Global anthropogenic CO₂ emissions and uncertainties as prior for Earth system modelling and data assimilation

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Abstract. The growth in anthropogenic carbon dioxide (CO₂) emissions acts as a major climate-change driver, which has widespread implications across society, influencing the scientific, political and public sectors. For an increased understanding of the CO₂ emission sources, patterns and trends, a link between the emission inventories and observed CO₂ concentrations is best established via Earth system modelling and data assimilation. In this study anthropogenic CO₂

- 15 emission inventories are processed into gridded maps to provide an estimate of CO₂ emissions for 7 main emissions groups: 1) energy production super emitters, 2) energy production standard emitters, 3) manufacturing, 4) settlements, 5) aviation, 6) other transport at ground level and 7) others, with estimation of their uncertainty and covariance to be included in the European Centre for Medium Range Weather Forecasts (ECMWF) Integrated Forecasting System (IFS). The emission inventories are sourced from the Intergovernmental Panel on Climate Change (IPCC) 2006 Guidelines for National
- 20 Greenhouse Gas Inventories and revised information from its 2019 Refinements, and the global grid maps of Emissions Database for Global Atmospheric Research (EDGAR) inventory. The anthropogenic CO₂ emissions for 2012 and 2015, (EDGAR versions 4.3.2 and 4.3.2_FT2015 respectively) are considered, updated with improved apportionment of the energy sector (decreased by 8 %) and the energy usage for manufacturing (increased by 18 %), and with newly generated diffusive CO₂ emissions from coal mines. These emissions aggregated into 7 ECMWF groups with their emission uncertainties are
- 25 calculated per country (considering its statistical infrastructure development level) and sector (considering the most typical fuel type) and use the IPCC recommended error propagation method assuming fully uncorrelated emissions. While the uncertainty of most groups remains relatively small (5 20 %), the largest contribution (usually over 40 %) to the total uncertainty is determined by the OTHER group with usually the smallest budget, consisting of oil refineries and transformation industry, fuel exploitation, coal production, agricultural soils and solvents and products use emissions, with
- 30 uncertainties more than 100 %. Several sensitivity studies are performed: 1) for country type by analysing the impact of assuming either a well or less-well developed statistical infrastructure, 2) for fuel type specification by adding explicit information for each fuel type used per each IPCC activity, and 3) for national emission source distribution by aggregating

all emission sources and evenly redistributing them over the country – highlights the importance of spatial mapping. Uncertainties are compared with United Nations Framework Convention on Climate Change (UNFCCC) and the

- 35 Netherlands Organisation for Applied Scientific Research (TNO) data. Upgraded anthropogenic CO₂ emission maps with their yearly and monthly uncertainties are combined into the CHE_EDGAR-ECMWF_2015 dataset (Choulga et al., 2020) available from <u>https://doi.org/10.5281/zenodo.3967439</u>. CHE_EDGAR-ECMWF_2015 consists of 11 global NetCDF files with gridded yearly and monthly upper and lower bounds of uncertainties in % and kg·m⁻²·s⁻¹ for each ECMWF group and their sum, and 1 Excel file with 16 spreadsheets with the same information listed per country (metadata, emissions,
- 40 uncertainties, statistical parameters).Bringing together the different pieces of the puzzle of a very different nature (measurements, reported statistics and models) it is of utmost importance to know their level of confidence and boundaries well.

Inversions disaggregate the variation in observed atmospheric CO_2 concentration to variability in CO_2 emissions by constraining the regional distribution of CO_2 fluxes, derived either bottom-up from statistics or top-down from observations.

- 45 The level of confidence and boundaries for each of these CO_2 fluxes is as important as their intensity, though often limited for bottom-up anthropogenic CO_2 emissions. This study provides a postprocessing tool CHE UNC APP for anthropogenic CO_2 emissions, to help assessing and managing the uncertainty of the different emitting sectors. The postprocessor is available under [10.5281/zenodo.5196190]. Recommendations are given for regrouping the sectoral emissions, taking into account their uncertainty instead of their statistical origin, for addressing local hot spots, for the treatment of sectors with
- 50 small budget but uncertainties larger than 100 %, and for the assumptions around the classification of countries based on the quality of their statistical infrastructure. This tool has been applied on the EDGARv4.3.2_FT2015 dataset, resulting in 7 input grid-maps with upper and lower uncertainty range for the European Centre for Medium-Range Weather Forecasts Integrated Forecasting System. The dataset is available under https://doi.org/10.5281/zenodo.3967439. While the uncertainty of most emission groups remains relatively small (5-20 %), the largest contribution (usually over 40 %) to the total
- 55 uncertainty is determined by the OTHER group (of fuel exploitation and transformation but also agricultural soils and solvents) at global scale. The uncertainties have been compared for selected countries to those reported in the inventories submitted to the United Nations Framework Convention on Climate Change and to those assessed for the European emission grid-maps of the Netherlands Organisation for Applied Scientific Research. Several sensitivity studies are performed to check: 1) the country dependence by analysing the impact of assuming either a well- or less well-developed statistical
- 60 infrastructure, 2) the fuel type dependence by adding explicit information for each fuel type used per activity from the Intergovernmental Panel on Climate Change, and 3) the spatial source distribution dependence by aggregating all emission sources and comparing the effect against an even redistribution over the country. The third impact highlights the importance of spatial mapping.

