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TNO-MACC_II emission inventory: a multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling

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Abstract

Emissions to air are reported by countries to EMEP. The emissions data are used for country compliance checking with EU emission ceilings and associated emission reductions. The emissions data are also necessary as input for air quality modelling. The quality of these "official" emissions varies across Europe.

As alternative to these official emissions, a spatially explicit high resolution emission inventory (7 km × 7 km) for UNECE-Europe for all years between 2003 and 2009 for the main air pollutants was made. The primary goal was to supply air quality modellers with the input they need. The inventory was constructed by using the reported emission national totals by sector where the quality is sufficient. The reported data were analysed by sector in detail, and completed with alternative emission estimates as needed. This resulted in a complete emission inventory for all countries.

For particulate matter, for each source emissions have been split in coarse and fine particulate matter, and further disaggregated to EC, OC, SO_4, Na and others using fractions based on literature. Doing this at the most detailed sectoral level in the database implies that a consistent set was obtained across Europe. This allows better comparisons with observational data which can, through feedback, help to further identify uncertain sources and/or support emission inventory improvements for this highly uncertain pollutant.

The resulting emission dataset was spatially distributed consistently across all countries by using proxy parameters. Point sources were spatially distributed using the specific location of the point source. The spatial distribution for the point sources was made year-specific.

The TNO-MACC_II is an update of the TNO-MACC emission dataset. Major updates included the time extension towards 2009, use of the latest available reported data (including updates and corrections made until early 2012) and updates in distribution maps.

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1 Introduction

Over the last decades, environmental problems such as acidification, eutrophication, air pollution and climate change have caused significant adverse impacts on human health and vegetation (EEA, 2010). Only part of the air pollution emission reductions set by the 2010 National Emission Ceilings have been achieved (EEA, 2012a), therefore transboundary air pollution remains a problem (EEA, 2010). All these environmental problems are directly related to the emissions of substances to air. Reliable emission inventories are a prerequisite to understand these environmental issues and to develop effective mitigation options.

For a good understanding of environmental problems, not only the magnitude of the sources but also their location is important. The spatially distributed emissions need to cover the complete domain, and describe the emissions in a consistent way, i.e. in all countries the same sources should be included, and these sources should be assessed as accurately and consistently as possible.

Emission inventories are typically developed by using a bottom-up approach, i.e. combining available statistics on fuel combustion, industrial production, etc. with the most appropriate emission factors. For a detailed description on how emission inventories are constructed we refer to EMEP/EEA (2013), IPCC (2006) and Olivier et al. (1999). This approach has been taken also by many countries that produce annual emission inventories for greenhouse gases and air pollutants, since they have to report their emissions under the various international treaties. Over time, as experience and expertise increased, the number, substances covered and quality of these inventories significantly improved (EMEP, 2013). These in-country systems take into account all country-specific information and national legislation and are therefore capable of providing a more accurate estimate of the emissions compared to a regional or global emission inventory.

When using regional chemical-transport modelling in policy studies, the use of these official inventories is often required. However, the official emissions do still contain

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a number of gaps and shortcomings, e.g. not all countries report according to the requirements (EMEP, 2013). This paper presents a complete, consistent and spatially distributed inventory, which has used the official reported emissions as basis where possible. This makes this inventory suitable for application in policy-related modelling and impact studies for air pollution. The TNO_MACC-II inventory is the successor of the widely used GEMS inventory for 2000 (Visschedijk et al., 2007) and the TNO_MACC inventory for the years 2003–2007 (Kuenen et al., 2011; Pouliot et al., 2012).

2 Methodology

2.1 Emission estimates

The Convention for Long-Range Transboundary Air Pollution (CLRTAP; http://www.unece.org/env/lrtap/) requires countries to report their emissions. Fifty-one countries in Europe and North America, including the EU as a whole, have to annually submit their emissions of air pollutants for the latest year and all historic years to EMEP (Co-operative Program for monitoring and evaluation of long-range transmission of air pollutants in Europe). The reporting follows well-defined Guidelines and asks countries to complete a pre-defined template with emissions by year, pollutant and sector (defined by the Nomenclature for Reporting; NFR). Countries are encouraged to set up their own inventory system and choose the best methodologies for emission estimation which fit their national situation. For larger sources, Parties have to use more advanced methodologies, with specific emission factors for each technology. When no specific national methodology is available, the EMEP/EEA Air Pollutant Emission Inventory Guidebook (EEA, 2013) provides default guidance on how to estimate emissions for each sector. The official submitted data for all countries are collected by the Centre for Emission Inventories and Projections (http://www.ceip.at/) and made available online. Because of the more detailed methodologies included in most inventories and the national focus of each of the inventories, the reported emissions often provide

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the most accurate estimate for a country. However, in many cases gaps and errors do exist in the reported emission data. Especially the consistency in emissions reporting for consecutive years is problematic.

In order to assess the quality of the reported emissions, we have downloaded the data from CEIP (CEIP, 2011) for CO, NO_x, SO_2, NMVOC, NH_3, PM_{10} and $PM_{2.5}$ and from EEA (EEA, 2012b) for CH_4 for all countries for the period 2003–2009. Before analysing the data in detail, we have first aggregated the NFR sectors to 43 individual sectors (link table available from Supplement). These 43 sectors were defined based on the SNAP (Selected Nomenclature for Air Pollution) at level 1 with one additional level of detail for most sectors. Industrial combustion (SNAP 3) and industrial process emissions (SNAP 4) have been aggregated to a new defined SNAP 34. This was done because there is often confusion between combustion and process emissions for a particular plant or facility, partly because countries may have slightly different definitions on where to draw the line or how to report. In an overarching European inventory this problem is effectively solved by merging both categories. An explanation of the SNAP source categories as used in this study is given in Table 1.

For this dataset we have analysed the time series between 2003 and 2009 in detail. Where the time series or the sector split of the total country emissions was not understandable (e.g. unexplainable jumps in the trend, multiple years of data missing, not understandable sector splits), the data were discarded.

