this FMF against the threshold of the coarse-mode category (FMF below  $0.35 \pm 0.05$ ) is used to assess whether the first guess can be retained or not.

**Page 20082: Line 11, 12 and Fig.1: Very strange distribution of fine-mode natural: Where are the year-round biogenic aerosols in the tropics?**

It is virtually impossible to distinguish anthropogenic and fine-mode natural aerosols from their AOD or FMF alone, since they are both in the fine mode. (Indeed, it is difficult to reliably pinpoint locations with fine-mode natural aerosols on a map of satellite-retrieved AOD.) We therefore chose to use anthropogenic fractions, derived from numerical modelling. Unfortunately, this simple choice means that anthropogenic and fine-mode natural aerosol distributions share the same patterns, which is unrealistic. This limitation is now mentioned in the paper, and will be improved upon in future versions of the MACC aerosol forcing products, where regional anthropogenic fractions will be replaced by gridded distributions, as stated in the conclusion.

**Line 19, 20: not just errors in forward modeling of AOD, but also errors in mod-**

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**eled/assumed component fractions.**

Agreed, sentence now reads “in the forward modelling of total and component AODs”.

**Line 27 – 29: If there is a factor of 2 uncertainty in the seasalt optical depth (based on quotes in Jaegle et al, above), how do you obtain a 10% uncertainty on anthropogenic AOD over ocean. Also, what is justification for uncertainty estimate over land?**

and

**Page 20083: line 1 - 7: how do you calculate the relative uncertainty? Something is being used to argue for small uncertainties here since 10% anthrop. over ocean / 21% total AOD over ocean is not 31%. It would seem Yu’s method is more reasonable estimate of uncertainty.**

The uncertainty analysis now uses a Monte-Carlo framework, and yields a 16% and 23% relative uncertainty on anthropogenic AOD over ocean and land, respectively. Those values are in fact smaller than relative uncertainties previously obtained by a simple “bottom-up” assessment of uncertainties on a global average. This decrease happened because, although local uncertainties may be large, they are not a systematic bias, and thus partially compensate on a global average.

**Your statement “The method used in this study only compares the FMF to a threshold and the impact of the FMF uncertainty is reduced.” Is not clear.**

The statement has been made clearer. The idea is that when the FMF is used in a mathematical expression, its uncertainty propagates directly to the value being computed. When the FMF is compared to a threshold, however, its uncertainty only propagates when it is large enough to bring the FMF below the threshold, which does not happen all the time, thus lowering the impact of the uncertainty.

**Line 15 – 18: “Here, the AeroCom anthropogenic AOD of 0.029, defined with C10921**

**respect to pre-industrial conditions, has been multiplied by 1.25 to correct for the pre-industrial aerosol distribution, following Bellouin et al. (2008) where aerosol and precursor emissions for the year 1860 were used to represent pre-industrial aerosols.” Please clarify this writing. Are you correcting the AeroCom values because their PI case was for 1850, whereas yours is 1750? Where did you get the factor of 1.25?**

Numerical models (and the IPCC) define “anthropogenic” as the difference between pre-industrial and present-day conditions. Observation-based studies (such as this paper) define “anthropogenic” as the fraction of present-day aerosols that are thought to be of anthropogenic origin. The difference is subtle but important, because there were aerosols of anthropogenic origin in pre-industrial times, most notably biomass-burning aerosols. Consequently, the “AeroCom anthropogenic” AOD has to be converted to the MACC definition of the term before attempting a comparison. The paragraph describes that conversion.

The writing has been clarified as follows: “On the modelling side, AeroCom models simulate an anthropogenic AOD of 0.029, or 25% of their present-day total AOD (Schulz et al., 2006). However, AeroCom defines anthropogenic AOD as the difference between total AOD in pre-industrial and present-day conditions: this is not the same as the present-day anthropogenic AOD derived in MACC. Bellouin et al. (2008) used the Hadley Centre climate model to find that the optical depth of present-day anthropogenic aerosols is 1.25 times larger than the change in total AOD between pre-industrial and present-day conditions. Using that factor, AeroCom models would simulate a present-day anthropogenic AOD of 0.031, 31% of their present-day total AOD.”

