CAMS Service Evolution

D5.3 Uncertainties in primary PM emissions from CAMS-REG at the grid cell level

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1 Executive Summary

This deliverable presents a comprehensive methodology to estimate uncertainties in gridded, regional primary PM emissions. The uncertainties data consist initially of country-level emission uncertainties, gridded uncertainties related to the spatial distribution, and spatial correlation lengths. These estimates are based on a wide range of data at the highest level of detail possible (generally per sub-sector and fuel) and aggregated to the GNFR sector using error propagation. We present an overview of the data that is used and how everything is combined. The results show that significant uncertainties exist in primary PM emissions, both at the national scale and in the spatial distribution.

Next, we demonstrate an optimization procedure, which is used to create consistency between country-level and gridded uncertainties. The results of the optimization procedure are scaled spatial correlation lengths and gridded uncertainties, which match with the country-level uncertainties. In this way, the gridded uncertainties can be used directly instead of having to combine them with country-level data. This is more user-friendly, but also increases consistency and statistical soundness.

Finally, we look into the uncertainty related to the PM split into different PM components. Because the PM split consists of fractions, which for each sector and country should sum up to one, there are additional constraints to consider. The results show a strong impact of methodological choices and whether or not we consider error correlations to exist between the different components. We conclude more effort is needed to get a handle on the PM component uncertainties and error correlations.

The data provided in this deliverable are gridded uncertainties of primary PM (PM2.5 and PM10), NMVOC, SOX, NOX and NH3 and spatial correlation lengths per sector, which match the CAMS-REG-AP_v6_1_Ref2_v2_1_emissions_year2019. We also provide aggregated uncertainties per country and sector. These data are intended for use in CAMEO WP6.

Table of Contents

2 Introduction

2.1 Background

Monitoring the composition of the atmosphere is a key objective of the European Union's flagship Space programme Copernicus, with the Copernicus Atmosphere Monitoring Service (CAMS) providing free and continuous data and information on atmospheric composition.

For monitoring atmospheric composition a combination of data and models is used to provide a starting point for the state of the atmosphere: fluxes of atmospheric pollutants (both anthropogenic and biogenic), atmospheric transport processes, and atmospheric concentrations of air pollutants. All this information is combined and weighted to optimize knowledge on air pollution and emissions.

The uncertainty in the data products and models can have a significant impact on the final data and information that is provided through CAMS, and as such on policy making. Therefore, an important challenge is to quantify the uncertainties related to all the data products and models in CAMS.

Emission uncertainties are an essential input for the optimization of emissions through inverse modelling, but also, for example, to better understand their impact on source-receptor calculations. One difficulty is the complexity of the data sets and the existence of error correlations. Therefore, a methodology needs to be developed to accurately and consistently describe uncertainties and error correlations in gridded emissions data.

2.2 Scope of this deliverable

2.2.1 Objectives of this deliverables

The objective of this deliverable is provide regional uncertainties in gridded primary PM emission data to support work done in WP6 on source-receptor calculations.

2.2.2 Work performed in this deliverable

In this deliverable the work as planned in the Description of Action (DoA, WP5 T5.3.1) was performed, adding to the milestone M10 extra sub-sectors and an update to the data to improve consistency. In addition to primary PM, data is also provided for NMVOC, SOX, NOX and NH3 as requested by the users. Finally, we have performed a first analysis of the uncertainties and error correlations in the PM split into different PM components.

2.2.3 Deviations and counter measures

No deviations have been encountered.

2.2.4 CAMEO Project Partners:

ECMWF	EUROPEAN CENTRE FOR MEDIUM-RANGE WEATHER FORECASTS
Met Norway	METEOROLOGISK INSTITUTT
BSC	BARCELONA SUPERCOMPUTING CENTER-CENTRO NACIONAL DE SUPERCOMPUTACION
KNMI	KONINKLIJK NEDERLANDS METEOROLOGISCH INSTITUUT-KNMi
SMHI	SVERIGES METEOROLOGISKA OCH HYDROLOGISKA INSTITUT
BIRA-IASB	INSTITUT ROYAL D'AERONOMIE SPATIALEDE

	BELGIQUE
HYGEOS	HYGEOS SARL
FMI	ILMATIETEEN LAITOS
DLR	DEUTSCHES ZENTRUM FUR LUFT - UND RAUMFAHRT EV
ARMINES	ASSOCIATION POUR LA RECHERCHE ET LE DEVELOPPEMENT DES METHODES ET PROCESSUS INDUSTRIELS
CNRS	CENTRE NATIONAL DE LA RECHERCHE SCIENTIFIQUE CNRS
GRASP-SAS	GENERALIZED RETRIEVAL OF ATMOSPHERE AND SURFACE PROPERTIES EN ABREGE GRASP
CU	UNIVERZITA KARLOVA
CEA	COMMISSARIAT A L ENERGIE ATOMIQUE ET AUX ENERGIES ALTERNATIVES
MF	METEO-FRANCE
TNO	NEDERLANDSE ORGANISATIE VOOR TOEGEPAST NATUURWETENSCHAPPELIJK ONDERZOEK TNO
INERIS	INSTITUT NATIONAL DE L ENVIRONNEMENT INDUSTRIEL ET DES RISQUES - INERIS
IOS-PIB	INSTYTUT OCHRONY SRODOWISKA - PANSTWOWY INSTYTUT BADAWCZY
FZJ	FORSCHUNGSZENTRUM JULICH GMBH
AU	AARHUS UNIVERSITET
ENEA	AGENZIA NAZIONALE PER LE NUOVE TECNOLOGIE, L'ENERGIA E LO SVILUPPO ECONOMICO SOSTENIBILE

3 Methods

3.1 Emission uncertainties

The baseline emission inventory used in this work is the CAMS-REG-AP_v6_1_Ref2_v2_1_emissions_year2019, which provides emissions of air pollutants for the European domain for the year 2019. The dataset was made as part of the CAMS emissions service and is an updated version of the inventory described in Kuenen et al. (2022). Official country reported emissions are used as a starting point for this inventory and spatially distributed using a range of spatial data sets. This inventory furthermore includes a specific science-based inventory for PM emissions from small combustion, instead of the reported emission data, which fully accounts for condensable organics in PM emissions (Simpson et al., 2022).

