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TNO report

Scale dependency of source receptor matrices for air pollution across Europe

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Summary

Within the EMEP TFMM an initiative was taken for a new model exercise to analyze the model performance of different chemical transport models as a function of resolution. However, the exercise does not address the impact on source receptor matrices (SRMs), although SRMs are a key input to the integrated assessment modelling and thereby policy support. In this study we expand upon the TFMM exercise and used the LOTOS-EUROS model to assess the impact of model resolution on calculated source receptor matrices.

The SRMs were calculated for three resolutions being 56, 28 and 14 Km. Impacts of the model resolution were illustrated for nitrogen dioxide, sulphur dioxide, sulphate, nitrate and anthropogenic PM10.

It was found that the model on a 14 Km resolution predicts larger contributions of a countries to the concentrations in its own territory than the current 56 Km resolution. The impact range from a few percent for large countries to tens of percents for smaller countries. The impact was found to be dependent on the components life time and its primary or secondary nature. For example, maximum fractional changes for NO2, PPM, SO2, SO4 are 1.81, 1.80, 1.56, 1.27, respectively

For large countries the impact of their emissions to surrounding countries becomes lower at higher resolution. This impact is smaller than the change for smaller countries on themselves.

The reason for the impacts is that the borders are much sharper represented at the higher resolution.

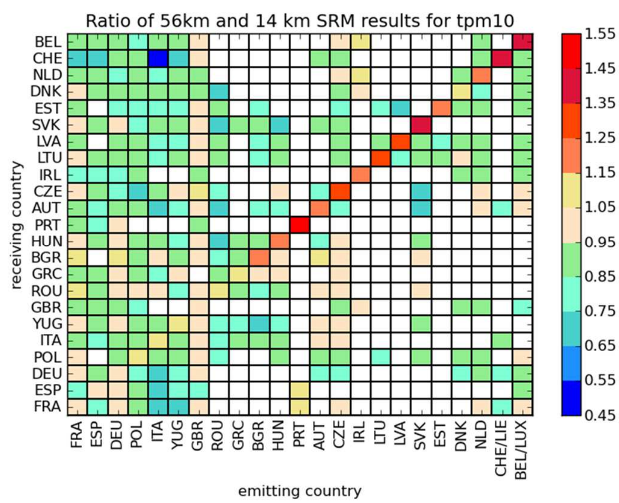


Figure 1. Ratio of the SRM at 14 and 56 Km resolution for anthropogenic PM10. On the x-axis the emitting countries are listed, whereas the y-axis represents the receiving countries. The countries were ordered going from large countries to smaller countries. Each cell represents the fractional change in the impact of a country on another. Positive numbers indicate the contribution at the 14 to be higher than at 56 Km resolution.

Contents

1 Introduction

Since 1999, the EMEP model has been run on a resolution of 50 * 50 km2 resolution. However, the last years, modification of the EMEP grid has been discussed. One of the important aspects of this change is the grid resolution. The input data (the most important being the emissions) need to be available on the same scale. A trade-off has to be made between the wish for as high resolution as possible and the computational costs. Therefore, it is important to determine the 'optimum resolution' – e.g. at which scale the improvement in resolution does not give improvement in performance any longer.

Within the EMEP TFMM an initiative was taken for a new model exercise to analyze the model performance of the different chemical transport models as a function of model resolution. However, the exercise does not address the impact on source receptor matrices, although SRMs are a key input to the integrated assessment modelling and thereby policy support. In this study we expand upon the TFMM exercise and use the LOTOS-EUROS model to assess the impact of model resolution on calculated source receptor matrices.

2 Methodology

In this study we use the LOTOS-EUROS model to assess the impact of model resolution on calculated source receptor matrices. Below we describe the general features of the LOTOS-EUROS model, the definition of the model simulations, the approach to assess the source receptor matrices (SRMs) as well as the approach the methodology to assess the model performance at the different resolutions.

2.1 LOTOS-EUROS

In this study we used LOTOS-EUROS v1.8, a 3-D regional CTM that simulates air pollution in the lower troposphere. Previous versions of the model have been used for the assessment of (particulate) air pollution (e.g. Schaap et al., 2004a; 2004b; 2009; Barbu et al., 2009; Manders et al., 2009; 2010). For a detailed description of the model we refer to Schaap et al. (2008), Wichink Kruit et al. (2012) and abovementioned studies. Here, we describe the most relevant model characteristics and the model simulation performed in this study.

The model uses a normal longitude–latitude projection and allows to specify the model resolution and domain within its master domain encompassing Europe and its periphery. The model top is placed at 3.5 km above sea level and consists of three dynamical layers: a mixing layer and two reservoir layers on top. The height of the mixing layer at each time and position is extracted from ECMWF meteorological data used to drive the model. The height of the reservoir layers is set to the difference between ceiling (3.5 km) and mixing layer height. Both layers are equally thick with a minimum of 50 m. If the mixing layer is near or above 3500 m high, the top of the model exceeds 3500 m. A surface layer with a fixed depth of 25 m is included in the model to monitor ground-level concentrations.

Advection in all directions is handled with the monotonic advection scheme developed by Walcek (2000). Gas phase chemistry is described using the TNO CBM-IV scheme (Schaap et al., 2009), which is a condensed version of the original scheme by Whitten et al. (1980). Hydrolysis of N2O5 is described following Schaap et al. (2004a). Aerosol chemistry is represented with ISORROPIA2 (Fountoukis and Nenes, 2007). The pH dependent cloud chemistry scheme follows Banzhaf et al. (2011). Formation of course-mode nitrate is included in a dynamical approach (Wichink Kruit et al., 2012). Dry deposition for gases is modeled using the DEPAC3.11 module, which includes canopy compensation points for ammonia deposition (Van Zanten et al., 2010). Deposition of particles is represented following Zhang et al. (2001). Stomatal resistance is described by the parameterization of Emberson et al. (2000a,b) and the aerodynamic resistance is calculated for all land use types separately. Wet deposition of trace gases and aerosols are treated using simple scavenging coefficients for gases (Schaap et al., 2004b) and particles (Simpson et al., 2003). The model set-up used here does not contain secondary organic aerosol formation or a volatility basis set approach as we feel that the understanding of the processes as well as the source characterization are too limited for the current application.

2.2 Model simulations

Four model simulations were carried out with different horizontal resolutions. The model simulations were specified following the specifications of the EC4MACS scale dependency exercise. The simulations were performed for the year 2009 for the EC4MACS domain encompassing Europe (Figure 2). Four resolutions were used doubling the resolution between each simulation. The spatial resolution ranges from a 56x56 Km resolution to 7x7 Km resolution. As the high resolution simulation is very demanding in terms of computing power the EC4MACS domain encompasses southern and central Europe completely, but cuts of the remote area in northern Scandinavia.

The anthropogenic emission input was harmonized by using a common EC4MACS emission dataset. The emission dataset was delivered by INERIS for all model resolutions separately. Except for SNAP 2, prescribed time profiles and height distributions were used following the EURODELTA protocol (REF). For SNAP2 daily gridded modulation factors were calculated based on temperature days (REF). For the SNAP 2 hourly variation the EURODELTA hour-of-the-day profile was used.

All other input parameters besides the anthropogenic emissions were not prescribed. For the present simulations the LOTOS-EUROS model was forced by ECMWF meteorology that was interpolated to the respective grid resolutions. Boundary conditions were obtained from …… For a more detailed specification of the model and its input data we refer to TABLE X.