In cases where reported data have not been used or were not available, emissions at the country level were taken from the GAINS model (IIASA, 2012). The GAINS model combines information on economic and energy development, emission control potentials and costs, atmospheric dispersion characteristics and environmental sensitivities towards air pollution (Schöpp et al., 1999). For a more detailed description we refer to Amann (2009) and Amann et al. (2011). The advantage of using the GAINS data is that it is consistent across countries and sectors and regularly updated. Emission data are available at sector and activity level, comprising more than 200 different categories

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for 5-yearly intervals. To obtain emissions for the years in between linear interpolation was used where necessary.

The GAINS model does not calculate emissions of CO. In case no country reported CO emissions of sufficient quality were available the CO emissions are gap-filled using a bottom-up emission inventory which has been developed at TNO for the year 2005. Like the GAINS model or the EDGAR inventory (JRC, 2011) this bottom-up inventory is built up using activities such as the energy statistics, industrial production figures, etc. as the baseline and combines this with the most appropriate emission factors. In the transport sector, this means that data from the TREMOVE model (De Ceuster et al., 2005) were used to disaggregate the energy use to detailed vehicle classes technologies for each country. These were combined with state-of-the-art emission factors for each technology for road transport (Ntziachristos et al., 2009) to calculate the emissions. If less detail was available for certain source categories, technology-specific emission factors have been applied to groups of countries with a similar technology level. Since this bottom-up inventory was originally only developed for the year 2005, emissions for the other years were estimated by scaling this inventory. Scaling factors for the different years were calculated from the EDGAR emission inventory v4.2 (JRC, 2011), which provides sector specific annual emission estimates for CO for each country in the world.

For the countries Armenia, Azerbaijan and Georgia, neither reported nor GAINS emission data were available. Therefore, EDGAR (JRC, 2011) data were used at SNAP level 1 (see Table 1) for these countries for all pollutants and all years. These were disaggregated to the same subcategories as the other countries by using the relative contribution of each subsector to the SNAP level 1 sector for Turkey (for each pollutant and each year) as a blue print.

To illustrate in more detail the extent to which each data source has been used, the Supplement includes a table which shows the main source of the emissions that was used, per country per pollutant. However, for underlying sectors the choice of which emission source to use may have been updated based on the checks that were

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performed. In the final dataset, the share of reported data in the total emissions varies between 40 % (for PM) and 70 % (for NH_3). In geographical terms, reported emissions were the primary data source for most EU Member States and EFTA countries, while for many former Soviet Union countries and some Balkan countries the use of GAINS or other alternative data sources was necessary.

2.1.1 Consistency between countries and across years

Emission data for certain years may be missing (Figs. 1 and 2, left panel), and countries may use different classifications or differ in what sources they report. To improve consistency between countries, a number of updates have been made to the resulting dataset which mainly affected the reported emissions data.

- Emissions of NO_x and NMVOC from agriculture have been removed for all countries, since reporting of this source is found to be very inconsistent between countries. Note that this does not apply to combustion emissions from agricultural machinery because this should be reported in the source category for non-mobile machinery (SNAP 8).
- Estimates for emissions from agricultural waste burning have been replaced by GAINS emissions because inter-country variation for this source was too large to be realistic.
- Emission estimates for national shipping were found to be very inconsistent between countries, most likely due to different definitions for the various sectors. For all pollutants, these emissions have been replaced with TNO estimates, which distinguish inland shipping and coastal shipping as separate sources.
- For particulate matter, numerous cases were found where reported $PM_{2.5}$ exceeded reported PM_{10}. These have been corrected by increasing PM_{10} emissions to $PM_{2.5}$ levels. This implies that in such cases the coarse fraction was set to zero and can be seen as a conservative correction.

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- Emissions from international shipping have been taken from CEIP (2012) for all years and pollutants.

- NMVOC, SO_2, NO_x, CO and PM shipping emissions for the 43 largest North Sea ports (including oil terminals) were additionally included. For the year 2009, these data were taken from MARIN (Cotteleer et al., 2011). For SO_2 and PM a strong decreasing emission trend for the period 2005 to 2010 is expected as a result of implementation of European sulphur reduction policies (EC, 2011). The assumed average sulphur contents in marine fuel used for the calculation of in-port emissions is presented in Table 2. SO_2 and PM emissions have been scaled accordingly from the 2009 emission data. Emissions of other substances are assumed to be constant at 2009 level for the 2003–2009 period. From the MARIN emission data, implied emission factors based on port turnover capacity were derived and applied to the 1200 other ports in Europe, for which capacity data was taken from the PAREST emission database (Denier van der Gon et al., 2011). Based on Google maps and visual identification of port activities the $1/8^{\circ} \times 1/16^{\circ}$ cells occupied by the 43 MARIN ports have been manually selected (e.g. the Port of Rotterdam occupies seven of such cells). Geographical distribution of emission within cells associated with a certain port is assumed to be uniform. The location of the centre point of the 1200 other ports in Europe has been taken from the PAREST emission database (Denier van der Gon et al., 2012).

To be suitable as model input the emissions need to be distributed on a grid (see Sect. 2.3), for which a more detailed sectoral breakdown is needed to allow for a different spatial distribution of different subsectors and fuels underlying the 43 sectors. Therefore, emissions have been disaggregated using the more detailed data available from the GAINS model and the TNO bottom-up inventory (for CO). For power plants, residential combustion and road transport (exhaust) the emissions are disaggregated to main fuel type (coal, gas, solid biomass, waste or light, medium or heavy liquid fuels). For some other sectors such as the iron and steel and non-ferrous metal industries,

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emissions are disaggregated to subsectors. An overview of the disaggregated sectoral classification is given in the Supplement.

2.2 Particulate matter composition

For particulate matter, the emissions have been further disaggregated from $PM_{2.5}$ and PM_{10} to various components in the coarse and fine mode. To calculate this PM split, more detailed sectoral information is needed, for example the fuel type used in combustion installations and the type of installation. Therefore, the emission data are first disaggregated to GAINS sector and activity combinations (more than 200 categories).