Uncertainties in the emission inventory occur from all input data. The country-level emission uncertainties can be calculated from uncertainties in the activity data and emission factors, whereas uncertainties in the gridding depend on the spatial proxies. We also calculate spatial error correlation lengths. A detailed description of the methodology is presented elsewhere (Super et al., 2024), but a summary is given below.

The country-level data and the spatial data are from independent sources and can therefore be inconsistent. However, the gridded uncertainties should ideally add up to the country-level uncertainties. For this purpose, we have developed an optimization methodology to ensure full consistency between the uncertainties from the different data sources.

3.1.1 Country-level emission uncertainties

Country-level emissions are based on activity data and emission factors. For greenhouse gases uncertainties in both parameters are reported in the countries' NIR reports, submitted annually to UNFCCC (UNFCCC, 2021). We use the data submitted in 2020 and extract country-specific activity data uncertainties per sub-sector/fuel (the level of detail differs per country). We assume that the activity data, at least for fuel combustion, is shared between all pollutants and greenhouse gases and therefore we create consistency when using these data for all species. A gap filling procedure is applied to ensure all sub-sectors/fuels are covered for all countries (Super et al., 2024). Additionally, we add non-combustion related uncertainties. For example, for road and brake wear we assume a similar uncertainty as for road transport fuel combustion, as both depend largely on the vehicle kilometres per vehicle type.

For the uncertainty in emission factors for air pollutants we make use of the EMEP/EEA air pollutant emission inventory guidebook 2019 (European Environment Agency, 2019). The guidebook provides emission factor ranges applicable mostly for Europe, so these ranges are applied to all countries in the domain. We use as much as possible Tier 1 (sub-sector) estimates per fuel, but sometimes a generic uncertainty range is assumed (based on a quality rating), or we select a dominant Tier 2 process for which an uncertainty range is provided, or we take an average of all Tier 2 processes if similar ranges apply.

The uncertainties (relative standard deviations) in activity data (AD) and emission factors (EF) are combined to get the uncertainty in emissions (E):

$$\frac{\sigma_E}{E} = \sqrt{\left(\frac{\sigma_{AD}}{AD}\right)^2 + \left(\frac{\sigma_{EF}}{EF}\right)^2}. \quad (1)$$

The emission uncertainties (absolute standard deviations) for sub-sectors (*sub*) can be propagated to get the uncertainty at the GNFR (*agg*) level (see Table 1 for an overview of GNFR sectors):

$$\sigma_{E,agg} = \sqrt{\sum_{s=1}^{n} \sigma_{E,sub,s}{}^2}. \quad (2)$$

Table 1. Overview of GNFR sectors in the CAMS-REG emission inventory.

GNFR sector	Sector name
A	Public Power
B	Industry
C	Other Stationary Combustion
D	Fugitives
E	Solvents
F1	Road Transport - gasoline
F2	Road Transport - diesel
F3	Road Transport - LPG
F4	Road Transport – non-exhaust
G	Shipping
H	Aviation
I	Off-Road
J	Waste
K	Agriculture Livestock
L	Agriculture Other

3.1.2 Gridded emission uncertainties

The country-level emissions (at the sub-sector level) are spatially distributed using ~40 different proxy maps. These proxy maps are based on spatial datasets, such as population density maps. Uncertainties in the proxy values are based on 1) metadata of underlying data or 2) literature in which the underlying data were compared to other datasets.

In addition, we try to define a representativeness error, which describes how well the proxy describes the spatial distribution of emissions of a certain sector. For road transport the location of roads is relatively well-defined, so this representativeness error is low. But population density is sometimes used as a default proxy when better data is lacking and in that case the representativeness error could be really large.

The two sources of error are combined following the equation:

$$\sigma_{total} = \sqrt{{\sigma_{proxy\ value}}^2 + {\sigma_{representativeness}}^2} \tag{3}$$

This means that there is one uncertainty value per combination of proxy map and sub-sector, which applies to all grid cells. However, by propagating these errors to the GNFR sector, taking into account the contribution of each sub-sector, we get spatially explicit uncertainties.

3.1.3 Spatial error correlation length

The spatial representativeness error causes gridded uncertainties to be correlated. For example, we know that heating demand per capita is lower in densely populated areas. If we do not correctly represent this relationship we make a similar error in areas with similar population density. So grid cells in a city centre have a correlated error. This correlation usually decreases with distance, following an exponential decay (see Eq. 5).

We estimate this correlation length for each proxy map using semi-variograms (Super et al., 2024). A semi-variogram describes the spatial autocorrelation as a function of distance, i.e., the degree of variability between points located at a certain distance from each other. In the case of the proxy maps points that are closer together are expected to be more similar, and therefore their errors are more strongly correlated.