**line 19-21 “There is therefore some agreement in the fraction of present-day total AOD that is anthropogenic between satellite-based, assimilation-based, and modelling-based estimates. However, free-running global aerosol models have lower total and anthropogenic AODs.” Yes, comforting, but clearly some**



**of this is just the choices for dust AOD and sea salt AOD you made, and other choices would not have provided a similar fraction. Also, the regional fractions may be totally different. Please provide these as well.**

The new uncertainty analysis suggests that the globally-averaged anthropogenic fraction (which has increased from 27% to 40% after the bug fixing) is relatively robust to choices in algorithm parameters, with a standard deviation of 4%. Regionally, differences with modelled estimates are expected to be larger, with the important caveat that anthropogenic fractions used by the MACC algorithm over land surfaces come from a numerical model, and are therefore not truly independent. This is now stated in the manuscript.

**Page 20084: line 3, 5: “The aerosol vertical profile is a key parameter in the long-wave spectrum.” Actually it is a key parameter in determining the effects of BC as well. If you cannot rely on it for the longwave, why is it ok in the shortwave?**

Agreed, the statement is removed. Note that we also do not rely on the modelled profile in the shortwave spectrum, we use prescribed profiles instead, whose impact on the uncertainty is difficult to assess (see Figure 2 and response to comment below).

**Line 6 – 8: Are the derived optical properties consistent with the optical properties used in the satellite algorithm to obtain the optical depth? This is key to determine, since otherwise you are relying on optical properties for radiative forcing that are not consistent with your optical depths based on satellite data. I think the answer you provide is a good start: providing one method of getting forcing, but a second evaluation using the optical properties from the satellite analysis should be made to obtain a range of results.**

There is no consistency between the aerosol optical properties used by MODIS, the ECMWF IFS, and the MACC aerosol forcing estimates because each of those distinguish different aerosol types. MODIS retrievals are based on aerosol models represen-

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tative of broad classes of ambient aerosols (and are now derived from an analysis of AERONET retrievals). ECMWF simulates sulphate, black carbon, organic matter, mineral dust, and sea-salt aerosols, and prescribes global optical properties for each of those components when computing AOD. MACC aerosol forcing products distinguish mineral dust, marine, anthropogenic, and fine-mode natural aerosols, and AERONET is used to characterise their optical properties. Overall, it is reasonable to expect that, in spite of inconsistencies, the different prescriptions of aerosol optical properties remain broadly similar.

In addition, maintaining consistency in aerosol optical properties may not be as advantageous as it first appears, because errors do not always compensate when going from top-of-atmosphere radiances to AOD, and then from AOD to radiative fluxes. The freedom of coupling component AODs to AERONET-derived regional optical properties is, however, a significant advantage, provided it is accompanied with an assessment of uncertainties, such as done in the paper where regional SSAs are given a standard deviation of 0.03 in the Monte-Carlo analysis.

**Table 1 only gives one value of single scattering albedo for each aerosol type and no value for mass extinction efficiency (needed for radiative transfer calculation of forcing).**

The radiative transfer calculations used for the MACC aerosol forcing products require only AOD, SSA, and phase function as inputs. Specific extinction coefficients are not explicitly required.

**Why doesn't the single scattering albedo change with relative humidity (RH)? Even if you are claiming it is fine to use the average boundary layer RH value of SSA over these large areas, I would think there should be a seasonal variation of RH over land areas at least. Also, the use of strongly absorbing anthropogenic aerosols (SSA = 0.86) over Africa for all ocean areas would seem to bias your calculation of anthropogenic aerosol effects over the oceans. Or since this ta-**

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ble refers to properties over land areas, perhaps you use something else over oceans? What is it? Also table heading states: “Representative AERONET sites refer to the sites studied by Dubovik et al. (2002). For the other components, optical properties are prescribed globally” What exactly are you prescribing globally? I gather you do not use the regional values globally (i.e. from GSFC, Creteil, . . .) Maybe you just refer to the dust and sea salt properties?

The caption of Table 1 has been made clearer by adding “over ocean and land surfaces” to the description of the prescription of aerosol optical properties, and spelling out that optical properties for mineral dust and marine aerosols are applied globally.

The SSA is taken from an analysis of AERONET retrievals (Dubovik et al., 2002) performed when the aerosol to be characterised was present in the atmosphere. It characterises the ambient aerosol, and therefore factors in the dependence in relative humidity. It is however true that applying properties from a specific AERONET site to large regions is a simplification. In future versions of the MACC aerosol forcing products, regional SSAs will be replaced by gridded datasets of fine-mode SSA, obtained from an analysis of a greater number of AERONET sites. In the present paper, regional SSAs are given a standard deviation of 0.03 in the Monte-Carlo analysis, and this uncertainty contributes to the total uncertainty in anthropogenic direct radiative effect and forcing.