For each GAINS category, a fractional split between 5 PM components (EC, OC, SO_4, Na and other minerals) was made both for the coarse and the fine mode. The fractional split is constructed in such a way that it adds up to 1, provided that OC is converted from a C-mass basis to full molecular mass (FMM). To convert to FMM, OC was multiplied by a factor 1.3 that accounts for other elements present on OC (e.g. O, N or S). It is known that the conversion factor of OC to FMM ranges between (1.1–1.8) but here a weighted average of 1.3 was used for all sources. Since the PM split provides fractions and not absolute emissions, this has no influence on total PM emissions. For EC and OC, the split is based on a recent bottom-up EC and OC inventory for the year 2005 (Visschedijk et al., 2009). This inventory involved creating "best estimates" per GAINS sector and activity combination for EC and OC fractions in PM, based on literature data and three earlier EC and OC emission inventories.

Particle-bound sulphate is mostly emitted through the combustion of high-sulphur fuels such as coal and residual fuel oil. In the LOTOS-EUROS model (Schaap et al., 2008) it is estimated that around 2 % of the sulphur is emitted in the form of particles. When particle mass is calculated based on the SO_2 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.

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The sodium fraction is relatively unimportant to the total PM but may be useful when looking at base cation deposition. The sodium content is based on reported sodium content for 40 PM sources calculated in Van Loon et al. (2005).

The fraction "Other minerals" contains other non-carbonaceous particles and is calculated as the remaining fraction after the other fractions have been calculated.

The fractions per GAINS category have been applied to the emissions of coarse PM (PM_{10}–$PM_{2.5}$) and fine PM ($PM_{2.5}$) for each GAINS category, and subsequently been aggregated to the 77 source categories which are used as input to the spatial distribution.

2.3 Spatial distribution

The final step in the inventory was the distribution of the complete emission dataset across the European emission domain at 0.125° x 0.0625° longitude-latitude resolution. For each of the 77 source categories for which emissions are available, one or more proxies were identified. These proxies provide the mapping of the emissions of a certain pollutant to the grid for a given sector and year. For each country, pollutant, sector and year the most appropriate proxy was chosen in a selection table. An overview of all the proxies used per sector is given in the Supplement.

For point sources, we have made use of the E-PRTR database (http://prtr.ec.europa.eu/) which provides information on the location (longitude, latitude) and emissions of major facilities in Europe. E-PRTR data was available on an annual basis from 2007 onwards, while data from the years 2001 and 2004 were available from its predecessor EPER (EC, 1996). For the intermediate years, data from the closest year available was used. Since the EPER and E-PRTR data only contain emissions from facilities above a certain threshold, using this data to distribute total emissions for a certain sector can only be done for those sectors comprised of large facilities, e.g. the cement and aluminium industry. Furthermore, a judgement has been made on the quality of the data before actually using it. For example, there are multiple facilities where the geographical location points to the administrative location (e.g. company headquarters)

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rather than the location where the actual emissions occur. For the other point sources, and also those in countries which are not covered by E-PRTR, TNO's own point source database (described in more detail in Denier van der Gon et al., 2010) was used as a proxy for the distribution of these point source emissions.

For non-point sources (e.g. residential combustion, transport sectors, agriculture), proxies were selected to distribute country total emissions over the grid. These proxies include a.o. total, rural and urban population, arable land, TRANSTOOLS road network (JRC, 2005). The proxies for the area sources were assumed to be static in time, e.g. changes in the population density are not taken into account. Most proxy maps were taken from Denier van der Gon et al. (2010) but a number of modifications and improvements have been made. A new population map for the year 2005 has been implemented at high resolution, and a special proxy has been developed for the distribution of residential wood combustion. The latter takes into account both the population density, but also the proximity to wood. Despite this modification for the distribution of residential wood combustion, an overallocation of the emissions in urbanized centres may well be present in the spatial distribution. This has previously been described by Denier van der Gon et al. (2010a) and Timmermans et al. (2013). However, a universal, representative and well-documented approach that justifies a modification of the spatial distribution between urban and rural areas for Europe does not exist at this moment.

For the actual calculation of the emissions grids, a SQL server system has been set up which performs all the calculations. Emissions that could not be distributed (e.g. because the proxy was not available for that specific country) are by default distributed using either total population, rural population or arable land. In a last step the gridded emissions are aggregated to SNAP level 1, primarily to reduce the size of the output emission grid file.

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3 Results and discussion

3.1 Analysis of reported emissions

To illustrate that consistency is an issue with reported emissions, Fig. 1 shows reported emissions for 5 selected combinations of country and SNAP level 1 source categories. It is shown that in a number of cases reporting only started somewhere during the time series. Also, some of the time series show unexplained trends (e.g. high emission in LVA SNAP 34 for 2004; very strong increase in Hungary SNAP 2 in the last years; small emission in Russian Federation from SNAP 2 compared to other countries in 2009).

Especially the issue of missing data for earlier years is important to the total emissions, as illustrated by Fig. 2. The left panel shows the time series for NOX and PM_{10} for all sectors, by country group, relative to the emission in 2009. For the EU15 countries (15 Member States of the EU as of 1995 plus Norway and Switzerland) the trend is a small decrease indicating improvements in technology and more abatement in later years. For the EU13 (New EU Member States joined after 1995, including Croatia) and the other (non-EU) countries, clearly a large part of the emissions is missing in earlier years.

The right panel of Fig. 2 shows the same trend data, but now for the final dataset. In this dataset, all missing emission data and erroneous time series were corrected and/or gap-filled by replacement with other data. NO_x and PM trends in EU countries are decreasing faster than in non-EU countries. In fact, for PM_{10} in non-EU countries the total emission is even slightly increasing.

EMEP (2011) provides an overview of submissions under the Convention of Long-Range Transboundary Air Pollution. The report shows that out of the 50 countries that have to report (excluding the EU as a separate Party), 42 countries actually did submit an inventory, while 34 submitted their inventory in time with the deadlines. Seven countries submitted an inventory without emission data for particulate matter (EMEP, 2011). For gridded data, data is to be reported every 5 years at a spatial resolution of 50 km × 50 km. As for the reporting in the year 2005, only 17 out of 48 countries in the

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EMEP area reported gridded emissions for the main pollutants, and only 15 countries reported gridded data for PM (EMEP, 2011).