The spatial correlation length is an important parameter, because it reduces the degree of freedom of the system. If not all grid cells are completely independent from each other we can use one observation to optimize multiple parameters, instead of just one. It also affects the summation of the gridded uncertainties to the country-level, which plays an important role in the optimization procedure described next.

3.1.4 Optimization of the covariance matrix

In the previous dataset the gridded uncertainties and country-level uncertainties were provided separately and they were not necessarily consistent. We consider the country-level uncertainties to be a reliable estimate of the actual uncertainty at this scale. Therefore, in an ideal case, the uncertainties in gridded emissions per country should add up to the country-level emission uncertainty, taking into account the spatial correlation length.

We can construct a spatial covariance matrix ***B*** as follows:

$$\boldsymbol{B} = \boldsymbol{S}\boldsymbol{C}\boldsymbol{S}^{\boldsymbol{T}}, \tag{4}$$

where ***S*** is a matrix containing gridded uncertainties (variances) and ***C*** is the spatial correlation matrix which describes the error correlation between grid cells using the following parameterization:

$$c_{i,j} = e^{-d_{i,j}/L}. \tag{5}$$

Following this equation, the correlation is a function of distance *d* between grid cells *i* and *j* and the spatial correlation length *L*. The sum of the gridded relative uncertainties belonging to a specific country and sector can be calculated as:

$$\sigma_{\eta,rel} = \frac{\sqrt{\boldsymbol{a}^{\boldsymbol{T}}\boldsymbol{B}\boldsymbol{a}}}{\sum_{i=1}^{n} E_i}, \tag{6}$$

where $\boldsymbol{a}^{\boldsymbol{T}}$ selects all cells belonging to a specific country and *E* is the emission per grid cell *i*. The standard deviation for a country is the sum of all elements from ***B*** where $\boldsymbol{a}^{\boldsymbol{T}}$ is not zero.

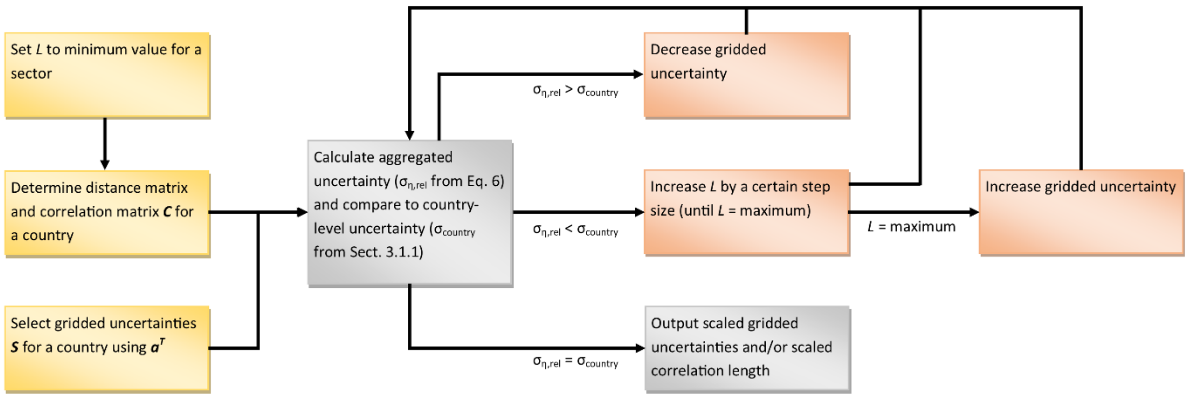

Figure 1. Flow diagram showing the steps taken for the optimization of the gridded covariances. Yellow boxes are related to the initialization and orange boxes to the optimization.

In most cases this standard deviation doesn't match with the uncertainty that was calculated for a country using the method described in Section 3.1.1. To match the two estimates we can optimize the covariance matrix ***B*** by scaling the gridded uncertainties ***S*** and/or by scaling the correlation length *L*. We assume that the correlation length has the highest uncertainty, as it strongly depends on the chosen methodology, and therefore our first option is to scale *L*. However, we set a lower (sector-specific) and upper limit for *L* to avoid getting physically unrealistic results. In some cases a match is impossible within these limits for *L* and in that case we optimize the gridded uncertainties. An example is when the result from Eq. 6 exceeds the country-level uncertainty following from Section 3.1.1. Adding/increasing the correlation length would further increase the aggregated uncertainty from Eq. 6 and a match is impossible without decreasing the gridded uncertainties. The whole process is also illustrated in Fig. 1.

3.2 PM split uncertainties

The primary PM emissions (PM2.5 and PM10) are split into 5 components (EC, OC, Na, SO4 and other minerals (hereafter OthMin)) following a sector- and country-specific profile. The sum of each profile adds up to 1 and is different for PM2.5 and PM10.

The PM split is provided per GNFR sector, but the underlying data contains more detailed sub-sectors and fuels. The fractions of EC and OC are taken from a wide range of scientific publications and datasets (e.g., (Bond et al., 2004; Kupiainen and Klimont, 2004; Schauer et al., 2006; Streets et al., 2001)). Expert judgement is used to select the most representative data from all these datasets. For Na a fixed fraction is assumed per GNFR sector, whereas for SO4 a fixed fraction is assumed per GNFR sector-fuel combination. OthMin gets the remainder and is therefore not based directly on any data.

