***Interactive comment on "Uncertainty analysis of a European high-resolution emission inventory of CO2 and CO to support inverse modelling and network design" by Ingrid Super et al.***

We would like to thank the reviewers for their enthusiasm about our study and for the comments on our work. The review comments have been helpful in reflecting on our work and pointing out parts that required further improvements. Below we address specific issues mentioned by the reviewers point by point. The manuscript has been updated accordingly (changes are highlighted, line numbers refer to the final manuscript).

***Anonymous Referee #1 Received and published: 18 October 2019***

*Review of "Uncertainty analysis of a European high-resolution emission inventory of CO2 and CO to support inverse modelling and network design" by Super et al.*

*This manuscript describes an effort to construct an anthropogenic CO2 and CO inventory for a portion of Europe with carefully constructed uncertainties. The authors also show some basic analysis of their results, comparing uncertainties in different sectors and between countries, and the effect of some uncertainties on concentrations on CO2 or CO in the atmosphere. It is well-written, relevant, and extremely thorough, and should be published in ACP. The only major comment I have is about the data availability statement. The data availability requirement for publication has not been met: data is only available by request to authors, which is not acceptable to this journal, I believe. Even if it is, I think the data (i.e. the inventory and uncertainties) should be made available publicly and without restriction, especially as I think this product would be of interest to many researchers.*

We thank the reviewer for this suggestion. We agree that the data is useful for many researchers and have made the data accessible through Zenodo (see Data Availability description).

*Otherwise, my comments are fairly minor, and detailed below.*

*Introduction: Please define TNO the first time to define the acronym for international readers.*

We have replaced TNO by 'the Netherlands Organisation for Applied Scientific Research (TNO)' in lines 35-36.

*L33 - How are the national numbers determined for reporting? These are also inventories, presumably of the scaled-up variety? perhaps the authors can make this section more specific to inventories that are spatially gridded and temporally downscaled, perhaps those commonly used for atmospheric studies?*

The reported country-level emissions are available from UNFCCC (https://unfccc.int/process-and-meetings/transparency-and-reporting/reporting-and-review-under-the-convention/greenhouse-gas-inventories-annex-i-parties/national-inventory-submissions-2019) and based on energy statistics and emission factors following IPCC guidelines and are calculated as national, yearly total. This has been clarified in lines 32-34. In some cases IPCC default values are used, but countries can also decide to use country-specific values (which are generally more realistic). There is no scaling involved here, except that the overall energy consumption for one sector is divided over several sub-sectors. This introduces some uncertainty, which is taken into account in the uncertainty definition for the activity data listed in Appendix A.

*L51: I am left wondering what a Tier 3 consists of in this regard, which the US EPA follows I believe.*

It should be noted that there are two types of calculations with respect to emission reporting: calculation of the emissions and calculation of the emission uncertainties. The IPCC describes only the Tier 1 and Tier 2 approach for calculating emission uncertainties. In contrast, there is also a Tier 3 approach for calculation of emissions, which uses country-specific data and models. The Tier 3 approach used by the US EPA is therefore related to calculation of the emissions, and not of the emission uncertainties. In short, there are 3 Tiers for emission calculation and 2 Tiers for uncertainties.

*L72: What is H2020?*

H2020 is short for Horizon 2020, a European Research and Innovation programme. This is clarified in lines 77-78.

*L72: Should be made public, not on request - Journal editors can decide on this but that is my understanding of current publishing policy.*

We agree and have made the data accessible through Zenodo (see Data Availability description).

*L70-76: These sentences are not actually very clear as to what the work is and confuse the reader. Are the 10 inventories part of this work, or only the new high-resolution inventory for the zoom region? No doubt this will be made clear later in the paper but should be outlined here.*

We have clarified that the methodology used to create the family of emission inventories is also used in the work described in this manuscript (lines 78-79).

*L81: Should read: ... (12-16h LT) emissions, which could be the only emissions optimized in a study with a small domain, such as a city, using only afternoon observations? [if a study is regional or the city is large, then using mid-afternoon observations will still allow optimization of early morning emissions for example, depending on wind speed and location of emissions relative to the measurement point, for example]. But I absolutely agree that looking at the temporal variability and whether that is correct can be crucial in an urban study as well as a regional one (as illustrated by Hu et al. Science Advances 2019 for continental work). It may be an issue even if the inversion is sensitive to all hours.*

For clarity this topic has been introduced earlier in the introduction (lines 66-70), so that more explanation can be given.

*L108. Comma should be a period.*

Done.

*L108: if it's not described later, an additional sentence on the temporal disaggregation would be nice (does it account for weekday/weekend effects for example?).*

The time profiles are described in more detail in Section 2.2.4 (lines 207-213). Although a weekly cycle is included, e.g. with lower traffic emissions during the weekend, the diurnal profile is the same for weekdays and weekends.

*L109: What is GNFR vs. NFR?*

NFR sectors are very detailed and aggregated to GNFR sectors, i.e. GNFR is the aggregated version of NFR used for delivering gridded inventory data (the "G" stands for Gridding) (line 119). This is now also mentioned in the caption of Table 2.

*L138: is the point source data also temporally explicit? I am specifically thinking of energy generation (e.g. gas or coal-fired power plants, whose hour-to-hour emissions can vary drastically with no predictable cycle, at least in the U.S.).*

The point sources get the same temporal distribution as the area sources, using the fixed time profiles. This has been made more clear in lines 148-149. We have previously studied daily activity from some major power plants and found that, indeed, the temporal variations in emissions from power plants is difficult to describe with environmental variables. Unfortunately, temporally detailed activity data is not broadly available and therefore not part of the emission inventory. This is an important point for the future, which is now also mentioned in lines 594-595.

*Fig. 2: I understand from the text that correlations between sub-sectors are accounted for, but as this (and the next) figure shows aggregated sectors and no off-diagonal terms (i.e. no correlations in the uncertainties between sectors), why show these in this manner? Is the color axis in units of emissions, or do they range from 0-1 because they are covariances? (I would think the former, or they would all be 1 on the diagonal?). Or am I missing something here. Please clarify.*

Error correlations exist between several sub-sectors that are part of the same aggregated (GNFR) sector. Therefore, no off-diagonal values are visible, i.e. there are no error correlations between GNFR sectors. The reason that the uncertainties are displayed in this manner (covariance matrix) is because this is the common way to describe prior uncertainties for inversion studies. They are indeed covariances, ranging from 0 to 1.

*L218: should read "it is important to ensure"...*

Done.

*L234 define MC as Monte Carlo earlier*

The abbreviation MC has been replaced with the full name 'Monte Carlo' throughout the manuscript.

*Fig 7 & 8: Captions should indicate left and right panels, for example "Contribution of source sectors to the total uncertainty in CO2 (left) and CO (right) emissions, summing to 100 %."*

*Same for Fig 9, it is easy enough to just say (left) and (right) in the caption here.*

These indications have been added consistently to all figure captions.

*To clarify for fig 10&11, these spreads in concentration are from the experiments using different random emissions maps, i.e. the model was run 500 times, correct?*

Yes, this is correct. This is now mentioned in line 293-294 and the figure captions.

*Fig 11 - I find this to be a very interesting analysis. It points to whether we expect an inversion to identify the true location of these 20 plants among all these scenarios. I.e. can some of the maps be shown to be false by the observed CO2? The large spread indicates maybe so, but then again, once all the other sources and their uncertainty are included, it would likely be pretty hard!*

Yes, we agree that it will be very challenging. Also given the uncertainty introduced by the model transport, which would likely be similar or larger than (depending on the complexity of the area) the spread caused by the different maps. Nevertheless, we show here that it is an important source of uncertainty for local inversions.

*Section 3.3: inversion usually does not only include time from 12-16, just because those are the observation times. You may be optimizing emissions from earlier in the day, depending on the domain size and wind speeds. This should just be noted.*

We have added a sentence that explains that our reasoning only applies to local studies (lines 420-421).

*Fig 12 and text related: this standard time profile (black) seems to have a monthly mean that is then also distributed hourly through the day? Is it hourly, or 3-hourly? weekday/weekend (for businesses vs. residences, assuming those are contributing to stationary combustion for on-site heating, e.g. burning of gas)? These details could be mentioned in the caption, I realize they are not necessary for describing the uncertainty method, but they are useful to know for users of the inventory emissions.*

For each sector there are three temporal profiles: a profile describing the seasonal cycle (monthly factors), a profile describing day-to-day variations (daily factors) and a profile describing the diurnal cycle (hourly factors). This is now described in more detail in lines 207-213.

*Fig 14, the text on the map, especially "Tirol" and "Hotspot", is hard to see here perhaps larger, or placed in a different section with an arrow to the appropriate box?*

Both Fig. 14 and Fig. 15 have been improved by increasing the weight of the boxes and the font size.

*L475: Does this statement refer to their methods for calculating emissions or uncertainties?*

Actually both, but in this case we mean the uncertainty calculation. This is clarified in line 503.

*Data Availability: See note at top, this data should be publicly available on a public-facing data portal.*

Data are now publicly available. The Data Availability section has been updated accordingly.

***Anonymous Referee #2 Received and published: 21 October 2019***

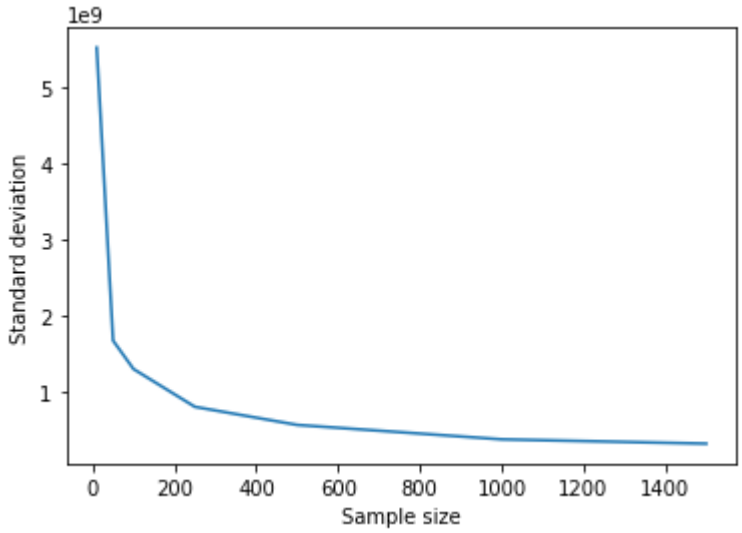
*This manuscript presents an assessment of the uncertainties in high-resolution emission inventories of CO2 and CO in 14 European countries. The uncertainties present in various underlying parameters of the inventories (e.g., absolute uncertainties in reported emissions, emission factors, spatial proxies, temporal profiles) are propagated using a well-described Monte Carlo simulation routine. The uncertainties are tracked to assess the importance of specific source sectors in introducing large uncertainties on both absolute and relative bases. Several factors are found to be playing an important role in contributing to the final emission uncertainties. For instance, spatial disaggregation of the emissions at a high spatial resolution results in large uncertainties at the local/city scale, which has important implications for inverse modeling studies operating within these smaller spatial domains. The authors find that because certain sectors with large overall contribution of CO2 and CO emissions are well-constrained (e.g., industrial sector in the Paris metro area), the relative uncertainties in these locations are far smaller than those in the immediate rural surroundings. Thus, future efforts to reduce absolute emissions of CO2 and CO may use the absolute uncertainties presented in this study to identify a network of key target areas/sectors.*

*Overall, I find this manuscript well-written. The methods are presented in sufficient detail that one could reproduce them. The methods section lacks some quality assurance, as I have described in my first major comment. The results are presented in an organized fashion, and the interpretations and conclusions are generally well-reasoned. My second major comment has to do with the framing of these interpretations with respect to observation-based literature, especially since the authors mention a motivation for this study is to facilitate inter-comparison of modeled and observed greenhouse gas concentrations. Once the authors have addressed these comments adequately, the manuscript should be ready for publication in ACP. In addition to my two major comments, I list several minor comments that are mostly typographical errors and/or suggestions to improve presentation of figures and tables.*

*Major comments:*

*1. A rationale for number of Monte Carlo (MC) simulations should be provided. For e.g., a plot with some metric of quality of results (total residual, total error, fullwidth half maximums of the distributions showed in Fig. 7, etc.) versus number of MC runs. I'd expect such a plot to have an exponential decay with respect to increasing MC runs, which would then help justify the choice of N = 500.*

Because the Monte Carlo simulations performed in this study are relatively cheap the sample size can be taken large enough to ensure a robust result. However, to support our choice for N=500 we used two methods described in literature: sampling statistics and bootstrapping. Both methods give similar curves, such as shown below. The curve indicates the spread in the standard deviations if we would repeat the Monte Carlo simulation multiple times for a specific sample size, i.e. it indicates how robust the uncertainty estimate is. From this figure we conclude that a sample size larger than 500 would not increase the robustness a lot.




We have added a statement that a sample size of N=500 is large enough to get robust results, based on the analysis shown here (lines 249-250). For completeness, we have added this figure to Appendix B.

Reference:

Janssen, H.: Monte-Carlo based uncertainty analysis: Sampling efficiency and sampling convergence. Reliability Engineering & System Safety, 109, 123-132, https://doi.org/10.1016/j.ress.2012.08.003, 2013.

*2. I think the presentation of the results could be better framed with respect to other literature. For instance, the authors show that the spread in their modeled CO2 and CO concentrations reduces over distances of 5–40 km from the source categories (Figures 10 and 11). How does this length-scale compare with other studies? Are there any monitoring studies that have shown similar fall-off length-scales? To say that road transport affects CO concentration as far as 40 km downwind seems excessive, if one were to compare it to, say, Figure 4A from Canagaratna et al.'s mobile monitoring study.*

We agree with the reviewer that we have not discussed this topic in our paper. Instead, we have focused our discussion on the estimated uncertainties in the (gridded) emissions, which is the main topic of our paper and covers most of our results. However, we have put some effort in improving the comparison of the model results with other literature, mainly to explain how these model results can be used for network design and inverse modelling (lines 499-501 and lines 570-585).

*Minor comments:*

*1. L24: I suggest using "abundant", instead of "important". In terms of warming potential, there are other gases more important than CO2 (e.g., CH4).*

We have replaced this word.

*2. L36: "report", not "reported".*

The sentence has been rephrased for clarification (line 37).

*3. L69: "atmospheric", not "atmospherics".*

Done.

*4. L64-65: "in contrast, if … are needed." This sentence is unclear. What is "prior" referring to? Please reword.*

In inverse modelling the word 'prior' refers to the initial emission inventory, containing information on the order of magnitude and location of emissions which is then updated using atmospheric observations. This has been clarified in line 65.

*5. L76-77: not sure what "European zoom region" means. Please clarify.*

We have clarified which region is meant in line 84.

*6. L81: suggest replacing "time profiles" with "temporal profiles", or "diurnal profiles".*

We have replaced 'time profiles' with 'temporal profiles' throughout the manuscript.

*7. L81-83: question 3 is somewhat unclear. It could be reworded for clarity, but also the motivation for this question was not set up in the introduction. This makes this question feel abruptly added.*

This topic has now been introduced in lines 66-70 for clarification.

*8. L86: is "partitioning" the right word? I suggest using "apportionment", instead.*

This word has been replaced in line 94.

*9. Table 1: for consistency with first usage in L26, please continue with the "FFCO", "FFCO2" naming conventions. The acronyms FF and BF should be declared in the Table caption. Also, is there a specific reason to use three-letter country codes, instead of simply country names?*

The names of the species have been updated and FF and BF are explained in the caption of Table 1. Also, the names of the countries have been given.

*10. L103: "gap-filled" (should be hyphenated).*

Done.

