Interactive comment on "Modelled and observed changes in aerosols and surface solar radiation over Europe between 1960 and 2009" by S. T. Turnock et al.

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We would like to thank both of the anonymous reviewers for their helpful and constructive comments. Below we have responded to each comment in turn and made alterations to the manuscript where appropriate (shown enclosed in "*speech marks and italic font*"). The referee comments are shown first in grey shading and the author's response is shown below in normal font.

Response to Anonymous Referee #1

General Comments:

1. The study focuses mostly on mass properties of atmospheric aerosol, but tells little about aerosol size distribution representation in the model. Aerosol size distributions have large effects on optical properties and aerosol-cloud interactions. In order to make the study stronger , I highly recommend including a figure or table comparing present day aerosol size distribution properties over the different European locations (or a few representative locations) as presented by Asmi et al., (2012). Asmi et al., (2012) presents a comprehensive analysis of present day aerosol size distributions in several locations across Europe and the data is freely available. The model results could help explain differences between modeled and observed AOD and shed some light on changes in CCN over Europe in the recent past. For example a simple comparison of the model ability to capture particle concentrations in the ranges 30-50nm, >50nm and >100nm, would complement the existing comparison. If the model represents well present day size distributions, changes over time in those parameters would be of special value to the community.

We have evaluated simulated aerosol number concentrations at sites across Europe using present day observational data from Asmi et al. (2011). The evaluation has used observational data for particles larger than 30 nm (N30), 50 nm (N50) and 100 nm (N100) dry diameter. An additional figure (see below for new Figure 11) shows the comparison between modelled and observed annual mean particle number concentrations at N30, N50 and N100 dry diameters across sites over Europe for the period 2008-2009. This figure shows that the model underpredicts particle number concentrations across all sizes over Europe by factor of 1.5-2.0. The model substantially underestimates the low particle number concentrations across all size ranges at the high latitude arctic sites of Zeppelin and Pallas. The model also underestimates larger size particles more than smaller sized ones, as indicated by the larger NMBF shown in the figure. These results are in accordance with the findings of the multi model comparison of global aerosol microphysical models to size distribution observational data undertaken by Mann et al. (2014). An underestimation of the aerosol size distribution across Europe could possibly account for some of the underestimation in simulated AOD and may also contribute to some of the discrepancies in the simulated surface solar radiation. However, factors other than aerosols might contribute to discrepancies between the modelled and observed AOD and surface solar radiation (e.g. humidity, clouds), which would require further work to investigate. We have modified the original manuscript to include a section and a new figure on the evaluation of the modelled present day aerosol size distributions. In addition, we have also included an amended Figure 4 showing the simulated change in annual mean particle number concentration across the different size fractions for Europe. This shows a large reduction (>50%) in aerosol number concentration over Europe across all sizes from 1970 to 2009. Taking into account the underestimation in present day aerosol number concentrations the simulated changes in aerosol number could be even larger, with potentially large impacts on climate.