Table 1 : Domains definition

Domain	**nx**	**ny**	**Lon. Res.**	**Lat. Res.**	**Kilometre scale**	**SW starting point Lon/Lat (grid centres)**
EC4M1	41	52	1.0	0.5	56x56	-10.000 / 36.125
EC4M2	82	104	0.5	0.25	28x28	-10.250 / 36.000
EC4M3	164	208	0.25	0.125	14x14	-10.3750 / 35.9375
EC4M4	328	416	0.125	0.0625	7x7	-10.43750 / 35.90625

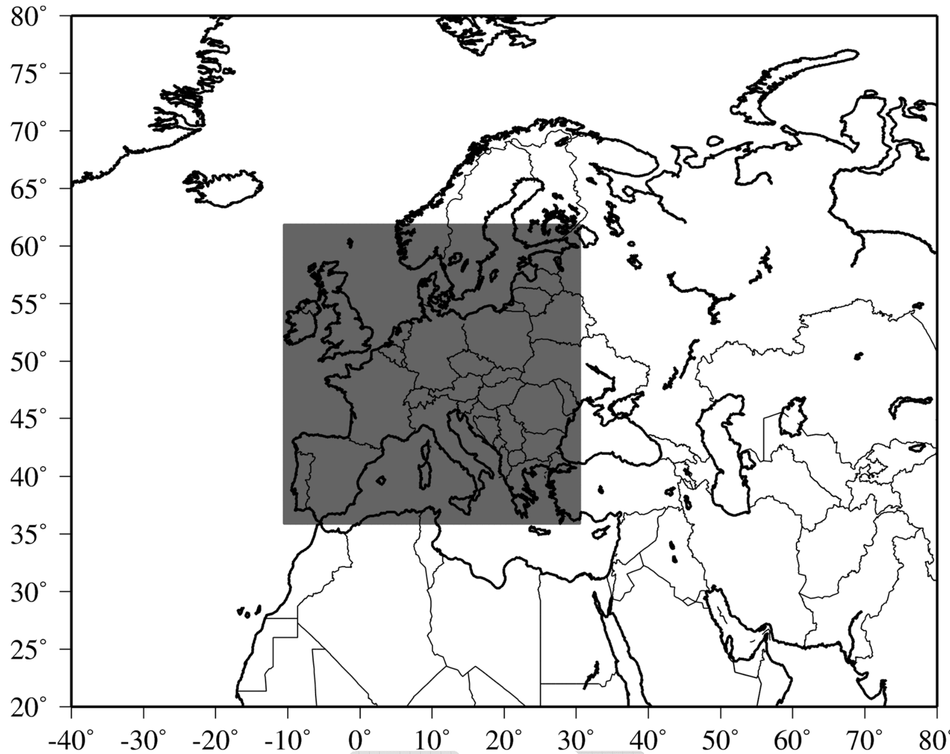


Figure 2 : Domain broken down in 4 resolutions types

2.3 Calculation of SRMs

The LOTOS-EUROS model is equipped with a dedicated labelling module to track the contributions of pre-defined source sectors throughout the model domain (Kranenburg et al., 2012). Besides the concentrations of all species the contributions of a number of sources to all components are calculated. The labelling routine is only implemented for chemically active tracers containing a C, N (reduced and oxidised) or S atom, as these are conserved and traceable. This technique is therefore not suitable to investigate the origin of e.g. O3 and H2O2, as they do not contain a traceable atom. The source apportionment module for LOTOS-EUROS provides a source attribution valid for current atmospheric conditions as all chemical conversions occur under the same oxidant levels. For details and validation of this source apportionment module we refer to Kranenburg et al. (2012). The system has been used to assess the particulate matter origin in the Netherlands by Hendriks et al. (2012).

In this exercise we use a slightly different way to visualize the SRMs than usually within the EMEP framework. Although the SRMs are country to grid maps, the SRMs were defined on the basis of countries. This means that the impact of each country was averaged for all the countries in the exercise. Hence, the concentrations due to the Netherlands in Germany were calculated as the mean over all cells that cover Germany. Cells containing borders were weighted according to the surface area of the countries in the cell.

To limit the required computation time 20 countries were chosen to be traced in the experiments described here. The selected countries are listed in Table 2. Countries labelled in this experiment. Selected countries had to be fully located within the EC4MACS domain. Therefore, emissions from several Scandinavian countries (Norway, Sweden, Finland) were not labelled. For convenience, a small number of countries were combined. For example, Switzerland and Liechtenstein as well as Luxemburg and Belgium were combined. The same was applied to all sea areas. To be mass consistent, all non-selected anthropogenic emissions, natural emissions and as well as the combined impact of initial conditions and boundary conditions were tracked as well.

Table 2. Countries labelled in this experiment

Country	**Country Code**
The Netherlands	NLD
Belgium and Luxembourg	BEL/LUX
United Kingdom	GBR
Ireland	IRL
Portugal	PRT
Spain	ESP
France	FRA
Italy	ITA
Germany	DEU
Denmark	DNK
Austria	AUT
Switzerland and Liechtenstein	CHE/LIE
Czech Republic	CZE
Slovakia	SVK
Poland	POL
Hungary	HUN
Romania	ROU
Bulgaria	BLG
Greece	GRC
Former Yugoslavia (Slovenia, Montenegro, Serbia, Kosovo, Bosnia and Herzegovina, Macedonia	YUG
Lithuania	LTU
Latvia	LAT
Estonia	EST

The labelling simulation for the 20 country labels requires about a factor 5 more computing power than a normal simulation. Moreover, it requires a lot of memory. Therefore, it is not possible to assess the SRMs at the highest resolution without going to a super computer for which the code has not been adapted. Therefore, the SRMs were calculated for the 56, 28 and 14 Km resolutions.

2.4 Model evaluation methodology

For the model evaluation methodology we refer to the TFMM exercise note.

3 Impact of model resolution

3.1 Modelled distributions

Figure 3 and Figure 4 show the annual mean concentrations of NO2, O3 and PM10 as calculated for the four different horizontal grid resolutions. For NO2 going from the coarse grid M1 to the fine grid M4 the concentration pattern increasingly reflects the underlying emission pattern. The calculated concentrations increase in particular in the high emission density areas. Overall, the increase in structure is tremendous. Especially going from 56 to 14 Km add a lot of detail, whereas the step to 7 Km does not show such a large change in the structure.

Also for PM10 the concentration pattern reflects increasingly the underlying emission pattern going to higher resolution, but much less pronounced than in the case of the NO2 concentrations, because the secondary aerosols, which provide a large part of the total PM10 mass, are much less affected by the grid size than the primary PM components. The calculated concentrations increase in particular in the high emission density areas.

Compared to NO_2 and PM_{10}, the effect of a decreasing grid size is small for ozone. In general, there are only small changes in rural areas. In urban areas, a decrease of the grid size leads also to a decrease of the calculated ozone concentrations as titration by local NO sources is enhanced.