*11. L103: suggest replacing "data was gap filled" with something more informative of which attributes of the dataset were missing, and how they were filled (i.e., with NaNs, or geospatial interpolated, etc.).*

The gap-filling refers to the reported emissions, which are sometimes missing for specific sectors. Also, in some cases the reported data is considered to be unreliable. In those cases, other sources of information are used to update and complete the emission inventory. This is now explained in more detail in lines 112-113.

*12. L105: acronym "AIS" not defined.*

The acronym has been defined in line 115.

*13. L108: comma is used instead of period.*

The comma has been replaced.

*14. L109: acronym "GNFR" not defined, and is also used in Table 2 without definition.*

GNFR is the aggregated version of NFR. This is now mentioned in the caption of Table 2.

*15. Figure 1: could the authors add a few landmarks or identify a few of the visible hotspots in these maps e.g., Paris? It'd be helpful for a reader not familiar with the placement of major urban areas of Europe.*

Figure 1 has been updated and indicates the location of some major urban areas (Paris, Ruhr area and Rotterdam).

*16. L148: which "differences" are being referred to in this sentence? Differences in uncertainties, I assume? Should be clarified.*

Indeed, this has been made explicit in line 159-160.

*17. Figures 2 and 3: the gridlines need to match the category labels. The current version of this figure is difficult to read easily.*

We choose to match the gridlines with the category labels, such that the intersection of two lines clearly indicates the value belonging to those categories. But if we understand correctly the reviewer would like to see the gridlines surrounding each entry in the matrix, instead of being in the middle of each row/column. This has been updated.

*18. L294-296: This sentence is confusing. Here is how I would calculate the "uncertainty in the total emissions": a) take the standard deviations of emissions from each sector (i.e., standard deviation of each box in Figure 7-left), b) calculate the average of the standard deviations from (a), and c) report this average from (b) as "uncertainty in total emissions". However, it seems the authors have used a different approach: a) take the standard deviations of emissions from each sector (i.e., standard deviation of each box in Figure 7-left), b) calculate the STANDARD DEVIATION of the standard deviations from (a), and c) report this STANDARD DEVIATION from (b) as "uncertainty in total emissions". Is this correct? If yes, this would not be the "uncertainty in total emissions", but rather would be an "uncertainty of the uncertainty in total emissions". Please justify/clarify/correct in the manuscript accordingly.*

The uncertainty in the total emissions is calculated from the Monte Carlo, similarly as the uncertainty for each sector. The Monte Carlo provides 500 solutions for each of the defined sub-sectors (NFR – fuel combination). These are summed to get 500 solutions for GNFR sector emissions. If we then sum the GNFR sector emissions we get 500 solutions for the total emission. What is reported here is the standard deviation of these 500 solutions. We have clarified this in lines 309-311.

*19. Figures 8, 9, and 16: As I indicated in my initial review prior to posting on ACPD, the legend needs to be reversed to be consistent with the order of stacking.*

The order of the legends has been reversed.

*20. L322-323: "Overall, the differences between countries are relatively small(Figure 9, right panel)." Instead of using a qualitative term like "relatively small", why not report the total uncertainty as done for Figure 7?*

We have presented the range of standard deviations, which is between 1.2 and 2.3% (lines 339-341).

*21. L339: "For CO2 (left panel) we see a concentration of about …" should be changed to "For CO2 (left panel), we see a spread in concentration of about …" There is no information in Figure 10 about the absolute CO2 concentrations, so seeing a concentration of 3 ppm anywhere in ambient air would be impossible. A related suggestion is to plot the absolute numbers on the right axis, to get a better sense of the absolute concentrations (especially for CO).*

We agree that these concentrations are not what you would measure in the atmosphere. Rather, they are the result only of the emissions of stationary combustion (CO2) and road transport (CO). This has been clarified in the text (lines 357-358).

*22. L343: what does "atmospheric signal" mean? Why not just say "modeled spread in concentration"?*

What we refer to is the concentration enhancement caused by the specified sector. We have replaced 'signal' by 'concentration enhancement' throughout the text.

*23. Figures 10 and 11: the x-axis labels are unevenly spaced. It is definitely not linear, but it doesn't seem log-spaced either. Please correct/clarify in Figure caption.*

The labels are unevenly spaced, because we only added labels for the distances that we have included in the model simulations. Nevertheless, the axis is linear.

*24. Figure 12: Please describe what the grey lines represent in the caption.*

The grey lines represent the spread in the temporal profile, resulting from the Monte Carlo simulation. This is clarified in the caption.

*25. L397: "in inverse modeling, often … transport". It'd be good to include a couple of references to support this statement.*

See lines 419-420 for the references.

*26. Figure 13: I know that time profiles used for modeling CO2 and CO emissions are the same, but it'd be good to rename the y-axis label to "normalized spread in CO2 and CO emissions", and remind the reader of this very briefly in the caption.*

We have added this comment to the figure caption.

*27. L447: "big cities like Paris, Berlin, and Brussels". I assume this is about the CO, and not the CO2 map? It'd help to point to these cities in Figure 15.*

Indeed, this part describes relative uncertainties in CO (as is also mentioned in the caption of Figure 17). We have added this to line 472. For clarity, we have replaced the current list with those urbanized areas now shown in Fig. 1, including a reference to this figure (line 471-472).

*28. L528: no need to define the acronym LTO, if it is only used once.*

The term LTO has been removed.

*Reference(s): Canagaratna, M. R.; Onasch, T. B.; Wood, E. C.; Herndon, S. C.; Jayne, J. T.; Cross, E. S.; Miake-Lye, R. C.; Kolb, C. E.; Worsnop, D. R. Evolution of vehicle exhaust particles in the atmosphere. J. Air Waste Manag. Assoc. 2010, 60 (10), 1192–1203.*

# Uncertainty analysis of a European high-resolution emission inventory of $CO_2$ and CO to support inverse modelling and network design

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**Abstract.** Quantification of greenhouse gas emissions is receiving a lot of attention, because of its relevance for climate mitigation. Complementary to official reported bottom-up emission inventories, ~~Q~~quantification ~~is often~~can be done with an inverse modelling framework, combining atmospheric transport models, prior gridded emission inventories and a network of atmospheric observations to optimize the emission inventories. An important aspect of such method is a correct quantification of the uncertainties in all aspects of the modelling framework. The uncertainties in gridded emission inventories are, however, not systematically analysed. In this work, a statistically coherent method is used to quantify the uncertainties in a high-resolution gridded emission inventory of $CO_2$ and CO for Europe. We perform a range of Monte Carlo simulations to determine the effect of uncertainties in different inventory components, including the spatial and temporal distribution, on the uncertainty in total emissions and the resulting atmospheric mixing ratios. We find that the uncertainty in the total emissions for the selected domain are 1 % for $CO_2$ and 6 % for CO. Introducing spatial disaggregation causes a significant increase in the uncertainty of up to 40 % for $CO_2$ and 70 % for CO for specific grid cells. Using gridded uncertainties specific regions can be defined that have the largest uncertainty in emissions and are thus an interesting target for inverse modelers. However, the largest sectors are usually the best-constrained ones (low relative uncertainty), so the absolute uncertainty is the best indicator for this. With this knowledge areas can be identified that are most sensitive to the largest emission uncertainties, which supports network design.

## 1 Introduction

Carbon dioxide ($CO_2$) is the most ~~important~~ abundant greenhouse gas and is emitted in large quantities from human activities, especially from the burning of fossil fuels (Berner, 2003). A reliable inventory of fossil fuel $CO_2$ ($FFCO_2$) emissions is important to increase our understanding of the carbon cycle and how the global climate will develop in the future. The impact of $CO_2$ emissions is visible on a global scale and international efforts are required to mitigate climate change, but cities are the largest contributors to $FFCO_2$ emissions (about 70% (IEA, 2008)). Therefore, emissions should be studied at different spatial and temporal scales to get a full understanding of their variability and mitigation potential.

One way of describing emissions is an emission inventory, which is a structured set of emission data, distinguishing different pollutants and source categories. Often, emission inventories are based on reported ~~country level~~ emission data (for example from the National Inventory Reports (NIR's) (UNFCCC, 2019)), which are national, yearly emissions based on energy statistics. ~~which~~ These country-level emissions ~~are~~ can be spatially and temporally disaggregated (scaled-down) to a certain level using proxies (e.g. the ~~TNO~~ inventories of the Netherlands Organisation for Applied Scientific Research (TNO) (Denier van der Gon et al., 2017; Kuenen et al., 2014)). Other emission inventories are based on local energy consumption data and reported emissions, which are (dis)aggregated to the required spatial scale ~~(scaled up)~~ (e.g. Hestia (Gurney et al., 2011, 2019)) or rely on (global) statistical data and a consistent set of (non-country specific) emission factors representing different technology levels (e.g. EDGAR (http://edgar.jrc.ec.europa.eu)). Most inventories, including the one used in this study, rely on a combination of methods, using large-scale data supplemented with local data. Gridded emission inventories are essential as input for atmospheric transport models to facilitate comparison with observations of $CO_2$ concentrations, as well as in inverse modelling as a prior estimate of the emission locations and magnitude.

During the compilation of an emission inventory uncertainties are introduced at different levels (e.g. magnitude, timing or locations) and increasingly more attention is given to this topic. Parties to the United Nations Framework Convention on Climate Change (UNFCCC) report their annual emissions (disaggregated over source sectors and fuel types) in a NIR (UNFCCC, 2019), which includes an assessment of the uncertainties in the underlying data and an analysis of the uncertainties in the total emissions following IPCC (Intergovernmental Panel on Climate Change) guidelines. The simplest uncertainty analysis is based on simple equations for combining uncertainties from different sources (Tier 1 approach). A more advanced approach is a Monte Carlo simulation, which allows for non-normal uncertainty distributions (Tier 2 approach). The Tier 2 approach has been used by several countries, for example Finland (Monni et al., 2004) and Denmark (Fauser et al., 2011).

These reports provide a good first step in quantifying emission uncertainties, but the uncertainty introduced by using proxies for spatial and temporal disaggregation are not considered. These are, however, an important source of uncertainty in the gridded emission inventories (Andres et al., 2016). Inverse modelling studies are increasingly focusing on urban areas, the main source areas of $FFCO_2$ emissions, for which emission inventories with a high spatiotemporal resolution are used to better represent the variability in local emissions affecting local concentration measurements. Understanding the uncertainty at higher resolution than the country-level is thus necessary, which means that the uncertainty caused by spatiotemporal disaggregation becomes important as well.

The uncertainties in emission inventories are important to understand for several reasons. First, knowledge of uncertainties helps pinpointing emission sources or areas that require more scrutiny (Monni et al., 2004; Palmer et al., 2018). Second, knowledge of uncertainties in prior emission estimates is an important part of inverse

modelling frameworks, which can be used for emission verification and in support of decision-making (Andres et al., 2014). For example, if uncertainties are not properly considered, there is a risk that the uncertainty range does not contain the actual emission value. In contrast, if uncertainties are ~~large~~ overestimated the ~~prior~~ initial emission inventory gives little information about the actual emissions and more independent observations are needed. Third, local inverse modelling studies often rely on daytime (12-16h LT) observations, which are easier to simulate. Given the small size of the urban domain these observations only contain information on recent emissions, which have to be extrapolated using temporal profiles to calculate annual emissions. Therefore, knowledge of uncertainties in temporal profiles helps to better quantify the uncertainty in these annual emissions. Finally, emission uncertainties can support atmospheric observation system design, for example for inverse modelling studies. An ensemble of model runs can represent the spread in atmospheric concentration fields due to the uncertainty in emissions. Locations with a large spread in atmospheric concentrations are most sensitive to uncertainties in the emission inventory and are preferential locations for additional atmospheric~~s~~ measurements. To conclude, emission uncertainties are a critical part of emission verification systems and require more attention.

To better understand how uncertainties in underlying data affect the overall uncertainty in gridded emissions, a family of ten emission inventories is compiled within the ~~H2020 project~~ $CO_2$ Human Emissions (CHE) project, which is funded by the Horizon 2020 EU Research and Innovation programme (see Data Availability). ~~These can be made available upon request.~~The methodology used to create this family of emission inventories also forms the basis for the work described here.

In this paper we illustrate a statistically coherent method to assess the uncertainties in a high-resolution emission inventory, including uncertainties resulting from spatiotemporal disaggregation. For this purpose, we use a Monte Carlo simulation to propagate uncertainties in underlying parameters into the total uncertainty in emissions (like the Tier 2 approach). We illustrate our methodology using a new high-resolution emission inventory for a European ~~zoom~~ region centred over the Netherlands and Germany (Table 1). We illustrate the magnitude of the uncertainties in emissions and how this affects simulated concentrations. The research questions are:

1) How large are uncertainties in total inventory emissions and how does this differ per sector and country?
2) How do uncertainties in spatial proxy maps affect local measurements?
3) How important is the uncertainty in ~~time~~ temporal profiles for the calculation of annual emissions from daytime (12-16h LT) emissions, which result from urban inverse modelling studies using only daytime observations?
4) What information can we gain from high-resolution gridded uncertainty maps by comparing different regions?

Inverse modelling studies often focus on a single species like $CO_2$, but co-emitted species are increasingly included to allow source ~~partitioning~~ apportionment (Boschetti et al., 2018; Zheng et al., 2019). In this study, we look into $CO_2$ and CO to illustrate our methodology, but the methodology can be applied to other (co-emitted) species.

## 2 Methodology

### 2.1 The high-resolution emission inventory

**Table 1: Characteristics of the high-resolution emission inventory TNO GHGco v1.~~0~~1 containing fossil fuel (FF) and biofuel (BF) emissions.**

| | |
|---|---|
| **Air pollutants** | FFCO~~_ff~~, BFCO~~_bf~~, NOx |
| **Greenhouse gases** | FF$CO_2$~~_ff~~, BF$CO_2$~~_bf~~, $CH_4$ |
| **Resolution** | 1/60° longitude x 1/120° latitude (~ 1x1 km over central Europe) |
| **Period covered** | 2015 (annual emissions) |
| **Domain** | -2° W–19° E, 47° N–56°N |
| **Sector aggregation** | GNFR (A to L), with GNFR F (Road Transport) split in F1 to F4 (total 16 sectors) |
| **Countries** | Complete: Germany, Netherlands, Belgium, Luxembourg, Czech Republic~~DEU, NLD, BEL, LUX, CZE~~<br>Partially: United Kingdom, France, Denmark, Austria, Poland, Switzerland, Italy, Slovakia and Hungary~~GBR, FRA, DNK, AUT, POL, CHE, ITA, SVK and HUN~~ |

The basis of this study is a high-resolution emission inventory for the greenhouse gases $CO_2$ and $CH_4$ and the co-emitted tracers CO and $NO_x$ for the year 2015 (TNO GHGco v1.0, see details in Table 1). In this paper we only use $CO_2$ and CO, which are divided over fossil fuel (FF~~ff~~) and biofuel (BF~~bf~~) emissions (no land use and land use change emissions are included). The emission inventory covers a domain over Europe, including Germany, Netherlands, Belgium, Luxembourg and the Czech Republic, and parts of Great Britain, France, Denmark, Austria and Poland (see also Figure 1).

