Response to referee comments for "Anthropogenic sulfur dioxide emissions: 1850–2005" by S. J. Smith, J. van Aardenne, Z. Klimont, R. Andres, A. Volke, S. Delgado Arias

We thank the referees for the many helpful comments that will significantly improve the paper. Responses to specific comments appear below.

Anonymous Referee #1

Received and published: 17 September 2010

Referee 1 comment: Anthropogenic sulfur dioxide emissions: 1850–2005 Smith, S. Jet al.

As the authors themselves points out, there exists already many global inventories of sulfur. The reasons why I still think it is enough new material in this paper to warrant publishing is in particular the attempt to include an uncertainty analysis and sector distribution.
We would like to point out that updated inventories using the latest information are needed for a wide range of research purposes. It is important that these not only be produced, but also that the methods and data used are documented. Publishing work in journals such as this one not only allows for wide access, but also ensures more through documentation and discussion of results through the peer review process. In addition to uncertainty analysis, this work extends global sulfur inventory data to 2005, with annual temporal resolution, and finer spatial resolution (country level inventory estimates, downscaled to 0.5 degrees) than available before. This work was also the source for the historical inventory data used for global model inter-comparisons exercises conducted in support of the IPCC AR5.

With respect to the latter I however puzzled about what sectors is actually included. I support the idea to rely on and include national emissions whenever available and judged to be of sufficient quality. Having said that, I have some concerns regarding the methodologies and presentation of results as detailed below.

Introduction Page 16112, Line 18. Sulfate aerosols are thought to have significant effects on climate. I don't think sulfate aerosols are only thought to have effects. There are multitudes of studies on this subject. Also state clearly that sulfur aerosols cause cooling.
We will change the wording in this section.

Methodology The methodology part of the paper is extensive (about half of the paper length), still it is rather difficult to grasp what is the innovative aspect of the approach. The authors state that an analysis of differences between inventories is beyond the scope of the current project. I propose however to include a broader analysis of differences between the current and earlier inventories from the first author in order to explain better the benefits of the most recent work. To me it is not enlightening enough to simply state that (P. 16130, L. 20): "The current estimate is somewhat below many recent estimates, particularly in the 1970s and 1980s, including the previous estimate by Smith et al. (2004) using a similar methodology, but at a more aggregate scale. The major differences between these two datasets over this time period are lower emissions estimates for China and the countries of the Former Soviet Union". The discussion could as well include the RCP inventory, and explain how a more detailed approach leads to lower emissions [if this is what the authors intend to say here].

We will move more of the methodological material to the supplement (as also suggested by the 2nd referee)

We will add a more extensive comparison between two inventories (likely to the supplementary material, and summarize that comparison in the main text). As discussed in the paper, new information that has been made available over the past several years is a particularly important factor in revisions to inventory values.

I would further add e.g. a float diagram to clarify and make more transparent the methodology and methodological choices. This could improve greatly the readability of the methodology chapter, by making references to such a figure at relevant places in the text. A well designed figure might also help to shorten the text.
We will consider how to add a figure, or perhaps a table, that summarizes the methodology in order to clarify the text.

The text refers to a multitude of adjustments, but nowhere is given an indication of the size and seldom the direction (lower/higher) of these. Please add information on which magnitudes we are talking about. The size of the adjustments could also be related to uncertainty of emissions. The countries with high quality recent emission inventories will in most cases also be the countries with more data available for input to the authors' calculations, and differences could perhaps be linked to uncertainty in methodology or specific input data.
The text will be edited to provide this information (perhaps in the supplementary material in some cases). We have, indeed, linked the uncertainty calculations to the quality of the input data in a general way, with lower uncertainty bounds used for years and regions where detailed inventory data exist, with higher uncertainty bounds used where data is less reliable, as indicated in Table 1 (as suggested elsewhere in the review, we will clarify our discussion of methodology on this point).

Page 16123, Line 15-22: Taken the stated lack of data into account. How did van Aardenne et al. split NOx emissions on sectors? I do not find that NOx is a very good proxy for SO2 sector distribution.

The primary goal of this calculation is to convert emissions by fuel into emissions by sector. Because data on fuel use by sector for early years was not available at the time these estimates were made, emissions splits from the Hyde data were used to estimate the sectoral breakdown. In the Hyde analysis used, emissions factors were not varied from 1890-1970, which is generally where the Hyde data were used, so any of the emissions species could have been used. NOx emissions were the most appropriate for this purpose since they arise almost entirely from combustion processes for the sectors considered. The derivation of the HYDE historical estimates is further described in Van Aardenne et al. (2001).

