Supplement of Atmos. Chem. Phys., 19, 5165–5186, 2019
https://doi.org/10.5194/acp-19-5165-2019-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

Contribution and uncertainty of sectorial and regional emissions to regional and global $PM_{2.5}$ health impacts

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S1 – TM5-FASST methodology

S1.1 – TM5-FASST regions

Figure S1 shows the 56 regions defined in TM5-FASST. Note that Middle East includes Israel, Jordan, Lebanon, Palestinian Territories and Syria; Rest of South Africa accounts for Lesotho, South Africa and Swaziland, while the Gulf region comprises Bahrain, Iran, Iraq, Kuwait, Oman, Qatar, Saudi Arabia, Yemen; Rest of Central Europe includes Albania, Croatia, Macedonia, Serbia and Montenegro; Rest of South Asia includes Afghanistan, Bangladesh, Bhutan, Nepal and Pakistan; Rest of South Eastern Asia includes Cambodia, Lao People's Democratic Republic and Myanmar. The 56 TM5-FASST regions were chosen to obtain an optimal match with integrated assessment models such as IMAGE (Eickhout et al., 2004; van Vuuren et al., 2007), MESSAGE (Riahi et al., 2007), GAINS (Höglund-Isaksson and Mechler, 2005) as well as the POLES model (Russ et al., 2007; Van Aardenne et al., 2007). The grouping of small countries was motivated by (a) finding a compromise between spatial resolution and computational effort required to obtain the set of source-receptor matrices for TM5-FASST and (b) avoiding inaccurate mapping of small individual countries that are represented by only a few 1°x1° grid cells. Most European countries are defined as individual source regions, except for the smallest countries, which have been aggregated.

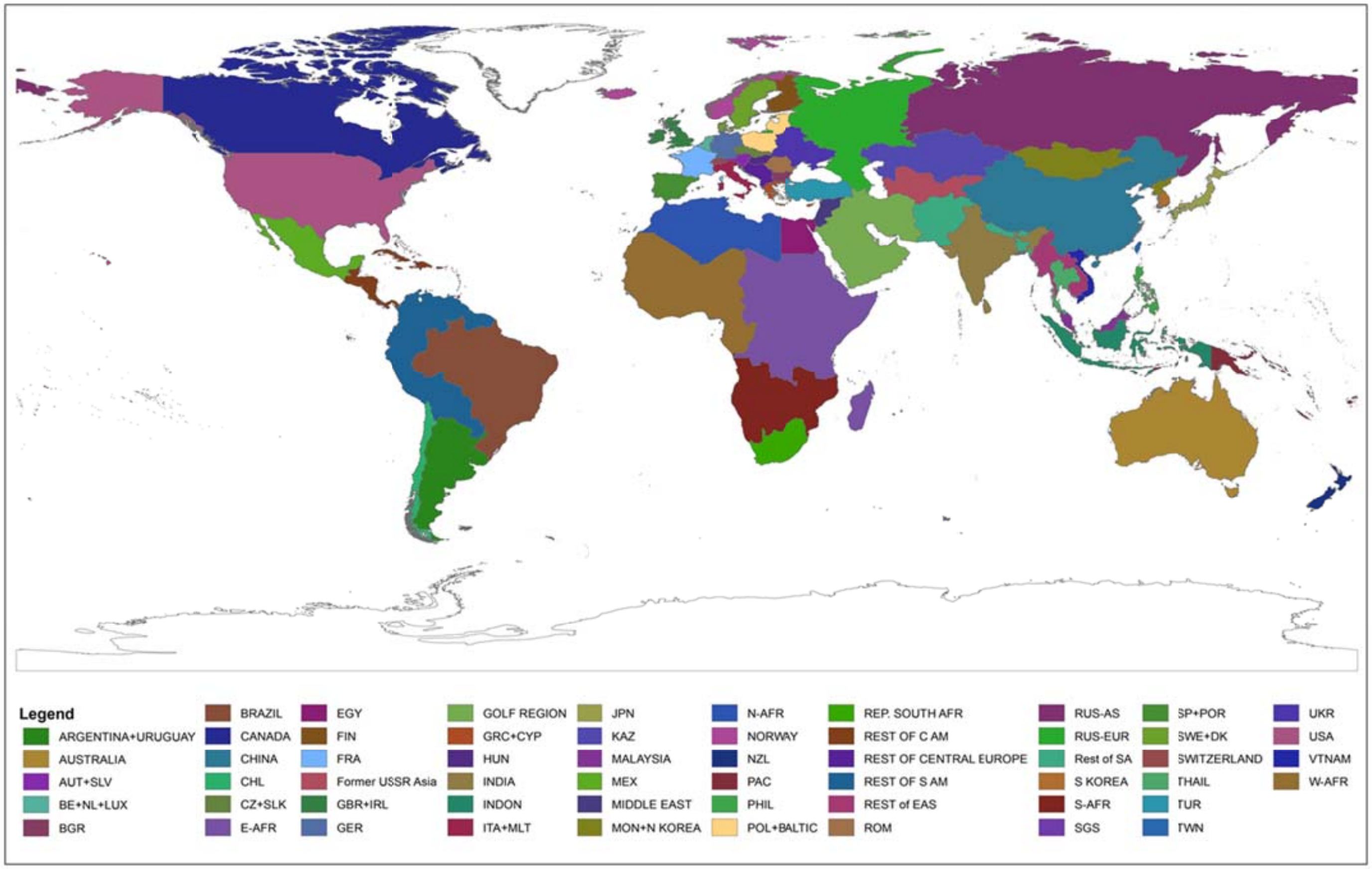


Figure S1.1 – Global map with the 56 TM5-FASST regions.

