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Improved definition of prior uncertainties in CO_2 and CO fossil fuel fluxes and its impact on multi-species inversion with GEOS-Chem (v12.5)

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S1. AD/EF uncertainties per sector-fuel combination

Overview of country-level relative uncertainties in activity data (AD) and emission factors (EF) at the highest level of detail (sector-fuel combinations). Solid, liquid and gaseous fuels refer to the non-biomass fraction only; the remainder is collected under biomass. Waste is only the non-renewable fraction.

The uncertainties in AD and CO_2 EFs are taken from the NIR reports and are therefore country-specific. We show here the median (lowest - highest) reported uncertainties for all countries, which represent 95 % confidence intervals. These values are used both as lower and upper limit and therefore a Gaussian uncertainty distribution is applied.

For CO and NO_x the uncertainties are taken from the EMEP guidebook and apply equally to all countries. The values represent the lower and upper limit of the 95 % confidence interval. For power production there are more fuel types separated in the EMEP guidebook than in the emission reporting and we use the EF uncertainty of the dominant fuel type in Europe, based on Eurostat data (Supply, transformation and consumption of solid fossil fuels).

The uncertainty calculations in the NIRs and EMEP guidebook may use different approaches. For GHGs we take the most detailed one (Tier 2 (Monte Carlo) if available, else Tier 1 (error propagation)). For many countries Tier 2 estimates are available, but often the differences with the Tier 1 estimates are small. For air pollutants we typically use the Tier 1 values that represent sub-sectors. Tier 2 and 3 values are related to specific combustion technologies, which are not considered in the sub-sector definition.

The level of detail in the reported uncertainties differs per sector, and for GHGs also per country, but in all cases gap filling is needed to cover all important sub-sectors. Missing sub-sector level uncertainties are gap filled using the uncertainty of a higher (more aggregated) sector level if available, e.g., for CO_2 the uncertainties for road transport (per fuel type) are used for all vehicle types. Otherwise, for CO_2 we apply gap filling with the median uncertainty for that sector based on all countries that do report an uncertainty for this sector. The variability between countries in reported uncertainties for AD and the EF of CO_2 is limited and therefore this approach is suitable. We also use this median uncertainty for countries with emission reporting, but without an uncertainty estimate. For air pollutants we make use of the generic uncertainty ranges (95 % confidence interval (CI)) provided in Chapter 5 of the EMEP guidebook to gap fill missing sectors. This uncertainty range differs per sector, but often amounts to an interval of -50 % and +100 %.

Table S1: Overview of country-level relative uncertainties in activity data (AD) and emission factors (EF). Uncertainties represent limits of the 95 % confidence interval.

Sector description	Sector (GNFR)	Fuel	AD	CO_2 EF	CO EF	NO_x EF
Public electricity and heat production	1.A.1.a (A)	Solid fuels	0.021 (0.01–0.083)	0.033 (0.01–0.080)	0.23–5.95	0.83–1.08
		Liquid fuels	0.019 (0.005–0.083)	0.02 (0.001–0.2)	0.40–0.40	0.42–1.31
		Gaseous fuels	0.020 (0.004–0.083)	0.023 (0.003–0.05)	0.49–0.54	0.51–1.11
		Biomass	0.035 (0.01–0.2)	0.096 (0.023–0.6)	0.50–1.00	0.51–0.98
		Waste	0.033 (0.01–0.2)	0.092 (0.023–0.6)	0.50–1.00	0.35–0.35
Oil and gas refining (comb)	1.A.1.b (B)	Liquid fuels	0.029 (0.006–0.107)	0.026 (0.001–0.25)	0.40–0.40	0.51–1.11
		Gaseous fuels	0.02 (0.005–0.107)	0.023 (0.003–0.120)	0.49–0.54	0.83–1.08
Oil and gas refining	1.B.2.a.iv (B)		0.05 (0.005–0.5)	0.087 (0.005–0.75)	0.67–1.89	0.67–1.89
Oil production (comb)	1.A.1.c (D)	Liquid fuels	0.02 (0.007–0.130)	0.025 (0.001–0.113)	0.50–1.00	0.45–1.00
		Gaseous fuels	0.022 (0.001–0.22)	0.025 (0.005–0.113)	0.50–1.00	0.45–1.00
Gas exploration (comb)	1.A.1.c (D)	Liquid fuels	0.02 (0.007–0.130)	0.025 (0.001–0.113)	0.35–0.35	0.35–0.35
		Gaseous fuels	0.022 (0.001–0.22)	0.025 (0.005–0.113)	0.35–0.35	0.35–0.35
Coke ovens (comb)	1.A.1.c (B)	Solid fuels	0.023 (0.01–0.130)	0.037 (0.01–0.389)	0.50–1.00	0.45–1.00
Solid fuel transformation	1.B.1.b (B)		0.040 (0.01–0.06)	0.323 (0.049–2)	0.78–3.59	0.78–4.11