The emission inventory is based on the reported emissions by European countries to the UNFCCC (only greenhouse gases) and to EMEP/CEIP (European Monitoring and Evaluation Programme/Centre on Emission Inventories and Projections, only air pollutants). UNFCCC $CO_2$ emissions have been aggregated to ~250 different combinations of NFR sectors (Nomenclature For Reporting) and fuel types. EMEP/CEIP CO emissions have been split over the same NFR sector-fuel type combinations by TNO using the GAINS model (Amann et al., 2011) and/or TNO data. In some cases, the reported data was gap~~-~~-filled~~,~~ or replaced ~~or (dis)aggregated~~with emissions from the GAINS model, EDGAR inventory or internal TNO estimates to obtain a consistent dataset. Next, each NFR sector is linked to a high-resolution proxy map (e.g. population density for residential combustion of fossil fuels or AIS (Automatic Identification System) data for shipping re-gridded to 1/60° x 1/120°), which is used to spatially disaggregate the reported country-level emissions. Where possible, the exact location and reported emission of large point sources is used (e.g. from the E-PRTR (European Pollutant Release and Transfer Register)). The third step is temporal disaggregation, for which standard ~~time~~ temporal profiles are used (Denier van der Gon et al., 2011)~~.~~, Finally, the ~~gridded~~ emissions are aggregated ~~per~~ to GNFR (gridded NFR) sectors (see Table 2) for the emission inventory. The final emission maps of $CO_2$ and CO are shown in Figure 1, together with two examples of a source sector map. Note that these maps do not clearly show the large point source emissions, while these make up almost 45 % of all $CO_2$ emissions and 26 % of all CO emissions.

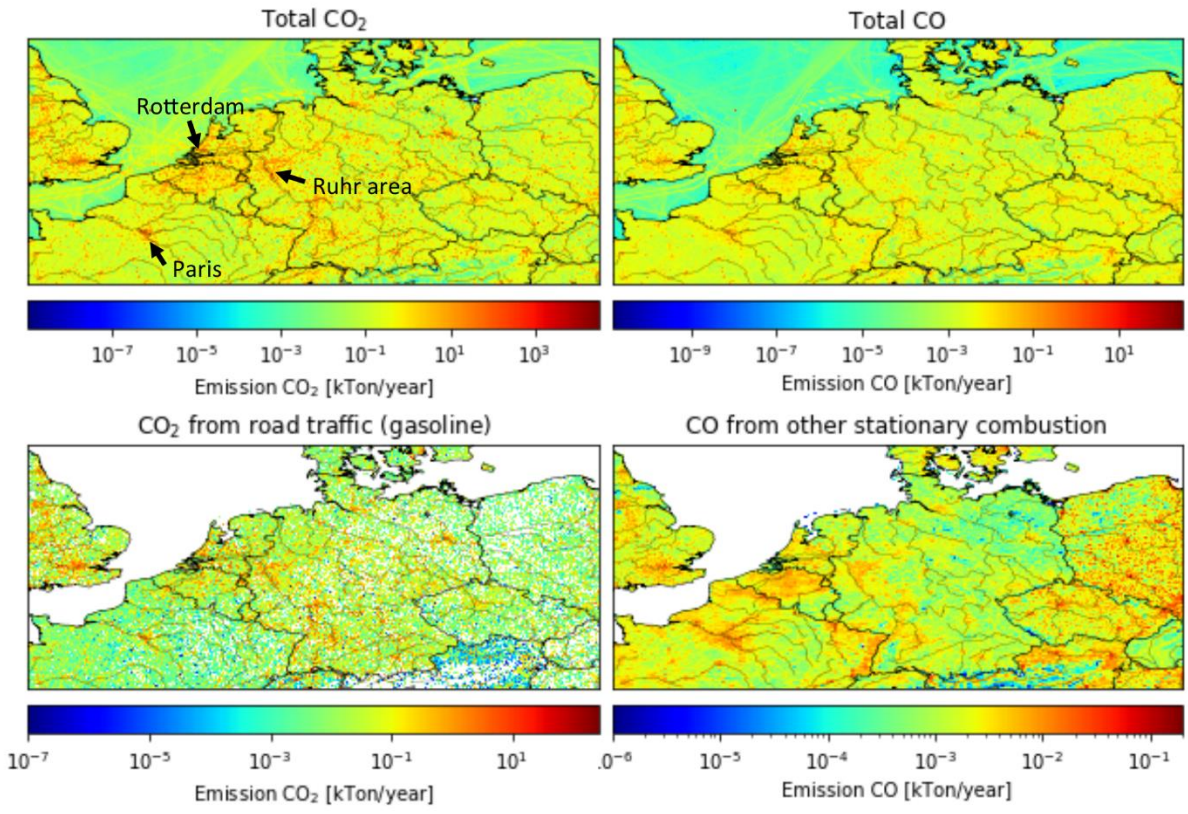


**Figure 1: Total emissions of $CO_2$ and CO, road traffic (gasoline) emissions of $CO_2$, and other stationary combustion emissions of CO for 2015 in kt $yr^{-1}$ (defined per grid cell).**

## 2.2 Uncertainties in parameters

The emission inventory is used as basis for an uncertainty analysis by assigning an uncertainty to each parameter underlying the UNFCCC-EMEP/CEIP emission inventories and further disaggregation thereof. Although the aggregation to GNFR sectors makes the emission inventory more comprehensible, we use the more detailed underlying data for the uncertainty analysis. The reason is that the uncertainties can vary enormously between sub-sectors and fuel types. Generally, the emission at a certain time and place is determined by four types of parameters: activity data, emission factor, spatial distribution and ~~time~~ temporal profile. The activity data and emission factors are used by countries to calculate their emissions. The spatial proxy maps and ~~time~~ temporal profiles are used for spatiotemporal disaggregation. All uncertainties need to be specified per NFR sector-fuel type combination that is part of the Monte Carlo simulation. In the following sections the steps taken to arrive at a covariance matrix for the Monte Carlo simulation are described. Tables with uncertainty data can be found in ~~the~~ Appendix A.

**Table 2: Overview of aggregated NFR (GNFR) sectors distinguished in the emission inventory**

| GNFR category | GNFR category name |
|---|---|
| **A** | A_PublicPower |
| **B** | B_Industry |
| **C** | C_OtherStationaryComb |
| **D** | D_Fugitives |
| **E** | E_Solvents |

| | |
|---|---|
| **F** | F_RoadTransport |
| **F1** | F_RoadTransport_exhaust_gasoline |
| **F2** | F_RoadTransport_exhaust_diesel |
| **F3** | F_RoadTransport_exhaust_LPG_gas |
| **F4** | F_RoadTransport_non-exhaust |
| **G** | G_Shipping |
| **H** | H_Aviation |
| **I** | I_OffRoad |
| **J** | J_Waste |
| **K** | K_AgriLivestock |
| **L** | L_AgriOther |

### 2.2.1 Parameter selection

The first step is to identify which parameters should be included in the Monte Carlo simulation. As mentioned before there are about 250 different combinations of NFR sectors and fuel types and including all of them would be a huge computational challenge. However, a selection of 112 combinations makes up most of the fossil fuel emissions (96 % for $CO_2$ and 92 % for CO) and therefore a pre-selection was made. This results in a covariance matrix of 224x224 parameters (112 sector-fuel combinations for two species). To further reduce the size of the problem, the emissions are partly aggregated before starting the Monte Carlo for the spatial proxies (mostly fuels are combined per sector, because they have the same spatial distribution). This results in a total of 59 NFR sector-spatial proxy combinations, which are put in a separate covariance matrix. The ~~time~~ temporal profiles are applied to the aggregated GNFR sectors, which make up the last covariance matrix. Note that the spatial proxies and ~~time~~ temporal profiles are the same for $CO_2$ and CO.~~,~~ Only ~~except for~~ the spatially explicit E-PRTR point source data can have a different spatial distribution for $CO_2$ and CO, but they also use the same temporal profiles.

### 2.2.2 Uncertainties in reported emissions

Country-level emissions are estimated from the multiplication of activity data and emission factors. Activity data consist for the most part of fossil fuel consumption data available from national energy balances. Some fuel consumptions are better known than others and uncertainties vary across sectors. An emission factor is the amount of emission that is produced per unit of activity (e.g. amount of fuel consumed). For $CO_2$ this depends mainly on the carbon content of the fuel. In contrast, CO emissions are extremely dependent on combustion conditions, ~~certain~~ choice of industrial processes and in-place technologies.

The NIR's for greenhouse gases (GHGs) provide a table with uncertainties in activity data and $CO_2$ emission factors on the level of NFR sector - fuel combinations. The uncertainties reported by each country are averaged to get one uncertainty per N~~R~~FR sector-fuel combination for the entire domain. Overall, the differences in reported uncertainties between countries are small. The uncertainties in activity data and $CO_2$ emission factors are relatively low and normally distributed.

The CO emission factors are mostly based on ~~basic~~ uncertainty ranges provided in the EMEP/EEA Guidebook (European Environment Agency, 2016) and supplemented by BAT reference documents from which reported emission factor ranges are taken as uncertainty range (http://eippcb.jrc.ec.europa.eu/reference/). The CO emission

factor uncertainties are generally expressed by a factor, which means that the highest and lowest limit values are either the specified factor above or below the most common value. Therefore, these uncertainties have a lognormal distribution and are relatively large.

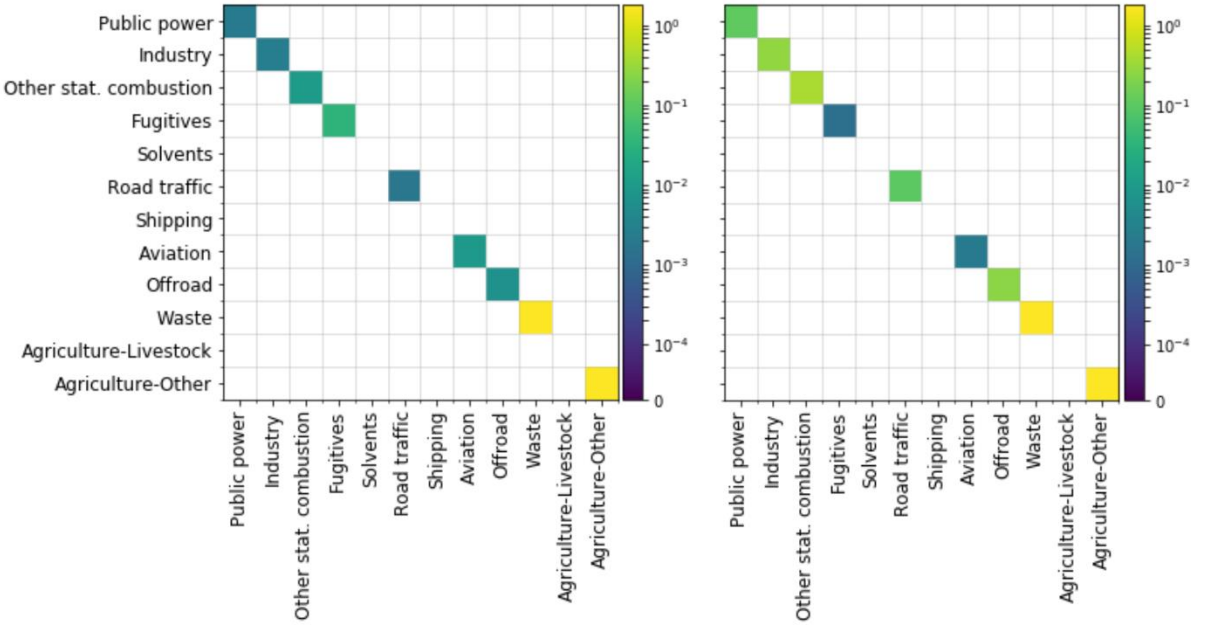


**Figure 2: Covariance matrices for total emissions of $CO_2$ (left) and CO (right) per aggregated source sector. A white space on the diagonal indicates this sector is not included in the Monte Carlo simulation.**

To estimate the overall uncertainty in the emissions per NFR sector-fuel combination, the uncertainties in the activity data and emission factors need to be combined (shown in Figure 2 for the aggregated GNFR sectors). When both uncertainties are of the same order and relatively small, as well as both having a normal distribution, the overall emission uncertainty is calculated with the standard formula for error propagation for non-correlated normally distributed variables (see Sect. 2.4). For most CO emission factors, uncertainties are much higher and have a lognormal distribution instead of normal. In that case the uncertainty of the variable with the highest uncertainty is assumed to be indicative for the overall uncertainty of the emission, which in general means the uncertainty of the CO emission factor determines the overall uncertainty of the CO emission, with the distribution remaining lognormal. The error introduced by fuel type disaggregation for CO is not considered.

Finally, for power plants and road traffic we assumed error correlations to exist between different sub-sectors per fuel type, and between different fuel types per sub-sector for other NFR sectors. In some cases, correlations also exist between different NFR sectors belonging to the same GNFR sector. The definition of correlations is important, because they affect the total uncertainties. For example, if emission factors of sub-sectors are correlated, deviations can amplify each other, leading to higher overall uncertainties. In contrast, the division of the well-known total fuel consumption of a sector over its sub-sectors includes an uncertainty which is anti-correlated (i.e. if too much fuel consumption is assigned to one sub-sector, too little is assigned to another). This has little impact on the total emissions, because uncertainties only exist at lower levels.

### 2.2.3 Uncertainties in spatial proxies

The proxy maps used for spatial disaggregation can introduce a large uncertainty coming from the following sources:

1) The proxy is not correctly representing real-world locations of what it is supposed to represent, either because there are cells included in which none of the intended activity takes place or cells are missing in which the intended activity does take place (proxy quality).
2) The proxy is not fully representative for the activity it is assumed to represent, for example if there is a non-linear relationship between the proxy value and the emission (proxy representativeness): a grid cell with twice the population density does not necessarily have double the amount of residential heating emissions, because heating can be more efficient in densely populated areas and/or apartment blocks.
3) The cell values themselves are uncertain, e.g. the population density or traffic intensity (proxy value).

We attempt to capture the second and third source of uncertainty in a single numerical indicator representing the uncertainty at cell level (see Figure 3 for the uncertainty per aggregated GNFR sector). The overall uncertainties are based on expert judgement and inevitably include a considerable amount of subjectivity. This type of uncertainty is often large and has a lognormal distribution, except for proxies related to road traffic and some proxies related to commercial/residential emissions sources. We assume no error correlations exist. The first source of uncertainty is also considered in one of the experiments (see Sect. 2.4 for a description of this experiment).

