Response to reviewers 1 and 2

We agree with both reviewers that there was a need for added information on the emission inventories described in this paper. We are therefore planning to included in the revised document the following tables

Table 1. Primary source of information for the various regional inventories used in the definition of the 2000 dataset.

Region	Ozone prec.	SO_2	OC/BC	NH_3
China	Cofala et al., 2007	Smith et al., 2010	Bond et al., 2007	EDGAR-v4
Europe	EMEP	UNFCCC	Bond et al., 2007	EDGAR-v4
United States	EPA	EPA	Bond et al., 2007	EDGAR-v4
Japan, Australia, NZ	UNFCCC	UNFCCC	Bond et al., 2007	EDGAR-v4
Canada	Env. Canada	Env. Canada	Bond et al., 2007	EDGAR-v4
Latin America	EDGAR-v4	Smith et al., 2010	Bond et al., 2007	EDGAR-v4
South America	EDGAR-v4	Smith et al., 2010	Bond et al., 2007	EDGAR-v4
Other regions	EDGAR-v4	Smith et al., 2010	Bond et al., 2007	EDGAR-v4

Smith et al. (2010) is now submitted to ACPD.

As the simulated CO 2000 distribution shows a negative bias in both models, we are also planning to include the following table to identify if there is a specific bias in the emissions.

Table 2. Regional and global estimate of year 2000 CO anthropogenic and biomass burning emissions (Tg(CO/year)).

Anthro.	EDGAR-FT2000	RETRO	GAINS	EPA-2006	EMEP-2004	TRACE-P	This work
Global	548	476	542	N/A	N/A	N/A	611
US	74	56	75	102	N/A	N/A	93
W. Europe	30	19	38	N/A	31	N/A	31
China	98	95	128	N/A	N/A	100	121

Bio. burn.	GFED-v2	GICC	This work
Global	427	467	459

References:

- EDGAR-FT2000: http://www.mnp.nl/edgar/model/v32ft2000edgar/docv32ft2000/
- RETRO: http://retro.enes.org/pub_reports.shtml
- GAINS: Cofala et al., 2007

- EPA-2006: http://www.epa.gov/airtrends/2006/emissions_summary_2005.html
- EMEP-2004: Vestreng et al., Technical Report NSC-W 1/2004.
- TRACE-P: Streets et al., 2003.
- GFED-v2: Randerson et al., 2007.
- GICC: Mieville et al., 2010.

This comparison for CO emissions thus shows that our anthropogenic regional estimates are in good agreement with reference inventories, with no clear indication of under-estimation in the global or regional amounts, whether anthropogenic or biomass burning emissions. In addition, information on the regional trend of emissions over the US is available from EPA so we are using this information as a point of comparison.

Table 3. Recent trend in US emissions (Tg(species)/year; NO_x expressed as NO_2).

1970	CO	NO_x	Total VOCs	SO_2
EDGAR-HYDE	84.2	16.6	19.4	N/A
RETRO	115.5	19.1	*	N/A
EPA-2003	N/A	N/A	N/A	N/A
EPA-2006	197.3	26.9	33.7	31.2
This work	79.9	16.5	25.8	27.0

1980	CO	NO_x	Total VOCs	SO_2
EDGAR-HYDE	90.2	19.0	22.4	N/A
RETRO	109.3	20.3	*	N/A
EPA-2003	105.7	22.0	23.7	23.3
EPA-2006	177.8	27.1	30.1	25.9
This work	118.8	19.8	25.4	22.2

1990	CO	NO_x	Total VOCs	SO_2
EDGAR-HYDE	95.1	21.8	24.2	N/A
RETRO	96.4	19.1	*	N/A
EPA-2003	89.2	21.8	18.9	21.3
EPA-2006	143.6	25.2	23.1	23.1
This work	112.1	20.6	23.8	19.0

2000	CO	NO_x	Total VOCs	SO_2
EDGAR-HYDE	N/A	N/A	N/A	N/A
RETRO	55.7	18.5	*	N/A
EPA-2003	98.4	22.4	18.3	16.2
EPA-2006	102.4	22.3	16.9	16.3
This work	93.0	19.6	15.2	14.8

References

- EPA-2003: http://www.epa.gov/oar/aqtrnd03/appenda.pdf
- EDGAR-HYDE: van Aardenne et al. (2001)

We would like to thank reviewer #1 for bringing up the point of comparing our emissions to the EPA values (for the CO transportation sector emissions). While we

would like to use the EPA estimates at face value, it is important to recognize 1) that those estimates vary a lot between revisions (EPA-2003 vs EPA-2006) and 2) that independent estimates (Parrish et al., 2006) are pointing towards very different trends in transportation than EPA presents. In particular, the Parrish et al. emission estimates present a much-reduced transportation sector in 1990 than the EPA-2006 estimates, especially for CO. We therefore believe that the reviewer statement on the low emission estimate for the US transportation CO emissions is not a clear indication of deficiencies in our dataset, as there is clear variability in those values amongst published estimates.