3.2 Resulting emissions

Table 3 lists the total emissions in each year per pollutant per year. The trend shows that emissions of all pollutants are decreasing in time. The decrease is most pronounced for CO and SO_2, for which emissions in 2009 are about 25 % reduced compared to 2003. However, the change in emissions is not uniform. Figure 3 shows the relative reduction in 2009 compared to 2003 by country group. It can be seen that for the EU15 countries (plus Norway and Switzerland) the highest emission reductions were achieved (up to 50 % for SO_2), and also for EU13 countries significant reductions were found. For the non-EU countries however, emission reductions were much smaller and even emission increases were found for CH_4 and particulate matter. On the Europeans seas, most emissions increased going from 2003 to 2009, most notably for NO_x, CO and NMVOC (Fig. 3).

In the Supplement, an Excel file is included which lists country total emissions by pollutant for each year between 2003 and 2009.

3.2.1 PM fractions

PM_{10} and $PM_{2.5}$ are broken down into components (EC, OC, SO4, Na and other minerals) using the developed PM split. Figure 4 shows the EC and OC emissions per SNAP category for the European domain. In terms of total mass, the particulate carbonaceous emissions < 2.5 µm were about 5 times higher than the coarse fraction (< 2.5–10 µm) emissions. The most important source of fine OC is residential combustion (SNAP 2), particularly related to wood combustion. For coarse OC however, agriculture is the most important source of emissions. For EC residential combustion and transport (diesel combustion) are the most important sources for fine EC, while for coarse EC power plants and industry are the main sources.

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The relative importance of source sector contributions varies substantially between countries. As an example, the EC emissions (for coarse and fine mode separately) for Poland and the Netherlands are shown (Fig. 5). In Poland, high EC emissions resulted from coal and wood combustion in the residential sector, which are much less relevant for the Netherlands. Total emissions from the road transport sector in the Netherlands and Poland are quite similar, the larger fleet size in Poland is more or less compensated by the lower share of diesel in the fuel mix.

3.3 Spatial distribution

The result of spatially distributing the emissions using the various proxies is shown for NO_x and EC (< 2.5 µm) for the year 2009 (Figs. 6 and 7, respectively). The major cities, major transport routes and shipping routes at sea can be identified as important sources in these maps.

Figures 8 and 9 show also NO_x and EC (< 2.5 µm), but now the difference from 2003 to 2009. Positive numbers (blue colour in the maps) indicate a decrease in emissions from 2003 to 2009, while negative numbers (red colour) show an increase in emissions. For NO_x, it is shown that most land-based emissions decrease, but in some countries in Eastern Europe an increase is seen, e.g. in road transport for Poland, Slovak Republic and Bulgaria.

For fine particulate EC emissions are decreasing in most countries, but also increases are found especially in Eastern Europe and at sea. Highest reductions are achieved in cities and urban areas, since the initial 2003 emissions in these regions were higher. Increases can be due to a growth in activity, e.g. for the Slovak Republic, the increase is due to higher reported emissions of $PM_{2.5}$ from road transport in 2009 compared to 2003. Emissions from international shipping increased on all seas (CEIP, 2012).

To ensure consistency at borders, we have chosen to use a generic spatial distribution methodology. To account for sudden changes in point source emissions, e.g., due to implementation of emission abatement measures, E-PRTR data was used on an

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annual basis for the distribution of the emissions over the various point sources. As an example, the share of each major power plant in the total SO_2 emission from the power plant sector in Spain is shown in Fig. 10. The largest emitters in 2003 have reduced their emissions drastically. This causes some of the less important plants to become relatively more important, even though their absolute emission did not change. It was confirmed that in these specific cases for Spain, the power plants switched fuel (using coal with less or no sulphur) or installed advanced control technologies for desulphurisation. The use of annual E-PRTR data for these large point sources enables us to reflect these changes from year to year. As mentioned earlier, for the years 2003, 2005 and 2006 no point source information was available and the closest year available has been used instead.

4 Conclusions

A model-ready emission inventory at high spatial resolution for UNECE-Europe for 7 consecutive years (2003–2009) was constructed, which combines the advantage of using official reported emissions to the extent possible. For air quality modelling and environmental impact assessment studies, a good understanding of the magnitude and location of the sources of pollution is of crucial importance for deriving policy conclusions. The main advantages of this inventory are:

- We use source sector specific data in a harmonized way, which allows both tracking of sources in the modelled data as well as trend analysis without artifacts such as differences between annual reporting years. For instance, NMVOC and NO_x from agriculture were excluded for all countries as reporting was found to be very inconsistent.
- The application of a consistent gridding methodology for all countries ensures patterns across borders do not show sudden changes or jumps; e.g. consistent land use and animal density maps to distribute agricultural emissions

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- To model particulate matter concentration and fate, models need to breakdown PM into components with different behaviour. We now provide such data which are not available from the official reporting.
- By using the point source data form E-PRTR and EPER, better locations of point sources were brought into the data base. Moreover, the point source gridding data is now year-specific, where earlier the 2005 distribution was used as a proxy for all years.
- To further improve allocation to point sources, OMI satellite data were used to check the SO_2 source strengths of point sources (Visschedijk et al., 2012)
- Emission in ports were added in a harmonized way for the whole of Europe.
- Elemental Carbon and/or Black carbon is increasingly important in discussion of health effects of PM exposure as well as climate discussion focussing on short-lived climate forcers. By bringing this component into the gridded emission datasets, the models are able to better cater for policy makers in this respect.
- Finally, since the data are as much as possible (given our quality criteria) from the official reported data, the data can be readily used for policy evaluation.

All in all, this paper presents a significantly improved spatially explicit emission dataset for the European domain. However, one should bear in mind the limitations of the European scale emission inventory. Since the spatial distribution of national emissions is done using a generic system of point sources and proxies, differences with other inventories may exist, especially when zooming in to the local scale such as a large city or urban area.

A next step would be to include the "semi-natural" sources in our emission inventory, which are not covered by official inventory data (e.g. resuspension of dust and NO_x emissions from soils). With decreasing emissions from most anthropogenic sources, these become increasingly important for the comparison between modelled and observed concentrations.