**Line 22 – 25 “Rather, it is assumed that natural aerosols are located in the first kilometre of the atmosphere, below a layer of anthropogenic aerosols. For the cloud-free DRE, the impact of this assumed vertical profile on shortwave radiative fluxes is small” But, these would not be small effects if you were taking RH variations into account. What is the impact of neglecting these variations?**

The MODIS retrievals that are assimilated in the MACC re-analysis, and the AERONET retrievals that are used to prescribe aerosol optical properties in the MACC aerosol forcing products, characterise the full aerosol column, with no information on vertical profile. Relative humidity dependence is implicitly included in those retrievals, so it is

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not neglected at all.

However, it is true that the prescription of the vertical profile is unlikely to be consistent with the relative humidity profile at the time of the retrieval. This uncertainty, along with other uncertainties in the radiative transfer calculation, such as the use of standard atmospheric profiles, is difficult to quantify. The manuscript now mentions this caveat.

**Page 20085: line 3 – 4: Figure 3: I still fail to understand a lack of maximum natural component of forcing in the tropics, and that is similar for all seasons.**

In MACC, we use anthropogenic fractions, derived from numerical modelling, over land surfaces. Unfortunately, this simple choice means that anthropogenic and fine-mode natural aerosol distributions share the same patterns, which is unrealistic. This limitation is now mentioned in the manuscript, and will be improved upon in future versions of the MACC aerosol forcing products, where regional anthropogenic fractions will be replaced by gridded distributions, as stated in the conclusion.

**Page 20086, line 4-6: “Uncertainties in component AOD, computed above, are translated to uncertainties in component DRE by multiplying by the component DREE.” These are not explained on page 20082, so I would not say you “computed” the uncertainty. The way this is stated, it looks like you just pulled a number of 15air. (see 20082 line 28)**

and

**“In addition, the uncertainty on the DREE due to uncertain prescribed optical properties has been quantified for the anthropogenic component by the Monte-Carlo approach of Bellouin et al. (2005).” What was the approach of Bellouin? Please do not make the reader go to another paper. You need to summarize this approach here. Especially, if one goes to the 2005 paper and finds numbers pulled out of the air, then we are building “science” out of thin air.**

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The uncertainty analysis has been reworked and uses a Monte-Carlo framework, whereby uncertainties in component AODs are directly propagated to direct radiative effects and forcings. Uncertainties are now effectively “computed”. The Monte-Carlo method is described in a dedicated section of the paper.

**Line 17: values are not usually “stronger”: use “larger”**

The sentence has been rewritten to read “The corresponding MACC radiative forcings, ...”. The use of “stronger” is convenient for a negative forcing because, mathematically speaking, stronger RF correspond to smaller (more negative) values.

**Page 20087, line 13 -14: how different would the DRF be if you use natural aerosols over anthropogenic aerosols (i.e. one way to indicate uncertainty).**

The uncertainty in the vertical profile is not included in the Monte-Carlo framework, but we agree it merits a mention in the paper and have performed additional radiative transfer calculations. Results are shown in Figure 2.

The top panels show that increasing the altitude of the anthropogenic aerosol layer linearly strengthens (if the aerosol is scattering) or weakens (if the aerosol is more absorbing) the direct effect at TOA. The change is due to the reduction in Rayleigh scattering above the aerosol layer with increasing aerosol altitude. The effect on daily-integrated DRE is small, at less than 2% per kilometre.

The bottom panels show the impact of placing the natural aerosol layer above the anthropogenic layer, instead of below. Natural aerosols weaken the anthropogenic DRF by increasing the reflectance of the underlying atmosphere if located below, or by increasing the extinction by the overlying atmosphere if located above. For scattering anthropogenic aerosols, the two effects have a similar impact on the anthropogenic DRF (bottom left-hand panel) and the relative location of natural aerosol is of secondary importance. However, it becomes important when aerosols are absorbing, as shown in the bottom right-hand panel. Absorbing anthropogenic aerosols are more

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likely to exert weak, or even positive, DRFs when located above a layer made more reflective by underlying natural aerosols. In contrast, natural aerosols located above the anthropogenic layer do not trigger this mechanism. The first paragraph of section 4.2 now ends with “The assumption that natural aerosols are located below the anthropogenic layer effectively increases the reflectance of the underlying atmosphere, and makes the DRF less negative than when other vertical profiles are used. The weakening of the DRF is large when anthropogenic aerosols are absorbing, and the MACC DRF is underestimated when natural aerosols in fact overlie the anthropogenic layer, for example in situations of long-range transport of mineral dust aerosols above a biomass-burning plume.”