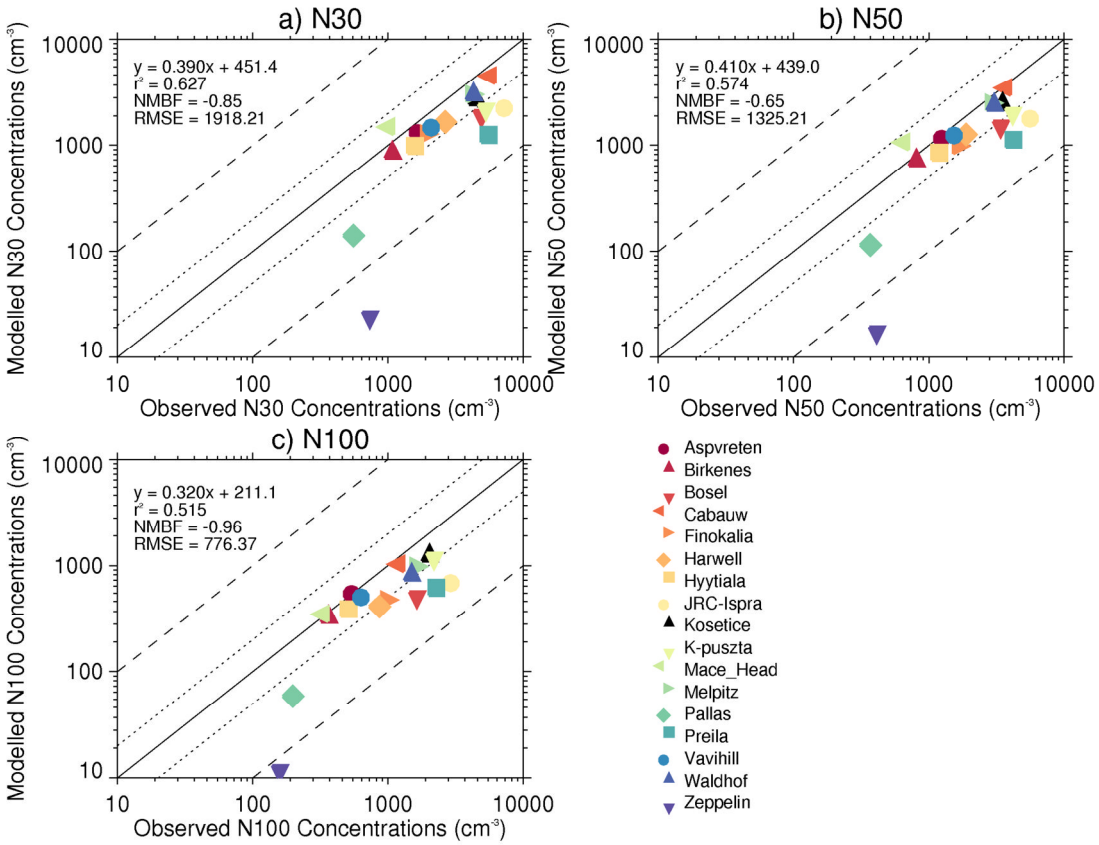

Figure 11. Annual mean observed vs modelled aerosol number concentrations for 2008-2009 over the size fractions a) N30, b) N50 and c) N100

The following sections of the original manuscript have been modified to include details of the evaluation of simulated aerosol size distributions.

An additional sentence on Page 13458, line 6 has been modified.
"Here we compare the HadGEM3-UKCA coupled chemistry-climate model for the period 1960 to 2009 against extensive ground based observations of sulfate aerosol mass (1978–2009), total suspended particle matter (SPM, 1978–1998), PM_{10} (1997–2009), aerosol optical depth (AOD, 2000–2009), aerosol size distributions (2008-2009) and surface solar radiation (SSR, 1960–2009) over Europe."

An additional sentence on Page 13458, line 13 has been included.
"The model also underestimates present day aerosol number concentrations at sizes larger than 30 nm (N30 NMBF = -0.85), 50 nm (N50 NMBF = -0.65) and 100 nm (N100 NMBF = -0.96)."

The sentence on Page 13463, line 2 has been modified.
"We evaluate the ability of the model to consistently capture observed changes in bulk in situ aerosol properties (PM, aerosol size distributions and chemical components) as well as radiative properties (AOD, SSR) over Europe."

The sentence on Page 13467, line 24 has been modified.
"Ground-based measurements of aerosols used in this study are listed in Table 1 and include aerosol mass concentrations (sulfate and total mass) from the EMEP network (http://www.emep.int), AOD from AERONET (http://aeronet.gsfc.nasa.gov/), aerosol size distributions from EUSAAR (European Supersites for Atmospheric Aerosol Research) and GUAN (German Ultrafine Aerosol Network) (Asmi et al., 2011) and SSR from the GEBA database (http://www.geba.ethz.ch/history/index)."

A new section has been added before section 2.2.2 on page 13468 to briefly mention the aerosol size distribution data.

"Section 2.2.2 Aerosol size distribution

Aerosol size distribution data from EUSAAR and GUAN ground based monitoring sites have previously been collected and processed by Asmi et al., (2011) over Europe for 2008-2009. Here we use the same 17 low altitude sites as in the multi model comparison by Mann et al., (2014) to compare modelled and observed aerosol number concentrations at three different size fractions (>30 nm (N30), >50 nm (N50) and >100 nm (N100) dry diameter) over the period 2008-2009."

Figure 4a has been revised to include an additional panel showing the simulated change in aerosol number concentrations for the total, N30, N50 and N100 size fractions over the period of 1960 to 2009. The paragraph starting on Page 13471, line 9 has been modified to the following.