Sector description	**Sector (GNFR)**	**Fuel**	**AD**	**CO_2 EF**	**CO EF**	**NO_x EF**
Exploration, production, transport	1.B.2.a.i (D)		0.05 (0.005–0.5)	0.087 (0.005–0.75)	0.50–1.00	0.50–1.00
Iron and steel industry (comb)	1.A.2.a (B)	Solid fuels	0.03 (0.015–0.151)	0.05 (0.02–0.389)	0.84–1.15	0.13–0.16
		Gaseous fuels	0.03 (0.008–0.151)	0.03 (0.003–0.5)	0.28–0.66	0.38–0.39
Iron and steel production	2.C.1 (B)		0.05 (0.005–0.1)	0.05 (0.005–0.25)	0.56–1.29	0.13–0.16
Non-ferrous metals (comb)	1.A.2.b (B)	Gaseous fuels	0.03 (0.008–0.104)	0.025 (0.003–0.5)	0.28–0.66	0.38–0.39
Aluminium production	2.C.3 (B)		0.02 (0.015–0.05)	0.05 (0.005–0.2)	0.17–0.25	0.50–1.00
Chemical industry (comb)	1.A.2.c (B)	Liquid fuels	0.03 (0.007–0.11)	0.028 (0.001–0.25)	0.39–0.41	0.40–0.40
		Gaseous fuels	0.03 (0.008–0.07)	0.03 (0.003–0.5)	0.28–0.66	0.38–0.39
Chemical industry	2.B.1 (B)		0.02 (0.01–0.5)	0.05 (0.015–0.5)	0.50–1.00	0.95–0.50
Pulp and paper industry (comb)	1.A.2.d (B)	Gaseous fuels	0.03 (0.008–0.089)	0.025 (0.003–0.5)	0.28–0.66	0.38–0.39
		Biomass	0.05 (0.01–0.193)	0.07 (0.03–1)	0.91–6.02	0.78–0.32
Pulp and paper industry	2.H.1 (B)		0.037 (0.005–0.07)	0.05 (0.05–0.05)	0.50–1.00	0.15–1.60
Food processing, beverages and tobacco (comb)	1.A.2.e (B)	Liquid fuels	0.03 (0.007–0.11)	0.03 (0.001–0.25)	0.39–0.41	0.40–0.40
		Gaseous fuels	0.03 (0.008–0.07)	0.03 (0.003–0.5)	0.28–0.66	0.38–0.39
Non-metallic minerals (comb)	1.A.2.f (B)	Solid fuels	0.037 (0.016–0.135)	0.05 (0.017–0.389)	0.84–1.15	0.13–0.16

Sector description	**Sector (GNFR)**	**Fuel**	**AD**	**CO_2 EF**	**CO EF**	**NO_x EF**
		Liquid fuels	0.03 (0.007–0.135)	0.021 (0.001–0.25)	0.39–0.41	0.40–0.40
		Gaseous fuels	0.03 (0.008–0.135)	0.021 (0.003–0.5)	0.28–0.66	0.38–0.39
		Waste	0.05 (0.01–0.193)	0.052 (0.02–0.6)	0.35–0.35	0.78–0.32
Cement production	2.A.1 (B)		0.02 (0.000–0.146)	0.025 (0.003–0.5)	0.50–1.00	0.73–2.76
Other non-metallic mineral production	2.A.2 (B)		0.028 (0.001–0.35)	0.025 (0.003–0.1)	0.50–0.40	0.40–0.40
Other manufacturing industry (comb)	1.A.2.g.viii (B)	Solid fuels	0.037 (0.004–0.41)	0.05 (0.009–0.389)	0.84–1.15	0.13–0.16
		Liquid fuels	0.03 (0.007–0.41)	0.028 (0.001–0.25)	0.39–0.41	0.40–0.40
		Gaseous fuels	0.03 (0.008–0.41)	0.03 (0.003–0.5)	0.28–0.66	0.38–0.39
		Biomass	0.05 (0.01–0.41)	0.053 (0.038–0.5)	0.91–6.02	0.78–0.32
Passenger cars	1.A.3.b.i (F)	Gasoline	0.03 (0.007–0.407)	0.03 (0.001–0.06)	0.42–2.18	0.49–2.42
		Gas/ diesel oil	0.04 (0.009–0.407)	0.03 (0.001–0.2)	0.38–1.46	0.14–0.07
		LPG	0.03 (0.01–0.55)	0.03 (0.01–0.2)	0.54–0.38	0.73–1.26
		Biomass	0.05 (0.01–0.55)	0.03 (0.01–0.3)	1.00–2.00	1.00–2.00
Light duty vehicles	1.A.3.b.ii (F)	Gas/ diesel oil	0.04 (0.009–0.407)	0.03 (0.001–0.2)	0.14–0.58	0.10–0.24
		Gasoline	0.03 (0.007–0.407)	0.03 (0.001–0.06)	0.55–0.56	0.75–0.93