Results Maybe this short text includes all there is to say about the results, but I would hope that some more interpretation linked also to the two figures (of 7 figures in total) included for this section could be added. Again the lower emissions in China are pointed out, but no attempt to explain the difference is given. Interpretation of emission trends from Figure 4 could be added, in particular reasons for the steep decrease in emissions from China (opposing to satellite observations). Also material from the first to paragraphs in the discussion section belongs in my opinion here.

These are good suggestions and we will add discussion of these points. The lower emissions from China in this inventory as compared to previous estimates are due to newer information, although uncertainty is still quite large. As noted in the discussion of Figure 7, the satellite data is quite consistent with the trend from 2000-2005 in this inventory. The trend from 1996-2000 in the inventory is not well determined. While our inventory estimate shows a small decline of 8% over this period, given the substantial uncertainty, either a slight increase or decrease would be consistent with the uncertainty band. Given trends in sectoral distribution (as discussed re: figure 7), it is difficult to resolve such small trends using remote sensing data.

Are the sectors listed in Figure 2 the sectors included in the study and gridded data, or what are these? If yes, I cannot see that sectors Petroleum Combustion and Coal Combustion is detailed enough for modeling purposes. Please clarify from which sectors emissions are estimated, and make a figure with actual sector data.
We will clarify the text. The sectors in Figure 2 are the primary inventory result. An additional breakout to end-use sectors is performed as a secondary calculation. We propose to add a figure with the secondary sectoral breakout (buildings, industry, etc.) to the appendix. Note that this second level of sectoral breakout has larger uncertainty than the primary calculation, so we focus on emissions by fuel for the analysis in this paper, as this breakdown is more reliable for emissions by fuel.

Uncertainty While it is appreciated that estimates of uncertainties are included, the way uncertainties are calculated is not very clear. I propose to add an equation.
This is a good suggestion. A set of equations will be added.

I also propose to explain better how the uncertainty bounds listed in Table 2 has been worked out. Page 16126, L. 25: "The set of uncertainty bounds given in Table 2 are applied to countries categorized depending on the estimated quality of the data used to construct the inventory values (see supplementary material)." Instead of estimated quality, I understand this is anticipated (authors' best judgment) quality. I propose to add an example of how different assumptions come about. E.g. what uncertainty is linked to different sorts of input data missing, countries' own estimates included, etc.
We will add some examples of how these categories are applied to clarify this calculation. For example, recent inventory data from OECD countries, as submitted to the UNFCCC, was judged to be of high quality. This is Category I in table 1. Older inventory data from OECD countries, for example the estimates of Mylona, were assigned Category II. Time periods in OECD countries where no inventory data was available were assigned category IIa.

Page 16127, L. 12: "An alternative calculation assuming no correlation between values at the country level results in lower uncertainty at a global level by 3–27%, depending on the year."
What do you want to say by this sentence? Global uncertainty is estimated to maximum 12%.
We will clarify this material.

This sensitivity calculation assumed that uncertainties were summed in quadrature at the country level, instead of at the regional level. This is equivalent to assuming no correlation between countries. This results in the absolute value for uncertainty that is lower by 3-27%. Because we believe that there is substantial correlation in sulfur content and other parameters within regions, we feel that this method would underestimate uncertainty.

Page 16128, L. 6 "To include the potential impact of such correlated effects, we add to the uncertainty estimate for each sector an additional uncertainty amounting to 5% of total emissions (half this value for countries with well-specified inventories), with the additional uncertainty combined again in quadrature between sectors."
I have difficulties to understand the methodology here. Why 5% and 2.5% ?
We will clarify the text. We argue that systematic uncertainty is potentially present in all inventory calculations, but this is likely smaller in countries where we judge inventories to be more reliable, which is why we use a smaller value in these cases. The choice of 5% and 2.5% is author's judgment, since systematic uncertainty is very difficult to quantify. These values, however, do serve to encompass most of the range shown in Figure 6.

Apparently you calculate uncertainty per sector. I propose to include these uncertainty figures in the paper. If they are not calculated, please include a judgment of sector uncertainty.
We will add material on sectoral uncertainty (perhaps a figure or table in the supplementary material, briefly summarized in the main text)

Discussions
This section concerns a summary of results, a comparison with both emission estimates and satellite data, and list of some improvements to be made. I do not really find so much discussions e.g about methodology, uncertainty analysis and results.
We will add discussion of these points.