"Figure 4b shows the change in simulated European surface aerosol number concentrations over the total, N30, N50 and N100 size fractions between 1960 and 2009. Aerosol number concentrations across all size fractions increase from 1960 to a peak in 1970 before declining to concentrations that are 50% lower in 2009.

Simulated European AOD (Fig. 4c) increases from 1960 to a peak in 1973 before decreasing till 2009 to an AOD that is lower than that simulated 1960. Figure 4d shows that simulated European SSR decreases from 1960 to 1980 ("dimming"), then increases until 2009 ("brightening")."

A new section has been inserted on Page 13476, line 1 before Section 3.2.3 Aerosol optical depth. Within this new section a new Figure 11 has also been included to show the model and observed aerosol number size distribution comparison (as shown above). The new section is as follows:

"Section 3.2.3 Aerosol size distribution

Figure 11a-c compares simulated and observed annual mean surface aerosol number concentrations averaged over the period 2008-2009 for particles of greater than 30 nm, 50 nm and 100nm in dry diameter. The model underestimates the observed aerosol number concentrations as all size fractions, with a slightly larger model bias in larger sized particles (N100 NMBF = -0.96, N50 NMBF = -0.65 and N30 NMBF = -0.85). The model substantially underestimates aerosol number concentrations over all size fractions at the high latitude Arctic monitoring locations of Pallas and Zeppelin. In addition, simulated aerosol number concentrations are underestimated to a larger extent at the more polluted, central European monitoring sites of Ispra, Preila, Bösel and K-Puszta.

The model is able to reproduce the observed aerosol number concentrations within a factor of 2 at the majority of European monitoring locations, which is in agreement with the recent intercomparison and evaluation of global aerosol microphysical models (Mann et al., 2014). In addition, Mann et al., (2014) also found that all of the evaluated models underestimated aerosol number concentrations in the Arctic.

This suggests that N50 concentrations (a proxy for CCN concentrations) are slightly underpredicted by the model in the present day but have declined across Europe (Fig 4b) by 50% since the 1970s, with a large potential impact on the European radiative balance. Although over the more recent period of 2001-2010 no discernible trend was found in aerosol number concentration by Asmi et al., (2013) across 3 central European monitoring sites."

A new sentence has been inserted on Page 13479, line 6.

"The model also underestimates aerosol number concentrations across the N30 (NMBF = -0.85), N50 (NMBF = -0.65) and N100 (NMBF = -0.96) size fractions in 2008-2009. Underestimation of surface sulfate, PM_{10} and aerosol number concentration is therefore not manifested in the simulation of AOD or SSR."

The sentence on Page 13481, line 21 has been amended to:

"We evaluated the model against European observations of sulfate aerosol mass, total suspended particulate matter (SPM), PM_{10} mass concentrations, aerosol number concentrations, aerosol optical depth (AOD) and surface solar radiation (SSR)."

A new sentence has been inserted on Page 13481, on line 27.

"In addition, the model underpredicts present day aerosol number concentrations over the size fractions of N30 (NMBF= -0.85), N50 (NMBF= -0.65) and N100 (NMBF= -0.96)."

Table 1 has also been modified to include the additional details as follows.

Table 1. Details of the ground based observations used in this study

Data Source	Measurements	Period	Total Number of Sites Available
EMEP	Total sulfate aerosol mass concentrations	1978-2009	97
EMEP	Total suspended particle mass concentrations	1978-2005	42
EMEP	PM_{10} Mass Concentrations	1996-2009	52
EUSAAR and GUAN	Aerosol number concentrations greater than 30 nm (N30), 50 nm (N50) and 100 nm (N100) in dry diameter	2008-2009	24
AERONET	Aerosol optical depth (AOD)	1994-2009	21
GEBA	Surface Solar Radiation (SSR)	1928-2009	50

2. Show, if possible, an additional plot similar to figure 12 of the seasonal SSR changes with time in addition to the annual values. It would be interesting to learn if the model captures the SSR seasonal cycle better than it does AOD and sulfate aerosol mass seasonal cycles, and the possible reasons why it does or does not.

