

Dear editor, colleagues,

Below we address the reviewer comments and questions raised during the open discussion of the paper "Sensitivity of air pollution simulations with LOTOS-EUROS to temporal distribution of anthropogenic emission". We would like to thank the reviewer for the time and effort reviewing the paper. We feel it has improved as a consequence of the constructive comments. We have listed all reviewer comments below and answers are provided in Italics.

Anonymous Referee #1:

p.1, abstract:

Why is the comparison with observations limited to Germany?

In general, limited attention has been given to temporal profiles of emission data. National emission reporting is aimed at establishing annual totals per country and, hence, no detailed information on timing is gathered or provided. This leaves modelers or teams generating model ready emission input to have to come up with the information themselves. Normally time profiles are generated based on information of a (group of) country and applied to all countries. This practice we did not want to follow as we assessed if an effort aiming at providing detailed, sector and country specific information on timing is beneficial. Hence, we focus on a single country here: Germany. We added some explanatory sentences to the text in the introduction section (p.19315, l.11): "In this study we test the sensitivity of the model performance for improved temporal emission information. For this purpose we focus on improving the emission variability for a selected number of components and sectors for Germany."

And to section 2.3 (p.19318, l.24): "The current emission time profiles per sector are old and often based on information representative for one or several countries (e.g. Dutch traffic count data for SNAP 7). However, they are applied to all countries in the model domain. In this study we constructed emission time profiles representative for Germany and evaluated the impact by evaluation against German monitoring data."

It might be helpful to give the information that the simulations are carried out for the complete year 2006 (Why?) with an hourly resolution and a horizontal resolution of about 25 km.

We will add this information to the abstract (p.19312, l.15). "The performance in comparison to observations for Germany was quantified for the pollutants NO₂, SO₂ and PM₁₀ and compared to a simulation using the default LOTOS-EUORS emission time profiles. The LOTOS-EUORS simulations were performed for the year 2006 with a temporal resolution of one hour and a horizontal resolution of approximately 25x25km². "

And to section 2.3 (p.19318, l.22): "The meteorological year 2006 was chosen because it was a very variable year in terms of air quality including clean and heavy polluted periods in Europe. Moreover, data to construct time profiles for SNAP1 (data from the REMix model) and SNAP7 (traffic count data) were available for this year."

p.2, introduction:

Even if it is not in the focus of this paper: the height distribution of emissions, in particular for SNAP 1 is also a part which might be more variable as assumed in emissions as used currently in the air

pollution models due to its dependency on activity/stack parameters (e.g. gas exhaust temperature) and ambient air (meteorology). One can think about to mention this at least somewhere in the text.

This is an interesting point, which is indeed not in the focus of the paper but worth to mention in the discussion section 5 in the outlook part, (p.19332, l.15): "Not within the scope of this study but another important issue for the release of emissions in the model is the vertical distribution to the model layers. Wang et al. (2010) showed in a study for East Asia that for example for NO₂ and SO₂ the vertical distribution of emission play an essential role."

It is known that the NO₂/NO_x ratio in traffic emissions is assumed to increase which might be one explanation for the observed trends in NO₂ concentrations in particular near streets. NO₂ concentrations do not show a clear trend despite the decreasing total NO_x (NO + NO₂) emissions during the last 20 years. Are NO and NO₂ emissions treated separately in the treatment of the emission data? Or is a fixed NO₂/NO_x ratio used for the different source categories (SNAP 1, 2, 7). This point is of particular importance for SNAP 7 (traffic).

The emission data are handled in terms of NO_x. Hence, the temporal profiles are applied to the NO_x emissions. Only in the final step before the emissions are added into the model domain the NO and NO₂ fractions are taken into account. For all sectors, 3% is emitted as NO₂ and the rest as NO, thus there is a fixed NO₂/NO_x ratio used in the model. The NO₂/NO_x ratio is currently under investigation. It is generally thought that the primary NO₂ fraction has an impact on the roadside concentrations but that regional and urban background concentrations are not affected. As no street locations are included in the analysis the impact of the fixed NO₂/NO_x is assumed to be not very relevant here.

p.4, Method and data

How are the boundaries of the models handled, lateral and upper boundary?

We will add information on this in section 2.1 (p. 19316, l.8) "The boundary conditions are obtained from the MACC near-real-time forecasts as produced by the IFS/MOZART coupled system (Flemming et al., 2009). These enclose the LOTOS-EUROS domain at the lateral as well as the upper boundary."