S1.2 – Sector and source region attribution using the TM5-FASST source-receptor relationships

S1.2.1 - Attribution by sector

The TM5-FASST methodology uses a local perturbation approach in the vicinity of a reference simulation, where the total concentration of component (or metric) j in receptor region y, resulting from emissions of all n_i precursors i in all n_x source regions x, is obtained as a perturbation on the base-simulation concentration (Van Dingenen et al., 2018). Hence, the $PM_{2.5}$ concentration in region y for an emission scenario different from the reference scenario is obtained as:

$$\mathrm{PM}(y) = PM_{ref}(y) + \Delta\mathrm{PM}(y) \qquad (1)$$

The perturbation term $\Delta\mathrm{PM}(y)$ is obtained from the linear scaling of the difference between scenario and reference emission (i.e. the emission perturbation):

$$\Delta\mathrm{PM}(y) = \sum_{j=1}^{n_j}\sum_{k=1}^{n_x}\sum_{i=1}^{n_i} A_{ij}[x_k, y] \cdot \left[E_i(x_k) - E_{i,ref}(x_k)\right] \qquad (2)$$

where the summation runs over n_i precursor species, n_j $PM_{2.5}$ components and n_x source regions, and $A_{ij}[x_k, y]$ is the source-receptor coefficient, expressing the emission-concentration response sensitivity in the vicinity of the reference conditions, evaluated from a 20% emission perturbation (see Van Dingenen et al., 2018):

$$A_{ij}[x, y] = \frac{\Delta PM_{ref}^{j}(y)}{\Delta E_{i,ref}(x)} \qquad (3)$$

with $\Delta E_{i,ref}(x)$=0.2$E_{i,ref}(x)$ and $\Delta PM_{ref}^{j}(y)$ the corresponding $PM_{2.5}$ component j response.

Eq. (2) can also be applied to attribute individual sector contributions to the pollutant concentration by setting the "emission perturbation" equal to the emission contribution of a single sector. The $PM_{2.5}$ contribution from the single sector S equals

$$\Delta\mathrm{PM}'_S(y) = \sum_{j=1}^{n_j}\sum_{k=1}^{n_x}\sum_{i=1}^{n_i} A_{ij}[x_k, y] \cdot \left[E_{S,i}(x_k)\right] \qquad (4)$$

Having obtained the marginal $PM_{2.5}$ contributions from the individual sectors, the total $PM_{2.5}$ can be re-composed as the sum from all n_S sectors S:

$$\mathrm{PM}'(y) = \sum_{s=1}^{n_S} \Delta\mathrm{PM}'_s(y) \qquad (5)$$

However, due to non-linearities in emission-concentration responses, the sum of all individual sector contributions may not exactly match the total $PM_{2.5}$ obtained from Eqs. (1) and (2) where we write $E_i(x_k)$ as the sum of the emissions by sector:

$$\mathrm{PM}(y) = PM_{base}(y) + \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} A_{ij}[x_k, y] \cdot \left[\sum_{s=1}^{n_S} \; E_{s,i}(x_k) - E_{i,ref}(x_k)\right] \tag{6}$$

$\mathrm{PM}'(y)$ from Eq. 5 and $\mathrm{PM}(y)$ from Eq. 6 are equivalent if

$$PM_{ref}(y) = \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} A_{ij}[x_k, y] \cdot E_{i,\mathrm{ref}}(x_k) \tag{7}$$

Using Eq. 3 this is equivalent to the condition that

$$PM_{ref}(y) = \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} A_{ij}[x_k, y] \frac{\Delta PM(y)}{0.2E_{i,\mathrm{ref}}(x_k)} E_{i,ref}(x_k) \tag{8}$$

or

$$PM_{ref}(y) = \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} 5.\Delta PM(y) \tag{9}$$

In other words, total $PM_{2.5}$ will be correctly reproduced as the sum of the individual sector contributions if and only if the $PM_{2.5}$ base concentration can be approached by 5 times the 20% perturbation response, implying a perfectly linear emission-concentration response for all precursors. Figure A1.1 shows the correspondence between regionally aggregated $\sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} 5.\Delta PM$ and PM_{ref}. The agreement is satisfactory although not perfect. In order to restore the closure between the total $PM_{2.5}$ and the sum of the sectors, we therefore rescale the sector contributions such that their sum corresponds to the total $PM_{2.5}$ obtained from the local perturbation calculation, i.e. we use the relative contribution by sector resulting from Eq. 5 and apply them onto the total $PM_{2.5}$ obtained from Eq. 6.

$$\Delta\mathrm{PM}_S(y) = \frac{\Delta\mathrm{PM}'_S(y)}{\sum_{s=1}^{n_S} \; \Delta\mathrm{PM}'_S(y)} \mathrm{PM}(y) \tag{10}$$

S1.2.2 Attribution by source region

The marginal contribution of an individual source regions (x) to the total $PM_{2.5}$ concentration in a given receptor region (y) is obtained (via Eq. 2) from

$$\Delta\mathrm{PM}'_x(y) = \sum_{j=1}^{n_j} \sum_{i=1}^{n_i} A_{ij}[x, y] \cdot E_i(x) \tag{11}$$