While I like the effort to compare the trends in emissions with satellite data, unfortunately the most interesting drop in Chinese emissions from 2007 onwards are not captured by the emission inventory. Is there any indication on how emissions behave in this period from potential additional data available in your database for China?
Unfortunately the current database only extends to 2005. The latest comprehensive energy data from the IEA are only just recently available up to 2008. Information on control measures and sulfur contents is also needed. Although there is work in progress evaluating such information for China, information from other countries is not always available. Because this was a global inventory project, we are limited to times where adequate data are available for most countries. We hope that an extension of this inventory to later years will be possible soon, but we cannot do that at this time.

Tables and figures
Table 1: Please quantify the contribution from sectors not included here.
We will add this material to the table.

Table 2: Difficult to understand even after having read the supporting material, which countries are included in which category. In "I. Recent-Country-Inventory" for example, USA is in this category from 1970, while Japan is included from 1980. All other countries are included from 1990-2005. What is this based on? Further, what does e.g. OECD (pre inventory) means? I suppose 10% means +- 10%? Are the uncertainties valid for all inventory years? Please update this table.

Yes, 10% means ±10%. We will clarify this material.

The uncertainty categories apply to the years as shown in supplementary table S-4. For example, a 5% uncertainty is applied to combustion emissions in the USA from 1970 through 2005. An uncertainty of 14% is used for these emissions from 1900 through 1950. These years correspond to the inventory data sources used (see Table S-2). Previous to 1900, a value of 21% is used. (As suggested earlier, we will add an equation to the main text for the uncertainty calculation, which will also help clarify this material.)

Figure 3. Consider to include the uncertainty estimates in brackets in table 1. Figure could still be kept, but please define East Asia and South- East Asia. Consider to sort legend according to emission size at e.g. around 1970 to ease readability.
This is a good suggestion. Since this table is already somewhat large, we may need to add a separate table of uncertainty estimates in the supplementary material.
We will clarify the region definitions here and in the region definition section of the supplementary material.

Figure 6. Please consider to add the recent Edgar 4 V.1 emission inventory data.
We will add these data now that they are publically available.

Editorial General, make sure abbreviations are explained first time. E.g. RCP
Reference REAS inventory
Page 16127, Line 6: delete "that" Page 16132, Line 27: Delete "be" Page 16134, Line 5: Insert "the" before fraction
We will make these changes.

Anonymous Referee #2
Received and published: 12 October 2010
General Comments
The authors assess sulfur dioxide emissions worldwide, considering all major sources and using country-level monitoring data in place of bottom-up mass balance calculations where the country-level data are judged to be reliable. Spatial resolution of the global SO2 inventor y is improved to 0.5 x 0.5 degree, in contrast to past work which has been mostly 1x1 degree resolution.
The relative amounts of text devoted to methods and results are not well-aligned with what will be most interesting and informative to ACP readers. In particular, the methods section needs to be shortened by moving details and long lists of references on data sources to supporting information. The results section should be expanded to show more intermediate results and guide the reader through more of the direct findings of the study.
We will move more methodological material to the supplement and add discussion as suggested.

1. It is suggested that the authors show time series plot of sulfur contents in key fuels: coal, residual fuel oil, distillate fuel, and gasoline, over the entire inventor y period (1890-2005). Separate results could be shown for Europe, North America, Asia, etc. if the authors are able and wish to show greater detail. In cases where country-level data

are available, fuel sulfur content could be back-calculated. This could help to expand the results section of the paper, although what would be shown could include more intermediate values rather than just final results for SO2.
We will add a figure as suggested (see Figure 3 in Smith et al. 2005 for an example). Note that what we will show is the implied emissions factor by broad fuel (coal and petroleum), which includes changes in sulfur content combined with other abatement measures. The actual sulfur content will depend on supplementary information (such as FGD sulfur removal) that is not available in many cases.

2. On page 16119, lines 7-8, there is excessive precision in reported fuel sulfur contents (2.86 and 1.25%). The authors should include error bars for these numbers. It is not plausible to define these values to three significant figures. While the residual fuel sulfur content seems reasonable, the reported distillate fuel sulfur content seems very high, unless the authors are somehow implicitly including blends of residual & distillate fuel (i.e., No. 4 fuel oil) into the reported value. The authors should review other data sources as a check on these values.
The display of these values has been changed. The distillate category is, indeed, actually "distillate and other" and we have clarified the text. We could not find statistically-based estimated of uncertainty for this quantity, but we did add a discussion of the source of sulfur content information.

3. The authors should briefly discuss coming changes to fuel sulfur contents, and the challenges this will pose for estimating SO2 emissions in future years. For example, mandates for Sulfur Emission Control Areas and Ultra-Low Sulfur Diesel Fuel will likely spread, with resulting decreases in SO2 emissions.
We will add this to the discussion.