Flemming, J., Inness, A., Flentje, H., Huijnen, V., Moinat, P., Schultz, M.G., Stein, O.: Coupling global chemistry transport models to ECMWF's integrated forecast system, Geosci. Model. Dev., 2 (2009), pp. 253–265.

p.7, Improved emission time profiles

The sentence "Note that a single diurnal profile is applied for all days of the week" is not completely clear to me. Are there different specific temporal profiles for each day of the week, i.e. different e.g. for Monday and Tuesday, ..., or is it the same profile for all days of the week, or is there only a differentiation between working days (Monday til Friday), Saturday and Sunday.

We will change the sentence to: "Note that the same diurnal profile is applied for all days of the week (Monday to Sunday) per SNAP category."

Looking at the different vehicle and road types: Is there a difference in NO₂/NO_x, VOC-split expected? How are the VOCs handled?

The VOC and NOX split are fuel and technology dependent. So yes, a variation between vehicle type and road type will be present. We kept them constant here for simplicity. This is mentioned in the emission database description. Moreover, we highlighted the issue at the end of section 3.1 (p. 19321, l. 24) where we added: "Although not in focus here, the additional detail also allows to specify the technology dependent composition of the emissions in more detail, e.g. VOC-split and NO2/NOx fraction." A separate study is presently ongoing to investigate the impact of primary NO2 fraction on modeled NO2 distributions. First results show a very low impact on regional levels.

p. 10, 11; SNAP 1

How is the height distribution of emissions from power plants? Does the EUROSLOTOS model resolve height dependencies in that case? How?

We will add information on this in section 2.2 (p.19318, l.9): "Emissions are distributed in the vertical following the profiles defined for the EURODELTA project (Thunis et al., 2008). For example, for SNAP1 8% of the total emissions are emitted in a height range between 170 and 310m, 46% between 310 and 470m, 29% between 470 and 710m and 17% between 710m and 990m. Since the height of the model layers depends on the mixing height the distribution of the emission to the model layers is re-computed every time step. Emission composition (e.g. VOC-split) was kept constant throughout all simulations."

Thunis. P., C. Cuvelier, P. Roberts, L. White, L. Post, L. Tarrason, S. Tsyro, R. Stern, A. Kerschbaumer, L. Rouil, B. Bessagnet, J., Builtjes, M. Schaap, G. Boersen, R. Bergstroem (2008). Evaluation of Sectoral Approach to Integrated Assessment Modelling including the Mediterranean Sea. Eurodelta II report. EUR 23444 EN . DOI 10.2788/87066. http://aqm.jrc.it/eurodelta/publications/EDII_finalreport.pdf

p.12, discussion of different model runs in chapter 4: if hourly values are available it might be interesting how sensitive the results of LOTOS-EUROS are with respect to the limit values as given within the EU air quality directives e.g. the number of exceedances of the 24h-average of 50 ug/m³ for PM10. This might help to show directly the importance of the temporal variations of emissions on measures currently considered as relevant within the EU directives.

As the annual mean concentrations for PM10 only differ very little between the different model simulations and they are highly underestimated it is not very likely that there is a remarkable impact on the number of exceedances of the 24h-average. Therefore we decided to not show results on this.

Why is the analysis limited to observations in Germany?

Please, see comment above.

Anonymous Referee #2:

The methodology by which the time profiles for SNAP sector 1 have been obtained seems to me as non optimal. An elaborate model is used for solar and wind energy (REMIX), while these represent 0.4% and 3.7% of the total electricity production respectively, while a zero hypothesis is made for nuclear and hydroelectric production, assuming constant production throughout the year and day. This hypothesis is defensible for nuclear power. However, it is well-known that hydroelectricity from mountain areas (lakes) is used by energy producer as a quickly adjustable energy source allowing to absorb the peaks in demand. Since the part of hydro power is 16% in Europe against 4.1% for other renewables, the hypothesis made seems not valid at European scale.

Using ReMIX in the paper for solar and aeolian energy (a total 4.1% of electricity production for Europe) suggests that the treatment of SNAP 1 is very accurate. While this may be the case for countries such as Denmark where the energy mix is massively dominated by fossil (and biomass) burning, aeolian and solar energy without a significant part of mountain hydroelectricity, this is certainly not the case for countries relying significantly on mountain hydroelectricity, including at least Switzerland, Austria, France, Norway, Italy. For these countries, the hypothesis made is certainly not justified and the results of the method presented are not reliable. For Germany in 2005, I found the corresponding figures:

Total production : 620.6 tWh

Total production through burning (coal + oil + biomass + waste burning): 384.7 tWh