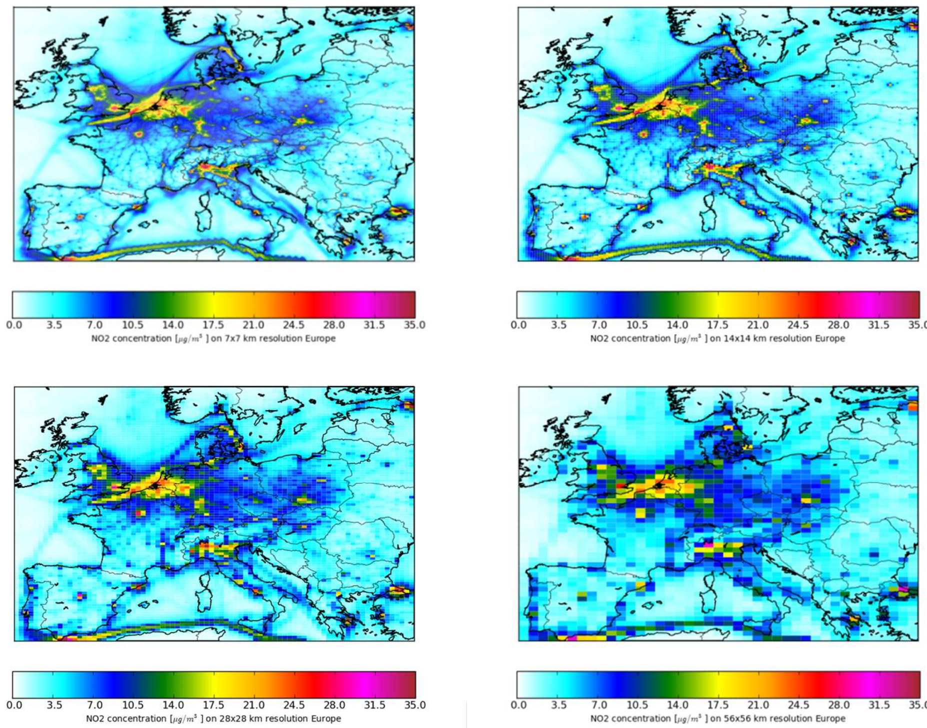


Figure 3. Modelled NO2 distribution for horizontal resolutions of 56, 28, 14 and 7 Km

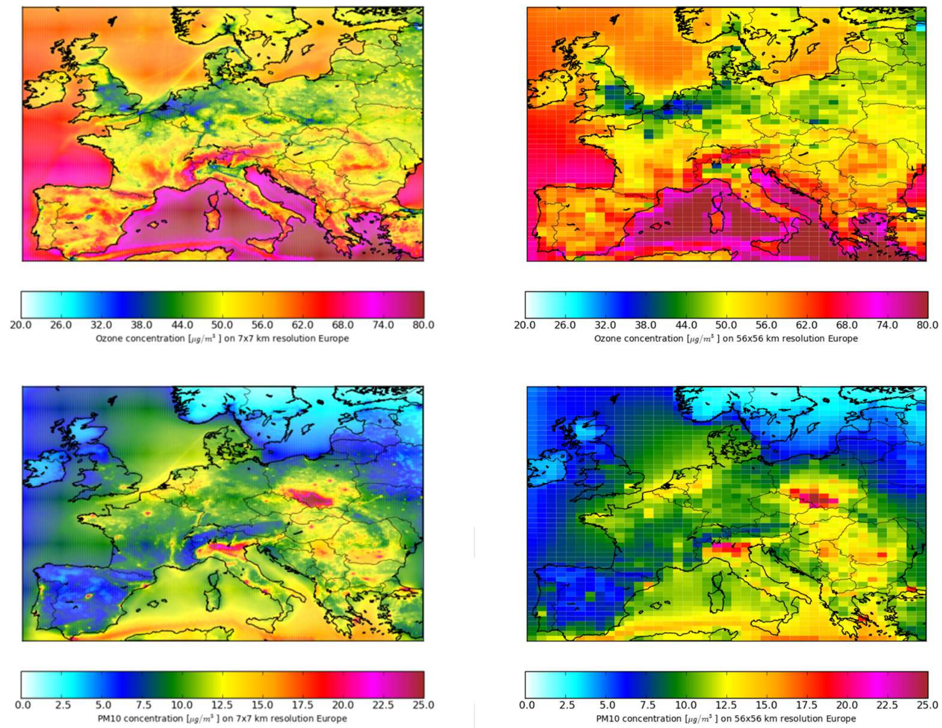


Figure 4. Modelled distributions at 7 and 56 Km resolution for ozone (upper panels) and PM10 (lower panels)

3.2 Model evaluation

For the model evaluation results we refer to the note on the TFMM exercise containing the evaluation for all participating model systems.

3.3 Source receptor matrices

In this section we investigate the impact of the model resolution on the calculated source receptor matrices. The source receptor matrices contain the impact of one countries emissions on the concentrations in the whole domain. For all countries the impact of their emissions to the concentration levels in all countries were calculated.

In Figure 5 and Figure 6 we show examples for two countries, i.e. France and Czech Republic and several components. For France the impact of resolution on the impact of French emissions is in general small. For NO2 the differences between the three resolutions is within 10 % for most countries. Only the impact of French emissions on Switzerland and to a lower extend Belgium show significant impacts of model resolution. In contrast to France, the impact of Czech emissions on the concentration in the country shows a strong dependency on model resolution. The highest difference is noticed for anthropogenic PM_{10} (38%), followed by SO_2 (37%), NO_2 (36%), and SO_4 (10%). The impact of Czech emissions on neighbouring countries is decreases with higher resolution. For instance, the impact on Slovakia and Poland decrease by about 20% going from 56 to 14 Km resolution. The reason for the differences is anticipated to be related to the size of the country as illustrated in the introduction. Below, we summarize the full SRMs to investigate if the impact of country size is systematic.

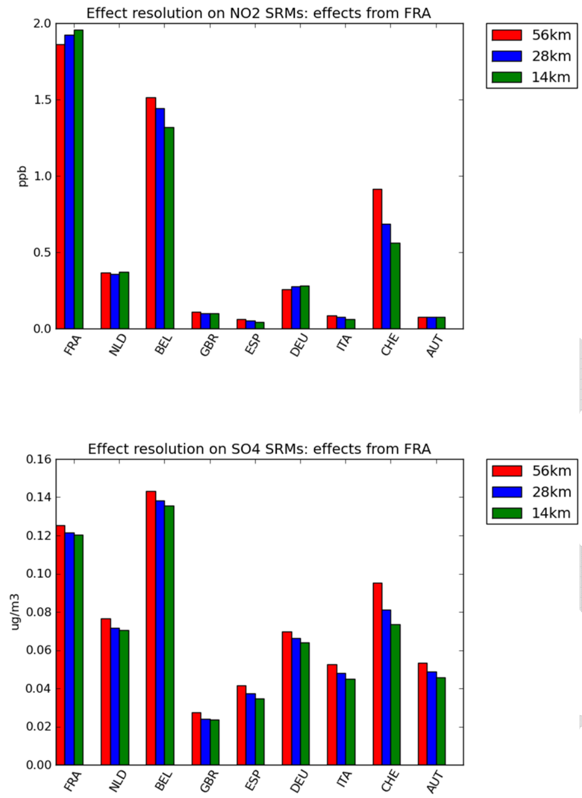
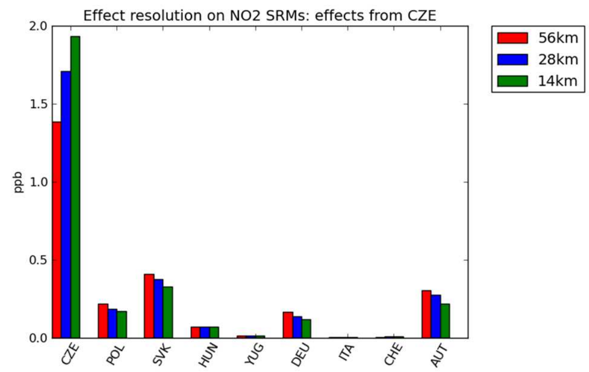


Figure 5. The impact of French emissions on surrounding countries for 56, 28 and 14 Km resolution. The examples given are provide for NO2 and SO4.