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Supplementary material related to this article is available online at http://www.atmos-chem-phys-discuss.net/14/5837/2014/acpd-14-5837-2014-supplement.zip.

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References

Amann, M.: Integrated assessment tools: The Greenhouse and Air Pollution Interactions and Synergies (GAINS) model, in: "Atmospheric Pollution and Climate Change: How to address both challenges effectively together?", 73–76, 2009.

Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Hoeglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schoepp, W., Wagner, F., and Winiwater, W.: Cost-effective control of air quality and greenhouse gases in Europe: modelling and policy applications, Environ. Modell. Softw., 12, 1489–1501, 2011.

Centre for Emission Inventories and Projections: Reported emissions by Parties under the Convention for Long-Range Transboundary Air Pollution, http://www.ceip.at/webdab-emission-database/officially-reported-emission-data/, (last access: 12 March 2012), 2012.

Cotteleer, A. and Van der Tak, C.: MARIN's emission inventory for North Sea shipping 2009: Validation against ENTEC's Inventory and Extension with Port Emissions, 2nd Final Report No. 25300-1-MSCN Rev. 2, Maritime Research Institute Netherlands MARIN, Wageningen, the Netherlands, 2009.

De Ceuster, G., Franckx, L., van Herbruggen, B., Logghe, S., van Zeebroeck, B., Tastenhoye, S., Proost, S., Knockaert, J., Williams, I., Deane, G., Martino, A., and Firello, D.: TREMOVE 2.30 Model and Baseline description, Final Report, Transport and Mobility Leuven, available at: http://www.tremove.org (last access: 16 February 2009), 2005.

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14, 5837–5869, 2014

Denier van der Gon, H. A. C., Visschedijk, A., Van der Brugh, H., and Dröge R.: A high resolution European emission database for the year 2005, a contribution to the UBA-project PAREST: Particle Reduction Strategies, TNO report TNO-034-UT-2010-01895_RPT-ML, Utrecht, 2010.

EMEP: Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe in 2009, EMEP Status Report 2011, EMEP Report 1/2011, 2011.

European Commission: Council Directive 96/61/EC of 24 September 1996 concerning integrated pollution prevention and control, Official Journal L 257, 26–40, European Commission, 1996.

European Commission: Directive 2012/33/EU of the European Parliament and of the Council of 21 November 2012 amending Council Directive 1999/32/EC as regards the sulphur content of marine fuels, Official Journal L 327, 1–13, European Commission, 2011.

European Environment Agency: The European Environment – State and Outlook 2010: Synthesis, European Environment Agency, Copenhagen, 2010.

European Environment Agency: Evaluation of progress under the EU National Emission Ceilings Directive, EEA Technical Report No. 14/2012, European Environment Agency, Copenhagen, 2012a.

European Environment Agency: Reported emissions by Parties under the Convention for Long-Range Transboundary Air Pollution and the UN Framework Convention on Climate Change, date of download 3 March 2012, 2012b.

European Environment Agency: EMEP/EEA Air Pollutant Emission Inventory Guidebook, 2013 edition, available at: http://www.eea.europa.eu/publications/emep-eea-guidebook-2013 (last access: 9 September 2013), 2013.

Intergovernmental Panel on Climate Change: IPCC Guidelines for National Greenhouse Gas Inventories, Intergovernmental Panel on Climate Change, Japan, 2006.

International Institute for Applied Systems Analysis: GAINS detailed emissions by source and activity, available at: http://gains.iiasa.ac.at/gains/EUN/index.login?logout=1 (last access: 19 June 2012), PRIMES baseline scenario 2009, 2012.

Joint Research Centre: TRANSTOOLS, Tools for transport forecasting and scenario testing, Version 1, available at: http://energy.jrc.ec.europa.eu/transtools/index.html (last access: 1 December 2008), 2005.

Joint Research Centre: Emission Database for Global Atmospheric Research (EDGAR) v4.2, available at: http://edgar.jrc.ec.europa.eu (last access: 24 May 2012), 2011.

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Kuenen, J. J. P., Denier van der Gon, H. A. C., Visschedijk, A., Van der Brugh, H., and Van Gijlswijk, R.: MACC European emission inventory for the years 2003-2007, TNO report TNO-060-UT-2011-00588, Utrecht, 2011.

Ntziachristos, L., Gkatzoflias, D., Kouridis, C., and Samaras Z.: COPERT: a European road transport emission inventory model, in: Information technologies in environmental engineering, Springer-Verlag, Heildelberg, 491–504, 2009.

Olivier, J. G. J., Bouwman, A. F., Berdowski, J. J. M., Veldt, C., Bloos, J. P. J., Visschedijk, A. J. H., Van der Maas, C. W. M., and Zandveld, P. Y. J.: Sectoral emission inventories of greenhouse gases for 1990 on a per country basis as well as on 1° x 1°, Environ. Sci. Policy, 2, 241–263, 1999.

Pouliot, G., Pierce, T., Denier van der Gon, H., Schaap, M., Moran, M., and Nopmongcol, U.: Comparing emission inventories and model-ready emission databases between Europe and North America for the AQMEII project, Atmos. Environ., 53 4–14, 2012.

Schaap, M., Sauter, F., Timmermans, R. M. A., Roemer, M., Velders, G., Beck, J., Builtjes, P. J. H.: The LOTOS-EUROS model: description, validation and latest developments, Int. J. Environ. Pollut., 32, 270–290, 2008.

Schöpp W., Amann, M., Cofala, J., Heyes, C., and Klimont, Z.: Integrated assessment of European air pollution emission control strategies, Environ. Modell. Softw., 14, 1–9, doi:10.1016/S1364-8152(98)00034-6, 1999.

Timmermans, R. M. A., Denier van der Gon, H. A. C., Kuenen, J. J. P., Segers, A. J., Honoré C., Perrussel, O., Builtjes, P. J. H., and Schaap, M.: Quantification of the urban air pollution increment and its dependency on the use of down-scaled and bottom-up city emission inventories, Urban Climate, 6, 44–62, 2013

Van Loon, M., Tarrason, L., and Posch, M.: Modelling base cations in Europe, MSC-W Technical Report 2/2005, available at: http://emep.int/publ/reports/2005/emep_technical_2_2005.pdf (last access: 1 December 2008), 2005.