We have calculated the seasonal mean statistics for SSR (see table below) and have amended a comment to the manuscript based on these results. The results show that as well as a small model bias in annual mean SSR (shown by small values of NMBF), the seasonal cycle in SSR is quite well reproduced by the model across all time periods (NMBF < 0.1). However, the model was previously shown to have larger seasonal biases in sulfate aerosol mass (DJF NMBF = -2.19 and JJA NMBF = 0.694), PM_{10} (DJF NMBF = -0.12 and JJA NMBF = -0.339) and AOD (DJF NMBF = 0.258 and JJA NMBF = -0.167). This suggests that like for annual means, seasonal model biases in aerosol properties are not manifested in the model simulation of the seasonal cycle of SSR. The seasonal cycle in SSR is probably more strongly determined by variations in factors other than aerosols e.g. clouds.

We have included the following with the manuscript to briefly mention the seasonal cycle in SSR. A new sentence on page 13477, line 8.

"An evaluation of seasonal mean modelled and observed SSR also showed small differences in the NMBF over each season for all time periods, although RMSE is larger and the spatial correlation is smaller in summer."

The sentence on page 13479, line 7 has been modified to:

"Underestimation of annual and seasonal mean surface sulfate and PM_{10} is therefore not manifested in the simulation of the annual mean and seasonal cycle of AOD or SSR."

Time Period	Season	Type	Absolute Change in SSR (W m^{-2})	S.D. of mean value	% change in SSR	Calculated linear trend (W m^{-2} yr^{-1})	-2 * S.E. trend	+ 2 * S.E. trend	r^2	NMBF	RMSE
		Observed	-0.79	2.40	-1	-0.011	-0.309	0.286	0.838	0.002	7.72
	Annual	Model + ARE	-1.67	1.89	-1	-0.088	-0.317	0.141			
		Model - ARE	-0.20	2.12	-0.2	0.069	-0.237	0.376	0.748	0.045	11.04
		Observed	-3.94	2.50	-10.6	-0.37	-0.601	-0.137	0.906	0.042	6.20
	DJF	Model + ARE	-2.60	2.51	-6.8	-0.20	-0.494	0.085			
		Model - ARE	-2.46	2.65	-6.4	-0.19	-0.500	0.124	0.847	0.108	7.45
		Observed	-0.73	4.76	-0.5	0.06	-0.527	0.650	0.552	-0.023	14.68
1960-74	MAM	Model + ARE	-1.53	4.06	-1.0	-0.03	-0.536	0.470			
		Model - ARE	-0.59	4.36	-0.4	0.06	-0.479	0.600	0.511	0.046	16.00
		Observed	2.40	5.94	1.2	0.39	-0.315	1.094	0.450	0.025	18.03
	JJA	Model + ARE	-3.32	4.02	-1.6	-0.13	-0.628	0.359			
		Model - ARE	0.48	4.73	0.2	0.26	-0.314	0.824	0.539	0.116	24.00
		Observed	-0.61	3.50	-0.8	-0.09	-0.522	0.339	0.906	-0.023	7.93
	SON	Model + ARE	1.50	2.59	2.1	0.11	-0.201	0.429			
		Model - ARE	1.77	2.97	2.4	0.15	-0.210	0.509	0.888	0.056	7.94
		Observed	-2.24	3.42	-2	-0.259	-0.658	0.140	0.803	0.029	8.43
	Annual	Model + ARE	3.12	3.06	3	0.304	-0.036	0.643			
		Model - ARE	1.89	3.50	1.6	0.187	-0.191	0.564	0.757	0.063	11.84
		Observed	-0.39	2.18	-1.1	-0.13	-0.393	0.127	0.905	0.099	6.92
	DJF	Model + ARE	1.06	1.87	2.8	0.03	-0.205	0.256			
		Model - ARE	0.79	1.96	2.1	0.00	-0.241	0.245	0.861	0.077	8.50
		Observed	-3.19	7.42	-2.1	-0.32	-1.221	0.584	0.485	0.000	14.00
1975-89	MAM	Model + ARE	2.87	5.95	1.9	0.30	-0.424	1.015			
		Model - ARE	1.87	6.06	1.2	0.22	-0.526	0.958	0.540	0.036	18.63
		Observed	-7.08	10.60	-3.5	-0.69	-1.947	0.569	0.450	0.046	20.03
	JJA	Model + ARE	5.06	9.70	2.5	0.56	-0.602	1.722			
		Model - ARE	2.01	10.73	-0.1	0.28	-1.041	1.603	0.637	0.102	30.40
		Observed	1.71	3.31	9.5	0.10	-0.302	0.511	0.879	0.016	7.87
	SON	Model + ARE	3.79	3.01	5.2	0.35	0.026	0.666			
		Model - ARE	2.90	3.02	2.6	0.25	-0.102	0.596	0.889	0.038	9.76
		Observed	5.78	3.47	5	0.369	0.153	0.584	0.896	0.043	8.00
	Annual	Model + ARE	3.98	3.04	3	0.316	0.125	0.507			
		Model - ARE	0.27	2.92	0.2	0.09	-0.185	0.366	0.828	0.053	10.23
		Observed	1.19	1.66	3.5	0.09	-0.037	0.214	0.930	0.078	5.91
	DJF	Model + ARE	0.27	2.01	0.7	0.02	-0.135	0.184			
		Model - ARE	-0.82	2.07	-2.2	-0.05	-0.211	0.116	0.890	0.030	7.31
		Observed	9.29	6.79	6.1	0.64	0.196	1.092	0.672	0.009	12.22
1990-09	MAM	Model + ARE	4.01	4.00	2.6	0.29	0.006	0.581			
		Model - ARE	0.25	4.29	0.2	0.06	-0.281	0.400	0.633	0.023	16.03
		Observed	6.07	7.70	3.0	0.36	-0.230	0.949	0.613	0.075	21.59
	JJA	Model + ARE	5.90	6.85	2.7	0.56	0.083	1.038			
		Model - ARE	-0.29	7.18	-0.1	0.18	-0.384	0.746	0.671	0.101	29.40
		Observed	6.58	4.58	9.5	0.38	0.066	0.701	0.920	0.034	6.74
	SON	Model + ARE	3.81	4.01	5.2	0.29	0.001	0.579			
		Model - ARE	1.92	4.16	2.6	0.17	-0.152	0.492	0.880	0.031	9.24