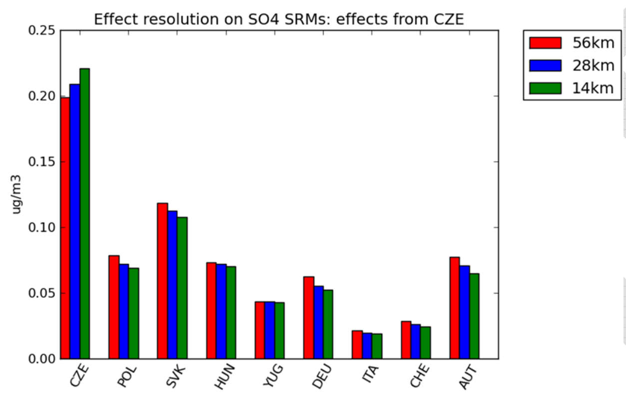


Figure 6. The impact of Czech emissions on surrounding countries for 56, 28 and 14 Km resolution. The examples given are provide for NO2 and SO2

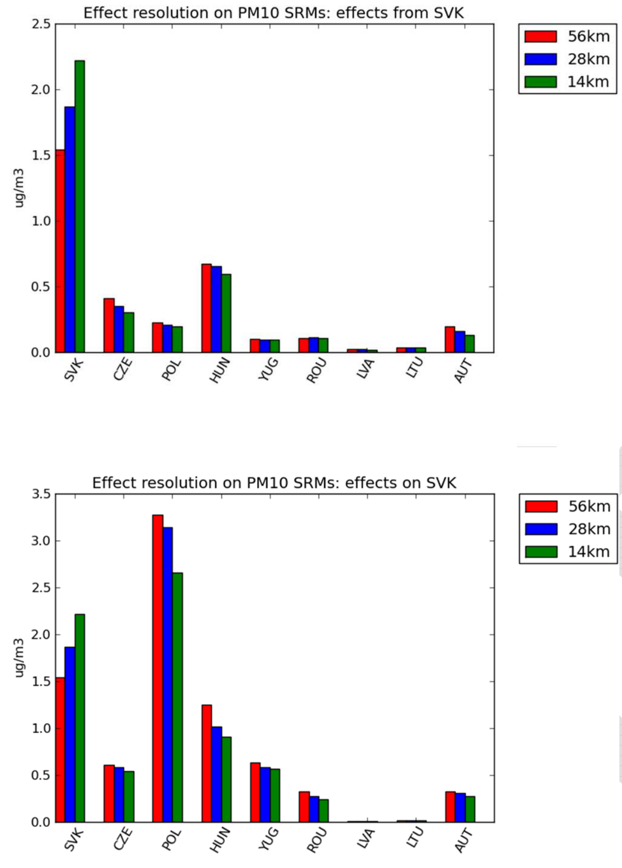


Figure 7. Impact of model resolution on calculated impact of the Slovakian emissions on surrounding countries (upper panel) as well as the concentrations received by the Slovakia from surrounding countries (Lower panel) for anthropogenic PM10.

The SRMs can be plotted as the impact of a country on the other countries. The other way around, also the impact of several countries to a receiving country can be assessed. This is illustrated for Slovakia and anthropogenic PM10 in Figure 7. Several interesting features are observed. For instance, the impact of the Slovakia on Hungary shows a low dependency on resolution with a small change from 28 to 14 Km resolution, whereas the impact of Hungary on the Slovakia shows a larger scale dependency and the largest change between the 56 and 28 km resolution. Notably, on a 14 Km resolution the labeled concentrations for Poland and Slovakia itself are not so far apart, whereas at 56 Km resolution the concentrations labeled Polish are twice those for Slovakia itself.

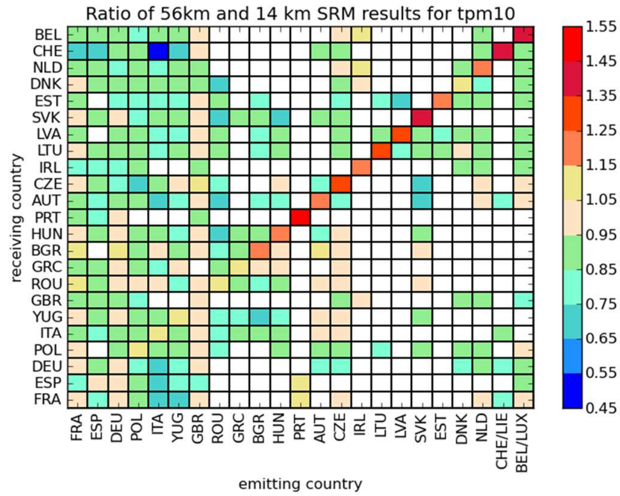
The impact of resolution on the source receptor matrices is summarized in Figure 8 to Figure 10 for a number of components. In these figures the ratio between all entries in the source receptor matrices between 14 and 56 Km resolution is plotted. On the x-axis the emitting countries are listed, whereas the y-axis represents the receiving countries. The countries were ordered to size going from large countries on the left to smaller countries to the right. Each cell represents the fractional change in the impact of a country on another. Positive numbers indicate that the contribution at the 14 Km is higher than at 56 resolution. To remove spurious ratios in which very small numbers are divided only significant country impacts are taken into account. The threshold for all species was set to 0.5% of the annual average concentration of that species in each country.

For all components the diagonal representing the contribution of countries to themselves draws attention. The values in the diagonal are systematically above 1. Maximum values for NO2, PPM, SO2, SO4 are 1.81, 1.80, 1.56, 1.27, respectively. Inspecting the results shows that the relatively small countries are generally associated with values significantly above 1. Going from 56 to 14 Km for anthropogenic particulate matter 12 countries show differences above 25% being Belgium, Ireland, Portugal, Austria, Switzerland, Czech Republic, Slovakia, Romania, Latvia, Lithuania, and Estonia. In contrast, the impact for large countries such as France, Germany and Spain is relatively small.

The impact of air pollutants outside a countries domain is generally lower at the higher resolution. The plots show that the impact to other countries is reduced, though in general with lower fractional changes than the positive impacts on the countries themselves. In contrast to the impact of the resolution for the smaller countries one observes that the impact of the larger countries to others is reduced. The number of countries impacted by the large countries is much larger than for the smaller countries, as indicated by the increase of white cells going from large to small countries. This feature is especially visible for the components with a short life time. Note that the impact of resolution to the SRMs for secondary inorganic species is very low, as illustrated for particulate nitrate and sulphate (Figure 9).

These summarizing “chess boards” are available for all species and combination of resolutions. In Figure 10 the comparison for the ratios to 28 and 14 Km is illustrated for NO2. For this short living component the impact from 56 to 14 km resolution is about twice the impact going from 28 to 14 Km.

Note that the threshold set for the valid combinations was on the low side. Hence, some noise is still present with valid ratios between contributions from countries far apart.