Eolian production : 27.2 tWh

Solar production : 1.2tWh

Hydroelectricity : 19.8 tWh

(figures found on a website http://energeia.voila.net/electri/allemande_nucle_renov.htm - , itself referring to AGEB (AG Energiebilanzen) 2013 and AGEE-Stats (AG Erneuerbare Energiern - Statistik) 2013

Therefore, the hydroelectric production, and its variations, are far from negligible even for Germany (same order of magnitude as aeolian)

While a detailed treatment of emissions from SNAP 1 would probably go beyond the proposed study, I recommend that the limits of the proposed methodology are explicitly stated:

* In the text, by explicitly stating that hydroelectric production is highly variable in time and that the assumption that it is constant is very crude, and not adapted for countries where mountain hydroelectricity is important, such as the ones listed above

* In the text, by analyzing the relative importance of all power sources in Germany (and if possible other major european countries) and stating that assuming constant hydroelectric production might lead to overestimation of emission peaks by fuel burning (hydroelectric power from mountain lakes is used to absorb part of the peaks in demand w/o recurring to fuel burning).

* In Fig. 1, besides the "LE-default profile" and "New profile" curves, by plotting a curve representing SNAP1 emissions calculated using the assumption that solar and wind energy are constants too like the hydro energy. This would permit the reader to evaluate the quantitative impact of using the ReMIX model on the modelled SNAP1 emissions.

Also, the impact of the other components of SNAP1 (liquid and solid fuel transformations) should be briefly addressed (is it neglected, supposed to have the same time profile as energy production?)

We addressed these points by rewriting large parts of the section 3.3, which is included completely in this document. Here, we would like to respond shortly on the major points of the above mentioned comments. We now show and discuss that the impact of accounting for the intermittent renewables sources (wind and solar) on the variability of new time profiles is only limited. This is illustrated by an additional curve in Figure 4b in which also solar and wind energy is assumed to be constant. Thus using the ReMIX model to construct the new time profiles does not add much information. Hence, following the country specific electricity demand had a much larger impact on the new time profiles than accounting for intermittent renewable electricity production.

We also added more information on the contribution and the temporal variability of hydroelectricity power production and the possible impacts of the crud assumption of constant hydroelectric production. As no data for the variability of hydroelectric power production over time were available, this assumption is made in this study, which is another reason why we focused on Germany, where hydro power is less important than in other countries.

The following text represents the revised section 3.3.:

3.3 SNAP1 – combustion in energy and transformation industries

The temporal variability of SNAP1 is assessed by focusing on the power generation sector, as the contribution of emissions from power plants dominates the total SNAP1 emission for all pollutants, except NMVOC. As for the other sectors the default emission profiles for the power sector (SNAP1) are assumed to be the same across all countries and invariable with meteorology. This may not be the best representation of reality, since e.g.:

- climate conditions may cause differences in seasonal profiles for countries across Europe;
- variations in electricity consumption (e.g. for heating/cooling) due to changes in meteorology during the year are not represented;
- variable social habits may induce shifts in diurnal cycles between countries.

Therefore, new time profiles for the power generation sector (SNAP1) were constructed for 2006 using electricity demand data from each country. The electricity demand data for the year 2006 have been obtained from the ENTSO-E, the European Network of Transmission System Operators for Electricity. These demand data (in MW) are country specific time series of hourly data.

In Europe on average, 54% of the electricity is generated using fossil fuels (<http://epp.eurostat.ec.europa.eu>). Nuclear power and hydroelectric power account for 25 and 16% respectively. Intermittent renewable sources only produce a minor part of the total electricity demand (3.7% for wind energy and 0.4% for solar power in EU27). Between countries large differences in the electricity mix exist. For example, France has a much larger share of nuclear than average, whereas the same applied to Norway, Austria and Switzerland with respect to hydro. For Germany, the contribution of power generated using fossil fuels on the total electricity demand is on average 63.5%, for nuclear and hydroelectric power it is 26% and 5.4%, respectively and the contribution of the intermittent renewable sources are for wind 4.8% and for solar 0.3%. Hence, intermittent sources represent a comparable small part of the electricity mix in Germany but should be considered also because they are expected to become more important.

As only fossil fuels cause emissions during electricity production, we made a limited effort to subtract the power which is generated from the non-fossil fuel sources from the total electricity demand. Specific attention was given to the role of intermittent renewables. For this purpose, we used the REMix model (Scholz, 2012). REMix is an energy system model which generates electricity supply systems based on electricity demand and an inventory of

the maximum installable capacity of different electricity generating types and technologies, potential power generation in each hour of a specific year (for intermittent sources like solar and wind), and the costs of technologies. Based on these input parameters, the model can calculate the optimal mix of electricity generation sources based on the user request, e.g. finding the most economical solution or by using a fixed percentage of renewables. For this study, the REMix was used to calculate the time series of electricity production from solar and wind such that the installed capacity meets the annual contribution of the sources for each country.