**Line 25-27: Models have shown that this area has positive forcing since 1998. This is expected theoretically for small SSA for biomass burning aerosols.**

We feel it is better to point out the recent advances in satellite retrievals of cloudy-sky aerosol RF. Before those advances, models were basically unconstrained. In addition, although the direct radiative effect is theoretically expected to be positive for absorbing aerosols overlying stratocumulus decks, not all models simulates those positive radiative effects, and diversity in AeroCom cloudy-sky forcing is large (Schulz et al., 2006). Diversity arises from different assumptions for biomass-burning aerosol absorption, and from varying skill at simulating stratocumulus decks in the right regions.

**Table 3: The uncertainty analysis is crucial here. You need to provide details. I also think you need to do this regionally, since the errors must depend on the regional character of the aerosols and the choices you’ve made.**

The uncertainty analysis is now documented in a dedicated section of the paper, and Table 3 is obsolete. In the Monte-Carlo framework, uncertain input distributions and parameters are varied either within each grid-box, or regionally, or globally, depending on how the corresponding parameter is prescribed and used by the algorithm.

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**Page 20089: line 15, 16:  $d\ln(Nd)/d\ln(\text{AOD})$  from the Quaas et al analysis can be very different than the change that a model would compute from  $d\ln(Nd) / d\ln(\tau - \tau_{\text{natural}})$ , i.e. the slope of the line computed from satellite data does not necessarily capture the change in droplets based on the full optical depth and the natural optical depth because the satellite data are unable to select only the natural data (i.e. even a low optical depth cannot be considered “natural”). What is the possible error/uncertainty due to this effect?**

We apologise if the manuscript has been unclear in this regard. The actual derivative is evaluated by statistical methods by using the present-day total (natural plus anthropogenic) aerosol optical depth. In order to make Equation 6 clearer, we have added a “.” sign between the term  $\frac{\partial \ln Nd}{\partial \ln \tau}$  and the term  $(\ln \tau - \ln \tau_{\text{nat}})$ . Uncertainties in cloud susceptibilities can be computed from the statistical method that provides the susceptibilities, and have been included in the Monte-Carlo analysis.

**Page 20090: line 2: the use of AOD as a proxy for aerosol number is known to be highly uncertain (better to use angstrom exponent (AE) times AOD. How different are your results if you use the latter? (The measurements by Andreae that you quote do not form a tightly fit 1:1 line.)**

We agree that it would be beneficial to explore metrics which are more elaborate, and we will do so in future versions of the MACC aerosol forcing products. In the framework of the present study, we continue to use AOD because this quantity is well constrained by the assimilation of satellite retrievals in the re-analysis, and because we can rely on published estimates for the sensitivities and their uncertainties.

**Line 9 – 10: actually the primary criticism of using  $d\ln Nd/d\ln(\text{AOD})$  which applies to both the use of AOD and the use of AOD X AE is that the slopes derived from satellite data for present day do not represent the slopes for present day – natural (see comment above).**

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This is true, and the statement in the manuscript is revised as follows: “... underestimation of aerosol indirect radiative forcing. Statistics sampled from present-day variability in AOD and CDNC may not be sufficient to sample the full difference between pre-industrial and present-day conditions (Penner et al., 2011).”

**Line 10 – 12: Grandey and Stier mainly fault the method based on using a large area average (shown also in a previous Quaas paper). And this may indeed lead to an overestimate. But this is no reason to suggest that the estimate given here is ok, considering it is faulty in both directions! Please use the information in these papers to estimate the uncertainty in your methods.**

The results by Grandey and Stier are interesting, but in our opinion merit further discussion. It is questionable why sampling just the temporal variability at one grid-point should be a method superior to sampling the spatio-temporal variability in a larger area. In any case, their results highlight that even lower sensitivities cannot be excluded. The standard deviations on sensitivity of CDNC to CCN perturbations used in the Monte-Carlo analysis include a large range of sensitivities.

**Line 20 – 21: the use of different regions/seasons for the partial derivatives here would suggest this also for the optical properties used for the direct effect (for balance, if not a more defensible estimate).**

Optical properties used for estimates of the anthropogenic direct radiative effect and forcing are regional (albeit over large regions). As for the seasonality, it is worth pointing out that anthropogenic optical properties are only applied when anthropogenic aerosols are identified, thus providing a seasonal variation in total aerosol optical properties.