### 2.2.4 Uncertainties in ~~time~~ temporal profiles

~~The~~ For each GNFR sector the emission timing is described using three ~~time~~ temporal profiles: one profile that describes the seasonal cycle (monthly fractions), one profile that describes the day-to-day variations within a week (daily fractions), and one profile that describes the diurnal cycle (hourly fractions). ~~currently consist of fixed monthly, daily and hourly fractions~~These profiles ~~that~~ are based on long-term average activity data and/or socio-economic characteristics and~~. These profiles~~ are applied for each year and for the entire domain, considering only time zone differences. In reality, the ~~time~~ temporal profiles can differ between countries, ~~and~~ from year to year and the diurnal cycle can vary between weekdays and weekends. For example, residential emissions are strongly correlated with the outside temperature, which follows a different pattern each year. ~~and therefore show a strong seasonal cycle. However, one winter can be very cold, whereas the next can have above-average temperatures.~~

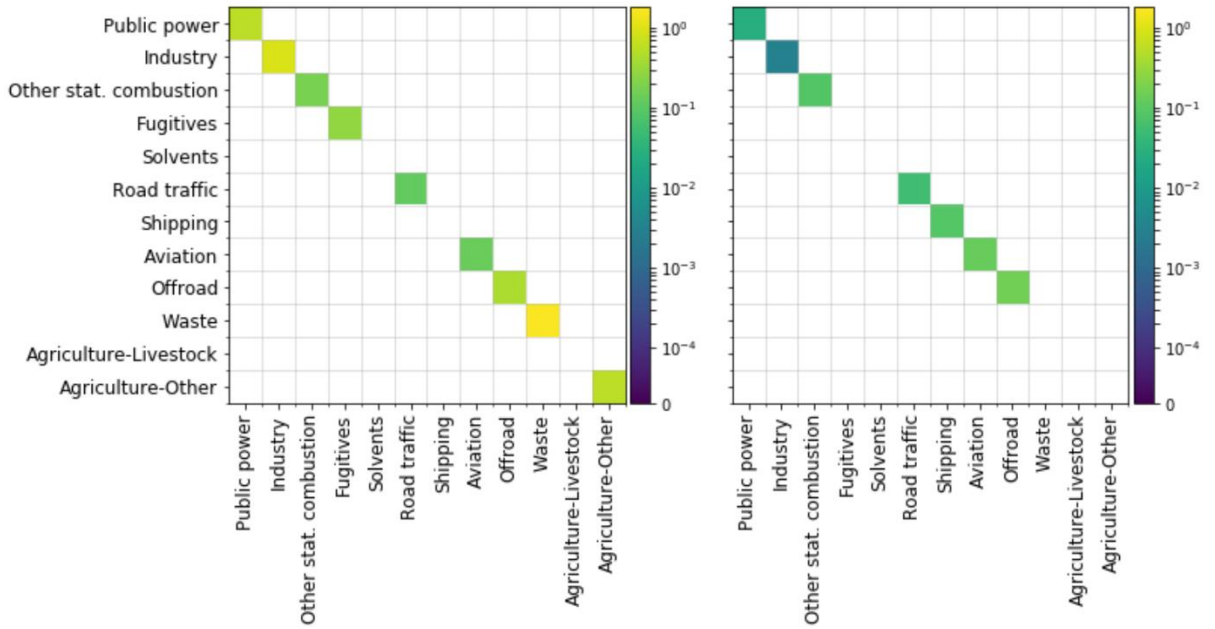

**Figure 3: Covariance matrices for spatial proxies (left) and time profiles (right) per aggregated source sector. These are the same for $CO_2$ and CO. A white space on the diagonal indicates this sector is not included in the Monte Carlo simulation.**

To quantify the uncertainty in ~~time~~ temporal profiles, a range of ~~time~~ temporal profiles (for a full year, hourly resolution) was created for each source sector based on activity data (such as traffic counts). These profiles can be from different years and countries, so that the full range of possibilities is included. These are compared to the fixed ~~time~~ temporal profiles to estimate the uncertainties, which are normally distributed (see Figure 3 for the uncertainty per aggregated GNFR sector). We assume no error correlations exist.

**Table 3: Percentage (%) of emissions of $CO_2$ and CO (~~fossil + biofuel~~FF + BF) that are attributed to large point sources (accounted for in databases) for source sectors public power and industry.**

| | $CO_2$ | | CO | |
|---|---|---|---|---|
| **Country** | **Public power** | **Industry** | **Public power** | **Industry** |
| **Netherlands** | 84.3 % | 80.4 % | 80.7 % | 86.0 % |
| **Belgium** | 65.4 % | 77.5 % | 99.5 % | 93.5 % |
| **Luxembourg** | 67.1 % | 67.2 % | 61.8 % | 94.2 % |
| **Germany** | 85.9 % | 74.1 % | 96.7 % | 87.9 % |
| **Czech Republic** | 89.2 % | 90.4 % | 79.3 % | 94.3 % |

### 2.3 The Monte Carlo simulation

Within a Monte Carlo simulation we create an ensemble (size N) of emissions, spatial proxies and ~~time~~ temporal profiles by drawing random samples from the covariance matrices described in Sect. 2.2. This creates a set of possible solutions in the emission space, reflecting the uncertainties in the underlying parameters. The entire process is shown in Figure 4. As mentioned before, not all sub-sectors are included in the Monte Carlo simulation and the non-included emissions are added to each ensemble member at the final stage. It is important ~~is~~ to ensure that the ~~time~~ temporal profiles and the spatial proxies do not affect the total emissions, so proxies should sum up to 1 for each country and ~~time~~ temporal profiles should be on average 1 over a full year. Before doing this, negative values are removed.

The source sectors that include point source emissions (mainly public power and industry) are treated separately. The large point source emissions and their locations are relatively well-known and available from databases (e.g. from E-PRTR), and therefore not included in the Monte Carlo. The remaining part of the emissions (non-point source or small point sources) from these sectors are distributed using generic proxies (e.g. industrial areas) and are calculated as the difference between the total emissions (activity data x emission factor) and the sum of the point source emissions. If negative emissions result from this subtraction of reported point source emissions, the residual is set to zero. Note that the spatial uncertainty of this residual part is often high. The fraction of the public power and industrial emissions that are attributed to large point sources are shown in Table 3 for several countries.

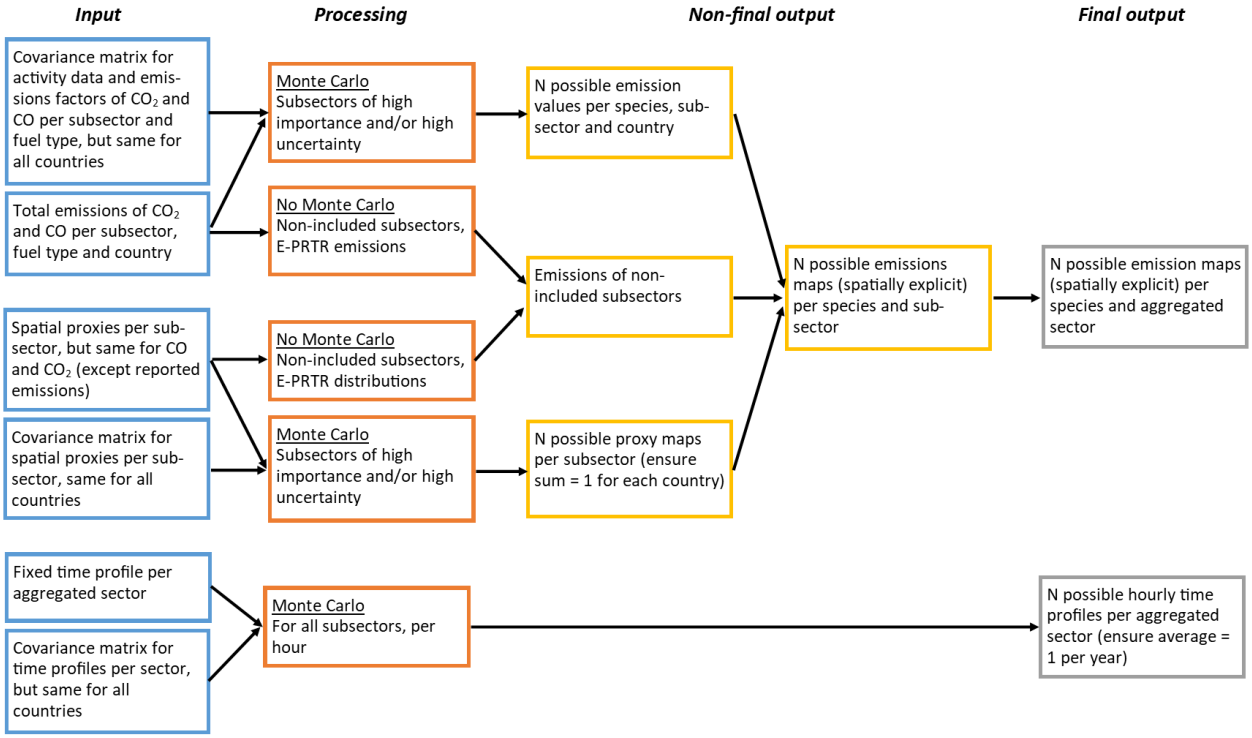


**Figure 4: Flow-diagram showing the input, processing and output of the Monte Carlo simulation.**

### 2.4 Experiments to explore uncertainty propagation

In this paper several experiments are performed to examine the impact of the uncertainties in different parameters on the overall emissions and simulated concentrations:

1) The first experiment uses a Monte Carlo simulation (N=500) to illustrate the spread in emissions per sector due to uncertainties in emission factors and activity data (no spatial/temporal variability is considered). This sample size is based on an analysis of the robustness of the uncertainty estimate (Janssen, 2013), which shows that a sample size of 500 is sufficient to get robust results (Appendix B). This experiment is used to show the contribution of specific sectors to the overall uncertainty and to illustrate how uncertainties vary between sectors and countries. For this experiment country totals are used, also for the countries that are partially outside the zoom domain shown in Figure 1. The results are presented in Sect. 3.1.
2) The second experiment uses a Monte Carlo simulation (N=500) to illustrate how the uncertainty in spatial proxy maps is translated into uncertainties in simulated concentrations (emissions are taken constant; no temporal variability is included). We use emissions of other stationary combustion ($CO_2$) and road traffic (CO) to illustrate the importance of having a correct spatial distribution for measurements close to the source area and further away. The results are presented in Sect. 3.2.
3) The third experiment compares two spatial proxy maps for distributing 'residual' power plant emissions (i.e. those not accounted for in point source databases) to illustrate the potential impact of spreading out small point source emissions when zooming in on small case study areas (emissions are taken constant; no temporal variability is included). The results are presented in Sect. 3.2.
4) The fourth experiment uses a Monte Carlo simulation (N=500) to illustrate the spread in ~~time~~ temporal profiles (emissions are taken constant; no spatial variability is considered). We use this information to determine the error introduced when extrapolating daytime (12–16 h LT) emissions (for example resulting

from an inversion) to annual emissions using an incorrect ~~time~~ temporal profile. Figure 5 shows two possible daily cycles, which have 46 % (blue) and 25 % (orange) of their emissions between 12 and 16 h LT. Therefore, both ~~time~~ temporal profiles will give a different total daily emission when used to ~~extrapolate~~ derive the daytime emissions. The results are presented in Sect. 3.3.

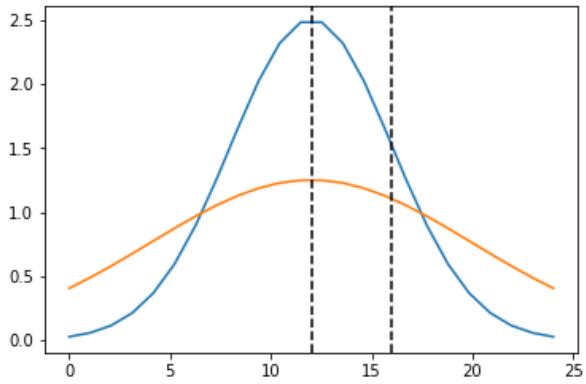


**Figure 5: Schematic overview of two possible ~~time~~ temporal profiles, which represent a different fraction of the total daily emissions during the selected period (12–16 h LT, illustrated by the dashed lines).**

5) For the final experiment, maps are made of the (absolute and relative) uncertainty in each pixel, including uncertainties in emission factors, activity data and spatial proxies (no temporal variability). For this we used a Tier 1 approach, using the following equations:

$$Total\ relative\ uncertainty = \sqrt{\sum standard\ deviations^2}/emission\ sum \quad (1)$$

for the summation of uncorrelation quantities (e.g. sectoral emissions), and:

$$Total\ relative\ uncertainty = \sqrt{\sum relative\ uncertainties^2} \quad (2)$$

for the multiplication of random variables, such as used to combine activity data and emission factors. Here, the (total) relative uncertainty is the percentage uncertainty (uncertainty divided by the total) and the standard deviations are expressed in units of the uncertain quantity (percentage uncertainty multiplied with the uncertain quantity). These maps are used to explore spatial patterns in uncertainties and examine what we can learn about different countries or regions. The results are presented in Sect. 3.4.

For experiment 2 and 3 a smaller domain is selected to represent a local case study (Figure 6). We used the Rotterdam area, which has been studied in detail before (Super et al., 2017b, 2017a). The domain is about 34x26 km and centred over the city, which includes some major industrial activity as well. To translate the emissions into atmospheric concentrations, a simple plume dispersion model is used, the Operational Priority Substances (OPS) model. This model was developed to calculate the transport of pollutants, including chemical transformations (Van Jaarsveld, 2004; Sauter et al., 2016) and was adapted to include CO and $CO_2$ (Super et al., 2017a). The short-term version of the model calculates hourly concentrations at specific receptor points, considering hourly variations in wind direction and other transport parameters. Although the model is often used for point source emissions, it can also handle surface area sources. This model was chosen because of its very short run time, which makes it suitable for a large ensemble. The model is run for each of the alternative emission maps.

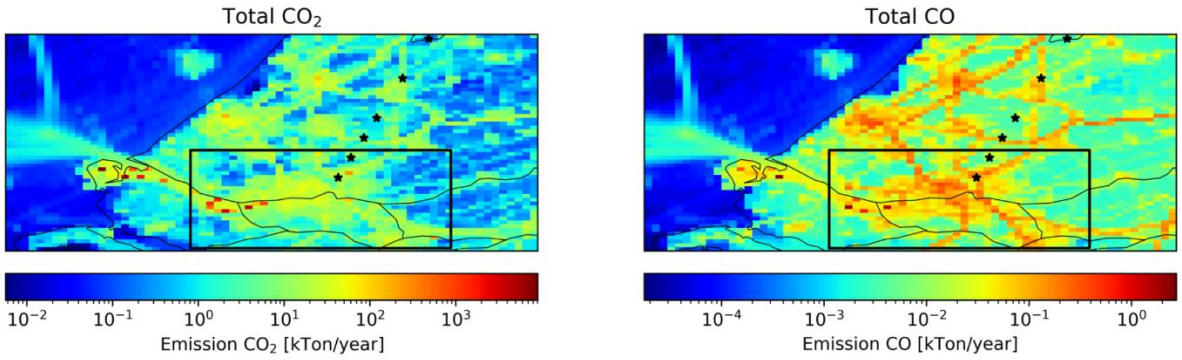


**Figure 6: Emissions of $CO_2$ (left) and CO (right) for part of the Netherlands, including the sub-domain (black rectangle) over Rotterdam. Black stars indicate the receptor locations.**

The OPS model is run for each ensemble member for 5 January 2014 from the start of the day until 16 h LT. On this day the wind direction is relatively constant at about 215° and the wind speed is around 6 m $s^{-1}$. We specify receptor points downwind from the centre of our domain at increasing distance (5, 10, 15, 20, 30 and 40 km). We use the last hour of the simulation for our analyses. We assume emissions from other stationary combustion and road traffic (experiment 2) to take place at the surface. The initial emissions of 'residual' power plants, smeared out over all industrial areas, are also emitted at the surface. However, we raise the height of the emissions to 20m when these emissions are appointed to specific pixels. This height is representative for stack heights of small power plants.

# 3 Results

## 3.1 Uncertainties in total emissions

Using the uncertainties in emission factors and activity data we can evaluate the uncertainty in the total emissions of $CO_2$ and CO per sector. Figure 7 shows the normalized spread in emissions per sector based on the Monte Carlo simulation (N=500). The $CO_2$ emissions have a relatively small uncertainty range and the uncertainty in the total emissions (~~all sectors together~~if we sum all emissions for each of the 500 solutions) is only about 1 % (standard deviation). The largest uncertainties are for fugitives and aviation, which are only small contributors to the total $CO_2$ emissions (1.3 % and 0.4 %, respectively). Therefore, their contribution to the total emission uncertainty is very small, as is shown in Figure 8. The largest uncertainty in the total $CO_2$ emissions is caused by the public power sector. Despite the relatively small uncertainty in the emissions from this sector, it is the largest contributor to the total $CO_2$ emissions (33 %) and therefore the uncertainty in the public power sector contributes about 45 % to the uncertainty in the total $CO_2$ emissions.