1 Introduction

- 65 Carbon dioxide (CO₂) is the most abundant greenhouse gas (GHG) (NOAA, 2019) contributing to the climate change. This study focuses on anthropogenic (man made) long cycle carbon CO₂ emissions (i.e. emissions from fossil fuel use and industrial processes: cement production, carbonate use of limestone and dolomite, non energy use of fuels and other combustion, chemical and metal processes, solvents, agricultural liming and urea, waste and fossil fuel fires (Janssens-Maenhout et al., 2019)), that occur on top of an active natural carbon cycle, and generation of a reliable uncertainty band globally for different emission types that can be used in Earth system modelling and data assimilation.
- The CO₂-growth rate varies from year to year with a tendency toward higher growth rates since the early 2000s. The added CO₂-has a long life time and only a portion of it transfers each year from the atmosphere to the oceans and to vegetation on land. The atmosphere exchanges carbon mainly between: (i) the terrestrial biosphere impact on growth rate through deforestation and other forms of land management; (ii) the oceans impact on growth rate through marine ecosystems
- 75 implications due to CO_2 in the form of carbonic acid absorption in surface waters and their mix with deep ocean waters; (iii) the fossil fuels and cement and other CO_2 process emissions — when around 1920 fossil fuel burning became the dominant source of anthropogenic emissions to the atmosphere, with a clear increase of 91 ppm in the past six decades (from 316 ppm in 1959 till 407.4 ± 0.1 ppm in 2018), according to NOAA (2019).

Accurate assessment of anthropogenic carbon dioxide (CO₂) emissions is important to better understand the global carbon

- 80 cycle. Efforts towards a global anthropogenic CO_2 monitoring and verification support capacity as described by Janssens-Maenhout et al. (2020), rely on atmospheric modelling and atmospheric observations (in-situ from-e.g., for example, the Integrated Carbon Observation System, ICOS, air-borne from-e.g., for example, aircraft campaigns, or space-borne from-e.g. the Orbiting Carbon Observatory, OCO-2, and the Greenhouse gases Observing Satellite, GOSAT). All measurements are assimilated by global tracer transport models to infer atmospheric CO_2 changes, or by flux inversion systems to estimate the
- 85 large-scale surface CO₂ fluxes. <u>The European Centre for Medium-Range Weather Forecasts (ECMWF)</u>, for example, applies both inverse modelling and direct modelling of global concentrations of CO₂ in the atmosphere assimilating several types of observations.

The global transport models require an initial best estimate of the CO_2 emission fields with uncertainties, the so-called "prior information-". The intensity of the emission fields is corrected through minimization of the difference between the modelled

- 90 and measured concentration values for CO₂. The uncertainty of these corrected CO₂ fluxes based on inverse modelling will be lower with the increase of CO₂ observations and its accuracy. The disentanglement of the fossil CO₂ emissions from the total atmospheric CO₂ emissions remains challenging, e.g.. For example in 2018 total anthropogenic CO₂ concentrations (5.4 \pm 0.4 ppm) represented only 1.3 % of the global atmospheric CO₂ concentration (407.4 \pm 0.1 ppm) (Friedlingstein et al., 2019), which states the need for a high accuracy of measurements (\geq 1.0 %).
- 95 Emission fields are often supplied through emission inventories. Bottom-up emission inventories start from human activity statistics-and-emission. Emission factors are defined for each activity and provided at international or country level (e.g.

National greenhouse gas Inventory Report, NIR). Such bottom-up inventories need to be gridded and characterised with uncertainties in order to represent a prior data set useful for numerical modelling. Table 1 shows examples of most commonly used global gridded CO_2 emission datasets, for more details see Andrew (2020), Janssens-Maenhout et al. (2019,

100 Table 3) and), Cong et al. (2018, Table 1). and Jones et al. (2021).

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There is strong evidence to suggest fixed annual emissions do not represent sufficient temporal variability, for example, natural gas consumption has two seasonal peaks, with consumption patterns predominantly driven by weather — the largest peak occurs during winter due to low temperatures when natural gas is used more to heat residential and commercial spaces, the smaller peak occurs during summer due to high temperatures when natural gas, coal or petroleum fired generators are used to generate more electric power for air conditioning (Bradley, 2015; Comstock, 2020). New version of EDGAR v5.0 is addressing high temporal disaggregation of emissions (Crippa et al., 2020).

Name			Source	
Carbon Dioxide	Spatial: 1.0°×1.0°	1751-2013	Use population density to disaggregate emissions,	Andres et al., 1996;
Information Analysis	Temporal: annual,		the mass-emissions data based on fossil-fuel	Andres et al., 2016
Center (CDIAC)	monthly		consumption estimates. Provide gridded annual	
	Sectoral: 1		and monthly uncertainty estimates for 1950-2013.	
Open-Data Inventory	Spatial: 1×1 km ² ,	1979-2018	First introduced the combined use of nightlight	Oda and Maksyutov,
for Anthropogenic	0.1°×0.1°		data and individual power plant emission/	2011; Oda et al. 2018;
Carbon dioxide	Temporal: monthly		location profiles.	ODIAC, 2020
(ODIAC)	Sectoral: 6			
Emissions Database for	Spatial: 0.1°×0.1°	1970-(year-1)	Based on international statistics, covers all IPCC	Janssens-Maenhout et
Global Atmospheric	Temporal: annual,		(2006) reporting categories, consistent	al., 2019
Research (EDGAR)	monthly		methodology applied to all the world countries.	
	Sectoral: 26			
Fossil Fuel Data	Spatial: 0.1°×0.1°	1997-2012	Provide gridded posterior uncertainty (version	Asefi-Najafabady et
Assimilation System	Temporal: annual		2.2); in addition, provide monthly, weekly, and	al., 2014
(FFDAS)	Sectoral: 2		hourly fractions from annual CO ₂ emissions.	
Community Emissions	Spatial: 0.1°×0.1°	1750-2014	Provide emissions of CO ₂ and other GHGs and	Hoesly et al., 2018
Data System (CEDS)	Temporal: annual,		pollutants <u>.</u>	
	monthly			
	Sectoral: 55			
Peking University Fuel	Spatial: 0.1°×0.1°	1960-2014	By request provide daily emissions and the results	Chen et al., 2016; Liu
combustion inventory	Temporal: monthly		of Monte Carlo simulation-based uncertainty	et al., 2015
(PKU-FUEL)	Sectoral: 6		analyses_	
Global Carbon Budget	Spatial: 0.1°×0.1°	1959-2018	National GHG inventories reported to UNFCCC	Jones et al. (2021)
Gridded Fossil	Temporal: monthly		are used for the GCP dataset, that is gridded with	
Emissions Dataset	Sectoral: 28		predefined grid-maps following EDGARv4.3.2	
(GCP-GridFED)			spatial distribution proxies; also provide gridded	
			sectoral uncertainties	