**Line 28: I am surprised the fractional cloud cover is thought to be well known from the IFS. How do these values compare to observations? We should at least be given an estimate of how “certain” they seem, since the paper makes this judgment.**

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The reviewer is right to question this assessment. Cloud cover uncertainty is now included in the Monte-Carlo uncertainty analysis, with a standard deviation of 0.03 for the global average, and a random error with a standard deviation of 0.1 at the grid-box level. The uncertainty on the global average is taken from the GEWEX cloud assessment (Stubenrauch et al., Bull. Am. Meteorol. Soc., in press, 2012, available online at [climserv.ipsl.polytechnique.fr/gewexca/papers/BAMS-D-12-00117.pdf](http://climserv.ipsl.polytechnique.fr/gewexca/papers/BAMS-D-12-00117.pdf)).

**Page 20091, line 1, 2: don't tell me you are just going to evaluate equ. 6 using seasonal averages and apply the range based on seasonal values as the uncertainty? What exactly do you do?**

The method was to run the algorithm again, this time using the largest and smallest of the four seasonal sensitivities in each region in order to bound the indirect RF forcing estimates – we did not simply use seasonal averages. However, the statement is now obsolete, since the uncertainty assessment has been fully revised and is now documented in a dedicated section of the revised manuscript.

**Line 10, 11: do these compilations address the issue noted above (i.e. that the primary criticism of using  $d\ln Nd/d\ln(AOD)$  which applies to both the use of AOD and the use of  $AOD \times AE$  is that the slopes derived from satellite data for present day do not represent the slopes for present day – natural)? Somehow, I do not think so, since they are all based on data for the total aerosol.**

They do not address these specific arguments directly, but stem from a large range of data sources and methodologies. As such, we expect that they cover a much larger range of variability, effectively encompassing the difference natural vs. present-day, and encompassing the smaller local vs. larger spatio-temporal variability. Also, they encompass a very large portion of the total physically plausible range (0 to 1). Those two points are now stated in the revised manuscript. In the Monte-Carlo uncertainty analysis, the compilations provided the standard deviations on regional cloud suscep-

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tibilities.

**Line 15: You need to provide a table with the uncertainties of the different factors that you include here and then deriving the 37%. Otherwise no one can truly know what you've done, and a scientific paper needs to have enough information for the next researchers to be able to reproduce your results.**

Thanks to the Monte-Carlo framework, uncertainties in anthropogenic AOD propagate fully and consistently to direct and indirect RF. The PDFs used in the Monte-Carlo analysis are clearly documented, which makes the results reproducible by a motivated reader.

**Line 20 – 23: It is nice that you get values that are somewhat larger than IPCC, but I suspect a true analysis of uncertainties would be far larger.**

Our revised estimate of direct RF (including both cloudy-sky uncertainty and scaling to a pre-industrial reference) is  $-0.4 \pm 0.3 \text{ Wm}^{-2}$ . The uncertainty in the pre-industrial reference is not included. The corresponding IPCC estimate is  $-0.4 \pm 0.4 \text{ Wm}^{-2}$ , so our estimate is slightly less uncertain. For the first indirect RF, our revised estimate is  $-0.6 \pm 0.4 \text{ Wm}^{-2}$ , or a 5–95% confidence range ( $\pm 2$  standard deviations) of  $-1.3$  to  $+0.1 \text{ Wm}^{-2}$ . The corresponding IPCC range is  $-1.8$  to  $-0.3 \text{ Wm}^{-2}$ : our estimated range is shifted towards weaker RF, but has a similar size.

The size of IPCC uncertainties in the direct and first indirect RF is debated, but our uncertainty analysis provides some support to the IPCC choice. It is worth noting that a “bottom-up” estimate of uncertainties is bound to produce a larger uncertainty range because it cannot account for compensating effects on local values (compensation between uncertain aerosol properties) or on the global average (compensation between regional uncertainties).

Caption of Figure 1: Probability density functions for all-sky shortwave direct and first indirect aerosol radiative forcing, and their sum, as obtained from the Monte-Carlo

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uncertainty analysis for the year 2003.