Sector description	Sector (GNFR)	Fuel	AD	CO_2 EF	CO EF	NO_x EF
Trucks (>3.5t)	1.A.3.b.iii (F)	Gas/ diesel oil	0.04 (0.009–0.407)	0.03 (0.001–0.2)	0.24–0.39	0.15–0.15
		Gasoline	0.03 (0.007–0.407)	0.028 (0.001–0.06)	0.55–0.56	0.75–0.93
Buses	1.A.3.b.iii (F)	Gas/ diesel oil	0.04 (0.009–0.407)	0.03 (0.001–0.2)	0.61–1.63	0.15–0.15
Motorcycles	1.A.3.b.iv (F)	Gasoline	0.03 (0.007–0.407)	0.03 (0.001–0.06)	0.33–0.34	0.70–0.62
Mopeds	1.A.3.b.v (F)	Gasoline	0.03 (0.007–0.407)	0.029 (0.001–0.06)	0.33–0.34	0.70–0.62
Civil aviation - LTO	1.A.3.a.i(i) (H)	Gasoline	0.098 (0.01–0.356)	0.05 (0.01–0.056)	0.50–1.00	0.50–1.00
		Kerosene	0.075 (0.010–0.356)	0.05 (0.002–0.056)	0.50–1.00	0.50–1.00
Railways	1.A.3.c (I)	Gas/ diesel oil	0.05 (0.01–0.2)	0.031 (0.01–0.2)	0.44–0.78	0.52–0.77
Domestic navigation, inland shipping	1.A.3.d.i(ii) (G)	Gas/ diesel oil	0.074 (0.01–0.825)	0.029 (0.01–0.091)	0.99–1.50	0.50–1.00
Agriculture/ Forestry/Fishing - Off-road vehicles and other machinery	1.A.4.c.ii (I)	Gasoline	0.05 (0.02–0.273)	0.06 (0.03–0.5)	0.50–1.00	0.50–1.00
		Gas/ diesel oil	0.05 (0.02–0.273)	0.06 (0.03–0.5)	0.50–1.00	0.50–1.00
Manufacturing industry - Off-road vehicles and other machinery	1.A.2.g.vii (I)	LPG	0.05 (0.01–0.41)	0.05 (0.038–0.5)	0.50–1.00	0.50–1.00
		Gasoline	0.05 (0.01–0.41)	0.05 (0.038–0.5)	0.50–1.00	0.50–1.00
		Gas/ diesel oil	0.05 (0.01–0.41)	0.05 (0.038–0.5)	0.50–1.00	0.50–1.00
Commercial/ institutional - Mobile	1.A.4.a.ii (I)	Gasoline	0.05 (0.02–0.35)	0.05 (0.016–0.2)	0.50–1.00	0.50–1.00
	1.A.4.b.ii	Gasoline	0.05	0.05	0.50–1.00	0.50–1.00

Sector description	**Sector (GNFR)**	**Fuel**	**AD**	**CO_2 EF**	**CO EF**	**NO_x EF**
Residential - Household and gardening	(I)		(0.02–0.35)	(0.003–0.2)		
		Gaseous fuels	0.05 (0.014–0.35)	0.032 (0.003–0.5)	0.50–1.00	0.50–1.00
Other mobile combustion	1.A.5.b (I)	Gasoline	0.03 (0.005–0.1)	0.03 (0.003–0.2)	0.50–1.00	0.50–1.00
		Gaseous fuels	0.035 (0.005–0.2)	0.023 (0.003–0.05)	0.50–1.00	0.50–1.00
Commercial/ institutional	1.A.4.a.i (C)	Liquid fuels	0.05 (0.007–0.35)	0.021 (0.001–0.1)	0.74–1.15	0.84–3.31
		Gaseous fuels	0.05 (0.014–0.35)	0.032 (0.003–0.5)	0.28–0.66	0.38–0.39
		Biomass	0.062 (0.02–0.35)	0.06 (0.016–0.2)	0.91–6.02	0.78–0.32
Residential	1.A.4.b.i (C)	Solid fuels	0.05 (0.02–0.35)	0.05 (0.003–0.5)	0.35–0.52	0.67–0.82
		Liquid fuels	0.05 (0.007–0.35)	0.018 (0.001–0.1)	0.31–0.62	0.39–0.41
		Gaseous fuels	0.05 (0.014–0.35)	0.032 (0.003–0.5)	0.74–1.15	0.39–0.39
		Biomass	0.05 (0.02–0.35)	0.05 (0.003–0.2)	0.75–1.50	0.40–2.00
Agriculture/ Forestry/Fishing	1.A.4.c.i (C)	Gaseous fuels	0.05 (0.014–0.273)	0.035 (0.003–0.5)	0.28–0.66	0.38–0.39
		Biomass	0.05 (0.02–0.273)	0.071 (0.03–0.5)	0.91–6.02	0.78–0.32
Field burning of agricultural residues	3.F (L)		0.2 (0.2–0.2)	0.2 (0.2–0.2)	0.43–0.43	0.22–0.26
Landfills	5.A (J)		0.135 (0.071–0.2)	0.071 (0.071–0.071)	0.50–1.00	0.50–0.50
Open burning of waste	5.C.2 (J)		0.320 (0.320–0.320)	0.4 (0.4–0.4)	0.67–2.00	0.67–2.00