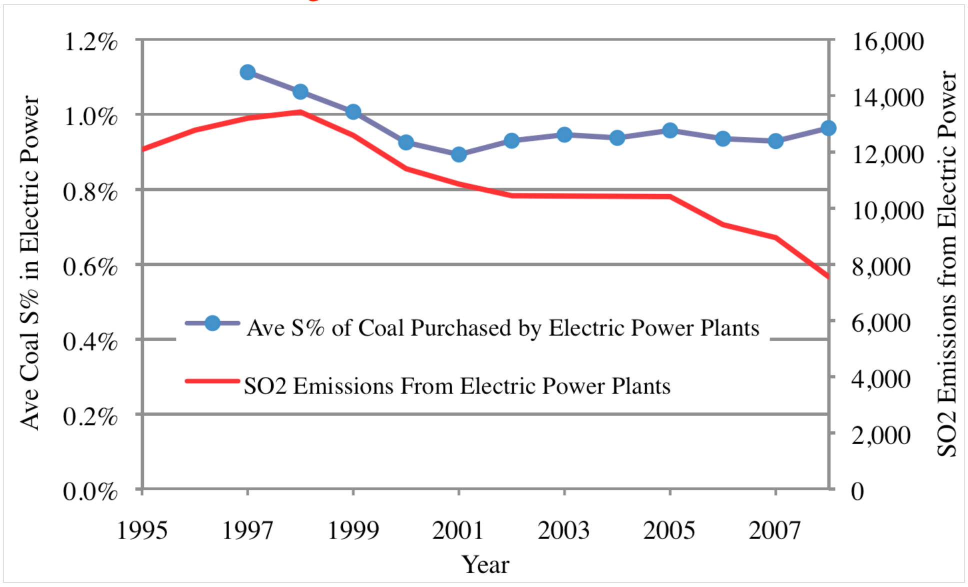
4. Some specific areas in the methods section where more concise exposition (and/or greater reliance on supporting information) would help include page 16114, lines 20-23; page 16115, lines 9-10 and lines 14-26, line 28; page 16117, line 27 through page 16118, line 4; page 16121, lines 27-29.
We thank the reviewer for pointing out these places where the text is unclear. We will clarify (and simplify as we can) text in these locations.

5. Important shifts have occurred in the use of natural gas versus home heating oil versus coal for winter season heating in the residential sector. The effects of the transition away from coal to natural gas are mostly on black carbon, but sulfur emissions from domestic fuel combustion are also affected. These transitions began around 1950 in US and some European countries. Former Eastern European nations made these transitions much later, including abandoing use of high-sulfur brown coal. It would be informative if the authors could discuss in more detail the consistency of their SO2 results in terms of known fuel transitions and timing for various countries or regions.
We thank the reviewer for pointing out these places where the text is unclear. We will clarify (and simplify as we can) text in these locations. These fuel shifts are, indeed, included in the calculations, although the sectoral split is increasingly uncertain in the past where sectoral energy consumption information is less detailed.

6. On page 16130, line 17, I dispute the contention that most of the SO2 reductions have been achieved through use of scrubbers. In the US at least, the reductions have been overwhelmingly due to switching to lower sulfur coal supplies.

The reviewer is correct that the source of SO2 reductions varies by region. Note that our text states that scrubbers are “a major driver of sulfur emission reductions”, not that they are the only source. The text also notes that lower sulfur fuels have also been a source of emissions reductions. For deep reductions, however, there is a limit to the amount of reductions that can be achieved by use of low sulfur coal. Ultimately, however, technologies such as FGD must be employed to reach the scale of reductions seen in Japan, Europe, and the United States.

The figure below (using data from the US EPA and US EIA) illustrates both effects for electric power plants in the US. For a few years, 1998-2001, emissions followed sulfur content, but since 2001 coal sulfur content has been relatively constant (or increased slightly) while the gap between sulfur content and emissions has increased since 2001, evidently due to sulfur removal technologies.



7. In section 2.2, prior work of Corbett and coworkers on ship emissions should be cited.

We apologize for the oversight. These is, indeed, important prior work for this section (and provides much of the foundation for the Eyring et al. results used here). We will add the appropriate citations and comparisons to this work.

Technical Corrections

Page 16112, line 15, sulfate acid should be sulfuric acid

Page 16117, line 7, Supplementary is misspelled

Page 16123, line 10, a word (from?) is missing between Emissions and smelting.

We will make the indicated changes.