Table 1: Examples of global gridded anthropogenic CO2 emission bottom-up datasets

110 Though there are global anthropogenic emission gridded datasets, most of them have scarce evaluation of uncertainties, which needs enhancement with the relative errors for sector specific country totals and the uncertainties in trends with the appropriate probability density functions. Only <u>3Only three</u> datasets from Table 1 provide uncertainty estimates, namely CDIAC, FFDAS and PKU-FUEL. CDIAC uncertainties have no sectors and include contributions from the tabular fossil

fuel CO₂ emissions (assigned per 7 country types, values are constant over time), geography map (power plant location), and 115 population map (has details both in time and space, isand used to distribute fossil fuel CO_2 emissions). Population map uncertainty strongly dominates in the generated gridded fossil fuel CO₂ uncertainties (Andres et al., 2016). CDIAC uncertainties have no sectoral distribution and are presented on 1.0°×1.0° grid. FFDAS provides only posterior uncertainties, which are based on a model inversion. These posterior uncertainties could be used as prior uncertainties for separate inversion systems, however, this would make the characterisation of uncertainty more complex if there were 120 similarities in the model and observations used. PKU-FUEL uncertainty estimates of CO₂ emission maps, associated with uncertain fuel data and uncertain activity data in the spatial disaggregation process, are based on Monte Carlo ensemble simulations. Input data was randomly sampled 1000 times from an a priori normal uncertainty distribution with a certain coefficient of variation: for fuel consumptions from ships/aviation, the sector coefficient of variation is set to be 20 %, for 125 was assigned for each country or subnational unit based on its size to consider uncertainty of spatial fuel data disaggregation (e.g. coefficient of variation for the largest subnational unit of the world Asian part of the Russian Federation is 1000 %).

Emission factor coefficient of variation was constant value of 5 % (% (Marland et al., 2003; Marland et al., 2006; Wang et al., 2013). PKU FUEL uncertainties were heavily based on subjective assumptions and rather; Oda et al, 2019). GCP-GridFED focusses strongly on the fuel disaggregation for the global CO₂ emissions, for which a detailed information of fuel
130 type, which makes is difficult to use for IPCC (2006) reporting categories. assessment of the uncertainty has not yet been

2 Methods

published.

2.1 Purpose and UNFCCC context

Intercomparisons of global greenhouse gas (GHG) emission inventories were carried out (e.g. Cong et al., 2019; Petrescu et

- 135 al., 2020) to better understand discrepancies and missing or lesser-known sources. The United Nations Framework Convention on Climate Change (UNFCCC) experts, reviewing national GHG inventories on a yearly basis, are keen to know which sectors or fuels need extra attention for an inventory that complies with the principles of transparency, accuracy, consistency, completeness and comparability (TACCC-principles). Discrepancies are often related to the different interpretations of definitions or to missing information (statistics and/or measurements). When focussing on global emission
- 140 datasets, which are calculated bottom-up following the Intergovernmental Panel on Climate Change (IPCC) 2006 Guidelines for National Greenhouse Gas Inventories, then the discrepancy using different definitions disappears, while the lack of information becomes strongly apparent for certain regions. More information costs time and effort, when compiling a global dataset in a consistent way. Therefore, it is of paramount importance to prioritise the additional information needs and the weaknesses in the inventory with sources of large uncertainty in intensity or variability.

145 IPCC has been addressing uncertainty from the beginning of its creation. Methodology, data, and data sources in this paper were taken from IPCC (2006) guidelines and its refinements (IPCC, 2019). Also, the assumptions are based on IPCC (2006) and all emissions are considered to be fully uncorrelated, for the calculation of the uncertainty as well as of the covariance matrices.

While the UNFCCC sticks to national inventories, the atmospheric modelling community needs spatially distributed data.

- 150 This adds an extra uncertainty to the emission grid-maps, not evaluated with the uncertainty of the proxy data but which needs an assessment of the representativeness of the selected proxies for distributing the emissions. The point sources, leading to large plumes, were prioritised for being treated separately with more data. These consisted of super power plants, which are defined as a large power plant or a group of closely located power plants (operating at maximum capacity and availability), causing CO₂ plumes from a single grid cell with a CO₂ flux $\geq 7.9 \cdot 10^{-6}$ kg·m⁻²·s⁻¹. According to expert
- 155 knowledge, the upper bound of annual uncertainty for super power plants is not larger than +3.0 %, whereas for small plants whose operation is decided on day-to-day needs, this can reach up to +15.0 %. In this paper, 30 grid-cells of 0.1°×0.1° from 12 countries were identified, representing these super power generators (896.7 Mt of the energy sector) and including large plants from China, Russia and India (for the detailed ranking of the power plant sites in function of their emission intensity, refer to the Supplementary Information, section S.1). The power plant coordinates were checked, to avoid the need for an
- 160 uncertainty related to their positioning. The remaining power plants (not super power generators), over 30000, could not be checked to the same extent and therefore are recommended in a second emission group.

