TNO-MACC_II emission inventory; A multi-year (2003-2009) consistent high-resolution European emission inventory for air quality modelling

Answer to reviewers, August 2014

Reviewer #1 comment:

The paper presents the results well, and the work represents substantial improvements in the availability of emissions on a fine spatial resolution across Europe. However, it is not appropriate to use a journal paper to simply log results. There is very little/no consideration of the strengths and weaknesses of the resulting data set. What are the main sources of uncertainty? How substantial are the resulting uncertainties? Are these assocaited with uncertainties in the emission estimates or the spatial distribution? As a minimum, some discussion of the resulting quality of the spatially resolved emissions needs to be added.

Answer to reviewer #1:

First of all, we thank the reviewer for providing comments to our paper.

The reviewer is correct that the article is mainly focused on describing the resulting dataset and how this dataset was derived. In order to address also the quality of the results, we have produced a revised manuscript where the we have included more description on the quality of the results. Part of this is a comparison between our inventory and other emission datasets, e.g. reported emissions data directly, GAINS and EDGAR.

Also, we have prepared an additional section on uncertainties associated with emission inventories. It will not be possible to provide a full uncertainty assessment due to lack of data. However, by providing information from other sources in our paper we think a good indication of the uncertainties associated with this emission inventory can be made.

A revised manuscript where the changes have been processed has been prepared.

Reviewer #2 comment:

I have read the paper "TNO-MACC_II emission inventory: a multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling" by Kuenen, et al. This paper reports on the construction of an inventory of anthropogenic emissions for the European region. The work is of high quality and will be of substantial value to the modeling and emissions community.

The work described to correct for inconsistent emissions across sectors is particularly valuable and, as mentioned below, providing more detail in this area would enhance the contribution of this work. This methodology not only likely produces a higher quality inventory (although issues of possible

double counting should be discussed) with better consistency over time, but is also a methodological advance in this area. More detail about these procedures and their results should be provided, as discussed below, so that this work will be even more useful to the broader community.

I recommend publication after consideration of the following specific comments.

The supplement supplies total emissions by country and year. It would make the dataset much more useful if the supplement could also provide emissions at a higher level of sectoral detail. It would seem feasible to provide these at the 2 digit SNAP level by country and species. This would appear to be about a 2000 line file if in a format similar to the current file "Resulting_emissions_by_country_and_year_clean.xlsx".

We have expanded the file with a second worksheet giving the information at sector level (2-digit SNAP detail).

Comparison, at the major country grouping level, of this emissions estimate with other estimates (specifically: EDGAR, GAINS, and original country inventory data) should be given and discussed. This is important context for these data.

We have made the comparisons and shown a few graphs in the paper. A new section in chapter 3 has been dedicated to this comparison.

PM2.5 is missing from the supplemental file "Resulting_ emissions_by_country_and_year_clean.xlsx".

There was a lookup error, which has been fixed.

EC and OC emissions should be added to the supplemental tables given the substantial interest in these emissions.

A separate Excel file prepared for EC/OC with emissions of EC and OC (both coarse and fine mode) by country and year.

It would be useful to see more quantitative detail about the issues raised in section 2.1.1, regarding differences between inventory estimates and values used by TNO. (some of this might best be in a text supplement). In general it would be very useful to report on the quantitative impact of each of these changes so that readers can have a sense of how important each of these issues were. Hopefully this will lead to improvements in the country inventories.

Including: On agricultural waste burning, were country inventory values generally less than GAINS, or were some higher and some lower? How much aggregate difference did this substitution make? Inland & coastal shipping. When the text says "very inconsistent between countries," was this mostly a matter of split (no concern for duplication) or were there inconsistent estimates (e.g., countries with known high levels of shipping activity reporting low emissions). There is a potential issue of double counting here (e.g., perhaps shipping or port fuel use might have been included in another category.) Again, how much difference did this substitution make in the inventory? "For particulate matter, numerous cases were found where reported PM2.5 exceeded reported PM10." How large a correction overall did this make in the final emissions? Were there particular sectors where this was the case?

We have addressed this by adding a more description with each of the changes to see the impact it made.

The issue of possible double counting of emissions should be discussed. This should always be a concern when additional data is added to an inventory. In inventories where there are discontinuities in the data, it is possible that those emissions might have been included in another category instead? Could this be, at least partially, excluded as a possibility in the cases where the corrections/additions increased emissions significantly (e.g., as in the cases shown in Figure 2).

We have added more description to the manuscript to address the possible risk of double counting, especially with those areas where additional data are added.

The text just generically refers to the supplement, while the supplement currently consists of 4 excel files. A text file should be added to the supplement that provides the file names for each file and briefly describes their contents. Giving each file a letter or number prefix (A., B., or S1., S2.) would also be helpful so that they can be appropriately referred to from the text.

Numbers added to the files and referenced in text

pg 5845, Line 10 "was made both for the coarse and the fine mode." clarify if separate splits were used for coarse & fine mode, or if the same split was used for both.