We do not aim to exactly quantify uncertainties in the PM components for now, but rather to get an idea of how important it is to consider these components separately and whether we need to consider error correlations between the components. The difficulty in working with fractions is that they interact differently, as the sum should always be equal to one. This can be done in different ways, which may affect the final uncertainties.

To understand the uncertainties in the individual PM components we did a literature review for OC and EC to collect ranges of EC/OC fractions for all sub-sectors. We made assumptions on the most important/representative sub-sectors within each GNFR sector to get one uncertainty value per GNFR sector. This is a very rough estimate, but sufficient for our goal. For Na and SO4, which are based on expert judgement, we assume one uncertainty value for all sectors.

These uncertainties form the basis of a hypothetical test, in which we assume 1) no correlations between the PM components or 2) a full negative error correlation (r = -1) between OC and EC and between OC and OthMin. These correlations are based on the notion that the OC and EC emission factors are related to each other and to the carbonaceous fraction of PM:

$$EF_{OC} = \frac{EF_{PM} \cdot f_{carb} - EF_{BC}}{f_{OM}}, \tag{7}$$

where BC is black carbon (represents EC), f_{carb} is the carbonaceous content of PM, and f_{OM} is the average fraction of organic molecular weight per carbon weight (Klimont et al., 2017). Hence, an error in the EC emission factor leads to a similar (but opposite) error in the OC emission factor. And an error in the carbonaceous fraction results in an error in the sum of OC and EC, which is to be assigned to OthMin (assuming Na and SO4 fractions are independent).

We create an ensemble of perturbations (N = 50) for each component following these assumptions and explore how different methodological choices and/or the inclusion of error correlations affects the component fractions.

The steps taken for the hypothetical test are as follows (summarized in Fig. 2):

- Draw a random sample from a lognormal error distribution (to avoid negative fractions) with an expected value of 1 and using the uncertainties determined for each component, sub-sector and fuel; note that for now we assume no uncertainty in OthMin.
    - If we assume error correlations exist, the perturbations of EC and OthMin are made opposite to the perturbation of OC (e.g., if the perturbation of OC is 1.3, the perturbation for EC and OthMin is set to 0.7). If this leads to a value lower than zero the perturbations are set equal to zero.

- Use these perturbations to calculate new component fraction: new fraction = old fraction * perturbation.
- Enforce that the sum of all fractions is equal to one:
    - In the absence of error correlations the OthMin fraction is not altered (it didn't get an uncertainty) and we may assume that this component will compensate for the others (as long as it remains a zero-positive value). Next, we divide all fractions by their sum to ensure a sum of one.
        - We can also choose to limit the fraction of OthMin to one, as it is not physically possible to have a fraction larger than one. But the other fractions can also become larger than one (we scale them in the end), so this is not needed. We test the impact of this methodological choice.
    - If we do assume error correlations we can argue that OthMin is already perturbed, so we only divide all fractions by their sum. For comparison, we also apply the methodology suggested for the uncorrelated data.
- All fractions are multiplied with the primary PM emissions per sub-sector, summed to GNFR sector and divided by the total primary PM emissions per GNFR sector to get the fractions at the GNFR level.
- We take the median value of all countries per component and GNFR sector to illustrate the results.

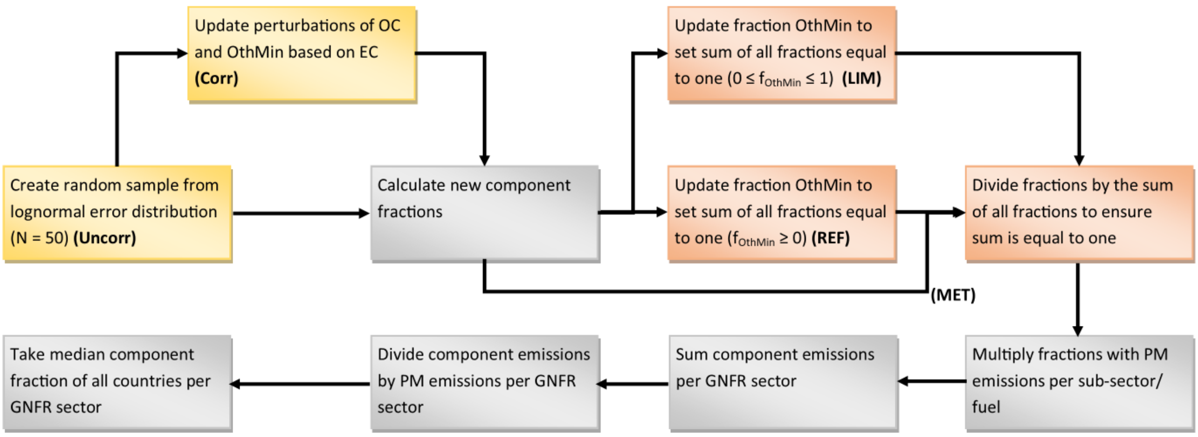

Figure 2. Flow diagram showing the steps taken for the hypothetical test on PM split uncertainties. Yellow boxes are related to the ensemble and orange boxes to the methodological choices.

4 Results

4.1 Emission uncertainties

To give an idea of the order of magnitude of the emission uncertainties we present some figures showing the total PM uncertainty at the country level for all sectors combined (Fig. 3), the total PM uncertainty per sector for all countries combined (Fig. 4) and the uncertainty in the spatial distribution (Fig. 5). Small differences in sector contributions are visible for PM2.5 and PM10, where the industry and other agricultural activities contribute relatively more to coarse particles.