Sector description	**Sector (GNFR)**	**Fuel**	**AD**	**CO_2 EF**	**CO EF**	**NO_x EF**
Other waste	5.E (J)		0.1 (0.1–0.1)	3 (3–3)	0.50–1.00	0.50–0.50

S2. Sensitivity analysis on error correlations in AD/EF

The errors in AD and EF between sub-sectors are not always independent. For example, at the country level the total gasoline consumption by road transport is well-known. However, the overall fuel consumption is divided over different vehicle types and road types. The error that is made in assigning the fuel consumption to each of these sub-categories needs to be compensated for by another sub-category. Hence, the error in the AD is in this case negatively correlated. Also for EFs error correlations may occur, for example for sectors that receive the same EF estimate because they use similar technologies. It is likely that if the EF of one of those sectors is overestimated that this is also true for the other related sector(s). In this case, the error correlation is positive.

Unfortunately, the error correlations are unknown. To better understand the impact of assuming a particular error correlation between AD and EF on the error in country-level emissions we run a Monte Carlo simulation (N=2000) for different correlations and compare the PDFs of the emissions. We assume negative error correlations in AD exist for:

- road transport: correlations between all vehicles using the same fuel type;
- biomass consumption for sub-sectors under GNFR C (other stationary combustion);
- liquid fuel consumption for sub-sectors under GNFR C (other stationary combustion);
- off-road transport and machinery using the same fuel type.

We assume positive error correlations in EF exist for:

- road transport for vehicle and fuel type groups;
- off-road transport per sub-sector.

Figure S1 shows the probability density function (PDF) of the emissions for each pollutant with different AD and EF correlation strengths. We find no significant difference between the set-ups. When we look at the different sectors (Fig. S2) we see no differences for the public power, industry and shipping sectors, for which we indeed do not assume any error correlations in AD and EF. For other stationary combustion error correlations only exist for fuels that are of minor importance, which results in no clear impact on the PDF. For road transport the inclusion of error correlations, irrespective of their exact value, seems to make a minor difference. For the other sector the impact is inconclusive, as including error correlations of -0.6 (AD) and +0.5 (EF) result in a similar PDF as when having no error correlations. Due to the large variety of sub-sectors this could be the result of random changes in the ensemble. Indeed, if we redo the Monte Carlo simulation the PDFs for this sector change and also shift with respect to each other. Hence, we conclude that the impact of error correlations in AD and EF is very limited, also compared to other sources of uncertainty.

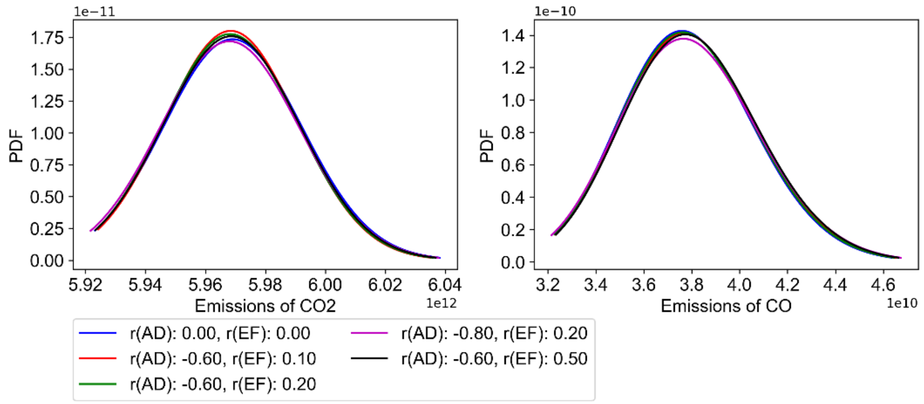


Figure S1: PDF of emissions of CO_2 and CO (kg a^{-1}) for the whole European domain resulting from a Monte Carlo simulation (N=2000) with different AD and EF correlation strengths (r).