We will also include the following tables for the global amounts of emission, anthropogenic and biomass burning for each species considered in this study

Anthro.	CO	NO_x	VOC	BC	OC	NH_3	SO_2
1850	63.04	1.24	10.16	1.05	4.63	6.55	2.02
1860	67.26	1.59	10.46	1.25	5.27	7.34	2.95
1870	76.60	2.23	12.15	1.49	5.83	7.40	4.62
1880	85.99	2.88	13.85	1.71	6.20	7.47	7.67
1890	95.66	3.55	15.62	1.99	6.51	7.54	12.61
1900	111.11	4.60	18.48	2.31	6.82	8.62	19.82
1910	132.79	6.24	20.99	2.79	7.50	9.20	30.09
1920	153.03	7.42	23.46	2.98	7.76	10.85	33.18
1930	182.76	9.18	27.41	2.81	8.07	12.20	41.29
1940	206.59	10.81	31.17	2.86	8.69	12.88	49.96
1950	277.91	17.24	43.35	2.91	8.78	16.97	56.96
1960	376.54	25.40	70.36	3.22	9.78	20.82	87.31
1970	474.43	36.49	101.16	3.34	10.25	26.48	117.65
1980	583.75	51.84	126.75	4.51	10.98	35.18	120.33
1990	626.76	59.24	137.50	4.81	11.91	42.61	116.11
2000	608.28	56.77	129.53	5.02	12.56	37.46	92.71

Bio. burn.	CO	NO_x	VOC	BC	OC	NH_3	SO_2
1850	322.55	10.36	51.81	2.03	17.99	6.14	2.45
1860	322.55	10.36	51.81	2.03	17.99	6.14	2.45
1870	322.55	10.36	51.81	2.03	17.99	6.14	2.45
1880	322.55	10.36	51.81	2.03	17.99	6.14	2.45
1890	322.55	10.36	51.81	2.03	17.99	6.14	2.45
1900	322.44	10.36	51.81	2.03	17.99	5.80	2.44
1910	315.18	10.04	50.78	1.97	17.60	5.85	2.43
1920	277.97	9.05	44.52	1.78	15.14	4.84	2.06
1930	276.30	9.07	44.15	1.79	14.94	4.68	2.01
1940	267.19	8.86	42.57	1.75	14.25	4.37	1.90
1950	260.79	8.74	41.43	1.74	13.70	4.12	1.82
1960	286.46	8.12	47.54	1.81	14.57	4.81	2.03
1970	333.81	9.40	55.51	2.10	16.86	5.68	2.37
1980	383.22	10.28	64.54	2.31	19.13	7.53	2.93
1990	470.86	12.20	80.00	2.75	23.31	10.20	3.79
2000	459.11	11.70	78.28	2.61	23.25	10.51	3.84

Finally, we have prepared the PDF files for all anthropogenic emissions generated in this exercise (we have included the file for CO in this response). This corresponds to a total of 112 pages (16 decades * 7 species) so it is not clear if this should be added as an appendix to the paper or make it available through the distribution web site. Feedback from the reviewers is appreciated.

Response to reviewer 1

In addition to the additions presented above (and the typos we will correct), we respond here to the specific points raised by Reviewer 1, as listed in the review

Page 4967, lines 24-26, are an unacceptable dismissal of the value of seasonal emission estimates. (Doesn't line 5 on the same page say that the target of the work is "to provide monthly emissions. . ."? Confusing.) Some of these species have large monthly variations in their emissions (BC, CO for sure), and these need to be taken into account if any comparison is to be done with observations; they will add to the seasonal variations in observations that result from transport variability. "no enough information of past emissions available" is not good English.

We will rephrase the discussion on the seasonal cycle. No seasonal cycle is indeed provided for the anthropogenic land emissions; however there is a seasonal cycle in biomass burning (a major driver in the seasonal and interannual variability), ship and aircraft emissions. We will make that point clear in the paper. In addition, we will make clear that this dataset has not been developed for use in regional AQ models where seasonal patterns for air pollutants matter probably more than for global climate models. And, indeed, we shall make it clear that this paper does not provide any new insights on seasonal distribution of emissions from most sources.