Similar as for the sector break-down, the emission perturbation has been replaced by an extrapolation of the SR coefficient over the total emission magnitude in a given source region, and non-linearities may lead to non-closure between the sum of all $\Delta\mathrm{PM}'_x(y)$ and total $PM_{2.5}$ obtained from the local perturbation as in Eqs. (1) and (2). In order to restore the closure we apply the same scaling procedure as in Eq. 10:

$$\Delta\mathrm{PM}_x(y) = \frac{\Delta\mathrm{PM}'_x(y)}{\sum_{k=1}^{n_k} \; \Delta\mathrm{PM}'_{x_k}(y)} \mathrm{PM}(y) \tag{12}$$

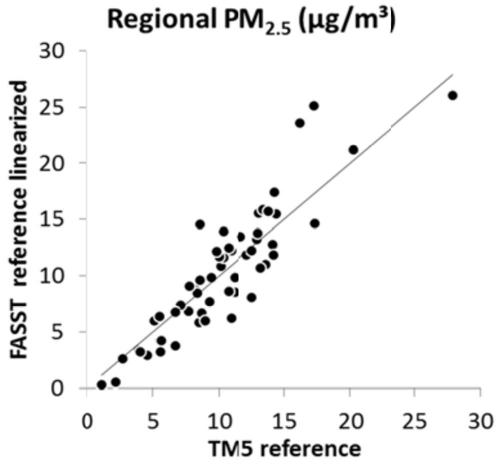


Figure S1.2 - Scatter plot of regionally aggregated $PM_{2.5}$ concentrations. Y-axis: FASST linearized extrapolation of a 20% emission perturbation towards 100%, versus the full TM5 computation, for the FASST reference emission scenario (RCP year 2000, see Van Dingenen et al., 2018). The Figure evaluates the validity of Eq. 9.

S2 – Within-region and extra-regional $PM_{2.5}$ concentrations

Table S2 reports the fraction of within-region vs. extra-regional $PM_{2.5}$ concentrations for the 56 TM5-FASST regions, although it should be recognized that results are influenced by the different size of the TM5-FASST regions (e.g. European countries vs. China or India), their emissions distribution, geographical isolation of the countries (e.g. Chile has very high local contribution to air pollution just because of its geographical isolation), etc., which can affect the transboundary export efficiency as discussed in the methodological section of the manuscript. Europe, China and India are discussed in detail in the manuscript, as well as other group of countries. Here we provide some peculiar features for few additional TM5-FASST regions. $PM_{2.5}$ concentrations in African countries are dominated by the local contributions coming from the continent itself, except for Northern Africa being affected by extra-regional transported pollution from shipping emissions and all Mediterranean countries (e.g. Italy, Spain, France). Argentina+Uruguay is the only country of Latin American being influenced by transboundary pollution, especially coming from Chile, Brazil and Rest of South America. Canada is affected for more than 50% by USA pollution, while South Eastern Asia countries are influenced by pollution of China and India (from ca 20% to 80%) depending on their location.

Table S2 – Within-region and extra-regional $PM_{2.5}$ concentrations for the 56 TM5-FASST regions. The extra-regional contribution represents the RERER metric.

Aggregated world regions	TM5-FASST regions	within-region	extra-regional (RERER)	$PM_{2.5}$ concentration ($\mu g/m^3$)	Population
Africa	**Northern Africa**	44.3%	55.7%	4.2	7.50E+07
Africa	**Egypt**	61.7%	38.3%	11.0	6.80E+07
Africa	**Western Africa**	76.7%	23.3%	4.0	2.60E+08
Africa	**Eastern Africa**	46.8%	53.2%	2.7	3.00E+08
Africa	**Southern Africa**	58.3%	41.7%	1.0	7.10E+07
Africa	**Rep. of South Africa**	96.5%	3.5%	6.1	4.80E+07
China+	**Mongolia+North Korea**	32.3%	67.7%	14.6	3.00E+07
China+	**China**	98.0%	2.0%	39.9	1.00E+09
Europe	**Austria+Slovenia**	34.1%	65.9%	8.4	1.00E+07
Europe	**Switzerland**	40.9%	59.1%	10.1	7.20E+06
Europe	**Benelux**	44.6%	55.4%	10.1	2.70E+07
Europe	**Spain+Portugal**	69.4%	30.6%	5.4	5.10E+07
Europe	**Finland**	57.1%	42.9%	2.6	5.20E+06
Europe	**France**	59.5%	40.5%	9.3	5.90E+07
Europe	**Great Britain+Ireland**	64.9%	35.1%	6.1	6.30E+07
Europe	**Greece+Cyprus**	39.1%	60.9%	7.6	1.20E+07
Europe	**Italy+Malta**	39.5%	60.5%	11.8	5.80E+07
Europe	**Germany**	51.3%	48.7%	9.3	8.20E+07
Europe	**Sweden+Denmark**	35.0%	65.0%	4.1	1.40E+07
Europe	**Norway**	72.6%	27.4%	2.4	4.80E+06
Europe	**Bulgaria**	36.7%	63.3%	10.6	8.00E+06
Europe	**Hungary**	25.0%	75.0%	9.2	1.00E+07
Europe	**Poland+Baltic**	54.3%	45.7%	7.9	4.60E+07
Europe	**Rest of Central EU**	46.5%	53.5%	9.3	2.30E+07
Europe	**Czech Republic**	33.1%	66.9%	10.3	1.60E+07
Europe	**Romania**	47.3%	52.7%	10.9	2.20E+07
India+	**Rest of South Asia**	50.0%	50.0%	29.3	3.00E+08
India+	**India**	86.8%	13.2%	34.7	1.00E+09
Latin America	**Brazil**	91.8%	8.2%	1.6	2.00E+08
Latin America	**Mexico**	80.4%	19.6%	4.2	1.00E+08
Latin America	**Rest of Central America**	79.6%	20.4%	2.0	7.00E+07
Latin America	**Chile**	98.5%	1.5%	13.7	2.00E+07
Latin America	**Argentina+Uruguay**	55.7%	44.3%	1.1	4.00E+07
Latin America	**Rest of South America**	92.4%	7.6%	2.4	1.00E+08
Middle East	**Turkey**	70.1%	29.9%	8.7	6.30E+07
Middle East	**Middle East**	40.8%	59.2%	9.2	3.40E+07
Middle East	**Gulf region**	72.0%	28.0%	7.8	1.40E+08