Visschedijk, A. J. H., Zandveld, P. Y. J., and Denier van der Gon, H. A. C.: A high resolution gridded European emission database for the EU Integrate Project GEMS, TNO report 2007-A-R0233/B, TNO, Utrecht, the Netherlands, 2007.

Visschedijk, A. J. H., Denier van der Gon, H. A. C., and Dröge R.: A European high resolution and size-differentiated emission inventory for elemental and organic carbon for the year 2005, TNO-034-UT-2009-00688_RPT-ML, TNO, Utrecht, the Netherlands, 2010.

ACPD
14, 5837–5869, 2014

Visschedijk, A., Denier van Der Gon, H., McLinden, C., Foletov, V., Identification and Cross-checking of large point source SO_2 emissions in Europe using OMI retrievals, 15th GEIA Conference, Emissions to Address Science & Policy Needs, 11–13 June 2012, Toulouse, France.

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14, 5837–5869, 2014

Table 1. Explanation of the SNAP source categories (SNAP 3 and SNAP 4 are merged to SNAP 34).

SNAP	Sector name
1	Energy industries
2	Non-industrial combustion
34	Industry (combustion + processes)
5	Extraction and distribution of fossil fuels
6	Product use
7	Road transport
8	Non-road transport and other mobile sources
9	Waste treatment
10	Agriculture

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Table 2. Sulphur content assumed in the fuel for the calculation of in-port emissions (in %).

Year	North Sea	Baltic Sea	Other EU(27)	Non-EU(27)
2005	1.335	1.335	1.335	1.335
2006	1.335	1.095	1.335	1.335
2007	1.305	0.975	1.335	1.335
2008	0.975	0.975	1.335	1.335
2009	0.975	0.975	1.335	1.335
2010	0.1	0.1	0.1	1.335

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Table 3. Overview of total emissions (kton) per pollutant and year for UNECE-Europe (including international shipping), and the overall reduction in the time period 2009–2003.

	2003	2004	2005	2006	2007	2008	2009	Reduction 2003–2009
CH_4	48 857	47 965	47 636	47 547	47 390	47 282	46 857	4 %
CO	48 642	47 602	44 905	43 271	41 608	40 739	38 157	22 %
NH_3	5786	5732	5675	5624	5645	5576	5543	4 %
NMVOC	15 744	15 367	14 936	14 525	14 123	13 577	12 943	18 %
NO_x	20 996	20 913	20 737	20 329	19 941	19 121	18 248	13 %
PM_{10}	5430	5432	5414	5328	5257	5142	5029	7 %
$PM_{2.5}$	3775	3779	3761	3695	3656	3590	3513	7 %
SO_2	17 921	17 353	16 689	16 144	15 815	14 483	13 189	26 %

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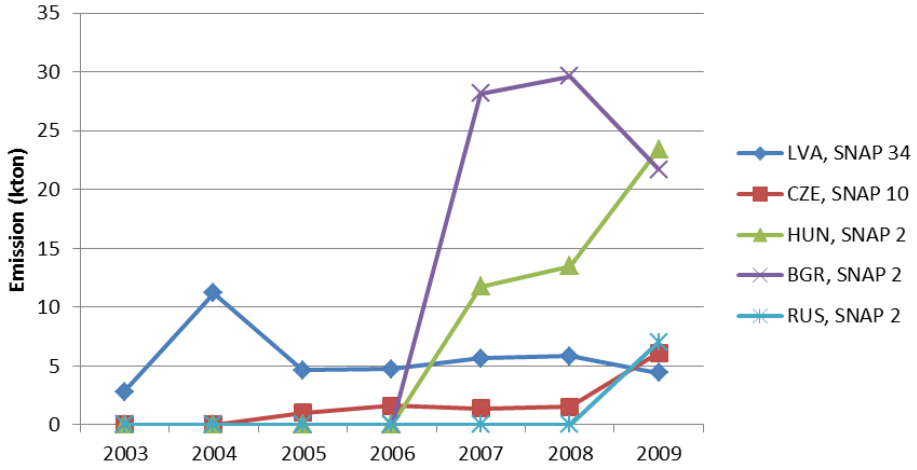

Fig. 1. Observed trends in PM_{10} reported emissions for selected countries and SNAP level 1 source categories.

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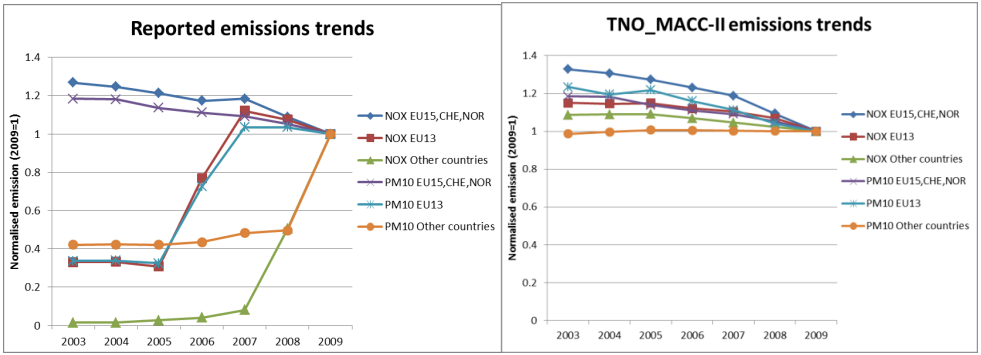

Fig. 2. Trends in reported emissions (left panel) and TNO_MACC-II (right panel) normalized to 2009 = 1.

