

General comments

The paper examines the impact of modifying the temporal variability of emissions from SNAP sectors 1, 2 and 7 on the realism of pollutant concentrations simulated with the LOTOS-EUROS CTM. This paper is written in a clear language and its structure is clear enough as well, except for some particular sections. It arrives at a time when a couple of recent articles addressing partly the same question have already been published (Menut et al. 2012, Bessagnet et al. 2012, De Meij et al. 2006), however this study is different from the above-mentioned studies and bring new aspects.

The authors propose alternative methodologies for the temporal redistribution of annual total emissions. For SNAP sector 1, this is based on the use of the time series for national total electricity demand, on one side, and on the other side on the modelling of electricity production by solar and eolian facilities, based on the ReMIX model. For SNAP sector 2 (essentially urban heating), the new profiles are based on a degree-day approach according to modelled meteorological fields, following Bessagnet et al. (2012). For SNAP sector 7, the new profiles are based on traffic-count data at a variety of stations (urban and highway and distributed between light and heavy vehicles). While the methods proposed for SNAP sectors 2 and 7 seem convincing, I have strong concerns about the method used for SNAP sector 1, essentially because the time-variability of hydroelectric energy production is neglected.

The authors perform 6 simulations on a European domain in order to evaluate the impact of the new profiles compared to constant profiles and compared to the actual ("default") profiles. By comparison to Airbase measurements for Germany, the authors show that the new profiles have a significant and positive impact on the model outputs, particularly for NO₂ (SNAP7 effect) and SO₂ (SNAP2 effect).

The main result is that small but significant added value can be obtained in a CTM simulation by updating the time-variation profiles of sectors 1,2 and 7 (extending the results of Bessagnet for sector 2 and Menut for sector 7), using country-specific factors. This study goes a little bit beyond the above-mentioned studies, first by examining SNAP sector 1 as well, also by examining separately and all-together the effect of these three SNAP sectors.

The strategy presented to evaluate traffic-related emissions using detailed countings separating highway and urban contributions, and Heavy-duty and high-duty contributions is also novel and interesting.

Another interesting and original aspect of this work is that using NO₂ measurements to validate model outputs is touchy since the measured time-series (daily cycle) depend highly on the measuring technology (photolytic vs. Molybdenum technique). While this result is not new (adequate reference is provided in the text, e.g. Villena et al. 2012), this result is important and of interest to the community of ACP readers.

However, some points of the methodology are not clear enough, particularly for SNAP sector 1 and, to a lesser extent, SNAP sector 2 (see below « Comments about the treatment of SNAP sectors 1 and 2 »).

I think this paper shall be published in Atmospheric Chemistry and Physics after major revisions, including a more thorough discussion on the hypotheses made in the treatment of SNAP sector 1 and their consequences, as well as complementary explications for the diurnal cycle in SNAP2.

- 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.

Comments about the treatment of SNAP sectors 1 and 2 :

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 AGEBA (AG Energiebilanzen) 2013 and AGE-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 adressed (is it neglected, supposed to have the same time profile as energy production ?)

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.

- 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.

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)

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 precised. They are potentially important since the model is of limited height (3.5km).

- 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

- 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"

- 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.

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 ? 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 ?

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

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...)

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.

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.

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

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

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

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.

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

Comments regarding the tables and figures specifically

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

Emission -> Emissions (second column)

"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.

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).

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

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.

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"