North America	**Canada**	44.8%	55.2%	4.3	3.00E+07
North America	**USA**	91.5%	8.5%	7.8	3.00E+08
Oceania	**Australia**	84.5%	15.5%	1.1	2.00E+07
Oceania	**New Zealand**	59.3%	40.7%	0.3	4.00E+06
Oceania	**Pacific Islands**	38.4%	61.6%	0.2	8.00E+06
Russia	**Kazakhstan**	32.3%	67.7%	4.9	1.00E+07
Russia	**Former USSR Asia**	59.7%	40.3%	7.5	4.00E+07
Russia	**Russia (EU)**	50.6%	49.4%	3.3	1.00E+08
Russia	**Russia (Asia)**	44.4%	55.6%	2.7	4.00E+07
Russia	**Ukraine**	62.0%	38.0%	7.8	6.00E+07
SE Asia	**South Korea**	30.0%	70.0%	13.8	5.00E+07
SE Asia	**Japan**	58.7%	41.3%	6.9	1.00E+08
SE Asia	**Taiwan**	29.0%	71.0%	6.4	2.00E+07
SE Asia	**Indonesia**	82.9%	17.1%	2.4	2.00E+08
SE Asia	**Thailand**	49.6%	50.4%	8.0	6.00E+07
SE Asia	**Malaysia**	51.1%	48.9%	3.1	3.00E+07
SE Asia	**Philippines**	76.5%	23.5%	2.0	8.00E+07
SE Asia	**Vietnam**	65.9%	34.1%	14.2	8.00E+07
SE Asia	**Rest of South Eastern Asia**	16.5%	83.5%	8.6	7.00E+07

S3 – Emission inventory uncertainty estimation

Table S3 summarizes region- and pollutant- specific emission uncertainties (σ_{EMI}) as calculated from Eq. 4.

Table S3 - Region- and pollutant- specific emission uncertainties ($\sigma_{EMI, \%}$).

TM5-FASST REGION	SO_2	NOx	CO	NMVOC	NH_3	Other $PM_{2.5}$	BC	Primary OM
Australia	49	115	156	135	298	61	82	71
Austria+Slovenia	17	20	25	36	200	73	101	120
Benelux	19	35	28	32	194	114	103	158
Canada	53	110	137	108	154	93	113	166
Switzerland	23	33	28	34	195	44	47	71
Spain+Portugal	35	129	98	136	290	69	111	73
Finland	11	22	31	32	188	106	63	151
France	17	36	37	35	193	67	68	111
Great Britain+Ireland	37	99	132	117	292	77	107	139
Greece+Cyprus	36	59	97	136	289	89	106	163
Italy+Malta	44	114	160	166	13	50	65	88
Japan	45	100	155	141	263	186	196	189

Norway	50	97	119	123	295	80	101	125
New Zealand	22	28	26	38	192	73	71	139
Germany	26	29	34	28	184	105	88	149
Sweden+Denmark	23	36	26	32	198	96	79	128
Turkey	27	25	31	39	188	63	57	108
Ukraine	49	93	175	138	281	46	78	57
USA	31	27	36	30	193	77	72	139
Bulgaria	48	122	110	108	284	115	111	162
Hungary	42	136	131	128	221	151	159	163
Poland+Baltic	25	36	32	32	195	120	86	156
Czech Republic	21	31	35	34	167	40	43	38
Former USSR Asia	40	48	88	138	300	83	120	141
Romania	49	61	144	131	290	55	73	99
Russia (Asia)	48	146	170	131	296	129	150	160
Russia (EU)	39	82	91	124	241	58	87	158
Argentina+Uruguay	45	92	160	136	232	53	83	103
Brazil	45	111	110	120	231	119	140	179
Chile	49	103	118	143	291	89	94	142
China	12	33	32	35	188	142	121	163
South Korea	14	27	26	33	203	91	89	133
Eastern Africa	49	58	157	130	296	169	185	177
Egypt	46	88	132	128	190	99	150	131
Gulf region	47	98	143	107	287	127	111	160
Indonesia	47	76	131	118	283	147	177	163
Kazakhstan	51	97	112	112	283	103	98	173
Middle East	49	108	99	113	284	142	113	174
Mexico	25	27	26	39	197	66	69	122
Mongolia+ North Korea	38	79	165	126	283	53	75	71
Malaysia	51	90	137	116	273	121	137	181
India	52	56	119	148	258	118	151	162
Northern Africa	36	124	167	125	295	102	125	127
Pacific Islands	48	114	151	143	235	150	152	176
Philippines	33	125	175	137	234	189	189	193
Rest of Central America	38	67	85	134	294	75	100	158
Rest of Central EU	38	67	85	134	294	75	100	158
Rep. of South Africa	34	111	139	144	240	173	189	186
Rest of South America	23	31	31	31	191	107	111	154
Rest of South Asia	44	98	106	115	221	53	79	69
Rest of South Eastern Asia	42	78	141	115	292	94	129	173
Southern Africa	41	106	155	122	176	52	71	68
Thailand	47	90	85	111	287	69	96	138

Taiwan	30	26	34	34	187	81	47	130
Vietnam	43	80	160	133	207	144	175	187
Western Africa	45	116	156	137	260	191	198	193

Figure S2 represents with more regional detail the uncertainty information reported in Fig. 6 and discussed in the manuscript.