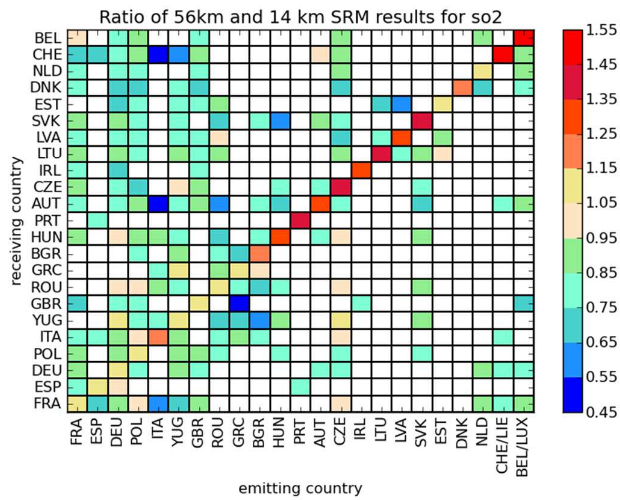
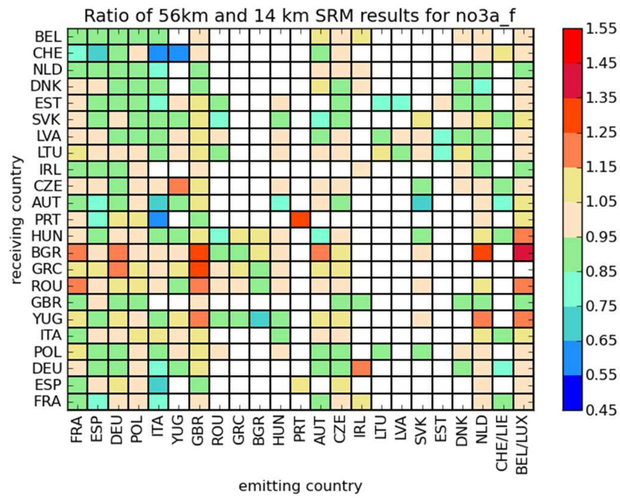


Figure 8. Ratio of the SRMs at 14 and 56 Km resolution for anthropogenic PM10 and SO2. On the x-axis the emitting countries are listed, whereas the y-axis represents the receiving countries. The countries were ordered to size going from large countries on the left to smaller countries to the right. Each cell represents the fractional change in the impact of a country on another. Positive numbers indicate that the contribution at the 14 Km is higher than at 56 resolution.



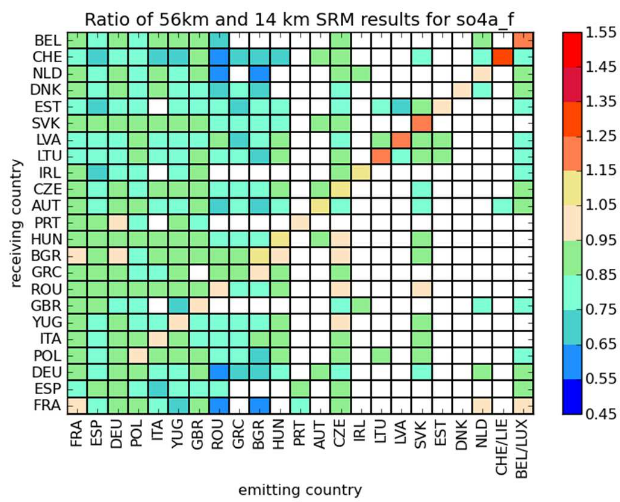
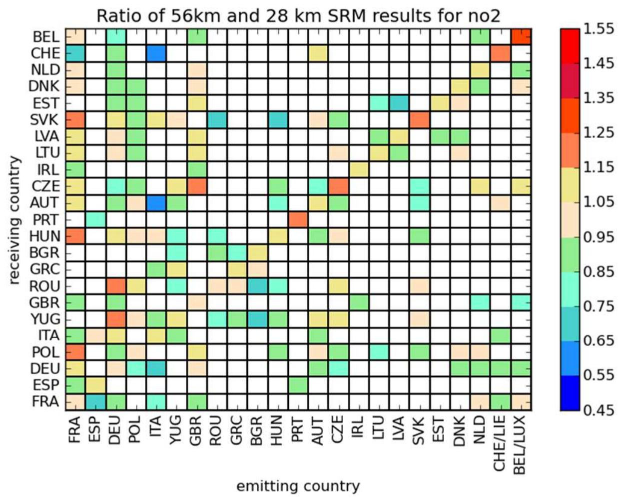


Figure 9. Ratio of the SRMs at 14 and 56 Km resolution for secondary nitrate and sulphate. On the x-axis the emitting countries are listed, whereas the y-axis represents the receiving countries. The countries were ordered to size going from large countries on the left to smaller countries to the right. Each cell represents the fractional change in the impact of a country on another. Positive numbers indicate that the contribution at the 14 Km is higher than at 56 resolution.



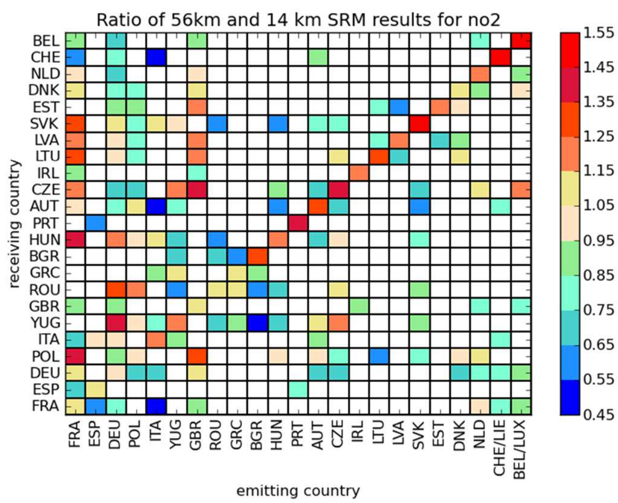


Figure 10. Ratio of the SRMs at 14 and 28 Km resolution (upper panel) and 14 and 56 Km resolution (lower panel) for nitrogen dioxide. On the x-axis the emitting countries are listed, whereas the y-axis represents the receiving countries. The countries were ordered to size going from large countries on the left to smaller countries to the right. Each cell represents the fractional change in the impact of a country on another. Positive numbers indicate that the contribution at the 14 or 28 Km is higher than at 56 resolution

4 Preliminary conclusions

In this study we successfully applied the LOTOS-EUROS system to assess the impact of emissions in one country to the concentrations in all other countries.

The source receptor matrices were calculated for three resolutions being 56, 28 and 14 Km.

Impacts of the model resolution were illustrated for nitrogen dioxide, sulphur dioxide, sulphate, nitrate and anthropogenic PM10.

It was found that the model on a 14 Km resolution predicts larger contributions of a countries to the concentrations in its own territory. The impact range from a few percent for large countries to tens of percents for smaller countries. The impact was found to be dependent on the life time of the component and the primary or secondary nature of the components. For example, maximum fractional changes for NO2, PPM, SO2, SO4 are 1.81, 1.80, 1.56, 1.27, respectively

For large countries the impact of their emissions to surrounding countries becomes lower at higher resolution. This impact is smaller than the change for smaller countries on themselves.