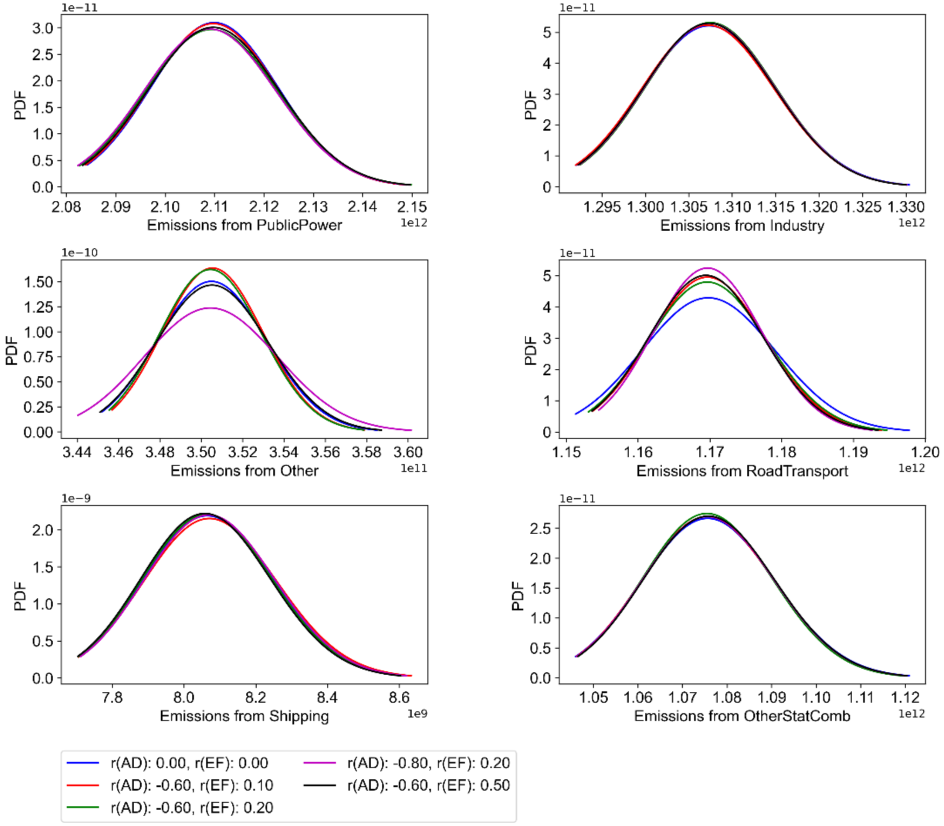


Figure S2: PDF of emissions of CO_2 (kg a^{-1}) for the whole European domain for different sectors resulting from a Monte Carlo simulation (N=2000) with different AD and EF correlation strengths (r).