2.2 Generating uncertainty input for transport models

The uncertainty calculation methodology and initial uncertainty values (i.e. activity data and emission factor uncertainties per CO₂-emitting activity) are both taken from IPCC (2006) and its refinements (IPCC, 2019). The following terminology is

165 <u>used to ease the explanation: "activity" – IPCC (2006) activities which result in anthropogenic CO₂ emissions in the yearly budget (a long-cycle carbon); "sector" – combination of different activities that are measured/reported together (that have emission budget data); "group" – combination of different "sectors", that have emission budget data, purely for modelling/comparison needs.</u>

In general, uncertainties are calculated in three steps: (i) "sector" uncertainties (based on emission factors and activity data

170 <u>uncertainties</u>), (ii) annual grouped uncertainties, and (iii) monthly grouped uncertainties. By default, all calculations are performed separately for upper and lower half-ranges of uncertainties and "sector"/"group" combined uncertainties, where upper and lower uncertainty half-ranges are in percent.

2.2.1 Calculating "sector" uncertainties

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The initial 92 IPCC (2006) activity uncertainties are combined into "sectors" for which the user has emission budget data¹, following Eq. (1) and Eq. (2):

$$UC_{activity_i} = \sqrt{EF_{activity_i}^2 + AD_{activity_i}^2}$$
(1)

where combined uncertainties $UC_{activity, i}$ per activity *i* were calculated using uncertainties for emission factors $EF_{activity, i}$ and activity data $AD_{activity, i}$ in percent provided in IPCC (2006) and its refinements (IPCC, 2019);

$$UC_{sector_j} = \sqrt{UC_{activity_1}^2 + UC_{activity_2}^2 + \dots + UC_{activity_n}^2}$$
(2)

180 where combined uncertainties UC_{sector j} per "sector" *j* were calculated with the error propagation method, taking into account particular for that "sector" activity combined uncertainties UC_{activity 1}, UC_{activity 2},..., UC_{activity n} used in percent.

2.2.2 "Group" annual uncertainties

This concerns the further grouping of the combined IPCC (2006) "sectors" according to the user needs into "groups" and calculation of "group" yearly uncertainties. Usually, there are computational restrictions for operational modelling: the

185 number of emission input fields read by the model can't be too large and some "sectors" need to be merged. In addition, instantaneous local emission data as an aggregated total might be rather uncertain and hard to evaluate for different emission types all over the world. IPCC (2006) and its refinement (IPCC, 2019) provide the best possible information on how certain emissions are on an annual national level.

"Sector" uncertainties have to be adjusted to consider a country's statistical system development level and its yearly

190 emission budget, log-normal distribution of non-negative emissions, and then further combined into "group" uncertainties for modelling/comparison purposes in the following way (by default all calculations are performed separately for upper and lower half-ranges of uncertainties):

$$FC_{sector_{j}} = \left[\frac{-0.7200 + 1.0921 \cdot UC_{sector_{j}} - 1.63 \cdot 10^{-3} \cdot UC_{sector_{j}}^{2} + 1.11 \cdot 10^{-5} \cdot UC_{sector_{j}}^{3}}{UC_{sector_{j}}}\right]^{2},$$
(3)
$$\left(UC_{sector_{j}}\right)_{corr} = \begin{cases} UC_{sector_{j}} \cdot FC_{sector_{j}}, 100\% \leq UC_{sector_{j}} \leq 230\% \\ UC_{sector_{j}}, UC_{sector_{j}} < 100\% \cup UC_{sector_{j}} > 230\% \end{cases}$$
(4)

195 where corrected uncertainties $(UC_{sector j})_{corr}$ per "sector" *j* were calculated to take into account large combined uncertainty (100 % $\leq UC_{sector j} \leq 230$ %), underestimation by the error propagation method in comparison to a Monte Carlo simulation, correction factor $FC_{sector j}$ is computed based on Frey (2003), also log-normal adjustment of the emission distribution is computed based on Frey (2003) as detailed in the Supplementary Information, section S.3;

¹Often, emission budgets are provided not per IPCC (2006) activity but for several activities together (usually due to measuring/reporting limitations), for which the user then needs to assume a lump sum activity, emission factor and uncertainties of those.

$$UC_{group,k} = \frac{\sqrt{\left[\left(UC_{sector,1}\right)_{arr}f_{group,k}^{-1} + f_{group,1}^{-1}\right]^{2} + \left(\left[\left(UC_{sector,2}\right)_{arr}f_{group,k}^{-1} + f_{group,k}^{-1} + f_{group,k$$

			GR	OUP 1				GR	OUP k		GROUP C			
		SECT	OR 1		SECTOR 2			SECTOR j			SECTOR B			
	ACTIV	ITY 1	ACTIV	ITY 2	ACTIVITY 3		ACTIVITY i		:		ACTIVITY A			
Uncertainty per ACTIVITY	Emission Factor ₁	Activity Data ₁	Emission Factor ₂	Activity Data ₂	Emission Factor ₃	Activity Data ₃		Emission Factor _i	Activity Data _i			Emission Factor _A	Activity Data _A	
peractiviti	Combi	ined ₁	Combi	ined ₂	Comb	ined ₃		Comb	ined _i			Combi	ined _A	
	Combined ₁			Combined ₂		Combined _j			Combined _B					
Uncertainty per SECTOR		Corrected for large values ₁			Corrected for large values ₂		Corrected for large values _j			Corrected for large values _B				
peroteron	LogarithmicLogarithmictransformation1transformation2						Logarithmic transformation _i				Logarithmic transformation _B			
Uncertainty			Com	bined ₁				Combined _k			Combined _c			
Uncertainty per GROUP	Parameters of logarithmic distribution ₁				Parameters of logarithmic distribution _k			Parameters of logarithmic distribution _c						

Figure 1: Yearly uncertainty calculation simplified roadmap.