For nuclear and hydropower, the electricity generation is assumed constant throughout the year. For nuclear electricity, this assumption is justified since nuclear power plants tend to run at constant capacity throughout the year, maintenance excluded. Hydroelectricity power production, however, is highly variable over time because of water availability for run off-river hydropower plants and because hydroelectricity plants taking water from weir reservoirs are often used as energy reservoir in times of low demand and electricity production is increased when demand peaks. The power output from hydroelectricity plants can be adjusted to meet demand easily. However, as no data for the variability of hydroelectric power production over time were available, the admittedly crude assumption of constant production was made in this study. Hence, this is another reason why we focus the evaluation on Germany, where hydro power is less important.

By subtracting the time series of solar and wind energy production as well as the constant contributions of other electricity sources from the hourly demand data, an approximation of the hourly pattern for the fossil fuel emissions remains. These data are used in a relative way to distribute the annual emissions from fossil fuel based power plants over all hours in the year. The new seasonal time profile for Germany show a stronger temporal variability between the months and weeks compared to the default LOTOS-EUROS profiles (Fig. 4b). The weekly cycle is more pronounced with higher amplitude caused by higher emission factors during the week and decreased factors on the weekend. This is especially pronounced in the summer months where emission at peak production is much higher than in the default profiles. Furthermore, the yearly minimum is shifted to spring and autumn months. Zooming in on a summer week, the daily cycle for the new timing shows peak values in the morning and late afternoon whereas the afternoon peak is not present in the base case (not shown).

In Figure 4b the impact of accounting for the intermittent renewable sources is quantified by comparing to the situation where also wind and solar are assumed to be constant. The day to day variability of the emission factors is slightly smaller when the renewables are assumed to have a constant production over the year, because meteorological characteristics causing the intermittency in renewable electricity production are not accounted for. The seasonal variability is hardly affected at all. Hence, following the country specific electricity demand had a much larger impact on the new time profiles than accounting for intermittent renewable electricity production.

For the generation of SNAP1 profiles, the implicit assumption made in this study is that the electricity generated within a country is used within the same country, i.e. no cross-border transport of electricity is assumed. Another major assumption is that storage of electricity is excluded, i.e. electricity which is produced in a certain hour needs to be used in that same hour. Combined with the assumption that hydroelectricity production is constant, this means that the variability in emission timing from fossil fuels used in this study is likely to be an overestimation of the variability in reality. Significant improvements in the time profiles for SNAP 1 are possible with a much more detailed energy model.

The contribution of emissions from power plants dominates the total SNAP1 emission for most pollutants (except NMVOC). As an example, according to the German official emission inventory for the year 2005, 227kton SO₂ and 9.79 kton primary anthropogenic PM₁₀ respectively, are emitted from power plants whereas 63kton SO₂ and 1.59kton primary PM₁₀

from other SNAP1 sources. In this study it is therefore assumed that the same profiles as constructed for the power plans also apply for the other sources.

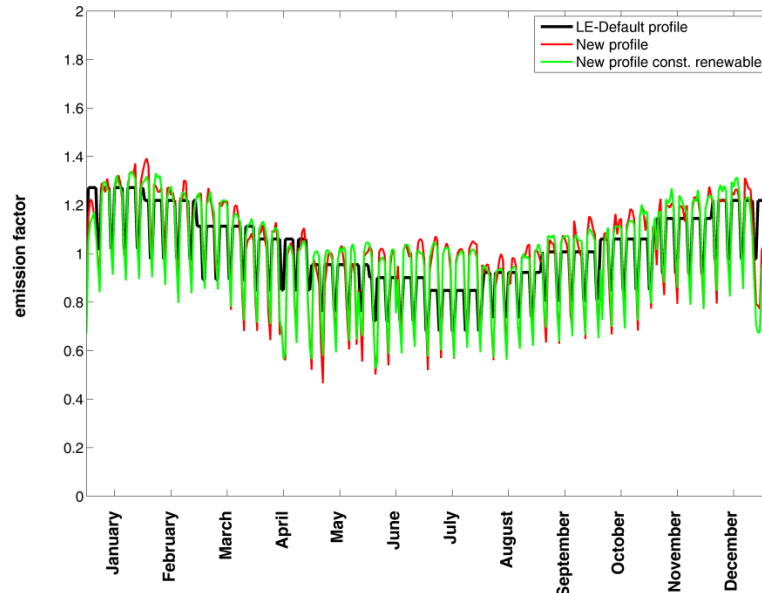


Figure 4. Comparison between the new and the default seasonal (daily) emission factors for SNAP2 (a) retrieved from model grid cells located in Germany (about lon = 10°E, lat =2.5°N) and in France (about lon= 2.75°W, lat = 43.75°N) and for SNAP1(b) for Germany. The green curve in figure 4b represents the time profiles when all renewable sources are assumed constant.