3. Please mention in the text the mean global radiative forcing induced by changes in European aerosol emissions between periods with largest and smallest aerosol loadings. This value makes a direct comparison to GHG global forcing direct, and tells how important the regional aerosol perturbation is.

In our simulations, global emissions were changed. Without running further simulations it is not possible to obtain the global mean radiative forcing changes solely induced by European aerosol precursor emissions. We would need to run additional simulations over 50 years whereby the emissions from Europe are fixed to be able to isolate their effect on the globe, which was considered outside the scope of work for this paper. However, changes in global aerosol radiative forcing (RF) due to changes in global emissions can be calculated from our existing simulations. We have added a new Table to the manuscript (Table 6, see below) where we compare changes in global mean RF with European mean RF that we already reported. This table highlights the importance of regional changes in aerosol radiative forcing when compared to global changes and also the magnitude of changes compared to greenhouse gas forcing between 1960 and 2010. In addition, if only a global view is taken then important regional changes with opposite signs can be masked.

We have also added the following comment to the manuscript that relates to the Table below.

The sentence on Page 13480, line 1 has been changed to the following.

"Figure 15 shows the changes in simulated European mean top of atmosphere (TOA) outgoing shortwave radiation, relative to a 1980 to 2000 mean, under all-sky (a) and clear-sky conditions (b) with the numbers presented in Table 6."

An additional sentence has been included on page 13481, line 9.

"Table 6 shows that the change in European mean aerosol RF is much larger than the change in global mean aerosol RF of +0.4 W m^{-2}. Shindell et al. (2013) reported a multi-model evaluation of aerosol RF over the period 1850 to 2100, with 6 of the 9 models reporting a positive increase in global mean aerosol RF between 1980 and 2000, qualitatively similar to the results presented here. At the global scale, our simulated change in all-sky aerosol RF between 1970 and 2009 is ~40% of the magnitude of change in global CO_2 radiative forcing over the same period. At the European scale, the simulated change in all-sky aerosol RF is more than three times the change in global mean CO_2 RF. This indicates the large regional impact that decreasing aerosol concentrations have had on the radiative balance and climate over Europe compared to other forcing agents."