Caption of Figure 2: Top panels: Direct radiative effect (DRE) as a function of aerosol altitude for an aerosol optical depth (AOD) of 0.5 and a single-scattering albedo (SSA) of 0.98 and 0.86 (left- and right-hand panels, resp.), on solstices and equinoxes. Bottom panels: DRE of an anthropogenic AOD of 0.5 and SSA of 0.98 and 0.86 (left- and right-hand panels, resp.) as a function of the AOD of a scattering natural aerosol located below (black) and above (red) the anthropogenic layer, on 21 June. DREs are computed in the shortwave spectrum at the top of the atmosphere, in  $\text{Wm}^{-2}$ , and given as 24h averages at a latitude of  $45^\circ\text{N}$ . AOD and SSA are given at  $0.55 \mu\text{m}$ .

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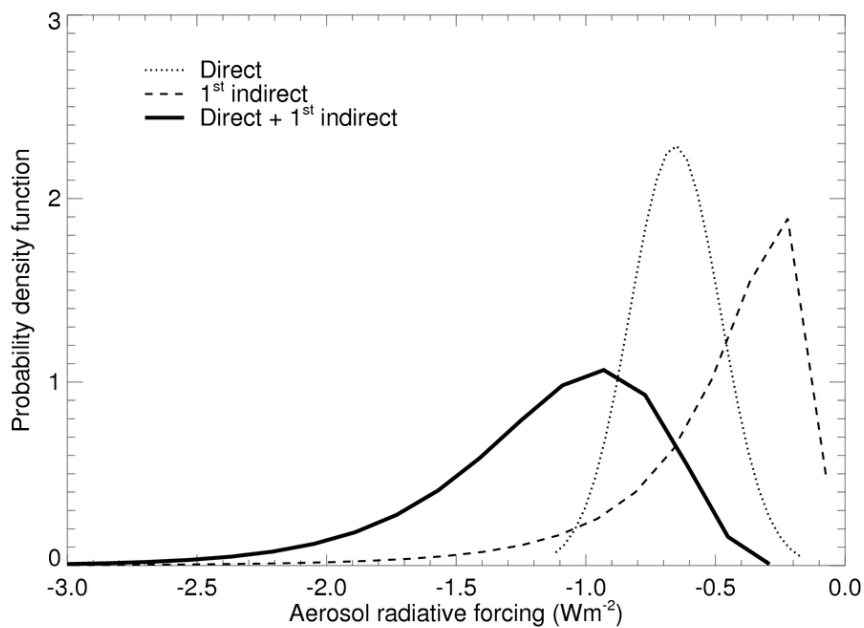


Fig. 1.

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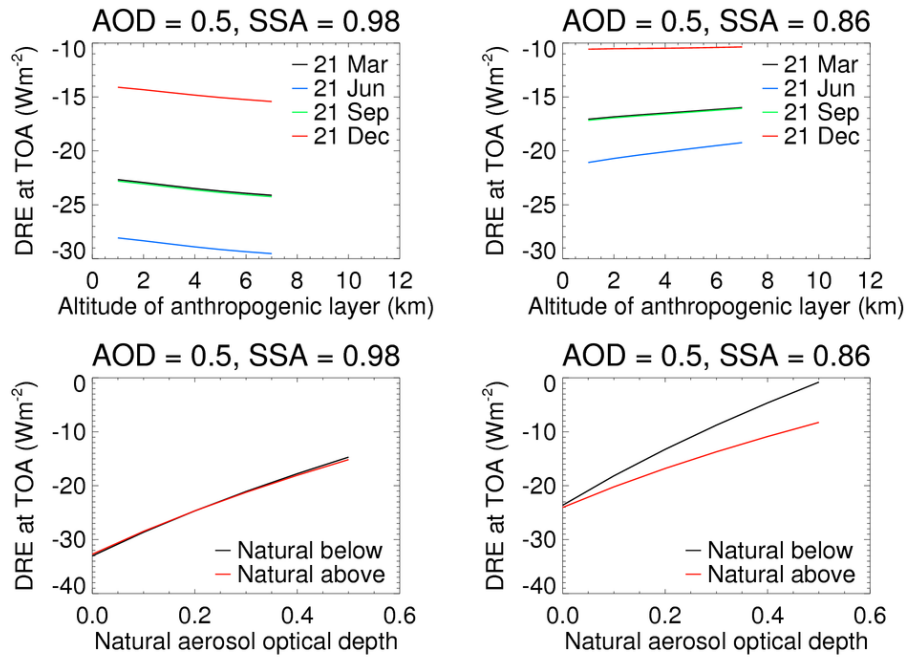


Fig. 2.