2.2.3 "Group" monthly uncertainties

225 The "group" monthly uncertainties are calculated starting from the yearly uncertainties, which can provide a more appropriate variation than the yearly timescale for operational modelling. In this way, yearly "sector" uncertainties are adjusted to represent monthly variability (no correlation between months is assumed), and further combined into "group" monthly uncertainties, by means of the following four steps:

the same steps as for annual uncertainty calculation are used but based on monthly emission budgets (i.e.
 uncertainties for IPCC activities are combined to "sectors" with the error propagation method, corrected for systematic

underestimation by the error propagation method, and adapted to have log-normal distribution;

2) the correlation α (an uncertainty boosting parameter) between yearly and monthly uncertainties is based on an analysis of the variations over the different months following Eq. (11). It's computed to enhance obtained monthly uncertainties as they are the same or even smaller than the yearly ones, because empirical equations applied use emission
 235 budgets, which are smaller for individual months compared to the yearly values:

 $(E_{YEAR} \cdot UC_{YEAR})^{2} = \alpha^{2} \cdot ((E_{MONTH1} \cdot UC_{MONTH1})^{2} + (E_{MONTH2} \cdot UC_{MONTH2})^{2} + \dots + (E_{MONTH12} \cdot UC_{MONTH12})^{2})_{(11)}$ where *E* and *UC* correspond to "sector" emission budget and uncertainty in kilotonne and % respectively, <u>YEAR,MONTH1,MONTH2,...,MONTH12</u> – yearly and monthly (January, February, ..., December) values. Eq. (11) is based on the rule for combining uncorrelated uncertainties under addition of the error propagation equation (see Eq. (5)) and the assumption that each month's uncertainty should be enhanced (boosted) by the same value;

3) the prior yearly "sector" uncertainties are multiplied by the boosting parameter (specific per country and emission "sector") and the results are used as a first guess of prior month "sector" uncertainties;

- 4) the calculation steps 1) to 3) are iterated to find the best boosting parameter as the best fit between yearly and combined 12-month uncertainties, with the incremental step below a given acceptable threshold from Eq. (11) for each
- 245 <u>country and emission "sector". With this optimum boosting parameter, monthly uncertainties per "sector" are calculated and</u> <u>then merged into "groups", with a log-normal distribution of CO₂ emissions.</u>

Detailed information on each Unix shell script included in the anthropogenic CO₂ emission uncertainty calculation tool CHE UNC APP (Choulga et al., 2021) is provided in the Supplementary Information, section S.4.

2.2.4 Remarks about the fuel dependence and assumptions concerning correlation

- 250 It should be noted that IPCC (2006) provide default emission factor values for different fuels in transport-related activities (e.g. railways, aviation, etc.). In this study, we focus on fossil emissions (from fossil fuel combustion, use and production, and process emissions from cement production and others such as glass, chemicals, urea) and we distinguish between point sources and sources with wider spatial distribution. The scope of this research is to generate a reliable uncertainty band on 0.1°×0.1° grid with global coverage based on emission type for the yearly and monthly emission budgets, that are the
- 255 composite of anthropogenic fossil fluxes, and that are aligned with updated IPCC requirements. Uncertainty characterisation is key for optimally combining the bottom-up inventories with the top-down data assimilation. Detailed fuel consumption information per IPCC activity, that result in a long-cycle carbon, was not available and instead the most typical and consumed (common) fuel type (or its emission factor value) was used:
 - Aviation cruise (1.A.3.a CRS), climbing & descent (1.A.3.a CDS), and landing & take off (1.A.3.a LTO) jet
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- Road transportation (1.A.3.b), and pipelines, off-road transport (1.A.3.e) most typical emission factor uncertainty;
- Shipping (1.A.3.d) composition of 80 % diesel and 20 % residual fuel oil;
- Railways (1.A.3.c) diesel.

kerosene;

It should also be noted that some uncertainty ranges for emission factors and/or activity data in IPCC (2006) and its refinements (IPCC, 2019) are not symmetrical and have higher uncertainty values for the lower bound than for the upper bound (or vice a versa), due to input from expert knowledge or available in-situ data, which then leads to the same pattern in final prior uncertainty bounds.

It should finally be noted that according to the IPCC (2006), all anthropogenic CO₂ emissions are assumed to be fully uncorrelated, hence the prior error correlations between grid-cell emissions from the same "sector"/"group" should be assumed negligible if country- and/or sector-specific information is lacking.

<u>3 Uncertainty calculation application</u>

The method explained above has been applied to the EDGARv4.3.2_FT2015 dataset to prepare prior uncertainty information for the ECMWF Integrated Forecasting System (IFS) model.