Actually, the improvement in correlation rates for SO₂ is much more significant in the LE_SNAP2 simulation than in the LE-SNAP1 simulation, even though SNAP1 contributions to SO₂ emissions is largely dominant compared to SNAP2. This should be commented in the text and, in my opinion, shows that the improvement brought by the new SNAP1 profiles compared to the LE_default simulation is moderate : either because the default profiles for SNAP1 are already fairly good, either because the methodology used to make the new profiles is not appropriate.

In order to address this aspect in the text, we will add the following sentences to the discussion in section 5 (p.19331, l.:16): "The improvement of the correlation coefficient for SO₂ is higher in the LE_SNAP2 than in the LE_SNAP1 simulations, which is counter intuitive as the SNAP1 contribution to total SO₂ emissions is higher than for SNAP2. A part of the explanation could be the application of the SNAP1 profiles to a limited number of point sources of which the impact at ground level is lower per unit emission than for area sources. In addition, the small improvement due to the new SNAP1 profiles hint that the default SNAP1 profiles may be quite reasonable for the sector. The latter is supported by the low change in profiles as illustrated in Fig. 4b."

– The methodology for estimating the diurnal cycle of SNAP 2 is unclear to me (Fig. 1a). I would expect heating-related combustion to be relatively constant throughout the day, which is not the case. While the peaks in the morning and evening may be related to heating of water and/or cooking, the very marked nighttime minimum seems hardly understandable to me. This should be discussed, since this diurnal cycle is also used for the revised profiles.

We will add the following explanatory sentences to section 3.2 (p.19321, l.26): "The default diurnal time profiles reflect the increased heating in the morning and evening to warm houses up as well as the cooking and warm water production activities when people are at home. Compared to residential houses, office buildings are more modern, cover a lower volume and are more efficient to heat and therefore emission amounts are assumed to be lower during the day. As the people turn the heating down at night and other activities relevant for SNAP2 are much lower during the night, the emission factors show a minimum at that point in time."

Other specific comments:

- p. 19316, l. 6: "the model used the dynamic mixing layer approach to determine the model vertical structure". I think that the following article, for example, should be referred to at this point in order to explain what is meant by "dynamic layer approach" (or another article where this methodology is described in more detail): Source apportionment using LOTOS-EUROS: module description and evaluation (by R. Kranenburg, A. J. Segers, C. Hendriks, and M. Schaap, *Geophys. Model Dev.*, 2013)

We will add this reference to the text.

l. 8: "the ECMWF model" is too vague, some precision is brought in Tab. 2, which is referred to only later (p. 19318). Some precisions on the meteorology should be brought at that point. Also, for a continuous 1-year run, one would expect that the ERA-interim reanalysis is used and not discontinuous forecast runs. A brief discussion on that point would be appreciated, stating what is the purpose of using forecast runs and not reanalysis data. The boundary conditions for chemical concentrations at the model top and model boundaries are not specified. They are potentially important since the model is of limited height (3.5km).

We will add some more information on the boundary conditions and meteorological input data in section 2.1 and refer to table 2, (p. 19316, l.8): "The model is driven by short-range meteorological forecasts (0-12 hour) from the ECMWF Operational Data stream (Table 2). Forecast data is used to ensure physical consistency in the data, which might be slightly lost during analysis, and because this data is available from the operational air quality forecasts in which the model is also used. For the experiments in this study the meteorological data is retrieved at a horizontal resolution of 0.50° longitude x 0.25° latitude and interpolated to the model grid if necessary; temporal resolution is three hourly, with linear interpolation applied to obtain an hourly resolution. The boundary conditions are obtained from the MACC near-real-time forecasts as produced by the IFS/MOZART coupled system (Flemming et al., 2009). These enclose the LOTOS-EUROS domain at the lateral as well as the upper boundary."

*J. Flemming, A. Inness, H. Flentje, V. Huijnen, P. Moinat, M.G. Schultz, O. Stein. Coupling global chemistry transport models to ECMWF's integrated forecast system *Geosci. Model Dev.*, 2 (2009), pp. 253–265.*

- p. 19317, l. 14: "Natural emissions are calculated on-line using the actual meteorological data". It should be stated which model/approach is used, referring to the corresponding publication.