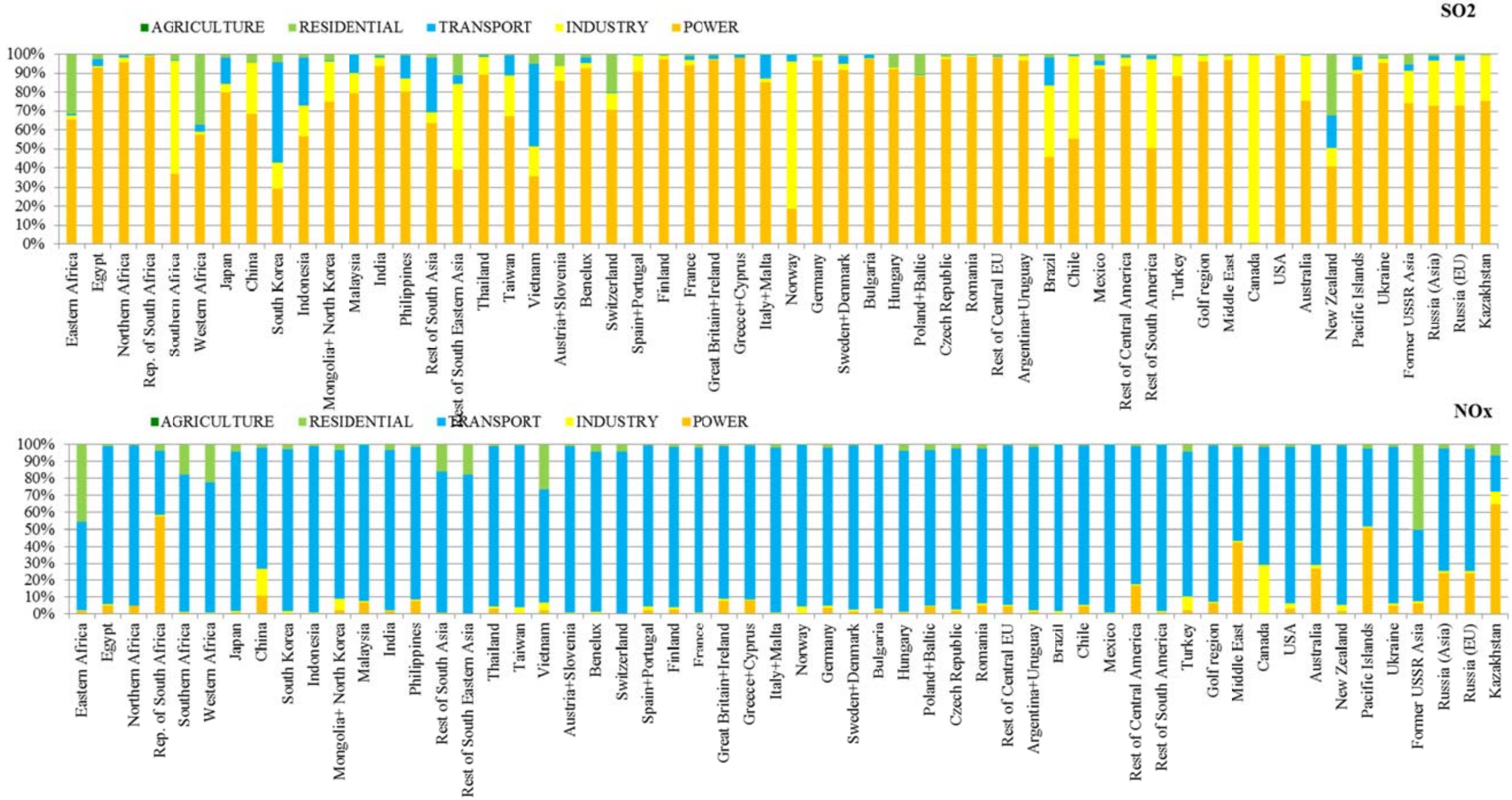
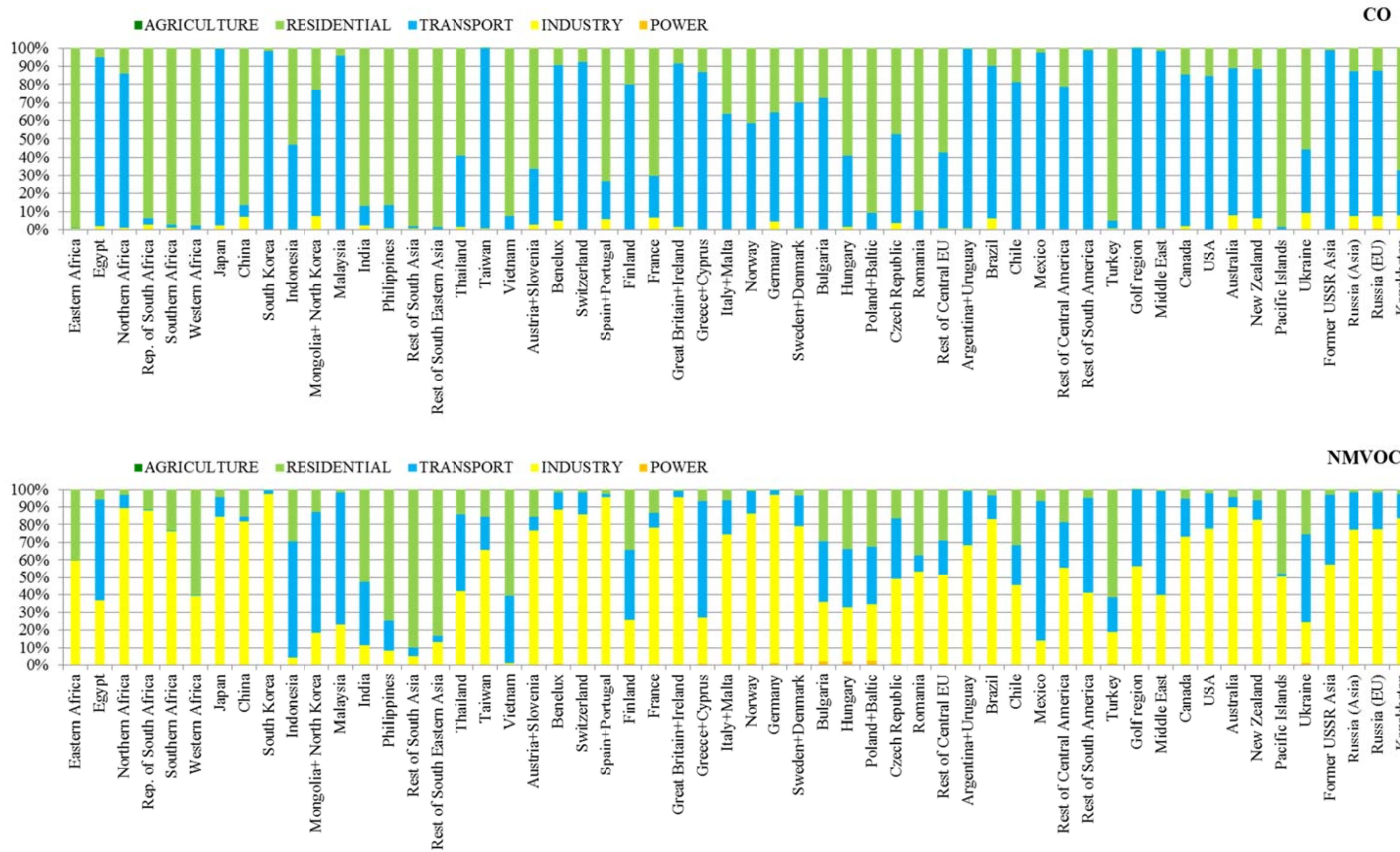