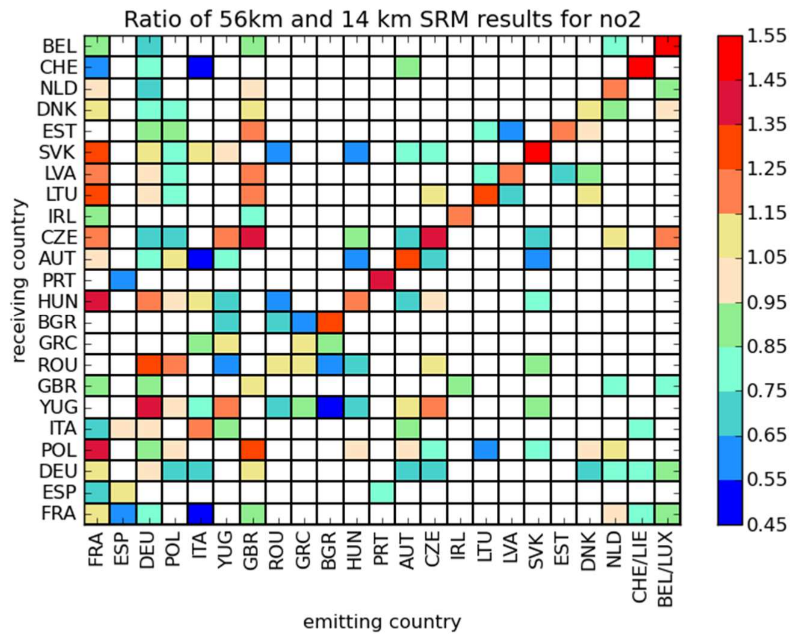
The reason for the impacts is that the borders are much sharper represented at the higher resolution.

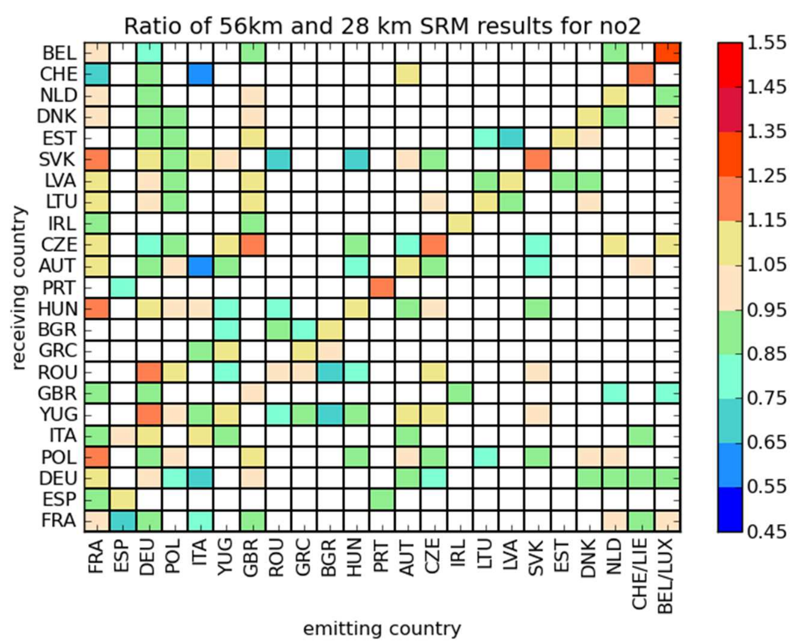
5 Signature

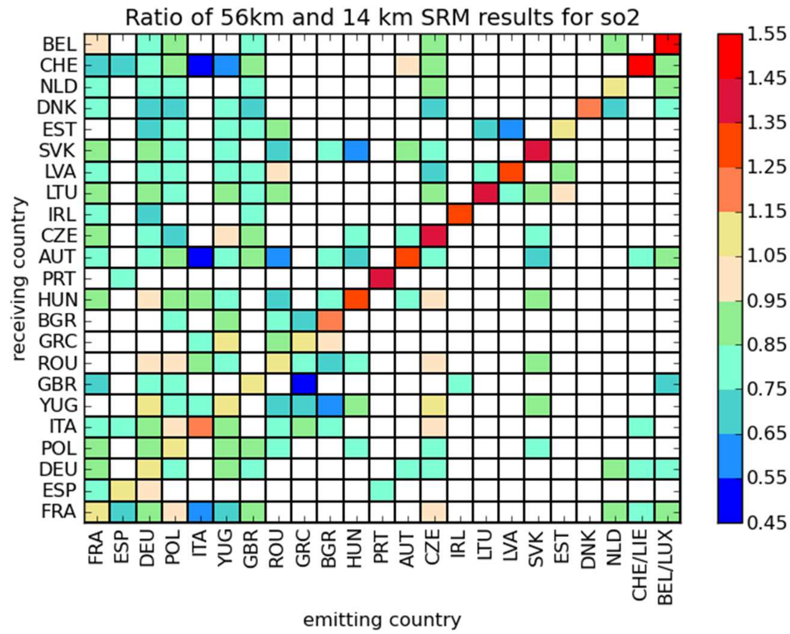
Utrecht, <datum>	Placeholder
<naam afdelingshoofd> Head of department	M. Schaap, C. Hendriks Author

Appendix A

Ratio of the SRMs at 14 and 28 Km resolution and 14 and 56 Km resolution for a number of components. For an explanation of the figures we refer to the main report.