In contrast, the CO emissions show a larger uncertainty bandwidth with many high outliers caused by the lognormal distribution of uncertainties in the emission factors. The uncertainty in the total emissions is 6 % for CO (standard deviation). Here, again the largest uncertainties are related to sectors (public power and road transport (LPG fuel)) that are relatively small contributors to the total CO emissions. The main contributor to the uncertainty in total CO emissions is other stationary combustion, which contributes about 31 % to the total emissions and is responsible for more than 60 % of the total uncertainty.

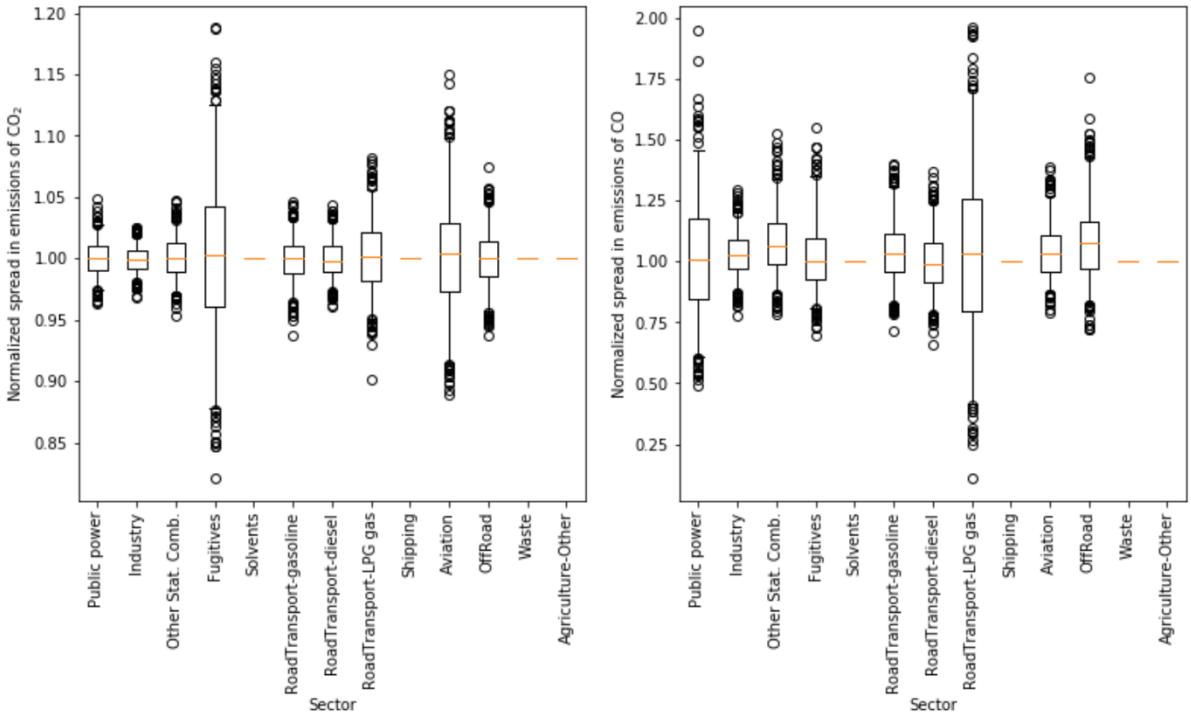


**Figure 7: Normalized spread in emissions of $CO_2$ (left) and CO (right). The box represents the interquartile range, the whiskers the 2.5–97.5 percentile, the lines the median values, and the circles are outliers. For sectors where no box is drawn there is no data included in the Monte Carlo simulation. Note the different scales of the y-axis.**

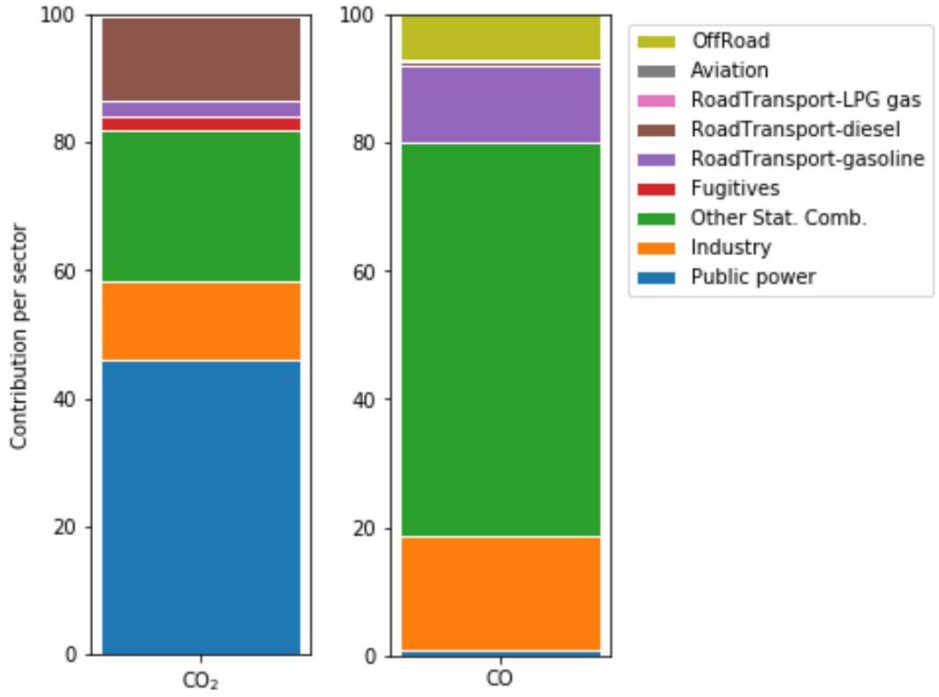


**Figure 8: Contribution of source sectors to the total uncertainty in $CO_2$ (left) and CO emissions (right), summing to 100 %.**

Although the uncertainty in each parameter is assumed to be the same for each country, how a sector is composed of sub-sectors can vary per country. Therefore, the uncertainty per aggregated sector can also vary per country. An example is shown in Figure 9 (left panel), which shows the normalized spread in $CO_2$ emissions of other stationary combustion for all countries within the domain. We find a much larger uncertainty in countries where

the relative fraction of biomass combustion is larger, because biomass burning has a much larger uncertainty in both the activity data and the emission factor. For example, the percentage of biomass burning in the residential sector is 54 % for the Czech Republic and 65 % for Denmark, compared to only 11 % and 9 % for the Netherlands and Great Britain. Thus, differences in the fuel composition of countries result in differences in the overall emission uncertainties, even if the uncertainty per parameter is estimated to be the same. ~~Overall~~For the total $CO_2$ emissions~~,~~ the differences between countries are ~~relatively~~ small, with standard deviations between 1.2 and 2.3 % (Figure 9, right panel).

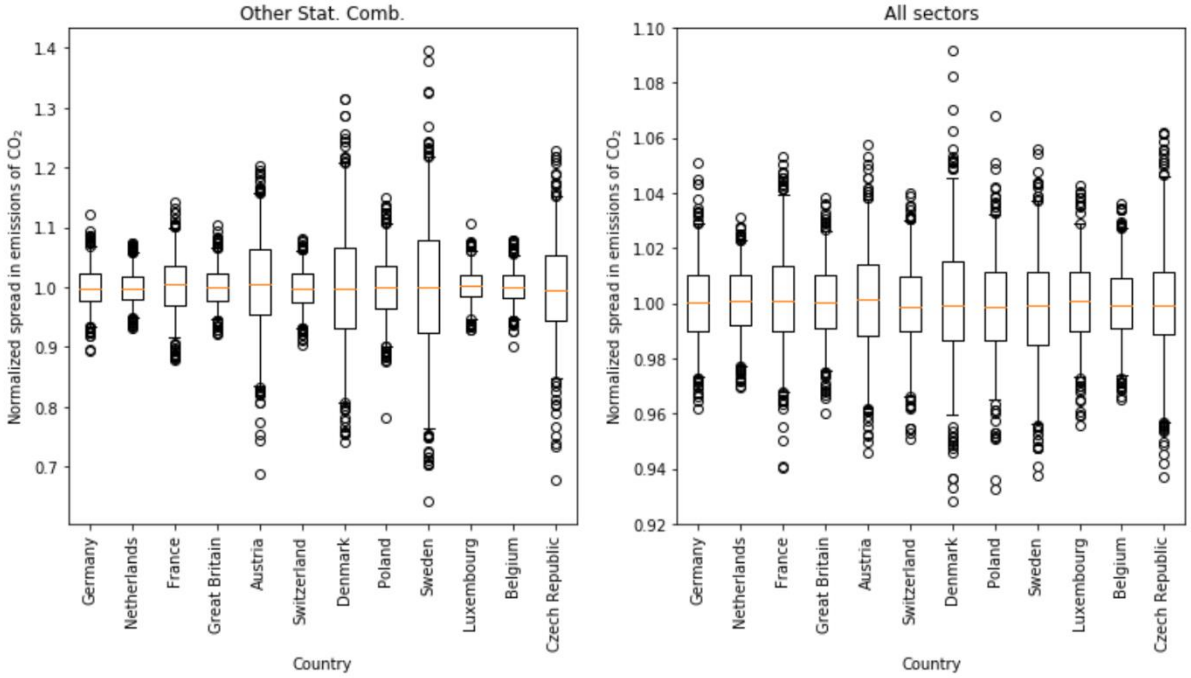


**Figure 9: Normalized spread in emissions of $CO_2$ for other stationary combustion (left) and all sectors combined (right) for a range of countries. The box represents the interquartile range, the whiskers the 2.5–97.5 percentile, the lines the median values, and the circles are outliers.**

### 3.2 Uncertainties in spatial proxies

We examined the impact of uncertainties in spatial proxies on modelled $CO_2$ and CO concentrations for major source sectors. For $CO_2$ we selected other stationary combustion (only commercial/residential, no agriculture/forestry/fishing). The largest fraction (>90 %) of $CO_2$ emissions from this sector is distributed using population density as proxy, which is used here (the remainder of the emissions is not considered). The uncertainty in this sector-proxy combination is estimated to be 50% (normal distribution), mainly due to the disaggregation to the 1x1 km resolution. For CO we selected road transport (all fuels, but only passenger cars). The spatial proxy for distributing passenger car emissions is based on traffic intensities compiled using Open Transport Map and Open Street Map, vehicle emission factors per road type/vehicle type/country, and fleet composition. The uncertainty in this proxy is estimated to be 30 % (normal distribution) due to a higher intrinsic resolution.

Figure 10 shows the resulting spread in atmospheric concentrations as a function of downwind distance from the source area. Note that the concentrations are enhancements caused by local emissions of the selected source sectors and do not include ambient concentrations or other sources. For $CO_2$ (left panel) we see a concentration of about 3.0 ppm at 10 km from the source area centre, but with a large uncertainty bandwidth. This signal is large enough to measure, but with this large uncertainty such measurements are difficult to use in an inversion. The

measurement at 5 km from the source area centre is slightly lower than the one at 10 km, because it is up~~down~~wind of a part of the emissions. At longer distances, the ~~magnitude of the atmospheric signal~~concentration enhancement decreases drastically, and so does the absolute spread in concentrations. The ~~signal~~ enhancement becomes too small compared to the uncertainties occurring in a regular inversion framework to be useful. The right panel shows a similar picture for the CO concentrations resulting from passenger car emissions. Again, the spread in concentrations is large close to the source area centre and decreases with distance, but also the ~~magnitude of the signal~~absolute concentration enhancement decreases. However, in this case the concentration at 5 km from the source area centre is larger, because it is very close to an emission hot spot (see also Fig. 6). Note that we focus here on a single source sector and the overall ~~signals~~ enhancements will be larger and therefore easier to use. Nevertheless, the large spread in concentrations shows that a good representation of the spatial distribution is important for constraining sectoral emissions.

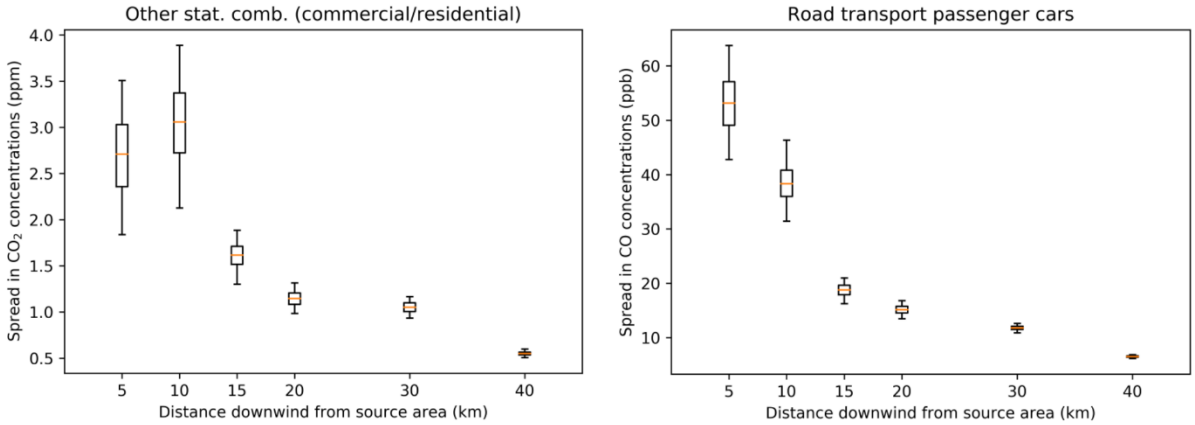


**Figure 10: Spread in simulated concentrations of $CO_2$ resulting from commercial/residential emissions due to uncertainties in the total population proxy map (left) and spread in concentrations of CO resulting from road transport (passenger cars) emissions due to uncertainties in the passenger cars proxy map (right). The box represents the interquartile range, the whiskers the 2.5–97.5 percentile, and the lines the median values of the full ensemble.**

Both proxy maps discussed here are the main proxy maps for the selected sectors. As mentioned before, some sectors have residual emissions that are distributed using an alternative proxy map. An example is public power. Large power plants are listed in databases, including the reported emissions (Table 3). The remainder of the country emissions is spatially distributed over all industrial areas. However, it is more likely that the residual emissions should be attributed to specific point sources (small power plants not listed in databases). That means that instead of spreading the emissions over a large area, leading to very small local emissions and a low concentration gradient, there could be relatively large emissions at a few locations. Therefore, the uncertainty in these sector-proxy combinations is assumed to have a lognormal distribution, in part because of the absence of a better estimation.

We illustrate the effect of this assumption by creating a new proxy map for residual (small) power plants. We find that for the Netherlands a total capacity of 3655 MWe by 676 combustion plants is not included as a point source (source: S&P Global Platts World Electric Power Plants database (https://www.spglobal.com/platts/en/products-services/electric-power/world-electric-power-plants-database)). At least 70 % of this capacity, attributed to 280 plants, is assumed to be in industrial areas. Given 4052 grid cells designated as industrial area in the Netherlands, this is just 7 % of the total amount of industrial area grid cells assuming no more than one plant per grid cell. The remainder is mainly related to cogeneration plants from glasshouses, which are located outside the industrial areas. Therefore, we create a new proxy map for power plants by equally assigning 70 % of the emissions from the

residual power plants to 20 randomly chosen pixels (7 % of the total amount of industrial area pixels in the case study area, i.e. the same density as for the Netherlands as a whole). As mentioned before, we also raise the height of the emissions from surface level to 20 m, which is a better estimate of the stack height for small power plants. The effect on local measurements is large (Figure 11). Instead of measuring a small and constant signal from this sector, the assumed presence of small power plants results in measuring occasional large peak concentrations. Thus, despite being relatively unimportant at the national level, for local studies the impact of the uncertainty in these 'residual' proxies can be large.