For the energy sector we also see a higher uncertainty for PM10, which is related to the dominant process (sub-sector) contributing to the total emissions. The different sector contributions also affect the aggregated uncertainty in the spatial distribution, which is dominated by important sectors with a high uncertainty in the spatial distribution (e.g., agricultural activities for PM10).

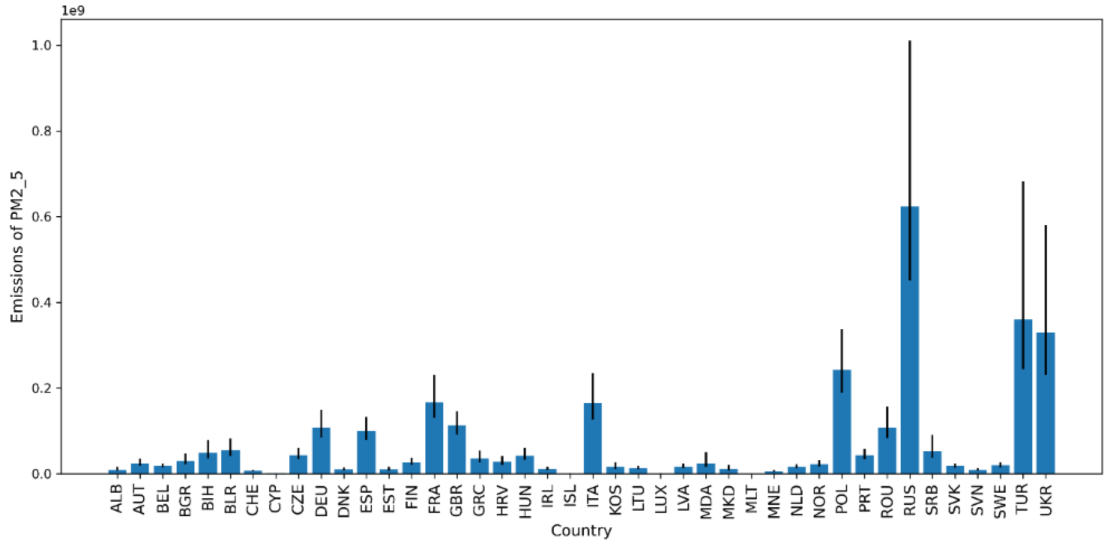

Figure 3. Emissions of PM2.5 (kg/yr) per country (for all sectors combined) and error bars representing the standard deviations.

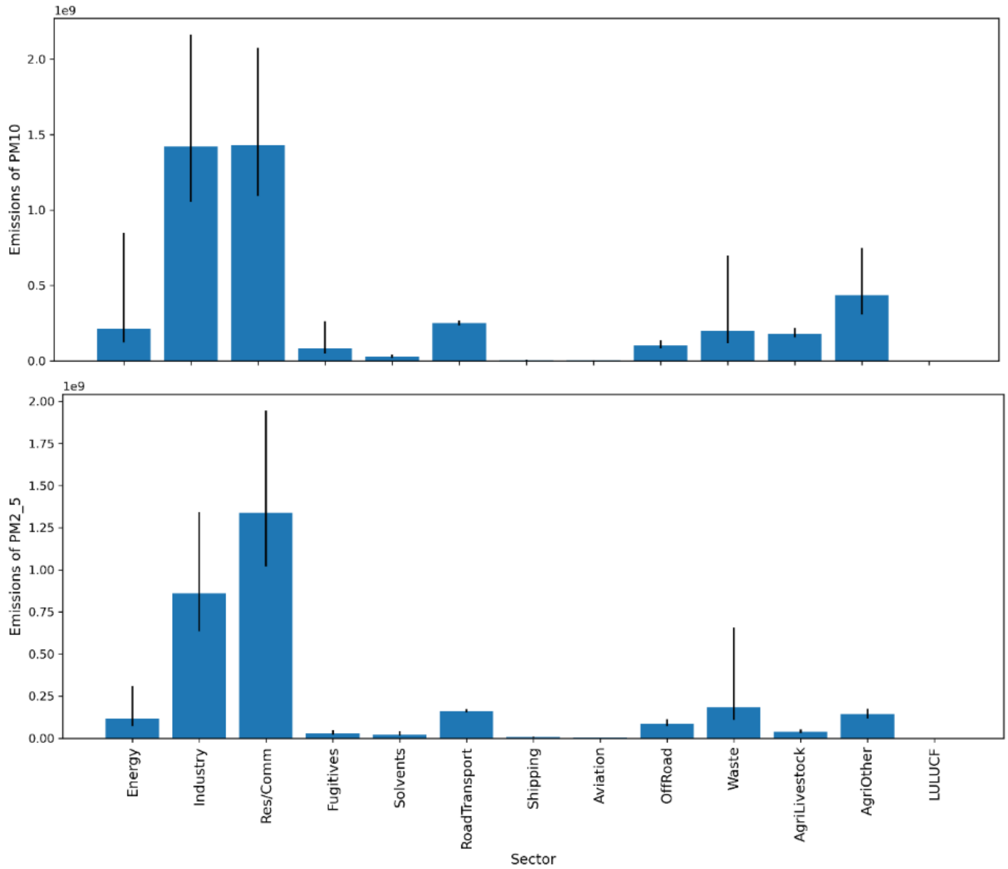

Figure 4. Emissions of PM10 and PM2.5 (kg/yr) per sector (for all countries combined) and error bars representing the standard deviations.