S3. CO:CO2 error correlations

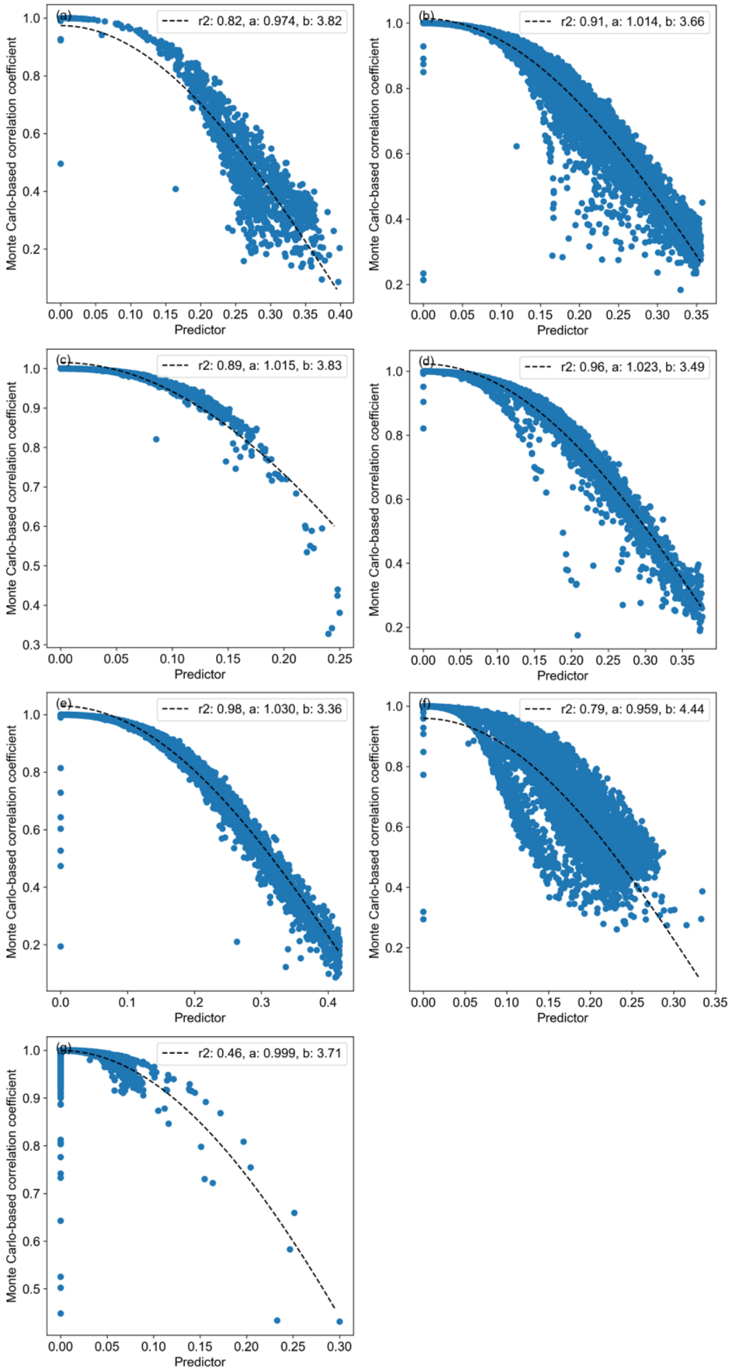


Figure S3: Scatter plots of Monte-Carlo (N=500) based correlation coefficient (r) per grid cell against the predictor calculated with Eq. 3 for (a) Netherlands, (b) Germany, (c) Czech Republic, (d) Italy, (e) France, (f) UK, and (g) Sweden for GNFR C. The correlation (R^2) and cosine function parameters are also shown.

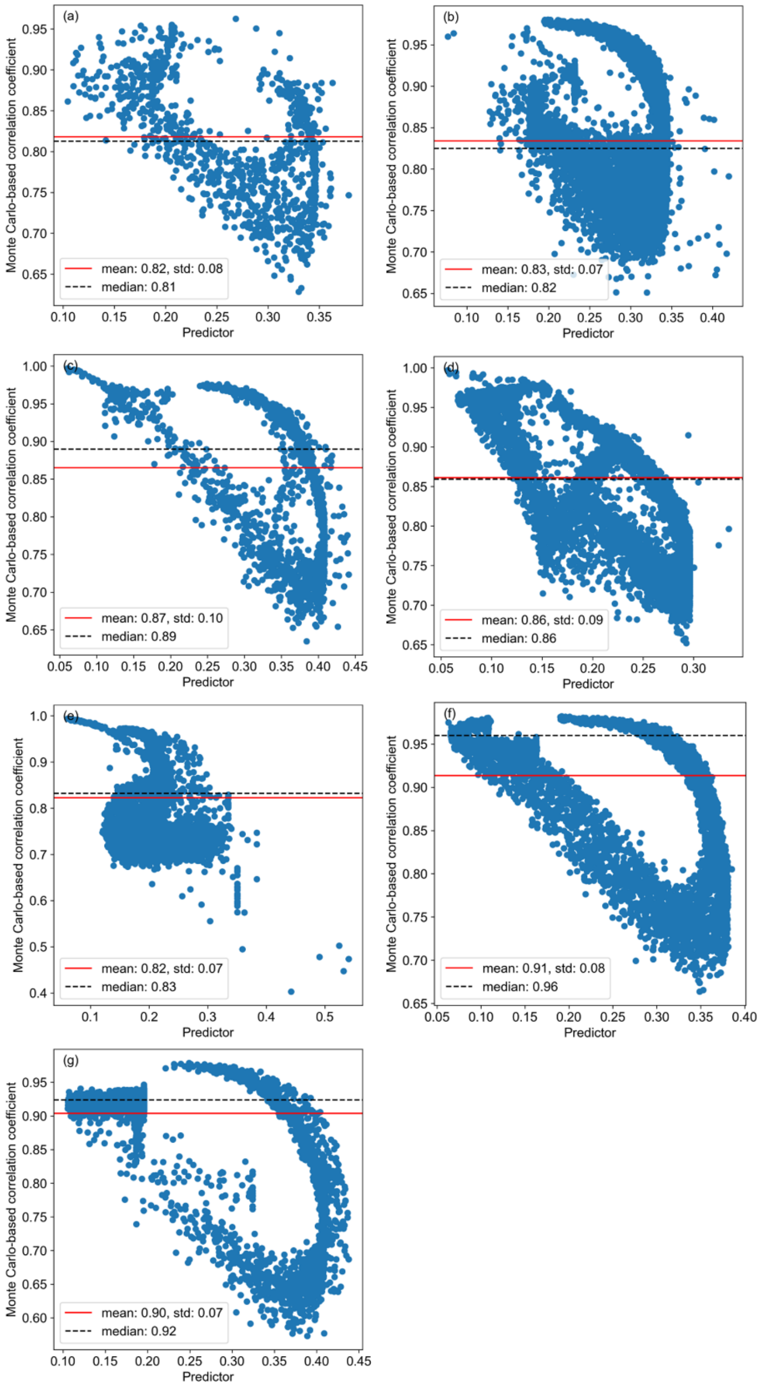


Figure S4: Scatter plots of Monte-Carlo (N=500) based correlation coefficient (r) per grid cell against the predictor calculated with Eq. 3 for (a) Netherlands, (b) Germany, (c) Czech Republic, (d) Italy, (e) France, (f) UK, and (g) Sweden for GNFR F. The mean, median and standard deviation (std) of the correlation coefficients are also shown.