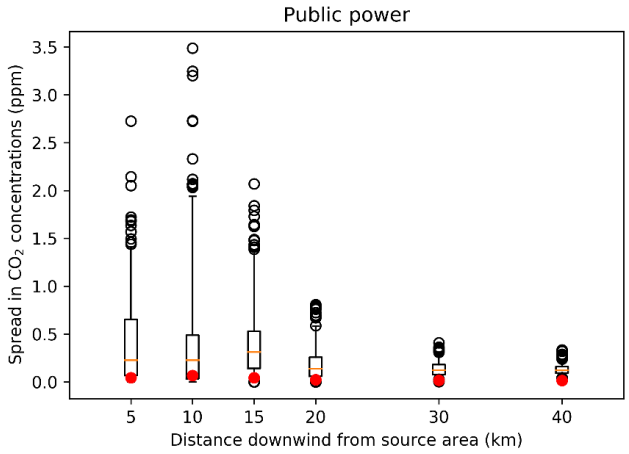


**Figure 11: Spread in simulated concentrations of $CO_2$ resulting from public power emissions due to differences in the proxy map: emissions are distributed using the new proxy map with only 20 randomly chosen pixels containing emissions. The box represents the interquartile range, the whiskers the 2.5–97.5 percentile, the lines the median values, and the black circles are outliers of the full ensemble. The red dots show concentrations of $CO_2$ when the original proxy map is used (industrial area).**

### 3.3 Uncertainties in ~~time~~ temporal profiles

The timing of emissions is important to interpret measurements correctly. During morning rush hour, a peak is expected in road traffic emissions, but the magnitude of this peak can differ from one day to the next. Also, the seasonal cycle in emissions due to heating of buildings can vary between years due to varying weather conditions. Yet, often fixed ~~time~~ temporal profiles are used to describe the temporal disaggregation of annual emissions. The range of possible values for the ~~time~~ temporal profile of other stationary combustion is shown in Figure 12. The range can be very large, especially during the winter. However, note that the average of each ~~time~~ temporal profile is 1.0 for a full year, so that the temporally distributed emissions add up to the annual total. Therefore, changes in the ~~time~~ temporal profile indicate shifts in the timing in the emissions and not changes in the overall emissions due to cold weather, which are accounted for by the activity data.

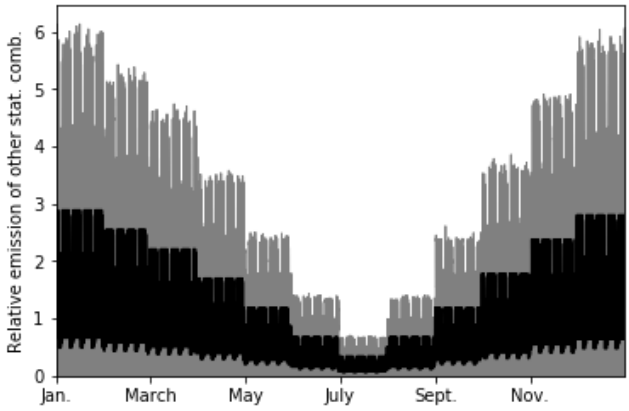


**Figure 12: Spread in ~~time~~ temporal profiles for other stationary combustion (N=500), resulting from the Monte Carlo simulation (grey shading). The black line represents the standard time profile.**

In inverse modelling, often well-mixed (non-stable) daytime measurements are selected (Boon et al., 2016; Breón et al., 2015; Lauvaux et al., 2013), because these are least prone to errors in model transport. For local studies, where transport times are short, this means that only afternoon emissions are optimized. The total annual emissions can then be calculated using a ~~time~~ temporal profile. However, if the ~~time~~ temporal profile is not correct, an incorrect fraction of the emissions can be attributed to the selected hours. We examined the impact of using an incorrect ~~time~~ temporal profile on the total yearly emissions by calculating yearly emissions for each ensemble member. Figure 13 shows the normalized spread in sectoral emissions for all ensemble members. The error in the total annual emissions, resulting from the upscaling of daytime emissions using an incorrect ~~time~~ temporal profile, can reach up to about 1–2 %. This is a significant source of error for country-level $CO_2$ emissions, but less important for CO as the other uncertainties for CO are much larger.

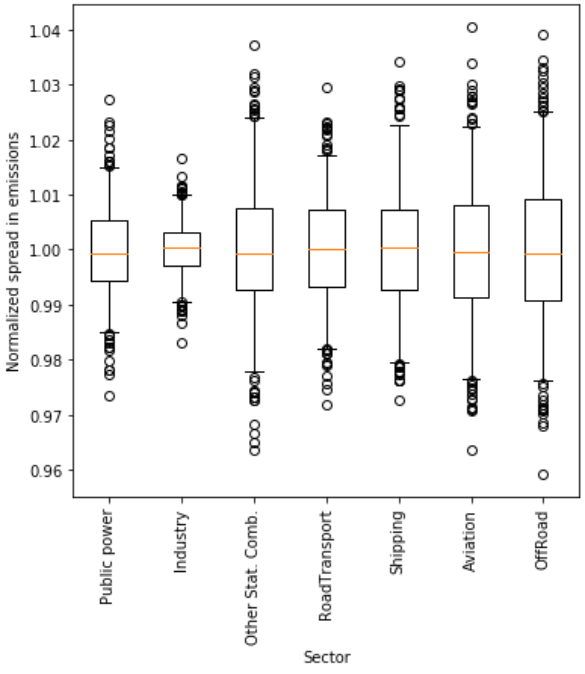


**Figure 13: Normalized spread in emissions of $CO_2$ and CO per sector due to uncertainties in ~~time~~ temporal profiles. The box represents the interquartile range, the whiskers the 2.5–97.5 percentile, the lines the median values, and the circles are outliers. The spread is the same for $CO_2$ and CO, because they have the same temporal profiles.**

### 3.4 Uncertainty maps and spatial patterns

As mentioned before, the uncertainty of the emission value in a grid cell is determined by the uncertainties in activity data, emission factors and spatial distribution proxies. The gridded uncertainty maps in Figure 14 and Figure 15 illustrate that countries or (types of) regions differ significantly in their emission uncertainty, both in absolute and relative values. Concerning the uncertainty in $CO_2$ and CO emissions, several observations can be made.

First, for both CO and $CO_2$ the road network is visible due to low relative uncertainties and high absolute uncertainties compared to the surroundings. This indicates that, despite having large emissions per pixel, the spread in road traffic emissions among ensemble members is relatively small. This is likely due to the small (normally distributed) uncertainty in the spatial proxies for road traffic, i.e. the location of the roads is well-known. The surrounding rural areas are dominated by other stationary combustion, which has a slightly larger spatial uncertainty.

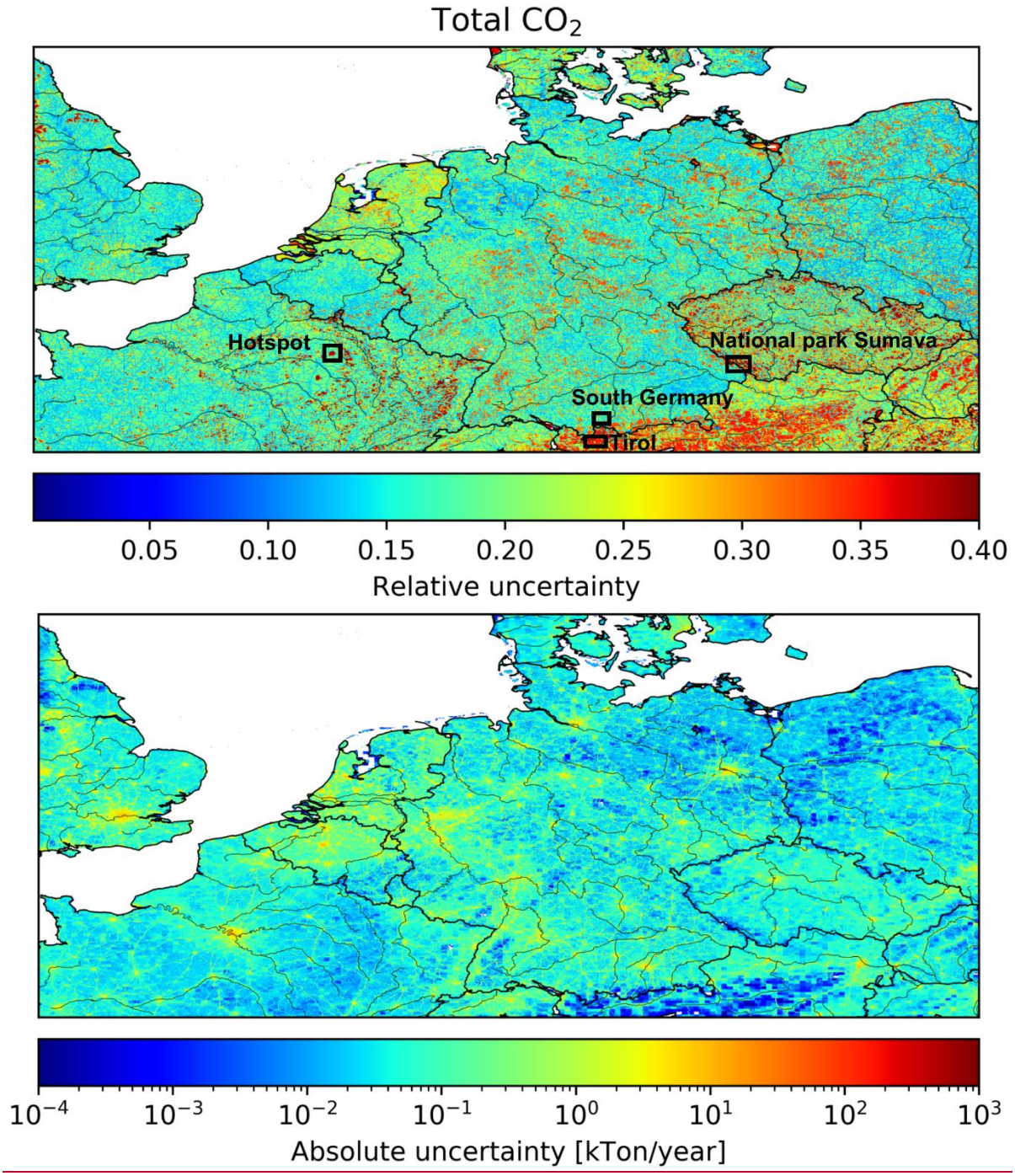


**Figure 14: Maps of the relative and absolute uncertainty in $CO_2$ emissions. Areas that are examined in more detail are outlined by black squares in the top panel.**

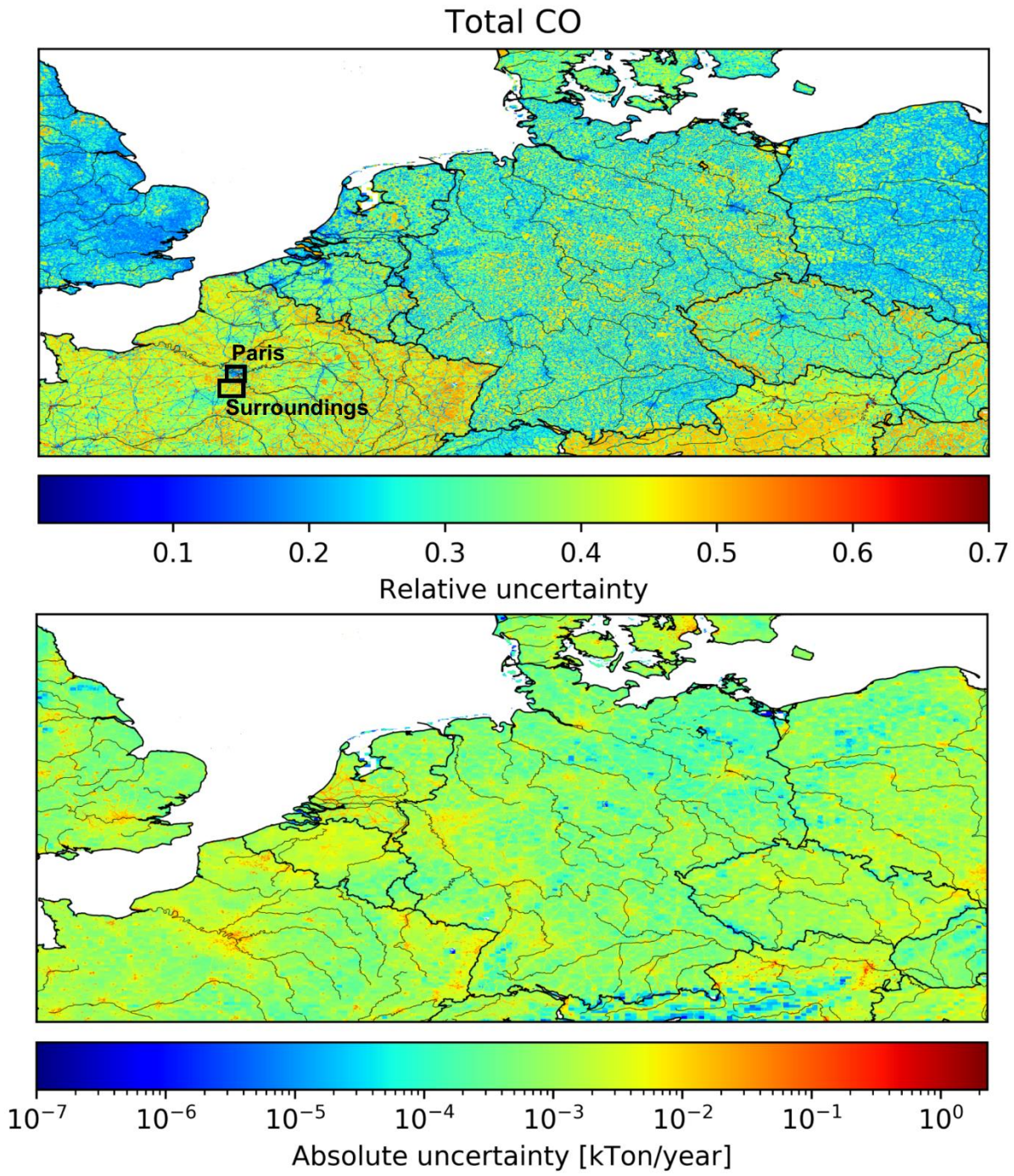


**Figure 15: Maps of the relative and absolute uncertainty in CO emissions. Areas that are examined in more detail are outlined by black squares in the top panel.**

Second, in Austria (Tirol mainly) a large area of high relative uncertainty in $CO_2$ emissions is visible (average pixel emission is 220 tonnes $CO_2$ $yr^{-1}$), which we compare to an area just on the other site of the border in southern Germany (average pixel emission is 495 tonnes $CO_2$ $yr^{-1}$). The uncertainty in both areas is dominated by other stationary combustion. Yet, in Austria a lot of biofuel~~s are~~ is used (52 % of the total emissions for this source sector) with a large uncertainty in the emission factor and spatial distribution, whereas in Germany only 20 % of the emissions in this sector are caused by biofuel~~s~~ combustion. On the other hand, the absolute uncertainty is very small in Tirol because of the low population density (and thus small emissions) in this mountainous area.