In the following lines of the text (ll. 15 -18) this is described in more detail with references. "Biogenic NMVOC and mineral dust emissions are prescribed following Schaap et al. (2009). Sea salt emissions are calculated following Mårtensson et al. (2003) and Monahan et al. (1986) from wind speed at ten meters. The MACC global fire assimilation system (Kaiser et al., 2009) is used on an hourly basis."

- p. 19318, l. 21-22: "Using emissions for the year 2005". Should probably rephrased something as "using the total annual emissions for year 2005 as described above"

We will change that in the text to: "All model simulations have been performed using annual emission totals for the year 2005 and the meteorology of the year 2006."

- p. 19319, at the end of the 2.3 paragraph: the choices made for averaging need to be precised further. In particular, one can observe that the annual mean of the hourly data (Tab. 3) is distinct from the annual mean of daily averaged data (Tab. 4), which is not a natural property. I guess that the daily average has been computed only for days for which there was no missing data, therefore retaining less data than for the hourly statistics. In any case, a quick precision should probably be added. It is also needed to state explicitly how many Airbase stations have finally been retained for rural and urban locations.

The annual mean of hourly and daily data are based on different stations, as on some stations hourly data and on others daily data were available. The annual mean for a station was only calculated if a minimum of 60% data coverage for 2006 was available. We will add the number of stations included to calculate the annual means to the captions of table 3+4. We will add/change the following text to the paragraph (p. 19318, l.24): "Air pollutant measurements at German stations with an hourly or daily time resolution from the AirBase database (AIRBASE, 2012) were selected and acquired. Only time series with a minimum of 60% data coverage for 2006 for an individual component and time resolution were chosen for the evaluation. Model data are neglected if no measurements are available on a specific hour or day in the time series. Note that for the hourly and daily time series of the individual components the (number of) stations included in the measured and modeled annual averages are variable. For the horizontal grid resolution of about 25x25km² mainly rural background stations are representative. However, because the focus of this study is also on SNAP7 and SNAP2, which are dominant in urban regions, also urban background stations are included in the statistic. This is despite the fact that the absolute concentration is highly underestimated at these stations. For the interpretation of the results the main interest is on the ability of the model to reproduce the temporal variability of the measured pollutant concentrations. Therefore, mainly the results for the correlation coefficient are discussed in the result section 4."

p. 19320, l. 16-17: "to obtain a profile representative for Germany as a whole." Is this profile the combination of annual, weekly and hourly profiles as in the earlier "default" approach?

This profile is a combination of annual and diurnal cycle per day of the week. This is in contrast to the default time profiles, which did not take into account differences between the daily cycle on weekdays and weekend. We will add some information on this in section 3.1. (p.

19320, l. 15): "Therefore, all traffic data were averaged across all urban and highways sites, respectively, to obtain profiles which are representative for all German urban and highway streets. These profiles are based on annual profiles representing the emission factors per month and profiles representing every hour of the week. The annual profiles are constructed by averaging the available traffic count data for the urban and the highways, respectively, per month and related them to the total traffic in the year. The same has been done for every hour of the week."

Also, I find Fig. 2 not very informative, particularly since one particular (and not precised) highway and one urban station have been picked, so the reader has no indication whether the presented time-series are representative for Germany. Also, why has year 2010 been chosen for Fig. 2, whereas year 2006 is studied in the rest of the study?

The figure 2 illustrates the impact of time profiles which are derived from actual traffic counts at highway and urban stations in Germany. With these profiles systematically more of the observed traffic counts at the two German stations can be explained in contrast to the default time profiles which are originally constructed for the Netherlands and from traffic counts at urban stations. We think that these figures stress very well the importance to consider the different street types for the emission time profiles and that this already has a considerable improving impact. As it is not possible to show the average effect for all stations we choose two example stations in the Ruhr area (Bottrop - urban station) and close to Hannover (Lauenau - highway station) in Germany. We will add this information to the caption of figure 2. There is not a special reason why the time series is shown for the year 2010 but as the time profiles are based on traffic counts of the years 2006-2010 and they are also considered to be representative for all years it is not of considerable importance which year is chosen.

"Figure 2. Time series of the differences between actual traffic counts and the application of the default and the new urban and highway time profiles at an urban street station (Bottrop) (a) and a highway station (Lauenau) (b) in Germany for the year 2010."

p. 19321, l.7-9: it seems important to me that the authors precise the source of the emission factors they have used.