Table 6. Global and European shortwave top of atmosphere all-sky and clear-sky aerosol radiative forcing, relative to a 1980 to 2000 mean. Values for 1972 are included as this is when the minimum aerosol radiative effect occurs over Europe. For comparison the global carbon dioxide radiative forcing (relative to 1750) from the IPCC fifth assessment report is shown in the last column (Myhre et al., 2013)

Year	All-Sky Aerosol RF (W m^{-2})		Clear-sky Aerosol RF (W m^{-2})		Estimates of CO_2 radiative forcing (Myhre et al., 2013)
	Global	Europe	Global	Europe	Global
1960	+ 0.9	+ 0.8	+ 0.4	+ 0.2	+ 0.7
1970	- 0.1	- 1.0	+ 0.01	- 0.1	+ 0.9
1972	- 0.1	- 1.4	+ 0.2	- 1.6	N/A
1980	- 0.4	- 0.6	+ 0.01	- 0.5	+ 1.1
1990	+ 0.01	+ 0.1	+ 0.04	- 0.1	+ 1.3
2000	+ 0.4	+ 1.7	- 0.1	+ 1.1	+ 1.5
2009	+ 0.3	+ 2.1	+ 0.1	+ 1.9	+ 1.8

Specific Comments:

Page 3(13459), line 26. How much have NOx, CO and BC emissions decreased between 1980 and 2010?

We have changed line 26 on Page 3(13459) to include the percentage change in NO_X, CO and BC emissions between 1980 and 2010 and it reads as follow:

“Anthropogenic emissions of oxides of nitrogen (NOx), carbon monoxide (CO) and black carbon (BC) have also decreased over Europe between 1980 and 2010 by 30%, 58% and 55% respectively (Granier et al., 2011).”

Page 13(13469), line 19. How was the interpolation made? Please add a short sentence explaining how this was done.

We have changed line 19 on Page 13(13469) to mention that the values from the four nearest model squares were used to linearly interpolate to the actual measurement location.

“Model values were linearly interpolated to each measurement site using the relative contribution from the four closest surrounding model grid squares.”

Response to Anonymous Referee #2

General Comments:

1. Discussion of the model evaluation is good, but could be presented more clearly. Comparison of modeled and observed 1978-2009 averages is a challenging task. Potentially significant details are lost in such a long-term average contains significant emission decreases. For example, according Figure 5, summer sulfate is underestimated early in the record is overestimated (quite significantly) before coming into closer agreement near the end of the record. Such details are likely lost in Figure 6, which very clearly depicts spatial biases, but how have those biases changed throughout the time period? This is important when assessing trends and impacts on SSR.

We use a combination of Figure 5 and 6 to show both spatial and temporal variability and trends in simulated and observed sulfate concentrations. Figure 5 shows the spatial distribution of annual mean sulfate concentrations in 1980, 1990, 2000 and 2009 as well temporal trends in annual mean, summertime and wintertime concentrations. Figure 6 shows summertime and wintertime NMBF at the European scale and also by region, with the range in NMBF reflecting variability in model bias over time. These show that wintertime sulfate is under estimated across all regions and for all of the period 1978 to 2009. Summertime sulfate is overestimated early in the record before coming into closer agreement towards the end of record, as indicated by the referee. Figure 6 shows that summertime concentrations are mostly underestimated in Northern and Southern Europe, but overestimated in central and eastern Europe. Combining Figure 5 and 6 together indicates that central European sites could be leading to the model overestimating summertime sulfate concentrations early in the record. The simulated trend in sulfate compared to that observed is dominated by discrepancies at these locations within different regions of Europe and over different time periods. In order to examine the reasons behind the changes in biases across the period 1978-2009 would require a more detailed investigation of simulated and observed summertime sulfate concentrations across monitoring sites within each region, which is outside the scope of the current work for this paper.

We have amended the lines 1-5 on Page 13472, line 1 to the following:

"*In summer,* sulfate is *underestimated at the start of the period before coming into closer agreement towards the end of the period (Figs., 5, 6). The model underestimates observed summertime sulfate across northern and southern Europe (NMBF between 0 and -1), and overestimates sulfate in central and eastern Europe (NMBF <1). The model consistently underestimates wintertime sulfate (NMBF of -1 to -6) across all the European regions, with the largest discrepancy occurring in northern Europe."*

2. The importance of seasonality needs to be further discussed, including in SSR. Differing errors in the sulfate simulation exist summer and winter. This is important when considering differing insolation and hygroscopic growth that impact AOD and forcing.

See response to REVIEWER 1, general comment number 2 on this point.