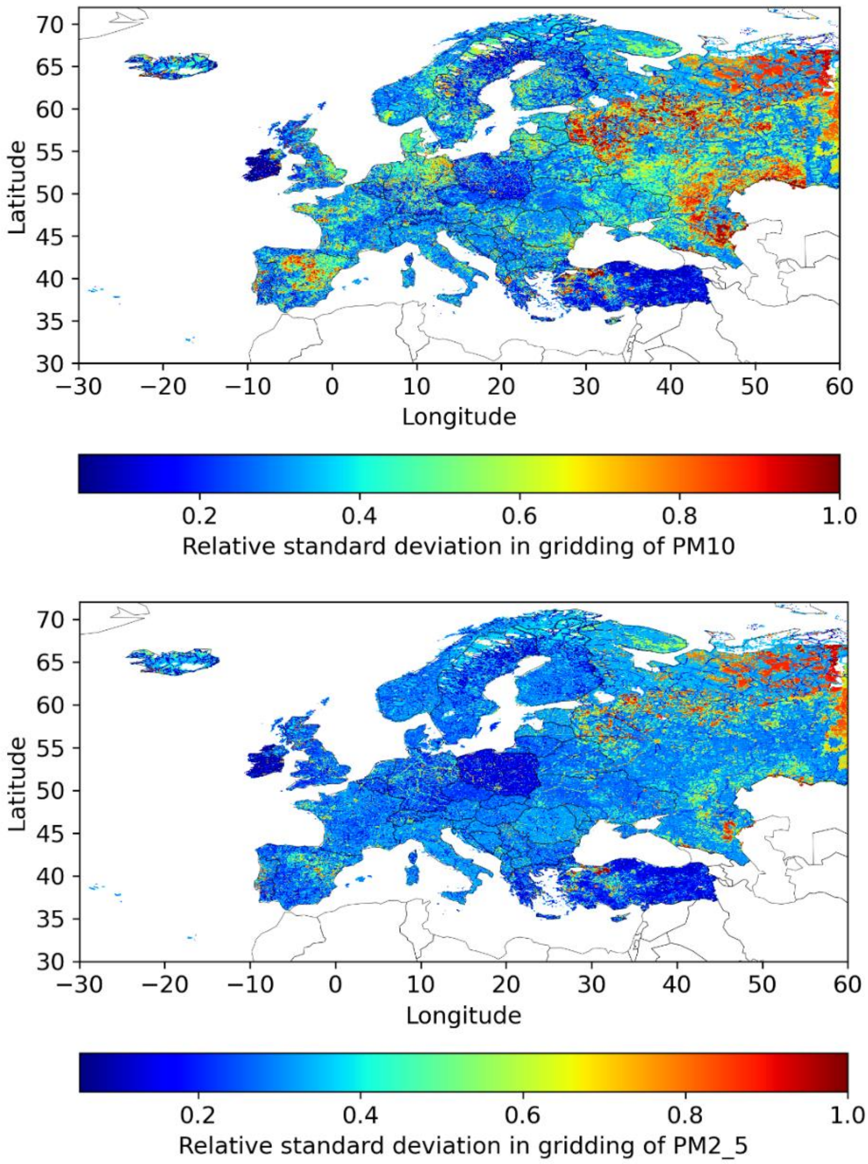

Figure 5. Relative standard deviations in gridding of PM10 and PM2.5.

4.2 PM split uncertainties

As a reference case (REF) we update the OthMin fraction (without setting a maximum value) for both the correlated and uncorrelated ensemble. The results for coarse and fine fractions of EC, OC and OthMin are shown below (Fig. 6). The boxplots show the spread in the ensembles based on the median of all countries.

The results show large differences between the correlated (Corr) and uncorrelated (Uncorr) ensemble for most sectors and components, suggesting that the inclusion of error correlations is important. The correlated ensemble generally shows a larger spread for OC and OthMin and mostly a smaller spread for EC.