Figure S2 – Sector relative contribution to total pollutant emission uncertainty for each region.



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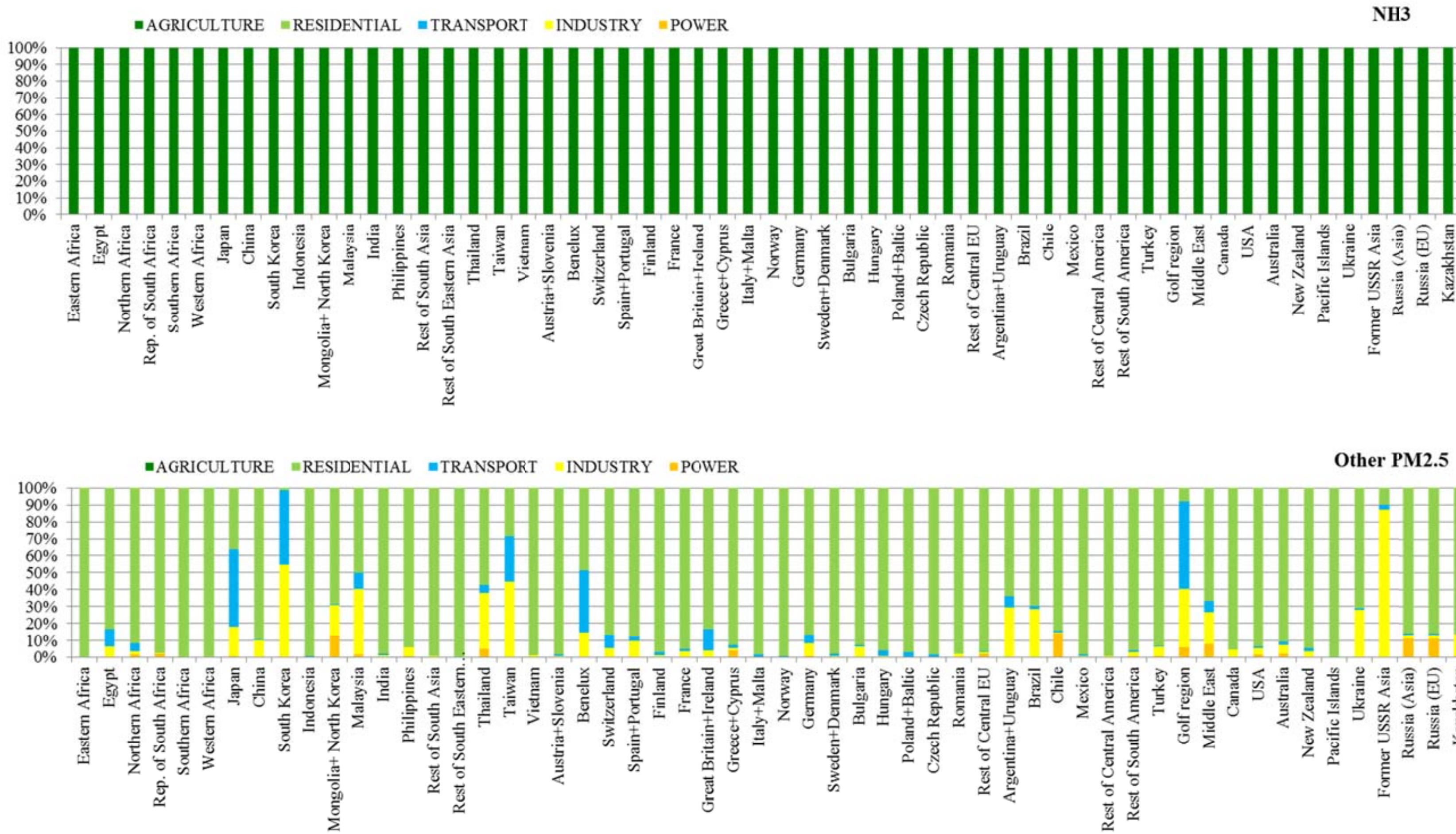