3.1 Data input

- 275 In this studyexample, 2015, the year of the Paris Agreement and reference for several Nationally Determined Contributions, is chosen as a base year to analyse anthropogenic CO₂ budgets (i.e. global, regional, national) from different sources (i.e. global statistics, national reports), benefitting the availability of observations (both in-situ ground and space-borne) as well as reported and verified emission inventories. Global CO₂ emissions from fossil fuel and industrial processes such as cement production reached a total of 4.6 ppm in 2015 according to EDGAR inventory version 4.3.2_FT2015 (Olivier et al., 2016a).
- 280 The use of energy represents by far the largest source of emissions (89 % share globally), and in particular the energy industry sector (38 % share) (including both combustion and fugitive gas releases from use but also production, processes, transmission and storage of fuels for energy and heat generation). Another reason for choosing 2015 is that it's the year of the Paris Agreement and the reference year for several Nationally Determined Contributions (NDCs). Countries have submitted their pledges to the United Nations (UN), setting out how far they plan to reduce their GHG emissions NDCs
- 285 (CarbonBrief, 2020). Yet concentrations are still growing. In 2015, the average concentration of CO₂ (399 ppm) was about 40 % higher than in the mid–1800s, with an average growth of 2 ppm/yr in the last ten years. Furthermore, according to JRC 2019 Report (Crippa et al., 2019) between 2015 till 2018, just in three years global CO₂ emissions have raised by 4.3 % (0.2 ppm/yr), of which the international component of CO₂ emissions (shipping and aviation bunker fuel) has even raised by 6.3 % (0.01 ppm/yr).
- Following the Intergovernmental Panel on Climate Change (IPCC) __(2006 Guidelines for National Greenhouse Gas Inventories) and revised information from its 2019 Refinementsrefinements (IPCC-TFI, 2019) we start), starting from the global fossil CO₂ grid-maps of EDGAR inventory versions 4.3.2 (Janssens-Maenhout et al., 2019) and 4.3.2_FT2015 (Olivier et al., 2016a), for 2012 and 2015 respectively, and derive an updated emission dataset as prior input to the ECMWF model: CHE_EDGAR-ECMWF_2015-(CHE stands for the CO₂-Human Emissions project (CHE,² (Choulga et al., 2020)).
- We improve the) is derived. The EDGARv4.3.2 dataset is improved by correcting the allocation of the autoproducers (autoproducers to the manufacturing sector instead of the energy sector. Autoproducers are defined by International Energy Agency (IEA) and include the energy (electricity and heat) generated by an industry for its own use, mostly for the manufacturing) to the manufacturing sector instead of the energy sector and by adding the diffusive CO₂ emissions from coal mines. We then aggregate the sectors in 7 emission groups while tracking 232 countries separately. Uncertainties are
- 300 calculated per country and sector considering the most typical fuel type using the error propagation method of the IPCC (2006) guidelines. According to the IPCC (2006) guidance all emissions are considered to be fully uncorrelated; this assumption is further used to calculate uncertainty and covariance matrices. The country based uncertainties and the share to the total uncertainty are presented for the 7 ECMWF emission groups, with calculations based on 20 EDGAR sectors for two distinct country types with well and less well developed statistical infrastructures. While the uncertainty of most groups (i.e.

² CHE stands for the CO2 Human Emissions project (CHE, 2020).

305 power industry, combustion for manufacturing, and road transport) remains small (5-20 %), the largest contribution (over 40 %) to the total uncertainty is determined by rather small but relative uncertain (more than 100 %) sectors (i.e. non energy use of fuels, chemical processes, fuel exploitation, and coal production) emissions.

This paper is organised as follows. Section 2 describes the data sources and includes the description of the anthropogenie CO₂ emission datasets used to calculate emission uncertainties, data pre processing, emission sectors and groups, and

310 geographical treatment of emissions. Section 3 discusses the uncertainty calculation methodology applied to the datasets, to calculate both yearly and monthly uncertainties. Section 4 provides details on the newly generated dataset. National sectorial emission budgets are compared in Section 5. The main results, a discussion and further research guidance are covered in the conclusion in Section 6. This paper also has Supplementary Information with details on methods and assumptions used.

2 Data

315 **2.1 Update of fossil CO₂ emissions as input for the ECMWF model**

Main requirements for datasets in order to be used in global numerical models are being global and gridded, and preferably with continuous update. In this study it was decided to use EDGARv4.3.2 (and EDGARv4.3.2_FT2015) because it is based on international statistics, mainly IEA data, has a unique global geo coverage with 228 countries/regions and continuous updates of the time series. The most relevant activity data for both EDGARv4.3.2 and CHE_EDGAR_ECMWF_2015 are the

- 320 energy statistics from IEA (2014), which has been corrected for few outliers and for the revised Chinese coal statistics of 2015. For the update from 2012 to 2015 we used the fast track approach of Olivier et al. (2016b), with IEA (2016) energy statistics and BP (2017) statistics. EDGAR distributes anthropogenic emissions for each source category over a uniform, global 0.1°×0.1° grid defined with lower left coordinates and provides annual and monthly global emissions grid maps. The bottom up emissions calculation methodology and emission factors, either defaults recommended by IPCC (2006) guidelines
- 325 or region specific ones justified by scientific evidence, are consistently applied to all countries in order to achieve comparability and full transparency.

We focus on long cycle carbon CO₂ and therefore consider the CO₂ from fossil fuel use (combustion and other use of 42 fossil fuels) and from industrial processes (cement production, carbonate use of limestone and dolomite, non-energy use of fuels and other combustion, chemical and metal processes, solvents, agricultural liming and urea, waste and fossil fuel fires).

330 Excluded are consumption of biofuels and short-cycle biomass burning (such as agricultural waste burning), large-scale biomass burning (such as forest fires, savannah burning, woodland and peatland fires) and carbon emissions/removals of land use, land use change and forestry (LULUCF)³. Based on the Global Carbon Budget 2018 findings this sector showed no significant trend since 1960s, only high year-to-year variability and high uncertainty (Bastos et al., 2020; Le Quéré et al.,

³ Following the UNFCCC national inventory reporting guidelines, emissions of biofuel combustion are only a memo item and have to be reported under the LULUCF sector. Together with all short cycle carbon emissions they are excluded from this study.