3. The potential role of nitrate needs to be further highlighted. Nitrate likely plays an important role, particularly in winter, and might be able to resolve some of the issues mentioned in the manuscript, but also may exacerbate other problems in simulating AOD/SSR.

Nitrate is an important aerosol component particularly in northern Europe over winter and in more recent years as the concentration of SO_2 and sulfate aerosol has reduced. Future versions of HadGEM3-UKCA will include nitrate aerosol but this version currently does not and therefore we are unable to include its effects. However, due to its seasonal and temporal variability nitrate is only anticipated to make a difference to more recent comparisons over northern and western Europe in wintertime and could aid in the reduction of model observational bias here.

In Section 3.2.2 on page 13475, lines 4-12 we state that the contribution from nitrate aerosol has been shown to be about 1-3 μg m^{-3} over Europe and could therefore account for some of the discrepancy in the comparison of the simulated aerosol mass to that observed (Fagerli and Aas, 2008; Bellouin et al., 2011; Pozzer et al., 2012). It is anticipated to have a much smaller contribution in 1980s due to the preference of ammonium sulfate aerosol formation over ammonium nitrate.

Therefore the inclusion of nitrate within the model would not aid in reducing the model bias in aerosol mass over this period. Additionally the inclusion of nitrate aerosol could further exacerbate the model evaluation with AOD in winter and also potentially SSR (although as discussed above other factors e.g. clouds are likely to be dominant). This would likely occur in more recent years and over more northern and western areas of Europe where nitrate is likely to be a dominant component whereas, over southern Europe the model evaluation is less likely to be affected as sulfate remains the dominant aerosol component (Aan de burgh et al., 2011).

We have added additional statements on the seasonal, spatial and temporal importance of nitrate at the following points.

The following sentence has been inserted on Page 13475, line 6.

"This could be particularly important in reducing the model bias in winter and over northern and north west Europe where nitrate concentrations are anticipated to be largest."

The following sentence has been inserted on Page 13475, line 12.

"Inclusion of nitrate aerosol would reduce the model bias in PM_{10} concentrations over the more recent years but have a smaller fractional impact in the 1980s-1990s due to the dominance of sulfate concentrations."

4. In general, the figures in this manuscript are well done. However, many are tightly cramped into the ACPD structure. Please ensure that the figures are of adequate size to best display their fine detail.

We will endeavour to ensure that the figures are of adequate size and clarity during the type setting process for ACP.

Specific Comments:

Page 13464, Lines 12-13 – The meteorology is as close as the model can simulate, but not necessarily a match. This should be clarified.

We have changed the sentence on page 13464, lines 12-13 to read as follows:

"By nudging to re-analysis data, we ensure that the model produces a realistic representation of the meteorological conditions under which aerosol observations were taken."

Page 13456, Lines 6-7 – This is potentially an important omission, particularly in northwestern continental Europe. Later in the manuscript the implications of this omission should be detailed.

See response to general comment 3.

Page 13466, Lines 1-10 – This is the standard procedure, but in this application it does not allow for segregation of the direct and first indirect effect.

As the reviewer has pointed out the double-call radiation configuration for all-sky and clear-sky effects does not allow for a direct separation of the direct and 1st indirect aerosol radiative effects. However, the radiative effects calculated under clear-skies are considered to represent the contribution from the direct aerosol radiative effect and thus an indication of the 1st indirect effect can be obtained by subtracting the radiative effects calculated under all-skies from those under clear-skies. From this calculation the change in cloud albedo effect over the period 1973-2009 has been stated on page 13480, line 20-24. Therefore we have included the following text within the methods section on page 13466, line 10:

"An estimate of the cloud albedo effect (first aerosol indirect) can be obtained by subtracting the radiative effect obtained under all-sky conditions from those under clear skies."

Page 13466, Line 14 – “Time-averaged sporadic” is an oxymoron. Can this be rephrased? Shouldn’t sporadic eruptions be allowed to occur at the appropriate place/time in the model to best reconstruct their impact?

The inventory of SO_2 emissions for explosively erupting volcanoes from the inventories by Andres and Kasgnoc (1998) and Halmer et al. (2002) does provide emissions from volcanoes at the appropriate location and date of occurrence. The sentence on page 13466, line 14 has been reworded to the following to improve clarity:

“Emissions from tropospheric volcanoes (both continuous and explosively erupting) are included from Andres and Kasgnoc (1998) and Halmer et al. (2002) using the AEROCOM recommendations.”