The Introduction does not mention the approach to gridding that was taken.

The gridding was performed following the grids defined in EDGAR-v4 (as mentioned on page 4970, from line 5). We will include that discussion in the Introduction as well.

Page 4968, lines 6-10, why is Bond et al 2007 not mentioned here or Junker and Li-ousse? There are more historical emissions papers than are mentioned here. Perhaps this par t is just not well written. What is the meaning of ". . ., or at least." (line 10)?

Those papers were not listed as the discussion was centered on ozone precursors. We will list the papers mentioned by the reviewer along with the existing list.

Page 4968, line 19, should be "East Asia" and possibly "South Asia", but not "South-east Asia". I think a rather poor case is made for choosing the base year of 2000, rather than 2005, say. There are ample emissions data in all par ts of the world now to do 2005. . . The point is that emissions changed A LOT between 2000 and 2005.

We will use South Asia. The point for using 2000 instead of 2005 is mostly due to timing issues related to the IPCC process. By the time we started this effort (September 2007), there was not enough information readily available for everything we needed; this evaluation was shared by all involved in this effort. We therefore decided, as indicated in the manuscript, to go for the year for which we had reliable data, i.e. year 2000. We agree with reviewer 1 that, as of today, we could and should indeed use 2005. At the time when the 2000 estimates were

needed by the Integrated Assessment Models for their projections (i.e. early 2008), this was simply not possible.

I think the argument made on Page 4969, lines 14-24, is flat out wrong, and it would have made a big difference if some of these other inventories had been included or at least compared. There should be no inference that these other inventories are somehow inadequate. In fact, a number of them are arguably better than what was chosen.

As discussed in the joint response, we will include in the revised version a comparison with available estimates of regional estimates (for year 2000). The inadequacy, as discussed in the manuscript, is not an indication of the validity or invalidity of the specific dataset; it is solely an expression of some of our specific needs. In addition, while indeed we did not use directly a number of specific papers presenting national or small region inventories many of them were included in the discussion of studies we relied on, e.g., REAS, Smith et al. (2010). Furthermore, some of the papers that appeared very recently were not available at the time when this emission set was developed for RCP.

Section 1.3, it is not clear to me if the methodology accounts for changing emission factors over the time period. Undoubtedly emission factors have changed radically for almost all source types, and are uniformly lower today than they once were. It seems to me that unless such changes were included in the original work, they are not included in this compendium. Yet, on page 4971, line 22-23, the text says changes in emission factors are captured. If so, how is this done? For all species? Can some examples be provided of emission factors assumed for key source types during the historical period?

It does implicitly; as described in the paper, the procedure we use for ozone precursors prior to 2000 is to use RETRO and EDGAR-HYDE as indicators of the rate of change of the emissions. We then apply this rate of change to the 2000 values (this is done for each sector and region separately). Therefore, implicitly, there is time evolution of both fuel use and emission factor, although we don't have explicitly used this information. This is what the discussion on page 4972 is describing, and it is unclear to the authors if that specific point was not well discussed in the original paper or if additional information was requested by the reviewer.

Page 4976, line 13, "(see Sect. 2)" cannot be correct, as this statement occurs in Section 2 itself. There is ver y little discussion of biofuel use, which is hugely important in the period 1850-1920 or so. How was biofuel use over time quantified and its emissions calculated? We need to see more data. . .

There was a problem in the edited document (conversion from Word to Tex) for which the numbering of the sections was reset wrongly by ACPD. Sorry about that. That explains the mismatch identified by the reviewer. In terms of biofuel, EDGAR-v4 has now included that emission in the domestic sector. Consequently, agricultural waste burning is specifically for the amount of biomass burned in a field

and is using EDGAR-v4 for 1970-2000 and the EDGAR-HYDE historical trend before that. This description will be included in the revised paper. Comparison with Yevich and Logan (2002) shows a good agreement on the 2000 global total.

Page 4987, lines 23 onward. The dismissal of uncertainty in this cursory way is disturbing.There have been many attempts to quantify uncertainty even in the studies that are used here and it is a shame to see the topic given such limited attention. Many current estimates are much better than a factor of two and some are much worse. How do the authors think the uncertainty increases as we go back in time? So many unanswered questions! By the way, Smith et al. 2010, is unpublished and not a good citation here.