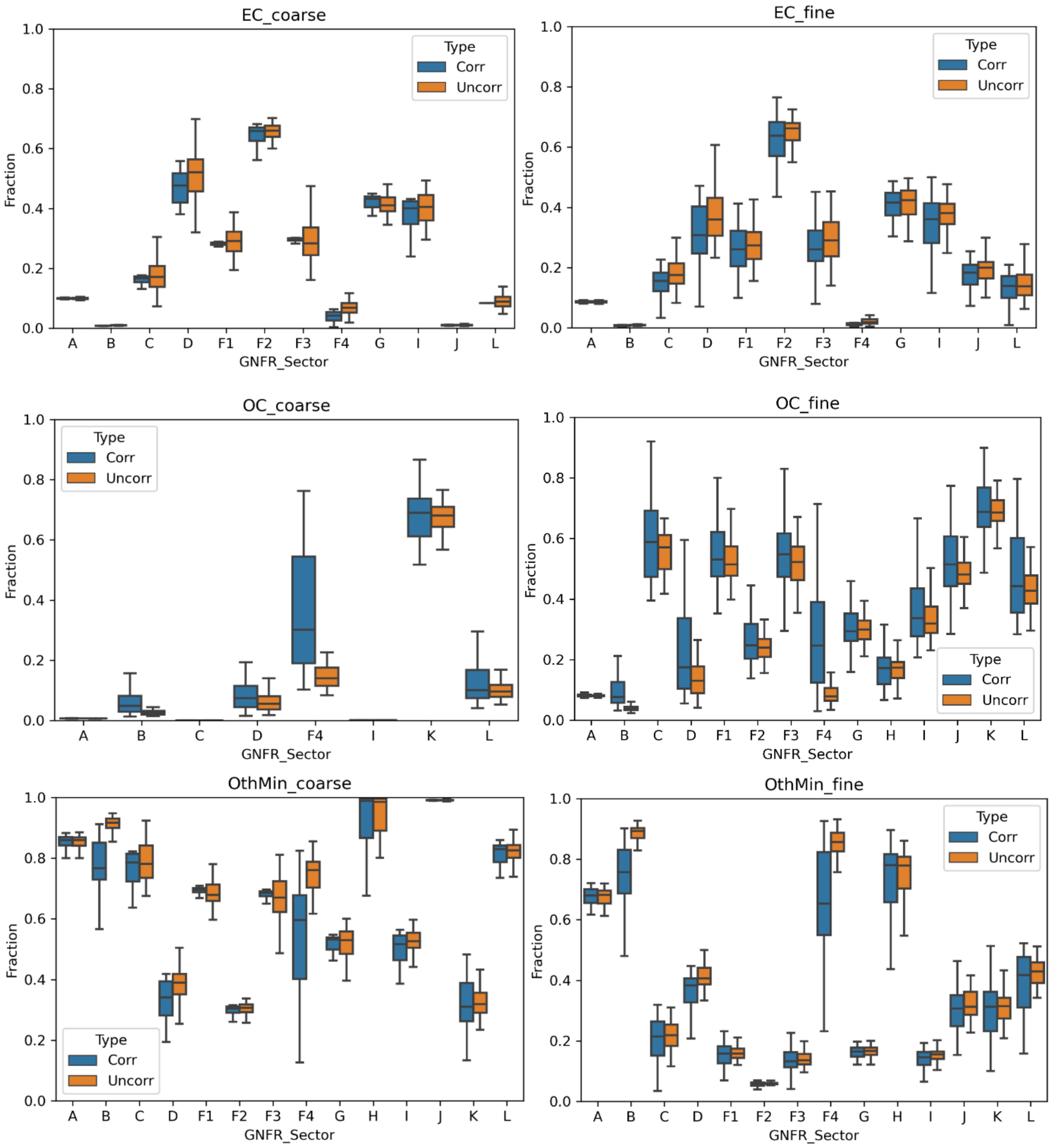

Figure 6. Boxplots (95% confidence interval) of the spread in the EC, OC and OthMin fractions in fine (PM2.5) and coarse (PM10) PM from the correlated (Corr) and uncorrelated (Uncorr) REF ensemble (N=50).

To test the impact of methodological choices we also perform an experiment in which we set the OthMin fraction to a maximum value of one before weighing the fractions (LIM) (Fig. 7). The spread in the ensembles becomes larger for both the correlated and uncorrelated ensemble, especially for the coarse EC fractions. The same pattern is visible for OC and OthMin (not shown).

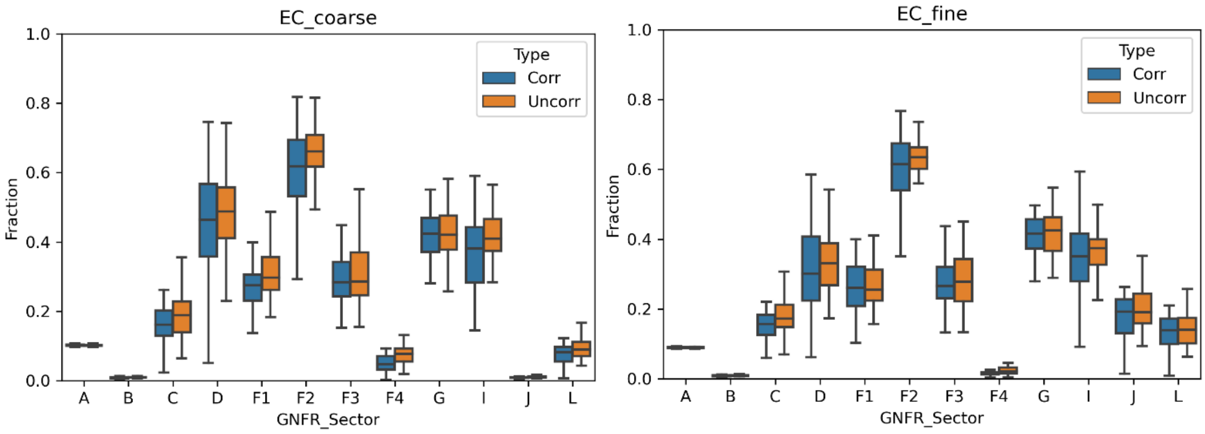

Figure 7. Boxplots (95% confidence interval) of the spread in the EC fraction in fine (PM2.5) and coarse (PM10) PM from the correlated (Corr) and uncorrelated (Uncorr) LIM ensemble (N=50).

Finally, we apply a different methodology to the correlated ensemble, as suggested above (MET) (Fig. 8). We maintain the maximum value for OthMin, so the uncorrelated ensemble is the same as in Fig. 7. The spread in the correlated ensemble becomes much smaller again and more similar to the REF ensemble. Hence, methodological choices have a significant impact on the results.