Page 13466, Lines 20-21 – Are the biogenic emissions not dependent on local meteorological conditions? Is this an acceptable approximation when looking at monthly mean values?

The inventory of Guenther et al. (1995) is used to provide monthly mean monoterpene emission fluxes here. This inventory has been derived based on a global model that simulates the flux of monoterpenes from natural sources, taking into factors such as light and temperature dependence. Therefore the fluxes used from this inventory are generated with local meteorological conditions taken into consideration.

Page 13467, Lines 13-15 – It is unclear what is meant here. Are ammonia emissions entirely omitted or just fixed to a constant value? This is an important detail.

Ammonia emissions are included within the model from the MACCity emissions inventory. Ammonia is used within the chemistry scheme and is available as an advected tracer for the aerosol scheme. However, due to the absence of ammonium nitrate within the aerosol scheme ammonia is not utilised by the aerosol scheme within HadGEM3-UKCA and it is assumed that sulfate aerosol is of the form ammonium sulfate.

For clarity we have amended page 13467 lines 13-15 to the following.

“The emissions of NH_3 across Europe (not used as part of the aerosol scheme in this study due to the absence of ammonium nitrate) increased continuously over the period 1960–2009, driven largely by the agriculture sector.”

Page 13469, Line 19 – How was the model data interpolated to each measurement site? A description of the method, even if just by name, or a reference is necessary since many model-observation studies simply compare the box average to measurements.

See response to reviewer 1, specific comment 2 for details on this point.

Page 13471, Line 15 – Are the values plotted in Figure 5 5-year averages or a single year?

The values in the contour plot on Figure 5 are single year annual mean concentrations for 1980, 1990, 2000 and 2009. We have slightly amended line 15 on page 13471 to reflect this as follows:

“Figure 5a–d compares simulated single year annual mean sulfate concentrations over Europe for 1980, 1990, 2000 and 2009 against observations.”

Page 13471, Line 24 – “slightly” should be omitted.

Deleted as requested.

Page 13472, Line 13-16 – Is wet deposition data available? Can it be added to the suite of observations used to validate the model?

Sulfate wet deposition data is available over Europe from EMEP but has not been processed and used as part of the current study.

Page 13476 – Is the discussion SSR clear-sky or all-sky? Is data available to evaluate COD?

The discussion is based on all-sky simulated and observed surface solar radiation data. Line 23, page 13476 has been amended below to reflect this:

"Figure 12 shows simulated and observed annual mean all-sky SSR anomalies across Europe between 1960 and 2009, relative to a 1980–2000 mean."

The model evaluation of cloud optical depth was considered outside the scope of this paper and its feasibility has not been investigated further.

Page 134678, Lines 3-12 – Can this be used as a critique of the emission inventory?

The uncertainties in emissions inventory will certainty contribute to the errors in the modelled representation of SSR over the period 1960-1974. However, there are other additional factors e.g. clouds, humidity, observational errors contributing to the SSR over this period. Therefore it would be difficult to say with any confidence (and additional observations) that the uncertainties in the emissions inventories are causing the errors in the SSR.

Page 13481, Line 1-2 – The percentage change is not what matters, but the absolute magnitude.

The sentence on page 13481, line 1-2 has been amended to include the absolute change in sulfate concentrations from the two studies. This shows that there is still a larger decrease in sulfate concentrations over Europe than the USA, which reflects the difference in the changes in aerosol radiative effects between the two regions.

"The smaller changes in aerosol RF reported by Leibensperger et al. (2012) are possibly related to the smaller reductions in sulfate aerosol mass concentrations observed over the USA of ~0.8 μg S m^{-3} (40 %), when compared to that observed over Europe of 1.2 μg S m^{-3} (70 %)."

Page 13501, Figure 3 caption – It is unclear what is meant by "within each particular network." I think that phrase can be removed.

Agreed and has been removed as requested.

Page 13503, Figure 5 – The legend is not clear, particularly the observation (solid) and model (dotted) lines beneath the annual, DJF, and JJA lines. The S.D. estimates could be shown as colored rectangles rather than lines.

The legends on Figure 5 and Figure 8 have been amended to reflect the above comments.

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