Smith et al. is now submitted to ACPD and will therefore be available for viewing and comments while our paper is being finalized. In terms of uncertainty, within the approach used in this study, there is no clear way to perform an in-depth analysis of uncertainty (as T. Bond has performed). We will include a discussion based on the regional and global comparisons with other inventories to provide a sense of uncertainty. This section will be rewritten and included earlier in the text.

Table 1. Can we identify the years of these cited studies (e.g., Bond et al, 2007)?

This will be done in the next version of the paper.

Table 2. I find the term "Asia-Stan" objectionable.

This follows the IMAGE naming (see http://www.pbl.nl/en/themasites/image/background/regions/index.html); however, we will change that to "Kazachstan region", as in their figure legend.

Table 3. Natural emissions are included? I am surprised, as I don't recall seeing any mention of natural emissions in the text.

No they are not; this is a placeholder in our work tables. We will remove it from the table in the paper.

Fig 1 is confusing to me. First, are there no EDGAR data after 1980 on the left panel? Why is that? Second, the green EDGAR line appears to stretch to 1990 on the right panel. Is that correct? Third, is the point on the right graph supposed to be the U.S. NEI value? This definitely should be explained on the graph. Fourth, avoid use of the term "actual" emissions. None of these values are actual emissions!
Absolute values, perhaps. I don't feel very comfortable with this illustration. EDGAR and RETRO are quite dissimilar, especially in the recent past. They appear to be trending in radically opposite directions during 1970-2000. RETRO in particular seems to be extremely different in 2000 to what I assume is the NEI value and the starting point for the historical reconstruction. Perhaps some time should have been taken to probe the reasons for the differences in the datasets used. The more I look at this figure, the more

uncomfortable I become. If you check EPA's emission trends reports for the transportation sector, you will find that CO emissions in 2000 are similar to what is shown on this graph, about 83 Tg. But the corresponding EPA value for 1980 (as far back as my version goes) is 146 Tg, far above any of the values in the right hand panel. Ouch.

What is shown is the rate of increase; in this case, the 1980 value is the rate of increase from 1980 to 1990. This left-panel figure seems to generate more confusion than explanation, so we are proposing to remove it. We will change "actual" to "absolute", as this was indeed the intended meaning. With respect to the EPA 1980 value for the transportation sector, we will include the table (listed in the joint response as Table 3) and discussion presented in the joint response section.

We think it is essential to keep the paper as a single entity since, in the end, it is the use of such inventories in chemistry models that will indicate their usefulness and validity. The comparison of the model results with observations can therefore identify specific issues; for example the low bias in CO around 2000 (while our CO emissions at the global scale are quite realistic), or the too-high ozone in the 1970s (which seems to indicate that our anthropogenic emissions are probably higher than in reality, at least in the Northern hemisphere).

Response to reviewer 2

We would like to thank W. Collins for his constructive comments and will respond to his comments as listed in his review.

With respect to the CMIP5 ozone dataset, there will be a specific paper (to be written by I. Cionni et al., see up-to-date information at http://www.pa.op.dlr.de/CCMVal/AC&CSPARC_O3Database_CMIP5.html), but the simulations described here are indeed the source of the tropospheric ozone portion of the CMIP5 dataset. Additional evaluation will be provided in the Cionni et al. paper.

It is unclear why the models are not capable of reproducing ozone trends (see below); and we believe there is a need for a concerted effort amongst the global chemistry community to tackle this issue and identify if this is an emission issue, a data issue or a modeling issue. This is a research topic however, well beyond the scope of this paper: a workshop on tropospheric ozone trends had been organized in Boulder in October 2009 (see published summary in EOS-March 2010) and this discussion will have to continue in the wider community.

Section 1: I would suggest starting this section with an introduction as to what gases and aerosols are needed by climate model, and which will be provided in this study.

We will include such discussion in the paper.

Page 4966, line 6: It needs to be made clearer that 1850 is not pre-industrial.

We will make it clear that 1850 is indeed not preindustrial.

line 8 "air pollutions": It is not all air pollutants that are important here, it is aerosols (and precursors) and ozone precursors.

This is correct; we will change the reference to “air pollutants” as suggested.

Line 25: I wasn’t sure if energy use in mobile sources included transport. If not, transport needs to be added to the list.

Mobile sources are indeed included in the transportation sector.