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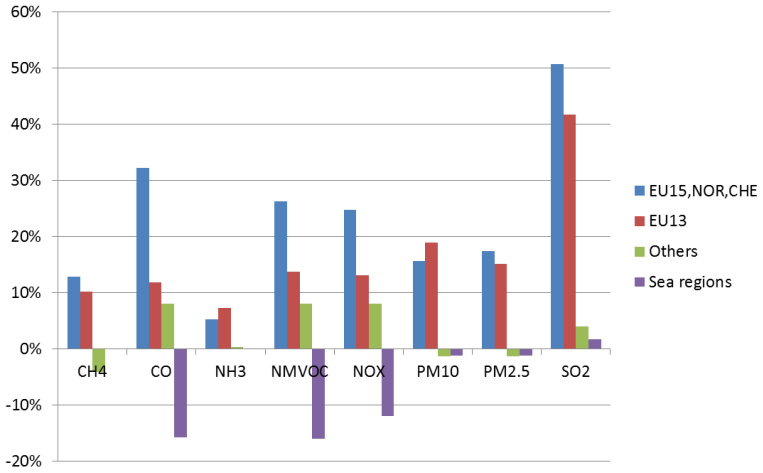

Fig. 3. Relative reduction in emissions per country group in 2009 compared to 2003.

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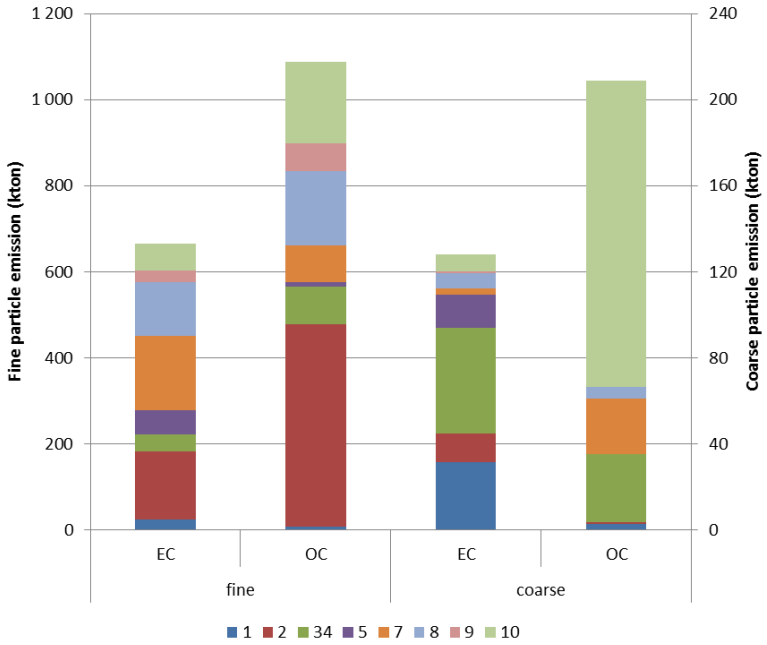

Fig. 4. Total EC and OC emissions per SNAP in coarse (2.5–10 µm) and fine mode (< 2.5 µm) for UNECE-Europe (including sea regions) for the year 2005. Note that fine EC and OC are plotted on the left *y* axis, while coarse EC and OC are plotted on the right *y* axis.

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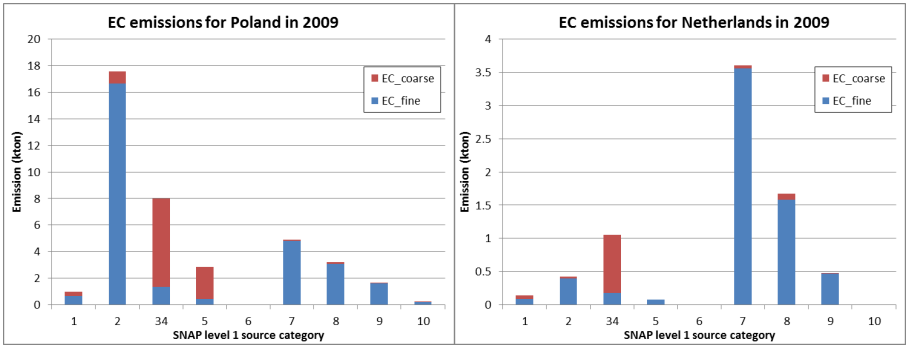

Fig. 5. EC emissions in Poland (left panel) and the Netherlands (right panel) per SNAP level 1 source category in 2009.

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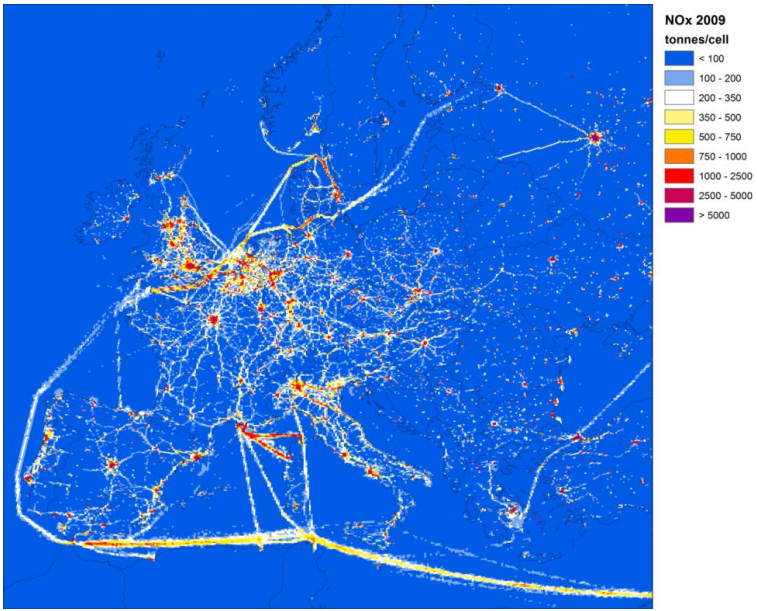

Fig. 6. Spatially distributed NO_x emissions from the year 2009 for all sources.