We added information to that in section 3.1. (p. 19322, l. 9): "This split has been made using the underlying data in the TNO_MACC inventory which is described in detail in Denier van der Gon et al. (2010). The split uses information from the IIASA RAINS model (Amann et al., 2005) for the differentiation in vehicle types and the TREMOVE model (De Ceuster et al., 2005) for differentiation in network types. The split takes implicitly into account different emission factors for different networks and vehicle types as identified by the IIASA GAINS model and the TREMOVE model. The TNO spatial allocation procedure contains major highways on grid and by traffic intensity, which makes it possible to distribute highway emissions according to traffic intensity over the major highways, while emissions from urban and rural roads are distributed using population. From this result, for each grid cell the share of vehicle types and network types were determined."

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., Firello D. (2005), TREMOVE 2.30 Model and Baseline description. Final report, 18 February 2005. Transport and Mobility Leuven. <http://www.tremove.org>.

Amann M., Bertok I., Cabala R., Cofala J., Heyes C., Gyarmas F., Klimont Z., Schöpp W., Wagner F. (2005), A further emission control scenario for the Clean Air For Europe (CAFE) programme, International Institute for Applied Systems Analysis (IIASA).

p. 19323, paragraph 3.3: apart from the comment above ("major comments"), more precision is needed here: where have the time series for electricity demand been obtained? Are they available for other groups? Are they on a day-per-day basis (in this case, what temporal profile is used for the hourly disaggregation?) or on a hourly basis? Sentence "The energy system model can also dimension power supply systems with high shares of renewable energy and calculate the least-cost operation of the system components, i.e. power generators, power storage and power transmission units": this seems out of the scope of this study, I think this sentence should be replaced by brief precisions about how ReMIX works (hypotheses, data...).

We add information on this in the section 3.3. (Please, see also above): "Therefore, new time profiles for the power generation sector (SNAP1) were constructed for 2006 using electricity demand data from each country. The electricity demand data for the year 2006 have been obtained from the ENTSO-E, the European Network of Transmission System Operators for Electricity. These demand data (in MW) are country specific time series of hourly data." and "As only fossil fuels cause emissions during electricity production, we made a limited effort to subtract the power which is generated from the non-fossil fuel sources from the total electricity demand. Specific attention was given to the role of intermittent renewables. For this purpose, we used the REMix model (Scholz, 2012). REMix is an energy system model which generates electricity supply systems based on electricity demand and an inventory of the maximum installable capacity of different electricity generating types and technologies, potential power generation in each hour of a specific year (for intermittent sources like solar and wind), and the costs of technologies. Based on these input parameters, the model can calculate the optimal mix of electricity generation sources based on the user request, e.g. finding the most economical solution or by using a fixed percentage of renewables. For this study, the REMix was used to calculate the time series of electricity production from solar and wind such that the installed capacity meets the annual contribution of the sources for each country."

p. 19324, l. 5-6: "stronger temporal variability between the months and weeks compared to the LOTOS-EUROS profiles": on Fig. 4b, I see that the variability is stronger between different weeks in the new profiles (by construction), but I would say that the variability between the months is similar between both models. The secondary peak in summertime (Fig. 4b) is very interesting and it would be nice to have an interpretation: increased demand due to climatisation utilities? Lower aeolian production in summertime? This was not captured by the previous profiles and is a very interesting feature in my opinion.

We changed and added the following to the section 3.3: "The contribution of emissions from power plants dominates the total SNAP1 emission for most pollutants (except NMVOC). As an example, according to the German official emission inventory for the year 2005, 227kton SO₂ and 9.79 kton primary anthropogenic PM₁₀ respectively, are emitted from power plants whereas 63kton SO₂ and 1.59kton primary PM₁₀ from other SNAP1 sources. In this study it is therefore assumed that the same profiles as constructed for the power plans also apply for the other sources."

p. 19326, the analysis of the absolute bias in Tab. 3 (bias of the order of 100% of modelled value for NO₂, SO₂ and PM₁₀ for urban stations) seem to indicate that, according to the relatively coarse model resolution, the modelled concentrations are representative only of rural background stations). The correlation coefficients for urban stations should therefore be examined more carefully, and these large biases for urban stations need to be commented in the text.

In order to comment this in the text the following sentences are included in section 2.3 (p.19318, l. 27): "For the horizontal grid resolution of about 25x25km² mainly rural background stations are representative. However, because the focus of this study is also on SNAP7 and SNAP2, which are dominant in urban regions, also urban background stations are included in the statistic. This is despite the fact that the absolute concentration is highly underestimated at these stations. But for the interpretation of the results the main interest is on the ability of the model to reproduce the temporal variability of the measured pollutant concentrations and the impact of the new time profiles on this. Therefore mainly the results for the correlation coefficient are discussed in the result section 4."