Page 4967, first paragraph: Again this paragraph might start with setting out the needs for climate modelling. This could then be used to justify the choices for spatial and temporal resolution and the choice of species. Note "All emissions necessary" is a bit strong, maybe "Emissions required by the current generation of coupled climate-chemistry models"

We will clarify what is actually needed by climate models. Indeed, not all emissions are needed in order to generate the fields necessary for the radiative forcing of climate models. We have a discussion of the justification of scales (especially time) in the conclusion section of the original version of the paper, but will include such discussion in the Introduction as well.

Section 1.2: This is much too short for a description of the 2000 emissions. These are the basis for the whole dataset. There needs to be at least a table listing which referenced datasets are used for the different regions, species and sectors. A table of the emission totals is needed too. For the BC and OC more detail is needed about what the updates are, how the data from the two studies are combined and how the emission factors are harmonised. For SO2 more detail is needed on what the updates are, which emissions are taken from UNFCCC, which are taken from regional inventories (with references to the inventories). It is not clear where the Smith et al. 2010 reference is available.

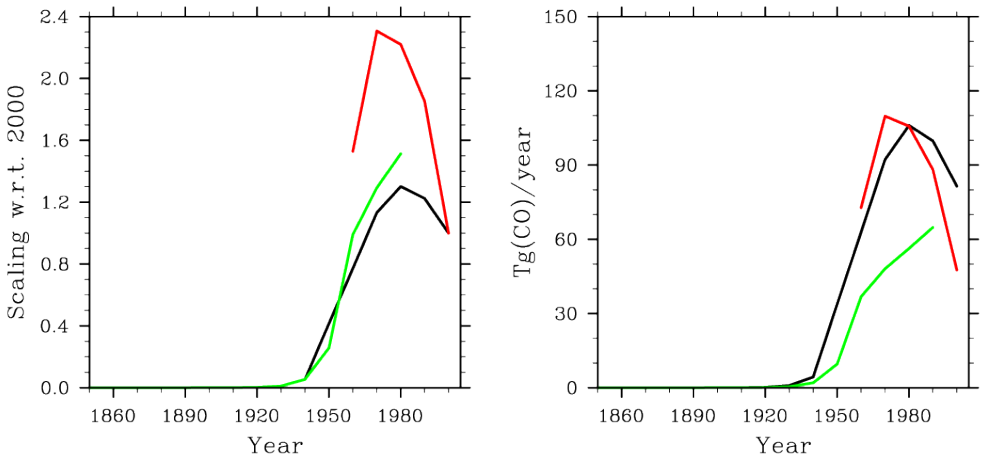
The discussion of year 2000 will be expanded, as presented in the joint response. We will have a limited discussion of the specific changes in OC/BC and SO2 from T. Bond and S. Smith respectively. The paper Smith et al. is now submitted to ACPD and should be available for viewing and comments soon. The discussion on BC will describe the updates T. Bond and C. Liousse have agreed to make to the original Bond et al. (2007) dataset. In particular, they performed a sector-by-sector comparison to resolve differences between the inventories of *Bond et al.* [2007] and *Junker and Liousse* [2008].

Section 1.3: It might be useful to have a summary of what the difference between RETROandthecurrentstudyisfortheyear2000.Isittheadditionoftheregionaldata?

Our inventory is quite different from RETRO (beyond fixing their known problem in CO emissions) as it uses different sources of information for specific regions (see Table 1 in the joint response).

Page 4972, first paragraph: The blue dot in figure 1 should be explained. I'm not quite sure why the black line in the right panel isn't simply a scaling of the black line in the left panel?

We have removed the blue dots. There was an error in the generation of the figure, leading to the noted discrepancy; because of the very large increase in 1980-1990 in RETRO (higher than indicated in the EPA estimates), we averaged (only for this sector and region) our estimate as being the mean of the EDGAR-HYDE value (itself closer to EPA) and our original estimate. We have corrected the figure (see below) and the text will be changed accordingly. We will however remove the left portion of the figure as suggested by Reviewer #1.



Page 4972, second paragraph: It is not clear why agricultural waste burning is excluded, or how it is subsequently treated.

Agricultural waste was excluded because the sectoral definition has changed, with biofuel being included in the domestic sector. We will include a description of the agricultural waste burning in the paper.

Page 4972, third paragraph: Why is the Bond inventory preferred over Junker and Liousse, why not take an average of the two? I couldn't see the CO estimate of agricultural waste burning that is referred to as "see above".

The OC/BC emission dataset in this paper is indeed a combination of the two, as generated by C. Liousse and T. Bond (both are co-authors). But most of the data are from the Bond paper (see response above).