Third, some large patches of high relative uncertainty in $CO_2$ emissions are visible in the Czech Republic and the northeast of France. The location of these patches seems to correspond to natural areas/parks. Therefore, absolute uncertainties are low in these areas given the low emissions (average pixel emission in the Sumava national park is 22 tonnes $CO_2$ $yr^{-1}$). The total uncertainty can be explained for 60 % by the uncertainty in other stationary combustion, mainly wood burning (Figure 16). Also, agriculture (field burning of residues) plays a significant role. In addition to these natural areas, there are also some very small dark red areas (relative uncertainty) in northern France. These areas are military domain and have a lower absolute uncertainty than their surroundings because very few emissions are distributed to these areas (average pixel emission is 250 tonnes $CO_2$ $yr^{-1}$). The public power and industrial emissions are probably too small to be reported, hence the large relatively uncertainty.

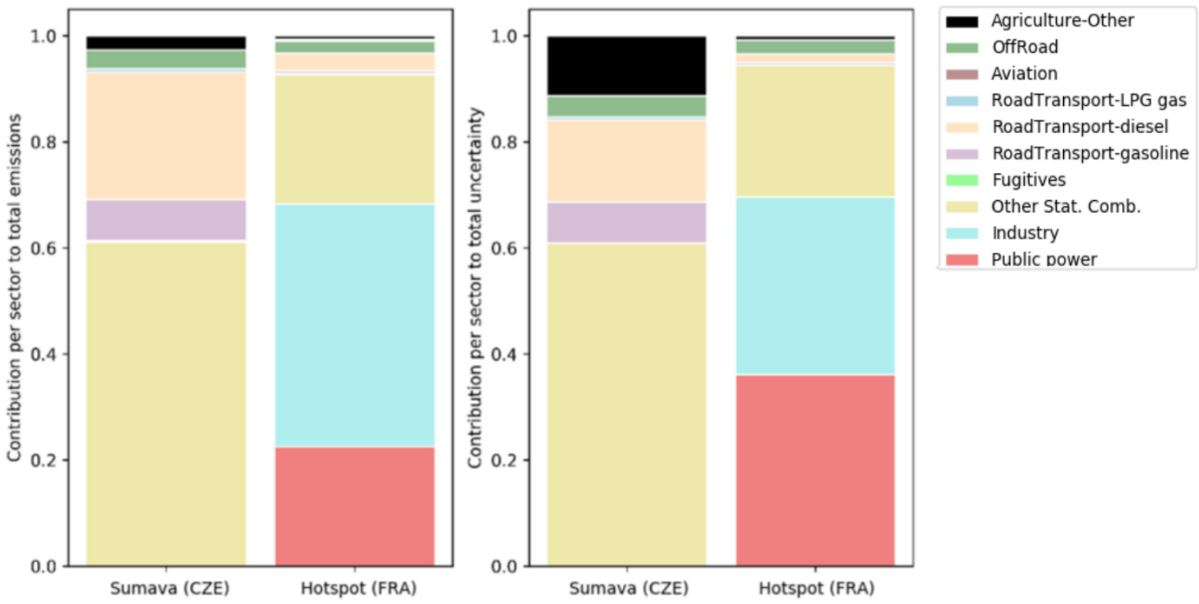


**Figure 16: Contribution of source sectors to the total emissions (left) and the total uncertainty (right) in $CO_2$ for the Sumava national park in the Czech Republic and a hotspot in France, summing to 100 %. See Figure 14 for the exact location of these areas.**

Fourth, ~~big cities~~strongly urbanized areas like Paris, ~~Berlin~~ the Ruhr area in Germany and ~~Brussels~~ Rotterdam (also see Fig. 1 for their locations) are clearly visible as areas where the relative uncertainty in CO emissions is lower than in the surrounding areas. Compared to its surroundings, the uncertainty in Paris is mainly determined by the industrial sector (Figure 17). Since industrial emissions are relatively well-known, the relative uncertainty is small. However, the absolute uncertainty is large for big cities because of the high emissions in these densely populated areas (average pixel emission is 64 tonnes CO $yr^{-1}$ for Paris). In the surrounding areas the emissions are again dominated by other stationary combustion, which has a larger uncertainty. Yet, the absolute uncertainty is smaller because of the lower emissions (average pixel emission is 12 tonnes CO $yr^{-1}$).

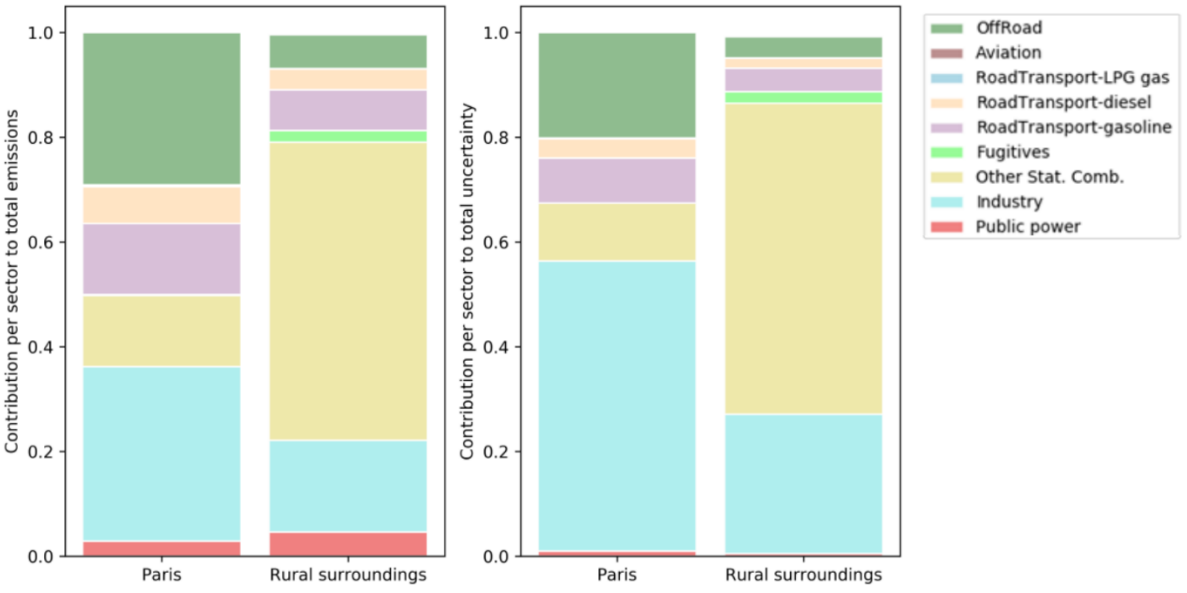


**Figure 17: Contribution of source sectors to the total emissions (left) and the total uncertainty (right) in CO for Paris and its surroundings, summing to 100 %. See Figure 15 for the exact location of these areas.**

Finally, the relative uncertainties seem to be consistently higher in some countries than in others. For example, the relative uncertainty in the total emissions of France and Great Britain (only pixels within the domain) are 39 % and 25 %, respectively. For France, the main sources of uncertainty are industry and other stationary combustion, whereas the off-road and road transport sectors have a significant contribution to the uncertainty in Great Britain (Figure 18). The main difference between the countries is again the amount of biomass used in the other stationary combustion sector (26 % in France and 8 % in Great Britain). This is likely to explain why in rural areas the relative uncertainty is much higher in France.

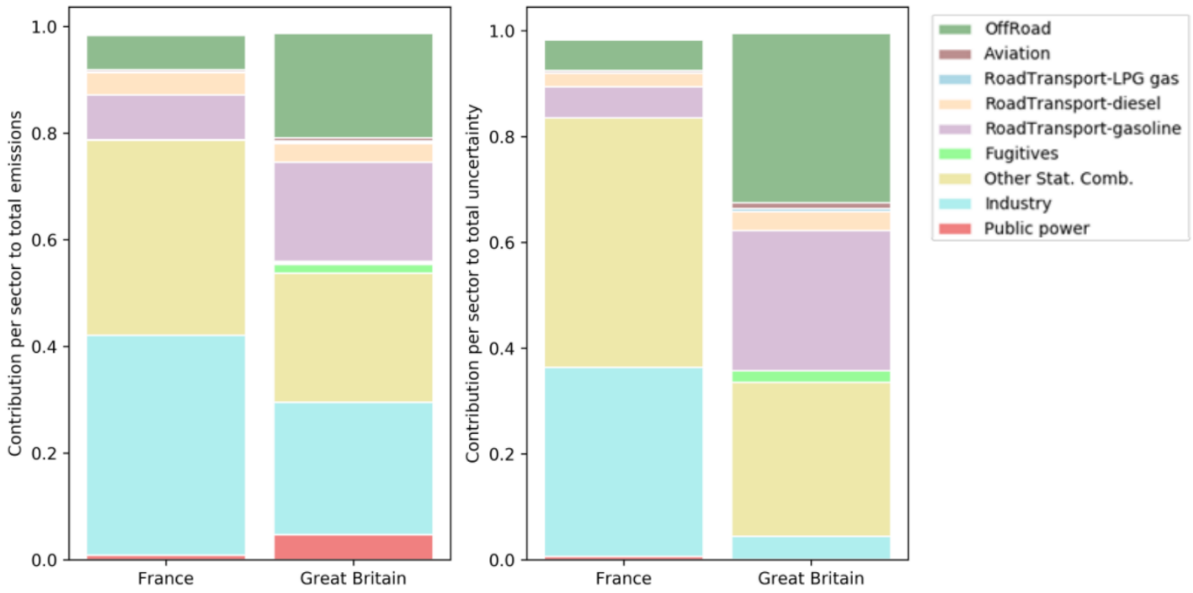


**Figure 18: Contribution of source sectors to the total emissions (left) and the total uncertainty (right) in CO for France and Great Britain, summing to 100 %.**

## 4. Discussion ~~and conclusions~~

Several previous studies have examined the uncertainty in emissions, either globally or nationally. For example, Andres et al. (2014) studied the uncertainty in the CDIAC emission inventory on a global scale, suggesting that the largest uncertainties are related to the fuel consumption (i.e. activity data). A similar concern was identified for China, for which the uncertainty in energy statistics resulted in an uncertainty ratio of 15.6 % in the 2012 $CO_2$ emissions (Hong et al., 2017). In the present study the uncertainties in activity data and emission factors are similar for $CO_2$, whereas the uncertainty in CO emission factors is much larger than the uncertainty in activity data. A possible explanation for this is that the energy statistics for the European countries included here are more reliable than for developing countries. The occurrence of large differences in the reliability of reported emissions between countries is also illustrated by Andres et al. (2014). In addition to these scientific studies, many countries report uncertainties in emission estimates in their National Inventory Reports (UNFCCC, 2019). Yet, their methods for uncertainty calculation differ and can even vary over time. Several scholars have examined the uncertainty in national ~~$CO_2$~~ greenhouse gas emissions in more detail. For example, Monni et al. (2004) (Finland) and Fauster et al. (2011) (Denmark) used a Tier 2 approach (Monte Carlo simulation) to determine the uncertainty in the total greenhouse gas emissions (in $CO_2$ equivalents). They found an uncertainty of about 5–6 % for the year 2001 for Finland and an uncertainty of 4–5 % for the year 2008 for Denmark, also considering non-normal distributions in uncertainties. Moreover, Oda et al. (2019) found a 2.2 % difference in total $CO_2$ emissions in Poland between two emission inventories, which is in agreement~~. These values agree~~ with our total $CO_2$ emission uncertainty~~ies~~.

Even fewer studies have focused on uncertainties in the proxy maps used for spatial disaggregation. Some studies compared emission inventories to get an idea of the spatial uncertainties (Gately and Hutyra, 2017; Hutchins et al., 2017), but these studies are likely to underestimate uncertainties due to systematic errors ~~caused~~ that occur when different emission inventories use similar methods and/or proxies for spatial allocation. Moreover, exact quantification of uncertainties is often limited, dependent on the spatial scale, and the uncertainties are not specified per source (i.e. total emissions and spatial disaggregation) (Oda et al., 2019). Sowden et al. (2008) used a qualitative approach to identify the uncertainty of different components of their emission inventory for reactive pollutants (activity, emission factors, spatial and temporal allocation and speciation) by giving each component a quality rating. They suggest that spatial allocation is an important source of uncertainty for residential burning, but not so much for point sources and road traffic. Indeed, the location of large point sources and roads is relatively well-known. However, we consider the allocation of emissions to pixels that include roads to have a significant (pixel value) uncertainty. Therefore, our results show that uncertainties in the spatial proxy used for road traffic can cause a significant spread in CO concentrations.

Andres et al. (2016) did a more extensive analysis of the spatial distribution in CDIAC, including uncertainties in pixel values (e.g. due to incorrect accounting methods or changes over time) and due to the representativeness of the proxy for the spatial distribution of emissions (also see Sect. 2.2.3). We considered these sources of uncertainty as well. However, Andres et al. (2016) also mention spatial discretization as a source of error, because they assign each pixel (1x1° resolution) to one country. The proxy maps used in this study include country fractions in each pixel, reducing this uncertainty. In contrast, we suggest another source of uncertainty, namely the fact that some pixels can include emissions while no activity takes place there or vice versa (proxy quality). Based on the listed uncertainties, Andres et al. (2016) found an average uncertainty ($2\sigma$) in individual pixels of 120 % (assuming normal distributions). Here, we find an average uncertainty ($2\sigma$) of 36 %. However, a small number of large

outliers occurs (less than 0.01 % of the pixels has an uncertainty of >1000 %) due to lognormal error distributions, although these are related to pixels with small emissions. A large part of the difference can be explained by the large pixel size of CDIAC and the large error introduced by spatial discretization (e.g. due to pixels that cover large areas of two different countries). Also, their emissions are spatially distributed based on population density, while we use a range of proxy maps depending on the source sector and use specific locations for large point sources. However, the uncertainty estimates are partially based on expert judgement and remain subjective. Moreover, the uncertainty related to the location of actual activities is not included in our uncertainty estimate, even though we have shown this can have a large impact locally.

The country-level $CO_2$ emissions used for our emission inventory are based on NIR's, which are assumed to be relatively accurate because of the use of detailed fuel consumption statistics and country-specific emission factors (Andres et al., 2014; Francey et al., 2013). The uncertainties reported in the NIRs were determined following specified procedures and are deemed the most complete and reliable estimates available. Yet, because of the use of prescribed methods and in some cases general emission factors, systematic errors can occur both in the estimate of parameters and in the estimate of uncertainties. We choose to average the uncertainties reported by several countries, because the uncertainty estimates are relatively consistent across countries. However, this would not eliminate such systematic errors. The effect of systematic errors could be analysed by comparing different sources of information. Additionally, we assume point source emissions are relatively certain, yet a recent study showed that significant uncertainties exist in reported emissions of US power plants (Quick and Marland, 2019). A similar study for Europe is recommended, not only to improve the knowledge for the European situation, but also to understand continental differences.

One source of uncertainty that is not considered in this study is the incompleteness of the emission inventory (i.e. if sources are missing) or double-counting errors. For example, during the compilation of the base inventory we found that in several cases the $CO_2$ emissions from airports were very low. The reason was that emissions from international flights are not reported in the NIR's and are therefore not part of the emission data used to create the inventory. Once discovered, this was corrected and ~~LTO (~~aircraft landing and take-off~~)~~ emissions from international flights were added in a later stage. Such discrepancies caused by reporting guidelines could be present for other source types as well. Although overall this error is likely to be small, locally the errors might be significant.

Finally, Sowden et al. (2008) mention (dis)aggregation as another source of error, i.e. the calculation of emissions on a different scale (spatially, temporally or sector level) than the input data. In principle, fuel consumption data is available on aggregated levels and then separated over different subsectors. This increases the uncertainty at the lower level, but on the aggregated level the uncertainties remain the same. A similar note was made by Andres et al. (2016) about the use of higher resolution proxy maps, which might increase the uncertainty due to lack of local data. However, when local data is available this might also decrease the uncertainties. For example, the EDGAR emission database uses non-country specific emission factors based on technology levels and sector aggregated energy statistics (Muntean et al., 2018). The reason is that the level of detail we used in this paper is not available globally. However, using generic emission factors can introduce large uncertainties when sub-sectoral chances occur. Therefore, regional/local studies could benefit from using a dedicated emission inventory for their region of interest instead of a global inventory.