2018; Arneth et al., 2017). We excluded also the fossil fuel fires, because we focus only on 2015, for which the Kuwait oil

- 335 fires of 1991 are of no importance and the coal mine fires data are considered to be very uncertain.
- Starting from EDGARv4.3.2_FT2015, the following updates were considered necessary for the derivation of the CHE_EDGAR-ECMWF_2015 dataset. Firstly, there was a need to reallocate the part of autoproducers in the energy sector to the manufacturing industry in line with UNFCCC reporting. The autoproducers' energy generated and used for industrial manufacturing was added to the manufacturing sector (causing an increase of 18 %) and taken away from the energy sector
- 340 (leading to a decrease of 8 %). The reallocation of the autoproducers part was done using the energy statistics reported by every country separately (IEA, 2016) but the correction remained limited to 30 % of the national total energy sector. More details are given in the Supplementary Information, section S.1.

Secondly, super power plants were considered to be treated separately, because they are expected to operate at full capacity with maximum availability. Super power plants are defined in this study as a large power plant or a group of closely located

- 345 power plants causing CO₂ plumes from a single grid cell with a CO₂ flux ≥ 7.9·10⁻⁶ kg·m⁻²·s⁻¹. According to expert knowledge the upper bound of uncertainty for super power plants is not larger than +3.0 %, whereas for small plants which operate based on day to day needs, this can reach up to +15.0 %. Currently 30 grid cells from 12 countries represent energy generated by the super power plants (7.1 % or 896.7 Mton of the remaining energy sector after autoproducers part separation 12705.5 Mton). Top 3 countries that produce energy using super power plants are China, Russia and India. For the detailed ranking of the power plant sites in function of their emission intensity, we refer to the Supplementary Information, section
- 350 ranking of the power plant sites in function of their emission intensity, we refer to the Supplementary Information, section S.1.

Finally, an<u>. An</u> extra emission source of fugitive CO_2 from coal mines <u>wasis also</u> added, following the recommendations from IPCC-TFI (2019). Even though this emission source is not that large globally, usually the coalseam gas is composed dominantly from of methane, (CH₄), but in some coalmines (in Australia, and also in Brazil) seam gas consists predominantly

(> 95 %) fromof CO₂ (Beamish and Vance, 1992), leading to significant atmospheric CO₂ concentration increases. An additional map for CHE_EDGAR-ECMWF_2015 with coal mining emissions from underground mines has been generated, following the IPCC-TFI (2019) default values and the coal mining activity of the methane (CH₄) emission grid-maps from hard and brown coal production of <u>in</u> EDGARv4.3.2. More details are given in (for more information refer to the Supplementary Information, section S.2, in which Table S3 lists all differences between EDGARv4.3.2_FT2015 and CHE_EDGAR_ECMWF_2015.

The detailed EDGARv4.3.2 spatial distribution is used for mapping the updated 2015 emission values (Janssens Maenhout et al. (2019) provide all special details on how emissions are spatially distributed and what proxies are used for that in EDGARv4.3.2). The relative changes per sector, fuel type and country). For the update from 2012 to 2015 are then applied on the EDGARv4.3.2 reference maps to obtain EDGARv4.3.2_FT2015. For non-energy use of fuels, chemical processes,

365 and solvents and products use we used directly the EDGARv4.3.2 maps. Also, the CO₂ emission maps from coal production are based on the 2012 maps of CH₄ from EDGARv4.3.2. Gridded monthly multiplication factors are obtained from 2010 monthly gridded emissions and applied to the final set of yearly emission maps of CHE_EDGAR_ECMWF_2015.

2.2 Aggregation of CO₂ emission groups for the ECMWF model

- 370 EDGARv4.3.2_FT2015 (as well as EDGARv4.3.2) has 20 global maps with anthropogenic long cycle carbon CO₂ flux values for energy, fugitives, industrial processes, solvents and products use, agriculture and waste involved sectors. In this study these sectors had to be grouped for the use of global flux inversion and ensemble perturbation systems. Grouping was done keeping in mind possible future evolution of present systems and sector common features: the fast-track approach of Olivier et al. (2016b) is used. The initial 92 IPCC activity type (point sources, 3D field, etc.), amount of knowledge for the
- 375 activity (uncertainty value), geographical distribution (e.g. over urban areas only), size of sector covariance matrix (computationally affordable size for the inversion system of the ECMWF model is covariance matrix of 7×7).uncertainties are combined into 20 EDGAR "sectors" for two distinct country types with well- and less well-developed statistical infrastructures (i.e. country's ability to register different emissions tabulate even very small emissions or only major ones, respectively). For the input to the IFS model the emission "sectors" are grouped in 7 "groups", with one "group" devoted to
- 380 <u>super power plants.</u> Table 2 shows <u>additionalactivity and "sector"</u> grouping-of 20 EDGAR sectors into 7 ECMWF groups, and emission budget difference between EDGARv4.3.2_FT2015 and CHE_EDGAR-ECMWF_2015 datasets due to reallocation of the autoproducers from the energy sector (-8 %) to the manufacturing sector (+18 %), and due to the extra emission source of diffusive coal mine CO₂.