Page 4973, first paragraph: I couldn't see any estimate of anthropogenic soil NOx on the Edgar v4 website - more detail should be given here. What is meant by "time evolution is based on Yan et al."? Is it just the seasonal cycle? It so, what meteorology is used to drive it? Or is a longer time evolution included, based on trends in precipitation and temperature over the 20th C?

The soil NO_x from EDGAR-v4 emissions were provided by J. van Aardenne (co-author). We use the Yan dataset, corrected for the natural (i.e. pre-1950) estimate, to define the long-term change in soil NOx. The seasonal cycle is taken from the year 2000 in Yan and applied for all decades. We will add those details in the revised paper.

Page 4976, first paragraph: Given that there is a noticeable decline in biomass burning from 1900-1950 in figure 4, it would be useful to understand why. Mieville et al. suggest it is artificial fire suppression in the boreal forests.

We are indeed using the Mieville et al. (2010, itself based on the Mouillot et al. paper) to provide the long-term reconstruction of biomass burning. So fire suppression is indeed the likely explanation.

Page 4981, section 4.2: I would suggest stopping the runs at 2000, since continuing to 2009 doesn't add any new information but can cause confusion. Is van Vuuren et al. in preparation, submitted or in press?

We will stop the plots at the end of year 2000. The paper by van Vuuren et al. is in preparation and will be submitted to Climatic Change in June or July 2010; this will be part of a special issue describing all RCPs.

Section 4.2.1: Discussion of the pre-1950 ozone is needed since these data will feed into calculations of the 1850-present forcing. Even if there are reasons not to believe the Montsouris data, the comparison needs to be shown. How do the values compare to the Gauss et al. 2006 estimate of the change from 1850-2000?

Our estimate of the change in tropospheric ozone in CAM-chem is 9 DU between 1850 and 2000, in very good agreement with the Gauss et al. model mean. As in all studies (except for Mickley et al., where specific tuning of emissions was performed to fit the observations), our Montsouris values (15-20 ppbv) are larger than "observed". Both points will be added to the paper.

It would be very useful to have a statistical analysis of the trends in this section to see how the models and observations differ. By eye it looks as if the G-Puccini trends are significantly less positive (or more negative) since 1980 than for CAM-Chem. There are more recent references to the observed trends - Parrish et al. 2009, Oltmans et al. 2008.

We plan to include the following table summarizing our linear trend estimates. In all cases, the trend is performed on the timeline of the observations, with GISS linearly interpolated to the time of the observations. This table does show that the models underestimate the largest long-term trends. Our ozone datasets are the same or extension of the paper listed.

	Observations	CAM-chem	G-PUCCINI
Hohenpeissenberg	0.32	0.18	0.22
Zugsptize	0.40	0.24	0.23
Mace Head	0.18	0.17	0.17
Arkona	0.36	0.14	0.16
US Pacific coast	0.33	0.21	0.19
Barrow	0.04	0.10	0.06
Mauna Loa	0.15	0.28	0.17
Samoa	-0.03	0.05	0.00
South Pole	-0.05	0.03	-0.20

Section 4.2.2: The G-Puccini CO trend from 1980 is less (more negative) than CAM-Chem. There may be a connection here with the ozone trend. I disagree with line 23 that there is a "very similar ozone change over the same period". This might be more obvious if the trends were calculated. The flux analysis is interesting. It would be useful to extend this to analyse the trends. Are the differences between the two model trends due to trends in CO removal, methane oxidation or NMVOC oxidation? Note that the Horowitz et al. paper (not in References and should be 2003) is another model study so doesn't rule out a systematic problem with modelled CO.

It is true that the ozone trends are NOT similar and we will correct that statement. The two models are actually quite similar in the NMVOC oxidation scheme, so it is possible that this could explain the CO behavior. Only with more users will we know the extent of this problem. Again, this is beyond the scope of this paper. Thanks for pointing the missing reference.

Page 4985, line 21: Are the natural contributions imposed as sulphate concentrations or precursor emissions? Even if the emissions don't change, the oxidation rates will change over the length of the model run.

Yes the natural contributions are included. It is true that the oxidation rates will change; we will correct the text accordingly.

Page 4989, line 7: This final phrase sits rather awkwardly as it is the only time in the paper that air quality is mentioned. I would suggest either removing it, or developing it to comment on whether this dataset will also be useful for simulating air quality.

We agree with that statement and will remove this sentence.

Grammatical/style comments

We will correct all the grammatical/typo errors as suggested.