Our results can be used to support network design and inverse modelling. The uncertainty maps are helpful to identify regions with large emission uncertainties, which can be the focus point of an inversion with the aim to optimize emissions in those regions. However, inverse modelling requires an observational network that is sensitive to the emissions from the regions of interest. A site is sensitive to specific emissions when it is often affected by them, taking into account the dominant wind direction and the magnitude of concentration enhancements, which should be larger than the uncertainties that affect model-observation comparison (e.g. measurement uncertainty and model errors). Plumes from emission hot spots can travel a long distance and sites up to 30 km downwind have shown to be able to detect urban signals (Super et al., 2017a; Turnbull et al., 2015). The concentration enhancement in these plumes is large and therefore easy to detect. In contrast, the concentration enhancements of a single source (sector) are much smaller, as shown in Fig. 10 and Fig. 11, and therefore they become undetectable at much shorter distances. For example, vehicle exhaust emissions were shown to decrease by a factor 2 at 200 m from a highway (Canagaratna et al., 2010), while power plants plumes have been detected several kilometres downwind (Lindenmaier et al., 2014). Dilution is strongly dependent on the atmospheric conditions and also the height of the measurement site plays an important role. To conclude, the optimal network design is strongly dependent on which question needs to be answered and the focus area and resolution needed to reach this goal.

## 5. Conclusions

In this work we studied the uncertainties in a high-resolution gridded emission inventory for $CO_2$ and CO, considering uncertainties in the underlying parameters (activity data, emission factors, spatial proxy maps and ~~time~~ temporal profiles). We find that all factors play a significant role in determining the emission uncertainties, but that the contribution of each factor differs per sector. Disaggregation of emissions introduces additional sources of uncertainty, which makes uncertainties at higher resolution larger than at the scale of a country/year and can have a large impact on (the interpretation of) local measurements. This is an important consideration for inverse modelers and our methodology can be used to better define local uncertainties for e.g. urban inversions. Inverse modelers should be aware that the use of erroneous ~~time~~ temporal profiles to extrapolate emission data could result in errors of a few percent, which for $CO_2$ is significant. In the future, the temporal profiles could be improved by using detailed activity data, e.g. from power plants. Moreover, we found that large regional differences exist in absolute and relative uncertainties. By looking in more detail at specific regions (or countries) more insight can be gained about the emission landscape and ~~what are~~ the main causes of uncertainty. Interestingly, areas with larger absolute uncertainties often have smaller relative uncertainties. A likely explanation is that large sources of $CO_2$ and CO emissions received more attention and are therefore relatively well-constrained, for example in the case of large point sources. Nevertheless, since we are most interested in absolute emission reductions the map with absolute uncertainties can be used to define an observational network that is able to reduce the largest absolute uncertainties. Finally, we believe that an uncertainty product based on a well-defined, well-documented and systematic methodology could be beneficial for the entire modelling community and support decision-making as well. However, specific needs can differ significantly between studies, for example the scale/resolution, source sector aggregation level, and which species are included. Therefore, the creation of a generic uncertainty product is challenging and needs further research.

## Data availability

The family of ten emission inventories ~~are~~ is available for non-commercial applications and research (https://doi.org/10.5281/zenodo.3584549). ~~Please contact Hugo Denier van der Gon ().~~

## Appendix A

**Table A1: Relative uncertainties (fraction) in activity data and $CO_2$ emission factors as taken from the NIRs (country-average) and in CO emission factors as derived from literature (assumed equal for all countries in the domain). The quoted uncertainty ranges are assumed to be representative for one standard deviation. Uncertainties in activity data and $CO_2$ emission factors are often relatively low and symmetrically distributed and normal distributions (Norm) are assumed for these activities. Compared to $CO_2$ emission factors, the uncertainty in CO emission factors is much higher, up to an order of magnitude. Uncertainties in CO emission factors are often lognormally distributed (Logn) and are assumed equal for all countries in the HR domain. The uncertainty in the activity of open burning of waste (not covered by the NIRs) is also assumed to have a lognormal distribution.**

| Sector (NFR) | Fuel type | Activity data | | $CO_2$ emission factors | | CO emission factors | |
|---|---|---|---|---|---|---|---|
| | | Average | Distribution | Average | Distribution | Average | Distribution |
| Public electricity and heat production (1.A.1.a) | Solid (fossil) | 0.018 | Norm | 0.030 | Norm | 0.149 | Logn |
| | Liquid (fossil) | 0.022 | Norm | 0.031 | Norm | 0.399 | Norm |
| | Gaseous (fossil) | 0.021 | Norm | 0.015 | Norm | 0.513 | Norm |
| | Biomass | 0.060 | Norm | 0.05 | Norm | 0.231 | Logn |
| Oil and gas refining (1.A.1.b & 1.B.2.d) | All | 0.038 | Norm | 0.048 | Norm | 0.402 | Norm |
| Oil production & Gas exploration (1.B.2 mainly flaring, 1.B.2.c) | All | 0.118 | Norm | 0.141 | Norm | 0.240 | Logn |
| Iron and steel industry (1.A.2.a & 2.C.1) | All | 0.044 | Norm | 0.056 | Norm | 0.240 | Logn |
| Non-ferrous metals (1.A.2.b & 2.C.2_3) | All | 0.031 | Norm | 0.029 | Norm | 0.208 | Norm |
| Chemical industry (1.A.2.c & 2.B) | All | 0.042 | Norm | 0.041 | Norm | 0.138 | Logn |
| Pulp and paper industry (1.A.2.d) | All | 0.027 | Norm | 0.016 | Norm | 0.138 | Logn |
| Food processing, beverages and tobacco (1.A.2.e) | All | 0.029 | Norm | 0.017 | Norm | 0.138 | Logn |
| Non-metallic minerals (1.A.2.f & 2.A) | All | 0.032 | Norm | 0.041 | Norm | 0.384 | Logn |
| Other manufacturing industry (1.A.2.g) | All | 0.029 | Norm | 0.014 | Norm | 0.138 | Logn |
| Civil aviation - LTO (1.A.3.a) | All | 0.089 | Norm | 0.040 | Norm | 0.231 | Logn |
| Road transport (all vehicle types) (1.A.3.b) | Gasoline (fossil) | 0.031 | Norm | 0.025 | Norm | 0.284 | Logn |
| | Diesel (fossil) | 0.032 | Norm | 0.026 | Norm | 0.319 | Norm |
| | Gaseous (fossil) | 0.039 | Norm | 0.027 | Norm | 0.320 | Logn |
| | LPG | 0.039 | Norm | 0.027 | Norm | 0.462 | Norm |
| Other transport (1.A.3.e & 1.A.4 mobile) | All | 0.067 | Norm | 0.023 | Norm | 0.384 | Logn |
| Other mobile (1.A.5.b) | All | 0.098 | Norm | 0.026 | Norm | 0.384 | Logn |
| Residential (1.A.4.b) | Gaseous (fossil) | 0.040 | Norm | 0.022 | Norm | 0.141 | Logn |
| | Liquid (fossil) | 0.048 | Norm | 0.024 | Norm | 0.404 | Norm |
| | Solid (fossil) | 0.085 | Norm | 0.041 | Norm | 0.141 | Logn |
| | Biomass | 0.163 | Norm | 0.055 | Norm | 0.384 | Logn |
| Commercial institutional (1.A.4.a) | Gaseous (fossil) | 0.043 | Norm | 0.022 | Norm | 0.138 | Logn |
| | Liquid (fossil) | 0.055 | Norm | 0.023 | Norm | 1.065 | Norm |
| | Solid (fossil) | 0.087 | Norm | 0.040 | Norm | 0.994 | Norm |
| | Biomass | 0.103 | Norm | 0.055 | Norm | 0.730 | Logn |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| Agriculture/Forestry/Fishing (1.A.4.c) | Gaseous (fossil) | 0.050 | Norm | 0.028 | Norm | 0.138 | Logn |
| | Liquid (fossil) | 0.051 | Norm | 0.029 | Norm | 1.065 | Norm |
| | Solid (fossil) | 0.095 | Norm | 0.048 | Norm | 0.994 | Norm |
| | Biomass | 0.096 | Norm | 0.09 | Norm | 0.730 | Logn |
| Other stationary (1.A.5.a) | Gaseous (fossil) | 0.097 | Norm | 0.023 | Norm | 0.138 | Logn |
| | Liquid (fossil) | 0.084 | Norm | 0.021 | Norm | 1.065 | Norm |
| | Solid (fossil) | 0.103 | Norm | 0.033 | Norm | 0.994 | Norm |
| | Biomass | 0.180 | Norm | 0.04 | Norm | 0.730 | Logn |
| Agricultural waste burning (3.F) | - | 1.609 | Logn | 0.2 | Norm | 0.429 | Norm |
| Uncontrolled waste burning (5.C.2) | - | 1.609 | Logn | 0.5 | Norm | 0.366 | Logn |

**Table A2: Relative uncertainties (fractions) at cell level resulting from the spatial distribution. The values listed represent the (one standard deviation) uncertainty of the emission per cell due to uncertainty sources 2 and 3 as listed in Sect. 2.2.3. All values in the table below are based on expert quantification and inevitably include a considerable amount of subjectivity. The data should therefore be considered as a first order indication only. Note that the natural logarithm (Ln) of the uncertainty fraction is given in case uncertainty has a lognormal distribution.**

| Sector name | Proxy name | Distribution | Uncertainty |
|---|---|---|---|
| Public electricity and heat production; Chemical industry; Food processing, beverages and tobacco (comb); Food and beverages industry; Other non-metallic mineral production; Small combustion - Commercial/institutional – Mobile | CORINE_2012_Industrial_area | Logn | 2.2 |
| Solid fuel transformation; Iron and steel industry (comb); Iron and steel production; Pulp and paper industry (comb); Pulp and paper industry; Non-metallic minerals (comb); Cement production | CORINE_2012_Industrial_area | Logn | 3.7 |
| Other manufacturing industry (comb); Other industrial processes; Manufacturing industry - Off-road vehicles and other machinery | CORINE_2012_Industrial_area | Logn | 1.4 |
| Oil and gas refining (comb); Oil and gas refining | CORINE_2012_Industrial_area | Logn | 3.7 |
| | TNO_PS for Refineries | Logn | 1.7 |
| Coal mining (comb) | CORINE_2012_Industrial_area | Logn | 4.6 |
| | TNO_PS for Coal mining | Logn | 1.7 |
| Oil production (comb) | CORINE_2012_Industrial_area | Logn | 1.7 |
| | TNO_PS for Oil production | Logn | 1.7 |
| Gas exploration (comb) | CORINE_2012_Industrial_area | Logn | 1.7 |
| | TNO_PS for Gas production | Logn | 1.7 |
| Coke ovens (comb) | CORINE_2012_Industrial_area | Logn | 1.7 |
| | TNO_PS for Iron and steel - Coke ovens | Logn | 1.7 |
| Non-ferrous metals (comb); Other non-ferrous metal production | CORINE_2012_Industrial_area | Logn | 3.7 |
| | TNO_PS for Non-ferrous metals - Other | Logn | 1.7 |
| Aluminium production | CORINE_2012_Industrial_area | Logn | 3.7 |
| | TNO_PS for Non-ferrous metals - Aluminium | Logn | 1.7 |
| Chemical industry (comb) | CORINE_2012_Industrial_area | Logn | 2.2 |
| | TNO_PS for Chemical industry | Logn | 1.7 |
| Passenger cars | RoadTransport_PassengerCars | Norm | 0.3 |
| Light duty vehicles | RoadTransport_LightCommercialVehicles | Norm | 0.3 |
| Trucks (>3.5t) | RoadTransport_HeavyDutyTrucks | Norm | 0.3 |

| | | | |
|---|---|---|---|
| Buses | RoadTransport_Buses | Norm | 0.3 |
| Motorcycles | RoadTransport_Motorcycles | Norm | 0.3 |
| Mopeds | RoadTransport_Mopeds | Norm | 0.5 |
| Civil aviation – LTO | Airport distribution for year 2015 | Logn | 1.4 |
| Mobile sources in agriculture/forestry/fishing | CORINE_2012_Arable_land | Logn | 1.4 |
| Other transportation, including pipeline compressors | Population_total_2015 | Logn | 3.7 |
| Small combustion - Residential - Household and gardening; Other mobile combustion | Population_total_2015 | Logn | 1.3 |
| Commercial/institutional | Population_total_2015 | Norm | 0.5 |
| | Population_rural_2015 | Logn | 1.3 |
| | Population_urban_2015 | Logn | 1.3 |
| | Wood_use_2014 | Logn | 2.2 |
| Residential | Population_total_2015 | Norm | 0.5 |
| | Population_rural_2015 | Logn | 1.3 |
| | Population_urban_2015 | Logn | 1.3 |
| | Wood_use_2014 | Logn | 1.4 |
| Agriculture/Forestry/Fishing | CORINE_2012_Arable_land | Logn | 1.4 |
| | Wood_use_2014 | Logn | 2.2 |
| Other stationary combustion | Population_total_2015 | Logn | 1.3 |
| | Population_rural_2015 | Logn | 1.3 |
| | Wood_use_2014 | Logn | 1.4 |
| Field burning of agricultural residues | CORINE_2012_Arable_land | Logn | 2.2 |
| | Population_total_2015 | Logn | 2.2 |
| Open burning of waste | CORINE_2012_Industrial_area | Logn | 3.7 |
| | Population_rural_2015 | Logn | 3.7 |

## Appendix B

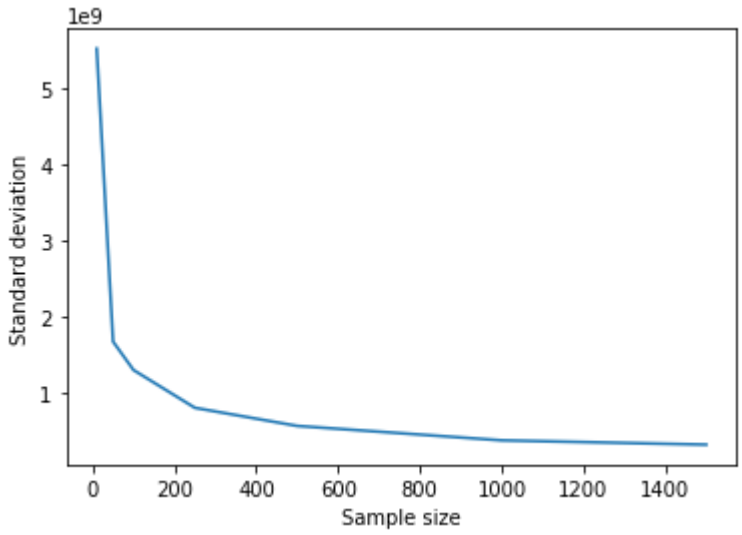


**Figure B1: Spread in the standard deviations if the Monte Carlo simulation were to be repeated multiple times for a specific sample size, based on a bootstrapping method.**

## Author contribution

A.J.H. Visschedijk assembled the uncertainty data used in this work. S.N.C. Dellaert and H.A.C. Denier van der Gon are responsible for the base emission inventory. I. Super designed the experiments, carried them out, and prepared the manuscript with contributions from all co-authors.

## Competing interests

The authors declare that they have no conflict of interest.

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