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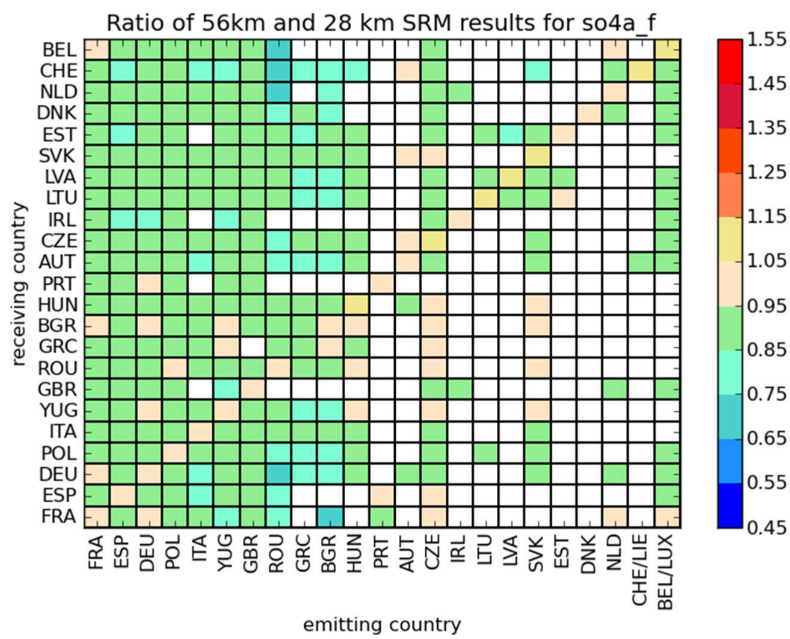
Ratio of 56km and 28 km SRM results for so4a_f
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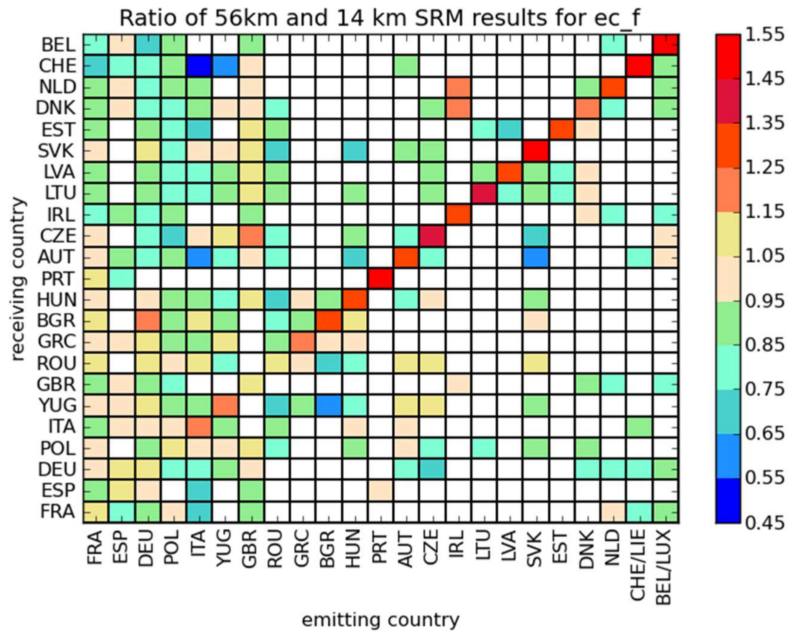
Ratio of 56km and 14 km SRM results for ec_f
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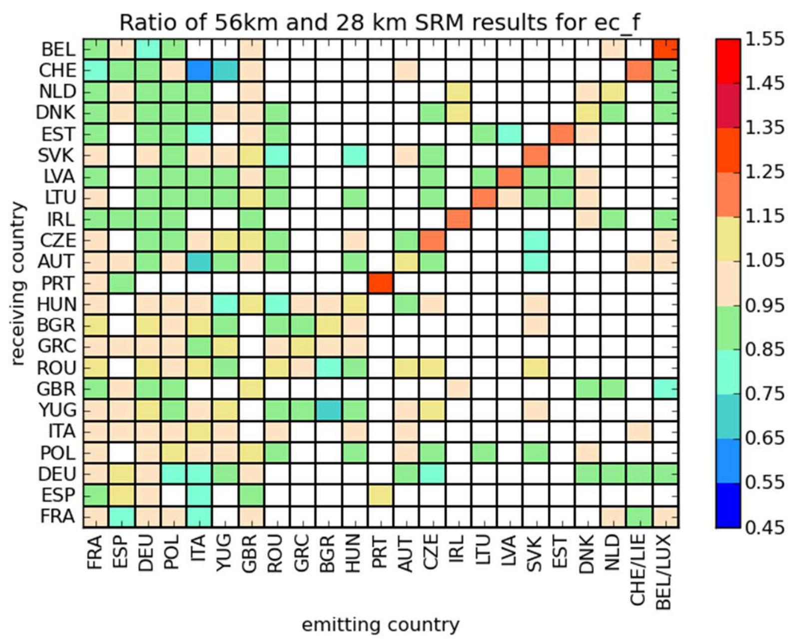
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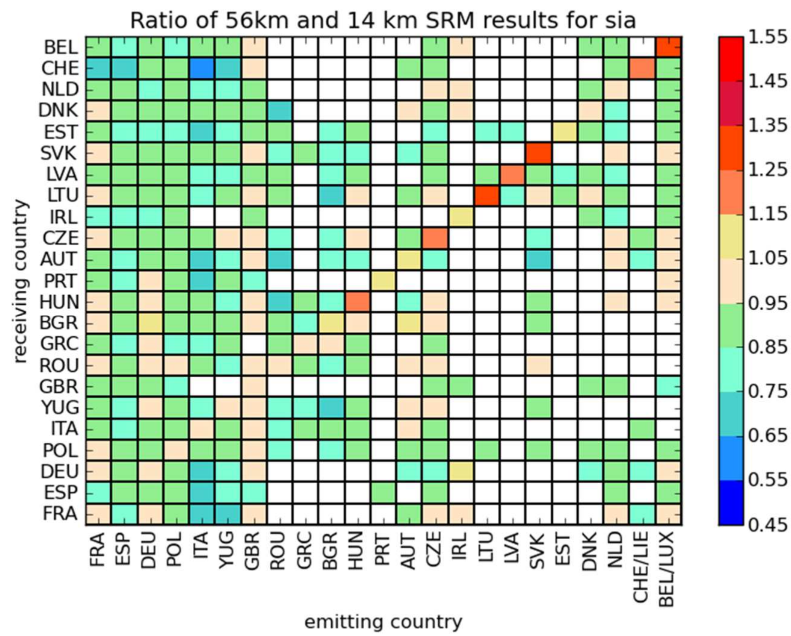
Ratio of 56km and 14 km SRM results for sia
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POL
DEU
ESP
FRA
emitting country
FRA
ESP
DEU
POL
ITA
YUG
GBR
ROU
GRC
BGR
HUN
PRT
AUT
CZE
IRL
LTU
LVA
SVK
EST
DNK
NLD
CHE/LIE
BEL/LUX
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1.35
1.25
1.15
1.05
0.95
0.85
0.75
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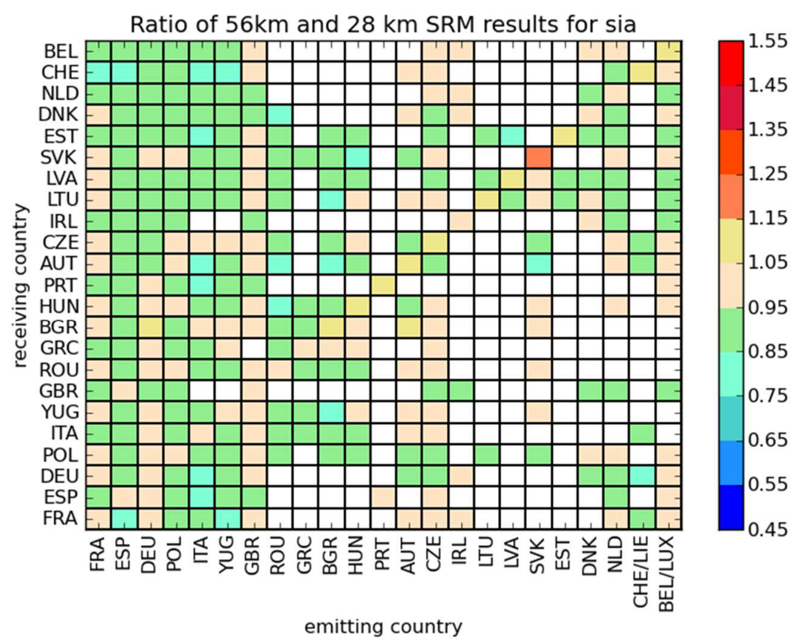
Ratio of 56km and 28 km SRM results for sia
receiving country
BEL
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SVK
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LTU
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AUT
PRT
HUN
BGR
GRC
ROU
GBR
YUG
ITA
POL
DEU
ESP
FRA
emitting country
FRA
ESP
DEU
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ITA
YUG
GBR
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GRC
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HUN
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CZE
IRL
LTU
LVA
SVK
EST
DNK
NLD
CHE/LIE
BEL/LUX
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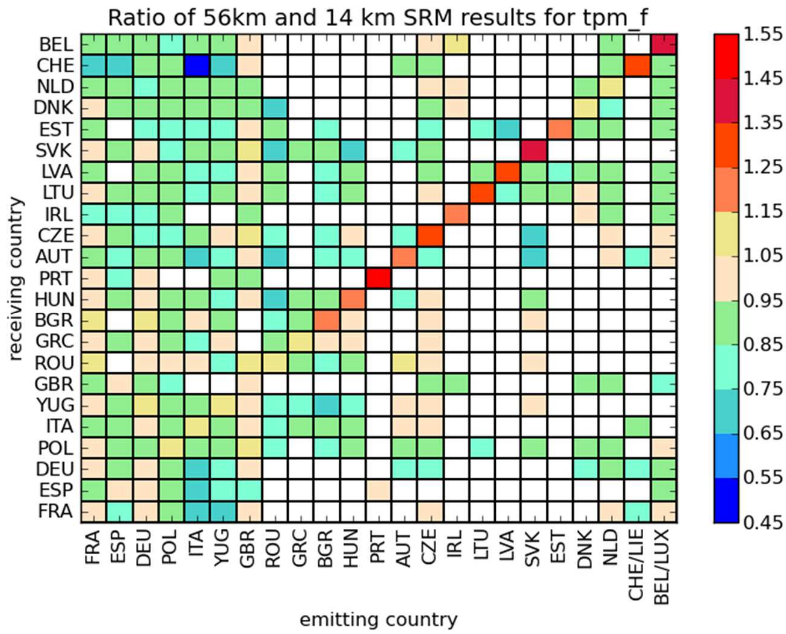
Ratio of 56km and 14 km SRM results for tpm_f