Figure S2 – Sector relative contribution to total pollutant emission uncertainty for each region.

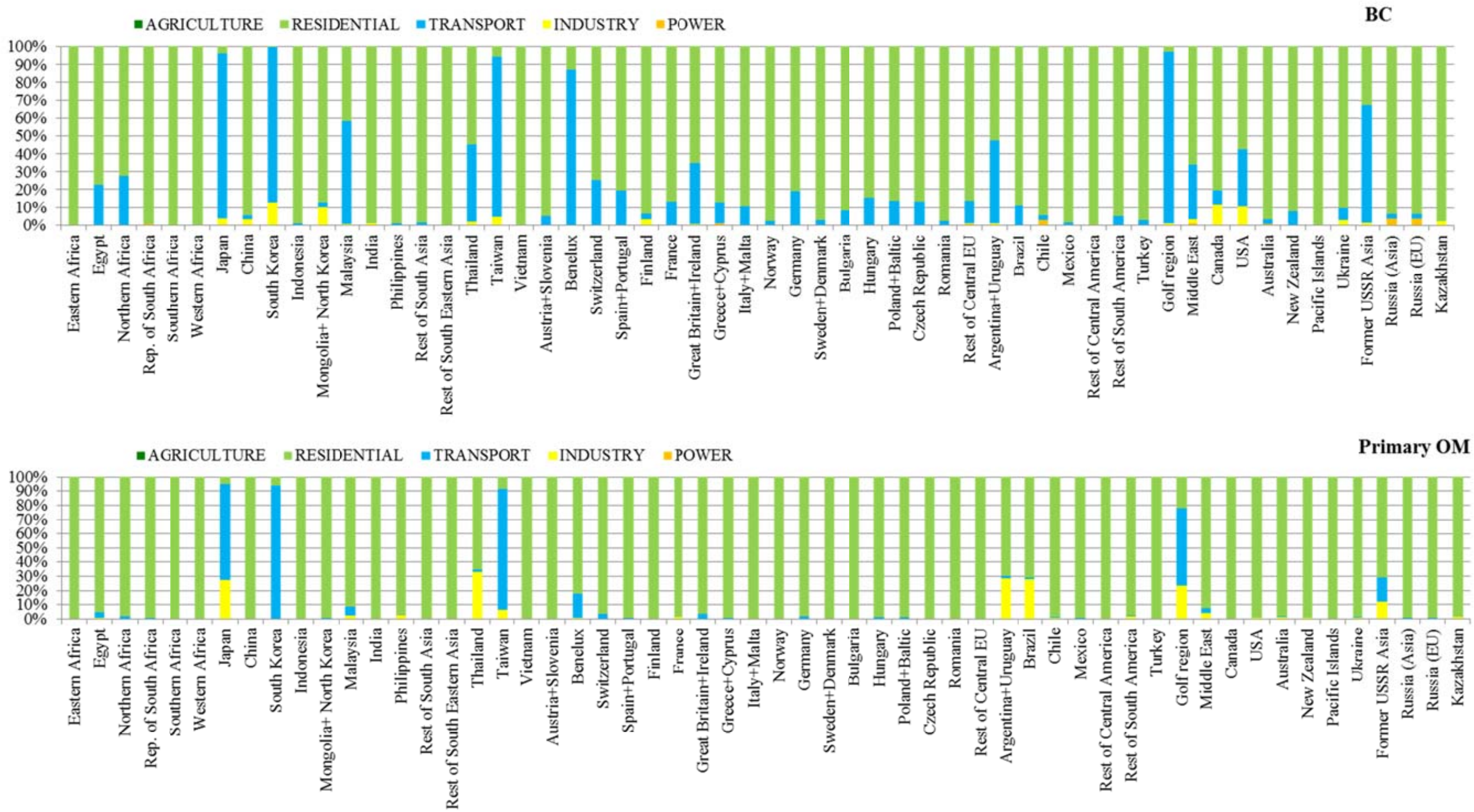

Figure S2 – Sector relative contribution to total pollutant emission uncertainty for each region.

Table S4 – Comparison of wood consumption estimates in the residential sector (AD) in 2010: TNO RWC estimates are evaluated using per capita consumption provided by Denier van der Gon et al. (2015), while the UNFCCC estimates are derived from the 2015 reporting.