Different splits for coarse and fine, clarified in the text

pg 5845, Line 25

The sentence "When particle mass is calculated based on the SO2 emissions, it is found that SO4 fractions in PM range from 0.1% for gasoline and diesel combustion in road transport to 10–20 % for coal and residual fuel oil combustion in energy and manufacturing industries and in shipping." is a bit confusing. If this is referring to the fraction of S that is emitted as particles, it would be clearer just to use the wording of the previous sentence, as follows. "... the fraction of sulphur emitted in the form of particles ranges from 0.1% for gasoline and diesel combustion in road transport to 10 20 % for coal and residual fuel oil combustion in energy and manufacturing industries and in shipping."

Sentence was updated

2) In this same sentence, "It was found", does this still refer to the LOTOS-EUROS model? Clarify.

Yes it does. Clarified in the text to prevent further confusion.

The "It was found" wording might be misleading. Generally the SO4/SO2 emissions factions are input into such models, they are not outputs of those models. (Unless a detailed plume chemistry model is included.)

Text updated to avoid confusion.

Section 3.1 This is one of the most important technical contributions of the paper. The "mosaic" type approach of combining emissions from different sources, giving precedence to officially reported data, is rather widely used. The authors show the importance of making sure consistent emissions datasets over time are provided. They construct a consistent dataset by considering emissions at a

moderately fine sub-sectoral level, and correcting for missing or in-consistent data. Given the importance of this contribution, it would be very useful to provide a supplemental table with detail by at least country, year, and sector (if not also fuel if this differs) of data type. Something similar to the table Resulting_emissions_by_country_and_year_clean.xlsx, but with a list of sectors for each country (e.g. Power plants, Refineries, Residential combustion, etc.) coded with the source for the data (R="Reported", L="Linearly Extrapolated", G="Gains", etc.) by year. This would be extremely valuable for the community in general, and would enhance the contribution of this work. It is important that, for any particular country and sector, that the source of the data used be clear.

Overview has been made and is available through the supplementary files (file 2), in addition to the more generic data source sheet that was provided earlier.

Section 3.2 The discussion noting that sectoral EC/OC ratios vary by country is interesting, but a more complete presentation of the data would be helpful. A table in the supplement that gives EC/OC by sector and country would be very helpful, particularly given the interest in black carbon reductions

Table provided in the supplementary files (file 5).

The conclusion says "To further improve allocation to point sources, OMI satellite data were used to check the SO2 source strengths of point sources" but this is not discussed in the text. This sounds potentially quite useful and interesting. Material discussing this procedure and its results should be added. How much did the original point data have to be modified to match the OMI observations?

We have made a first comparison but results are not integrated in this inventory, but this is definitely an interesting development for checking emissions independently. We will flag this as an emerging technique for the future.

Answer to reviewer #2:

We thank the reviewer for his positive feedback and constructive comments. We believe the comments are useful for the manuscript, and therefore we have produced an updated manuscript. In the manuscript, the comments are addressed as follows:

As to the supplementary data file with total emissions by country and year: we have further detailed this file by also including the emissions at the level of SNAP level 1. This means the total number of data in the file will become 9 times larger. We will also include EC and OC emissions by country and by year in the supplementary material based on the PM split presented in this paper. A readme text file to the supplementary data describing the contents of each of the supplementary files. Also we will update the names of the files such that they can be easily referred to, as suggested by the reviewer.

A comparison with reported data, EDGAR and GAINS has now been included in the paper, including a discussion of the main findings. Furthermore, we include an additional section on uncertainties. Despite that a full uncertainty assessment is not possible due to lack of data, information from other

sources can be used to get a good feeling of the uncertainties associated with an emission inventory like the TNO_MACC-II inventory.

PM2.5 was missing from the supplementary file due to a lookup error. It is now also included in the supplementary file, there was unfortunately a mistake.

Regarding each of the changes described in section 2.1.1, we included one or two sentences describing the impact on emissions and also on the risk of double counted or missed emissions where possible. In case double counting possibly takes place as a result of our approach, we describe this in the paper. We highlighted the impact of the PM2.5>PM10 issue, although in most cases the impact was only small.

The various types of shipping are treated quite differently in different countries, since definitions are not always followed directly. In the reporting, there is a split between national and international inland shipping, which is not always easy to make. By replacing this with other data we believe to enhance the consistency of our dataset, at the same time avoiding double counting because all shipping data are replaced.

We agree with the reviewer that it is important to clearly list the source of emission data used for each individual year, pollutant, country and sector, on the other hand the number of combinations is very large. We have therefore further detailed the Choice_of_emission_source.xlsx supplementary file to include this information also at the sectoral level.

The comparison between large point source and OMI observations for SO2 is not something that has been included in this work, but this is a very interesting development for the future to verify emission inventories (especially large point sources). Although it has not been taken up in this inventory yet, we aim to include this type of verification in future emission inventories.