receiving country
BEL
CHE
NLD
DNK
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SVK
LVA
LTU
IRL
CZE
AUT
PRT
HUN
BGR
GRC
ROU
GBR
YUG
ITA
POL
DEU
ESP
FRA
emitting country
FRA ESP DEU POL ITA YUG GBR ROU GRC BGR HUN PRT AUT CZE IRL LTU LVA SVK EST DNK NLD CHE/LIE BEL/LUX
1.55
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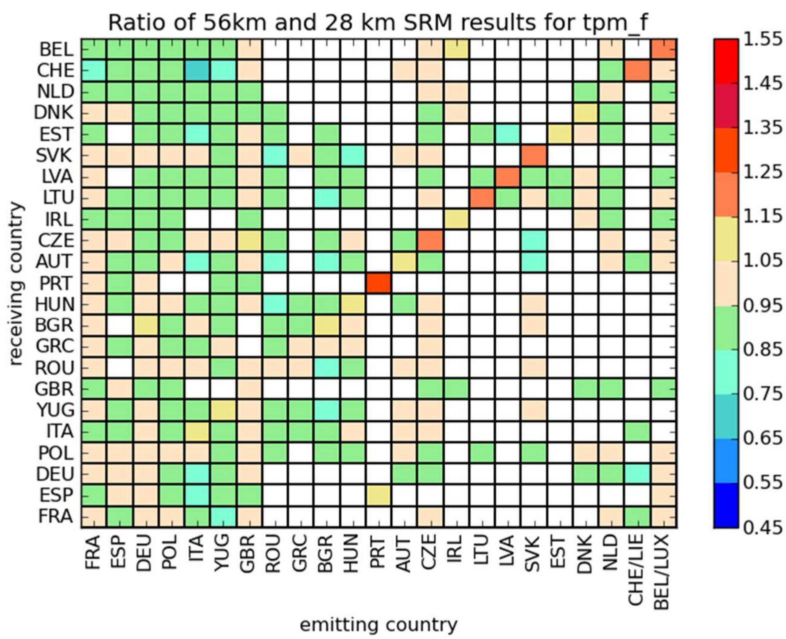
Ratio of 56km and 28 km SRM results for tpm_f
receiving country
BEL
CHE
NLD
DNK
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GRC
ROU
GBR
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ITA
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ESP
FRA
emitting country
FRA ESP DEU POL ITA YUG GBR ROU GRC BGR HUN PRT AUT CZE IRL LTU LVA SVK EST DNK NLD CHE/LIE BEL/LUX
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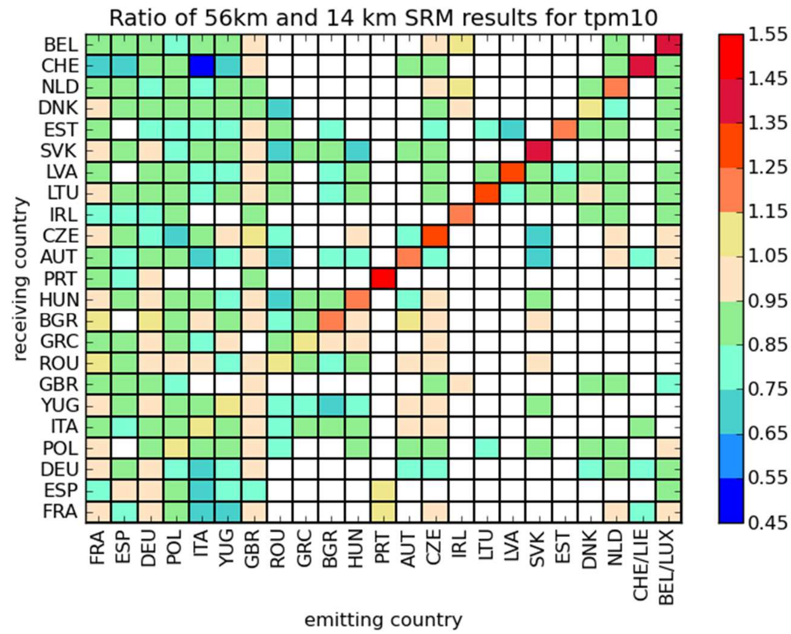
Ratio of 56km and 14 km SRM results for tpm10
BEL
CHE
NLD
DNK
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SVK
LVA
LTU
IRL
CZE
AUT
PRT
HUN
BGR
GRC
ROU
GBR
YUG
ITA
POL
DEU
ESP
FRA
receiving country
FRA ESP DEU POL ITA YUG GBR ROU GRC BGR HUN PRT AUT CZE IRL LTU LVA SVK EST DNK NLD CHE/LIE BEL/LUX
emitting country
1.55
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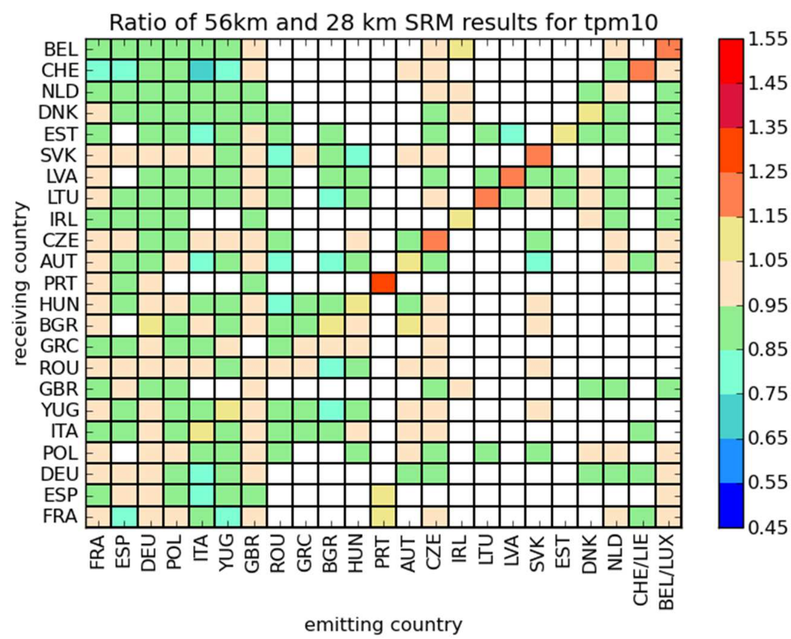
Ratio of 56km and 28 km SRM results for tpm10
BEL
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FRA
receiving country
FRA ESP DEU POL ITA YUG GBR ROU GRC BGR HUN PRT AUT CZE IRL LTU LVA SVK EST DNK NLD CHE/LIE BEL/LUX
emitting country
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0.75
0.65
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0.45