S4. Comparison of Monte Carlo approach and uncertainty propagation

There are several methods to propagate uncertainties through a model to estimate the uncertainty in the output. In this work we use relatively simple uncertainty propagation equations, as explained in the manuscript. The disadvantage of this approach is that it is more difficult to deal with error correlation or non-Gaussian uncertainty distributions. With more complex data a Monte Carlo approach is often more suited, but also computationally demanding. A Monte Carlo simulation produces a range of possible outcome values for any variable based on its probability distribution. It relies on random sampling from the probability distribution, which allows for different distribution shapes (e.g., lognormal, uniform). Another advantage of the Monte Carlo simulation is that it can deal with many interdependent input variables.

Here we compare the results of both approaches for the country-level emissions and spatial distribution. The uncertainty propagation approach is described in detail in the main manuscript. For the Monte Carlo approach we create an ensemble of country-level emissions or spatial distributions using Eq. 10–12 from the main manuscript.

First, we compare the normalized spread in total country-level emissions for the European emissions (Fig. S5). The box plots show the spread in the ensemble (N = 2000) created with the Monte-Carlo approach, with the whiskers representing the 95 % CI. The red dots show the 95 % CI from the simple uncertainty propagation method. For CO_2 we generally see a very small range of just a few percent and the ranges appear very similar for both methods. For CO the spread is larger and more skewed towards high values. This is the result of the EF uncertainties which are often strongly lognormal. Again, the simple uncertainty propagation method seems to mimic this relatively well, although the lower limits show stronger deviations.