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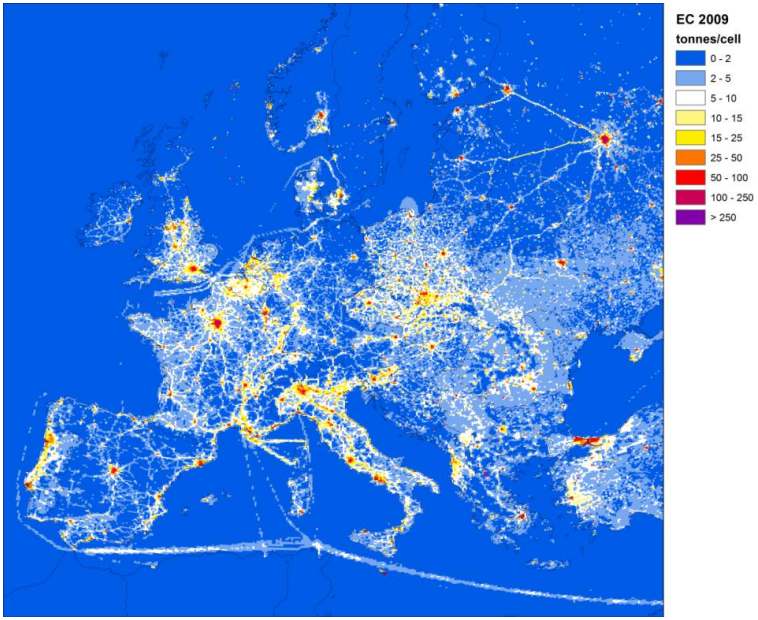

Fig. 7. Spatially distributed EC emissions (fine mode) from the year 2009 for all sources.

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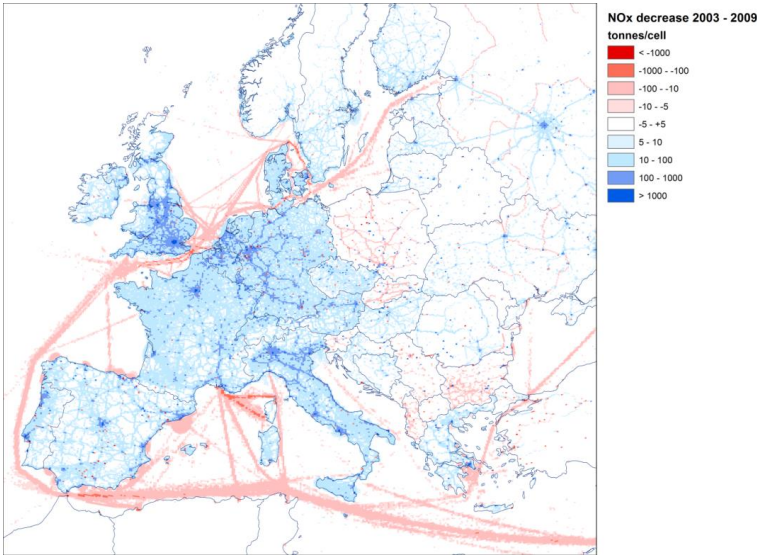

Fig. 8. Change in NO_x emissions between 2003 and 2009 in Europe, for all sources.

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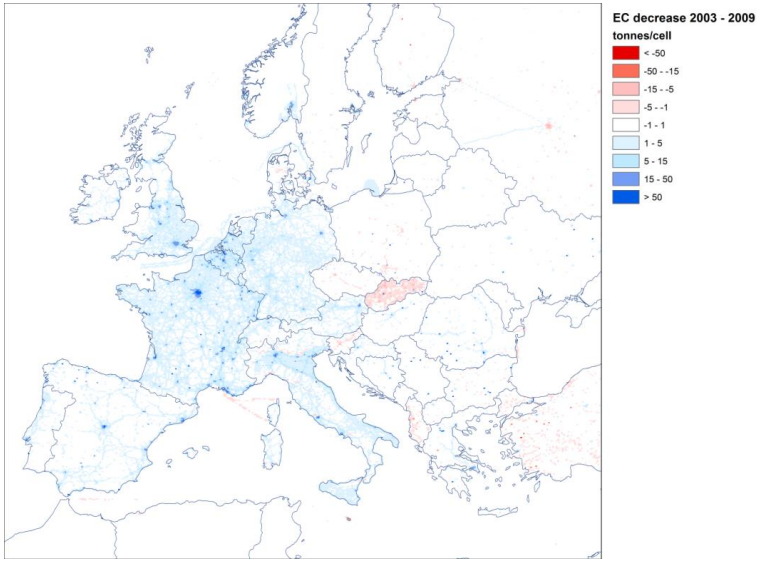

Fig. 9. Change in EC (< 2.5 µm) emissions between 2003 and 2009 in Europe, for all sources.

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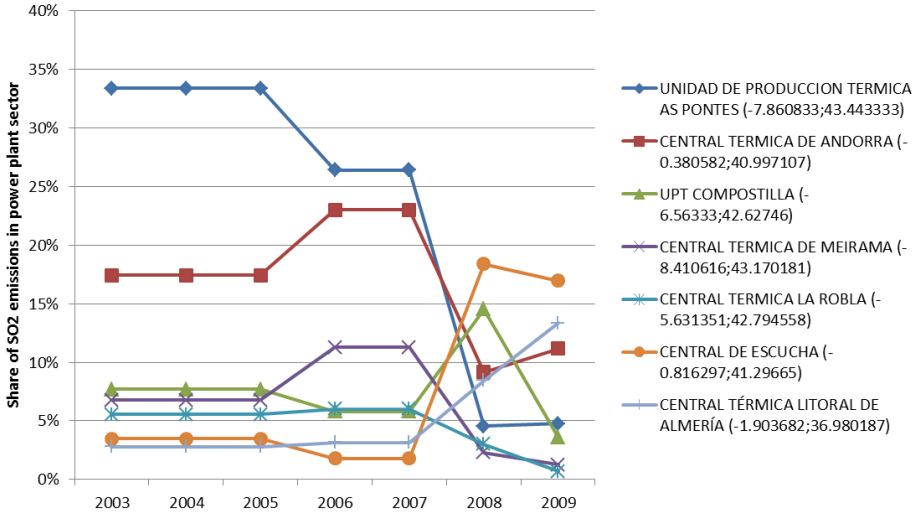

Fig. 10. Contribution of the top-7 SO_2 emitting power plants in Spain in 2003 to the annual total SO_2 emissions from the power plant sector.

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