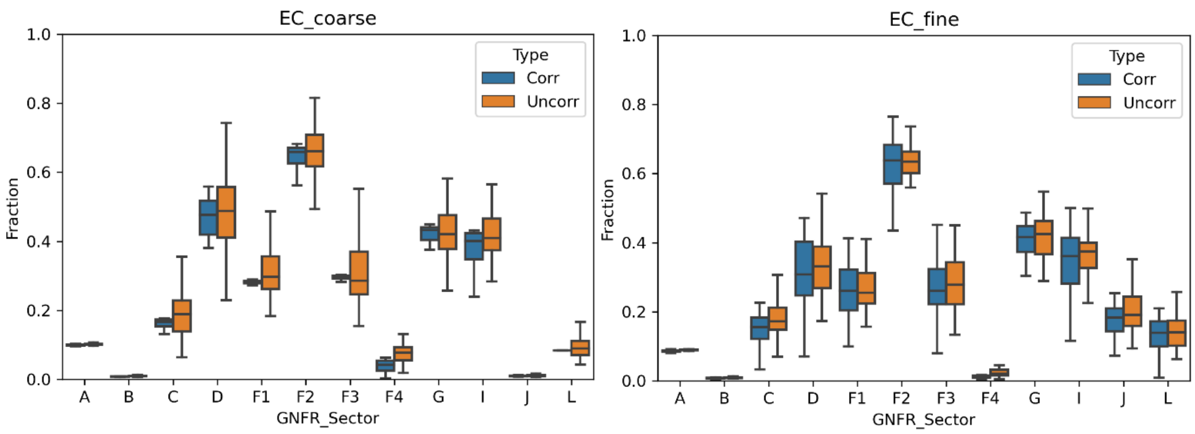

Figure 8. As Fig. 7, but for the MET ensemble.

5 Conclusion and outlook

The PM emission uncertainties presented in this report clearly show the importance of a detailed uncertainty calculation, given the large differences between sectors and countries, but also between fine and coarse particles. Uncertainties can be significant for specific sectors and regions and are therefore important to consider in decision making processes. It is important to note that the uncertainties in the coarse and fine PM fractions are likely to be correlated, but it is outside the scope of this deliverable to investigate this further.

The newly developed optimization procedure is still to be tested in practice, but should make the use of the emission uncertainties more user-friendly compared to the previous version of this dataset. Users now only need to use the gridded uncertainties and spatial correlation lengths to populate a covariance matrix, to create an ensemble or to use directly as a perturbation in source-receptor calculations. Improvements to the optimization procedure are foreseen to reduce computational resources for large countries. In the future we also want to take into account error correlations between species and more realistic boundaries for the correlation length (e.g., country-specific). Finally, we want to optimize all species simultaneously to have a fixed correlation length per country and sector.

The examination of the PM split uncertainties shows a large impact of methodological choices. The main challenge here is that we work with fractions, which form an extra constraint. In the future we want to explore the possibility to work directly with emission factors to calculate the uncertainties. Another point of concern is that we do not really know how strong the correlation is between, for example, EC and OC. In an ideal case, when we base OC directly on observations of PM and EC, this correlation is very strong and negative. However, the shares used in our database are estimated from multiple sources, are aggregated and/or averaged, so possibly this correlation is weaker. This requires more investigation and therefore we do not provide any data on this for now.

6 Data availability and usage

A total of six csv-files is provided:

1. Uncertainties_per_country_2019.csv: Contains per country and pollutant the total emissions (‘Emission_kg’), the relative standard deviation in that emission (‘Rel_stdev’), and the absolute lower and upper limit of the 95% confidence interval related to that emission uncertainty (‘Low_lim_95CI’ and ‘Upp_lim_95CI’).
2. Uncertainties_per_sector_2019.csv: Same as uncertainties per country, but aggregated per sector.
3. Uncertainties_per_country_per_sector_2019.csv: Same as uncertainties per country, but aggregated per country and sector.
4. Uncertainties_per_gridcell_2019.csv: Contains for each pollutant total emissions per grid cell and country (‘emis_kg_[pollutant]’), the relative standard deviation in that emission (‘Rel_stdev_[pollutant]’), and the absolute lower and upper limit of the 95% confidence interval related to that emission uncertainty (‘Low_lim_95CI_[pollutant]’ and ‘Upp_lim_95CI_[pollutant]’). Data are provided for point (P) and area (A) sources (‘SourceType’).
5. Uncertainties_per_gridcell_per_sector_2019.csv: Same as uncertainties per grid cell, but aggregated per country and sector.
6. Spatial_corr_lengths_2019.csv: Contains for each pollutant an optimized spatial correlation length per country and sector (‘length_opt_[pollutant]’).

The combination of the gridded uncertainties per sector with the spatial correlation lengths is consistent with the uncertainty per country and sector as provided in a separate file. The correlation length represent an e-folding distance and should be applied only to area sources. Point sources have no spatial error correlation.

Data can be downloaded from an FTP server:

Server: web-ftp81.tno.nl

Username: CAMEO@ftp0015.web-ftp81

Password: KxJusngPjYqCGKGfFVDd

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Document History

Version	Author(s)	Date	Changes
0.1	I. Super (TNO), D. Mathas (TNO), B. Jonkheid (TNO), J. Kuenen (TNO)	14-8-2024	Initial version
1.1	I. Super (TNO), D. Mathas (TNO), B. Jonkheid (TNO), J. Kuenen (TNO)	11-9-2024	Final version (added information on data availability and review comments were processed)

Internal Review History

Internal Reviewers	Date	Comments
K. Sindelarova	1-9-2024	The Deliverable clearly and thoroughly describes methodology for estimation of gridded uncertainties for PM emissions in the CAMS-REG inventory. I find the report to be very well written and have no further comments.
J. Flemming	11-9-2024	No comments.