Fuel wood use in residential sector (TJ)	EDGARv4.3.2	TNO RWC	UNFCCC (2015)
Austria	7.0E+04	8.4E+04	8.5E+04
Belgium	1.0E+04	9.8E+03	3.1E+04
Bulgaria	3.0E+04	2.6E+04	3.0E+04
Cyprus	8.4E+01	2.4E+03	7.0E+02
Czech Republic	4.8E+04	4.2E+04	5.2E+04
Germany	2.6E+05	1.8E+05	3.5E+05
Denmark	4.0E+04	3.3E+04	4.5E+04
Spain	1.0E+05	1.0E+05	1.1E+05
Estonia	1.8E+04	1.4E+04	1.8E+04
Finland	5.9E+04	4.9E+04	7.1E+04
France	3.3E+05	3.5E+05	3.3E+05
Great Britain	1.4E+04	1.6E+04	2.6E+04
Greece	2.4E+04	3.4E+04	1.2E+04
Croatia	1.3E+04	1.4E+04	1.4E+04
Hungary	2.8E+04	2.0E+04	3.3E+04
Ireland	1.1E+03	2.3E+03	1.8E+03
Italy	1.4E+05	6.0E+04	1.8E+05
Lituania	2.4E+04	1.6E+04	2.6E+04
Luxemburg	7.4E+02	6.1E+02	9.8E+02
Latvia	3.1E+04	3.1E+04	3.6E+04
Netherlands	1.2E+04	1.2E+04	2.4E+04
Poland	1.1E+05	1.3E+05	1.4E+05
Portugal	3.0E+04	5.3E+04	3.1E+04
Romania	1.5E+05	1.1E+05	1.5E+05
Slovakia	1.8E+03	2.4E+04	1.8E+04
Slovenia	1.8E+04	1.3E+04	1.8E+04
Sweden	2.6E+04	4.7E+04	5.6E+04
Rep. of Macedonia	7.5E+03	6.2E+03	0.0E+00
Switzerland	2.0E+04	2.4E+04	2.8E+04
Armenia	0.0E+00	8.9E+03	0.0E+00
Arzebaijan	3.0E+03	1.6E+04	0.0E+00
Belarus	2.1E+04	2.7E+04	2.6E+04
Georgia	1.3E+04	2.1E+04	0.0E+00
Moldova	2.1E+03	6.1E+03	0.0E+00
Russia (EU)	4.1E+04	4.6E+05	5.6E+04
Ukraine	3.7E+04	9.1E+04	2.0E+04
Albania	7.5E+03	9.3E+03	0.0E+00

Bosnia and Herzegovina	7.5E+03	6.9E+03	0.0E+00
Malta	2.6E+01	7.4E+02	2.7E+01
Serbia and Montenegro	5.9E+04	3.1E+04	0.0E+00
Turkey	1.9E+05	2.5E+05	1.9E+05
Norway	3.0E+04	2.8E+04	3.1E+04

Table S4 shows the comparison of wood consumption estimates in the residential sector in 2010 provided by the TNO RWC inventory (Denier van der Gon et al., 2015), EDGARv4.3.2 (Janssens-Maenhout et al., 2017, in prep.) and UNFCCC 2015. The TNO RWC estimates are evaluated using per capita consumption provided by Denier van der Gon et al. (2015), while the UNFCCC estimates are derived from the 2015 national emission inventory reporting. Note that EDGARv4.3.2 does not have information on the wood consumption in the residential sector for several countries of the Former Soviet Union (e.g. Azerbaijan, Armenia, Georgia, Moldova), but also for Albania, Bosnia Herzegovina, Serbia. Wood consumption in the residential sector is very low for Malta and Cyprus accordingly with the EDGARv4.3.2 and UNFCCC data while higher values are reported by Denier van der Gon et al. (2015). In addition the wood consumption in Russia estimated using the per capita information provided by Denier van der Gon et al. (2015) is 11 times higher compared to the activity data available in EDGARv4.3.2 and UNFCCC. Being one of the world top producers of crude oil and natural gas, the wood consumption in Russia for the residential sector is estimated to be quite low in particular in urban regions due to the use of district heating and natural gas in the household sector in this region, as reported by Nejat et al. (2015).

Table S5 – Uncertainty of the residential emissions for each pollutant and region including the uncertainty of wood consumption in the household sector in the European domain (TNO RWC vs. EDGARv4.3.2).

pollutant	σ_{EMI,RES_bio} (Annex I countries)	σ_{EMI,RES_bio} (non Annex-I & EIT)
SO2	63%	80%
NOx	107%	155%
CO	107%	155%
NMVOC	107%	155%
CH4	204%	302%
NH3	204%	302%
PM10	204%	302%
PM2.5	204%	302%
BC	204%	302%
OC	204%	302%

Emission uncertainties are calculated following the methodology described in Sect. 3.4.3 through the comparison of the TNO RWC wood consumption estimates and the corresponding EDGARv4.3.2 data (refer to Table S5) and through the comparison of the TNO RWC wood consumption estimates with the UNFCCC 2015 reports for the year 2010 (refer to Table S6).

Table S6 – Uncertainty of the residential emissions for each pollutant and region including the uncertainty of wood consumption in the household sector in the European domain (TNO RWC vs. UNFCCC).

pollutant	σ_{EMI,RES_bio} (Annex I countries)	σ_{EMI,RES_bio} (non Annex-I & EIT)
SO2	78%	92%
NOx	116%	161%
CO	116%	161%
NMVOC	116%	161%
CH4	209%	306%
NH3	209%	306%
PM10	209%	306%
PM2.5	209%	306%
BC	209%	306%
OC	209%	306%

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