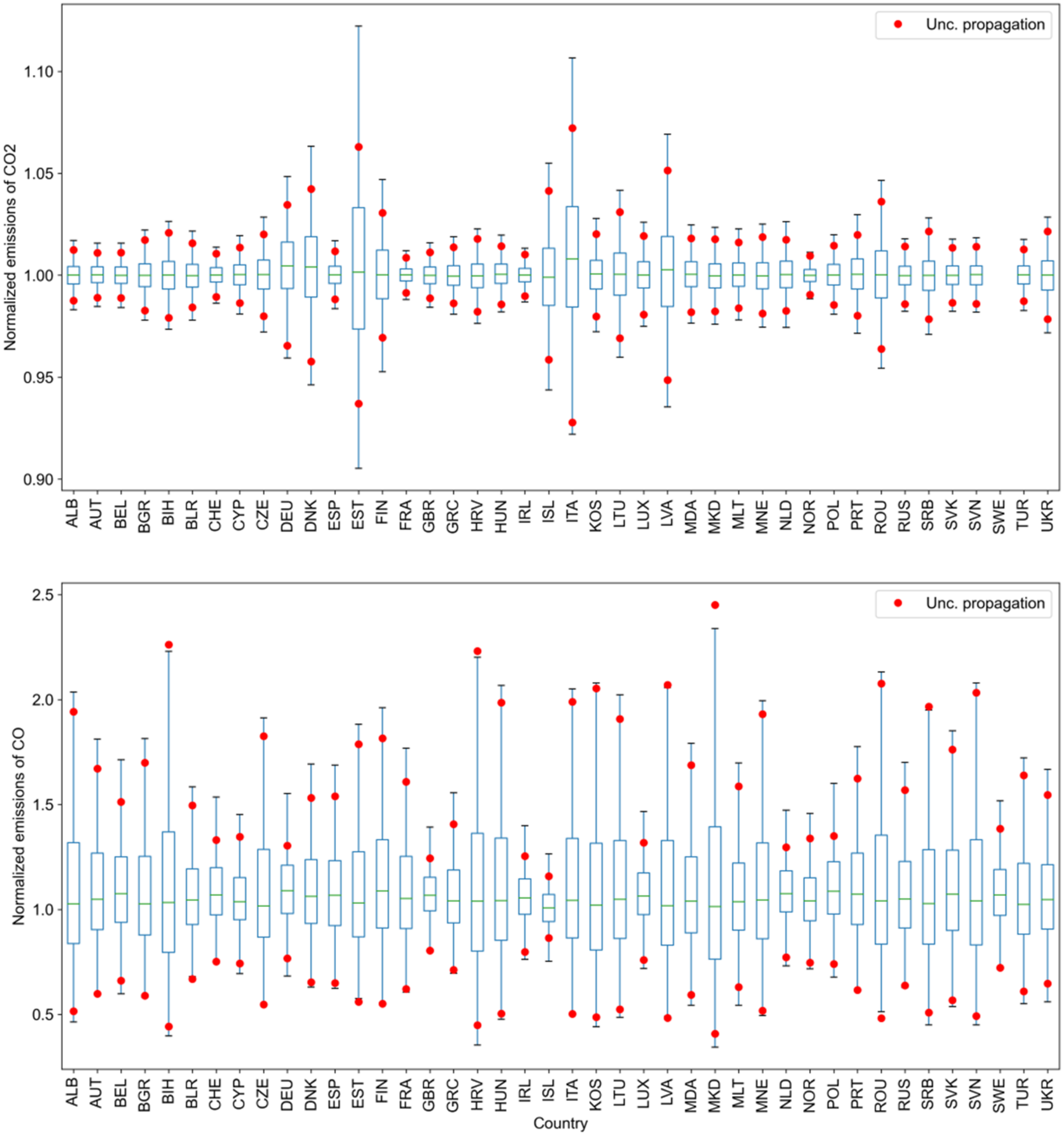


Figure S5: Normalized spread in emissions of CO_2 and CO for European countries resulting from a Monte Carlo simulation (N=2000, no error correlations) and the 95 % confidence interval as calculated from the uncertainty propagation method (red dots).

Secondly, we create a Monte-Carlo ensemble (N=200) of spatial distributions for the Dutch road transport sector. We compare the results against the uncertainty propagation method, from which we also create an ensemble using the Monte Carlo simulation with the aggregated uncertainties and the averaged proxy map. The reason for this is that we have a relatively small ensemble size (N=200) to reduce computational costs and this may not be sufficient to compare to the uncertainty propagation method directly. We compare the standard deviation in the emissions of each grid cell in the two ensembles. The result is shown in Fig. S6. We see a strong correlation in the standard deviation per grid cell. This suggests that the uncertainty propagation method gives a good estimate of the overall grid cell uncertainty and variations between grid cells.

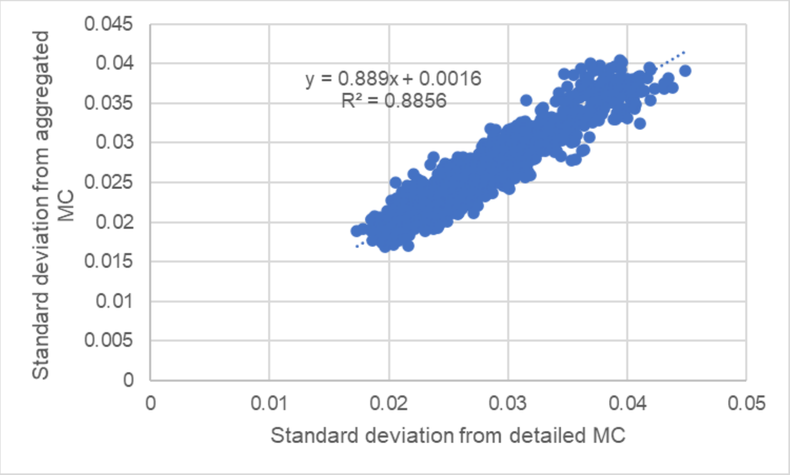
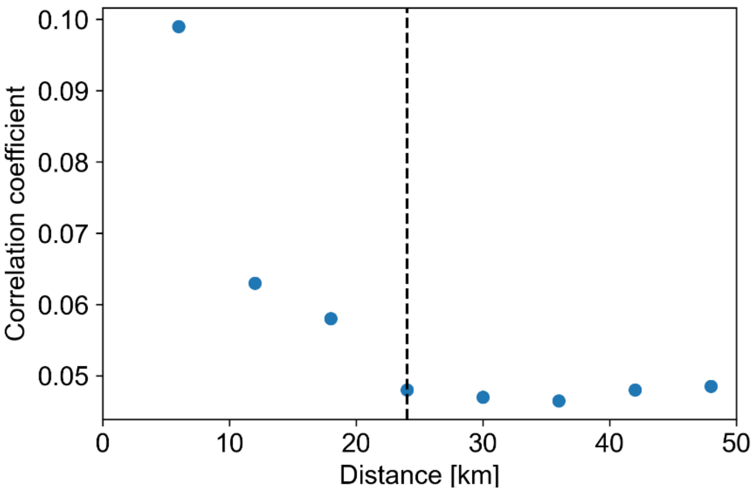


Figure S6: Scatter plot of standard deviation per pixel with road transport emissions (N = 1099) in the ensemble resulting from the detailed Monte Carlo simulation (x-axis) and the Monte Carlo simulation with aggregated uncertainties from the uncertainty propagation method (y-axis).

Finally, we look at the error correlation length in the aggregated sector emissions. We calculate the correlation coefficient between two grid cells from the full Monte Carlo ensemble. The average correlation coefficient per grid cell distance (6 km) is shown in Fig. S7. From the uncertainty propagation method we estimate an average correlation length of approximately 24 km for road transport (dashed vertical line in Fig. S7). This matches well with the decay in the correlation coefficient.



110 **Figure S7: The average correlation coefficient between pixels at certain distance from each other, as calculated from the detailed Monte Carlo ensemble.**