The aspect of the horizontal grid resolution and its effect on the results, especially at urban stations, is also mentioned in the outlook (section 5, p. 19332, l. 17) now: "Another important aspect is to test the impact of the time profiles on the model performance as a function of the horizontal grid resolution. The representativeness of urban background stations increases with the grid resolution and therefore also the impact of more detailed emission time profiles could increase especially for the SNAP categories 2 and 7."

p. 19328: for SNAP2, it would be interesting to mention the increase in correlation rates for the winter months (e.g. Dec.-Jan.-Feb. Mar.): this would permit to isolate even better the increased value of the new profiles for winter pollution, since the impact in summer can be assumed almost negligible.²

We add some information and number in section 4.3. (p. 19328, l. 12): "The difference between the default and the new constructed time profiles for SNAP2 is highest during the months January to March (Fig. 4). At urban stations the average correlation coefficient for SO₂ for this period is smaller for both the LE_Default (0.57) and the LE_SNAP2 (0.6) simulations compared to the results for the whole year presented in Table 3. Thus the increase of the correlation for this short period is comparable to that for the whole year."

p. 19330, l. 18-20, "the maximum difference ... larger than the impact of the improved emission profiles", this sentence is vague, it should be either removed or written more precisely and backed with a reference.

We will change the sentence to: "The following examples, taken from the literature, show that the maximum difference of correlation coefficients between individual CTMs is normally larger than the impact of the improved emission profiles. However, but these model comparison studies often show several models with very similar correlation coefficients."

p. 19331, l. 11, probably add "for SNAP7" after "profiles".

We will add that in the text.

l. 11-12: the current formulation suggests that national profiles have been generated for several countries in this study, while this has been presented only for Germany.

We will change that sentence to: "Replacing the default (Dutch) profiles with national (German) representative profiles yielded important improvements of the explained variability over the weekdays as well as the diurnal cycle, which was also found by Pierce et al. (2010) and Menut et al. (2012)."

p. 19332, l. 13-15: the Skjoth et al. study is already mentioned just before with a similar sentence (p. 19331, l. 22-25), one of these mentions should be removed

We will remove the sentence in l. 22.

Comments regarding the tables and figures specifically

table 2: the domain definition (10W-40E) does not correspond to the text (15W-35E)

We will change the domain definition to 15°W – 35°W in the table.

Emission -> Emissions (second column)

We will change that in the table.

"for SNAP 7 the new profiles were used for Germany and the Netherlands": is there a reason to apply these profiles to these 2 countries and no other countries ? This probably needs to be briefly discussed in the text.

We constructed the time profiles for SNAP7 specifically with German traffic data only for Germany. So far such country specific emission time profiles were also applied for other countries. In this study we decided to only use the time profiles for the country they are based on. But to test the performance of the more detailed time profiles on another country they were also applied for the Netherlands in the LE_SNAP7 and LE_SNAP127 simulations. As this is not relevant for the results described in the study we will remove the sentence from the table.

Tables 3-4: the bias of the simulated values relative to measured values should be given, not the reverse. Therefore, the sign of all the columns titles "Bias" should be changed (a value below the observation is considered as negatively biased).

We will change that in the tables.

Fig. 2: precise the name and location of the stations used for the plot.

We will add this information to the caption of figure 2: "Figure 2. Time series of the differences between actual traffic counts and the application of the default and the new urban and highway time profiles at an urban street station (Bottrop) (a) and a highway station (Lauenau) (b) in Germany for the year 2010."

Fig. 4a, the legend ("four locations in different countries") does not seem to fit the text. Please check, precise the location (coordinates) within France and Poland, and add the time series one or two sites in Germany which is the most useful here.

The referee is right, the caption of the figure 4 is now changed to: "Figure 4. Comparison between the new and the default seasonal (daily) emission factors for SNAP2 (a) retrieved from model grid cells located in Germany (about lon = 10°E, lat = 52.5°N) and in France (about lon= 2.75°W, lat = 43.75°N) and for SNAP1(b) for Germany.". There is also a mistake in legend of the figure it should be Germany instead of Poland. So for the rest the figure stays the same.

Technical changes

p. 19316, l. 15: "below cloud" ->-> "below-cloud"

p. 19319, l. 24: "our" -> "hour"

p. 19329, l. 22: Increased -> Improved ?

p. 19331, l. 8: This -> Therefore

p. 19332, l. 2: check "Across per"

We will change that in the text.