385 Table 2: Grouping of anthropogenic long-cycle carbon CO₂ emission <u>EDGAR</u>-<u>"sectors"</u> into <u>ECMWF</u>-<u>"groups</u>,", note provides main information and typical fuel type, global emission budgets for 2015 in <u>MtonMegatonne</u> provides values for EDGARv4.3.2_FT2015 and CHE_EDGAR-ECMWF_2015; *italics* – values with biggest differences, * – values that were replaced from EDGARv4.3.2

Nº	ECMWF	IPCC (2006) activities per	Note	Emission budget 2015, <u>MtonMt</u>				
JIS	group "Group" <u>name</u>	EDGAR- <u>"</u> sector <u>"</u>	- <u>sector</u>		CHE_EDGAR- ECMWF_2015			
1	ENERGY_S	1.A.1.a (subset)	Power industry (without autoproducers): super emitting power plants	13704.0	896.7			
2	ENERGY_A	1.A.1.a (rest)	.1.a (rest) Power industry (without autoproducers): standard emitting power plants		11671.6			
	_	4.C	Solid waste incineration	137.2	137.2			
		1.A.2	Combustion for manufacturing (including autoproducers)	6182.8	7320.4			
		2.C.1, 2.C.2	Iron and steel production	233.6	233.6			
3	MANUFACTURING	2.C.3, 2.C.4, 2.C.5, 2.C.6, 2.C.7	Non-ferrous metals production	91.4	91.4			
		2.D.1, 2.D.2, 2.D.4	Non energy use of fuels	24.7*	24.6			
		2.A.1, 2.A.2, 2.A.3, 2.A.4	Non-metallic minerals production	1748.8	1749.0			
		2.B.1, 2.B.2, 2.B.3, 2.B.4, 2.B.5, 2.B.6, 2.B.8	Chemical processes	678.8*	677.0			
4	SETTLEMENTS	1.A.4, 1.A.5.a, 1.A.5.b.i, 1.A.5.b.ii	Energy for buildings	3321.9	3322.7			

		1.A.3.a_CRS	Aviation cruise; typical fuel: jet kerosene	412.2	412.2
5	AVIATION	1.A.3.a_CDS	Aviation climbing & descent; typical fuel: jet kerosene	305.5	305.5
		1.A.3.a_LTO Aviation landing & take off; typical fuel: iet kerosene		97.7	97.7
		1.A.3.b	Road transportation; typical fuel: most typical emission factor uncertainty	5530.2	5530.6
6	TRANSPORT	1.A.3.d Shipping; typical fuel: composition of 80 % diesel and 20 % residual fuel oil		819.0	819.1
0	TRANSPORT	1.A.3.c, 1.A.3.e	Railways, pipelines, off-road transport; typical fuel: railways – diesel, off-road transport – most typical emission factor uncertainty	255.2	255.2
		1.A.1.b, 1.A.1.c, 1.A.5.b.iii, 1.B.1.c, 1.B.2.a.iii.4, 1.B.2.a.iii.6, 1.B.2.b.iii.3	Oil refineries and Transformation <u>transformation</u> industry	1917.4	1917.8
7	OTHER	ER 1.B.2.a.ii, 1.B.2.a.iii.2, 1.B.2.a.iii.3, 1.B.2.b.ii, 1.B.2.b.iii.2, 1.B.2.b.iii.4, 1.B.2.b.iii.5, 1.C Fuel exploitation		258.4	258.4
		1.B.1.a	Coal production	0.0	7.0
		3.C.2, 3.C.3, 3.C.4, 3.C.7	Agricultural soils	99.0	99.1
		2.D.3, 2.B.9, 2.E, 2.F, 2.G	Solvents and products use	168.7*	168.3

390 **3-Uncertainty calculation**.<u>2 Model constraints</u>

The operational IFS model is used to provide global CO_2 forecasts using the gridded prior emissions previously described (Agusti-Panareda et al., 2014; Agusti-Panareda et al., 2019). A prototype 4D-Var inverse modelling system is currently under development to monitor anthropogenic CO_2 emission using the IFS. There is also an on-going development to extend the window length beyond 24-hours using an ensemble-based methodology.

- 395 The uncertainties derived for the 7 "groups" described here have been used to generate an ensemble of forecasts for 2015 based on the operational IFS ensemble system (McNorton et al., 2020). This provides a representation of the model uncertainty and an estimation of the expected signal-to-noise ratio for a future inverse modelling system. Random seeds for each "group" and country were applied to the normalised log-normal mean μ^{ln} and standard deviation σ^{ln} to generate emission scaling factors, which were then used for 50 ensemble members.
- 400 Primarily, the derived emission uncertainties presented here are envisaged for use as prior errors within atmospheric inversion frameworks. Aggregation of emission "sectors" into 7 "groups" is required for computational efficiency and to reduce the dimensions of the inverse problem. To resolve collocated emissions, further information is required about spatial correlations and/or co-emitted species (e.g. nitrogen oxides (NO_x)). Within the IFS inversion prototype, the log-normal normalised standard deviation outlined in the previous section is used to provide the uncertainty values to prevent negative scaling factors.

3.3 CHE EDGAR-ECMWF 2015 output

The new CHE_EDGAR-ECMWF_2015 dataset with anthropogenic fossil CO_2 emissions and their uncertainties was compiled and tested at ECMWF. The fossil CO_2 emissions include all long-cycle carbon emissions from human activities, such as fossil fuel combustion, industrial processes (e.g. cement) and products use, but excludes emissions from land-use

- 410 <u>change and forestry.</u> Human CO₂ emission inventories were processed into gridded 0.1°×0.1° resolution maps to provide an estimate of prior CO₂ emissions, aggregated in 7 main emissions "groups": 1) energy production by super-emitters, 2) energy production by standard-emitters, 3) manufacturing, 4) settlements, 5) aviation, 6) other transport at ground level and 7) others, with an estimation of their uncertainty and covariance. Aggregation of the IPCC activities and "sectors" into "groups" was based on similarities between the magnitude of uncertainty, the spatiotemporal correlation and co-emission
- 415 factors of each "sector". It is assumed that each emission "group" is fully correlated with itself and fully uncorrelated with any other "group" (only diagonal values of the 7×7 "groups" covariance matrix for the atmospheric transport model are non-zero and equal to log-normal variance). The CHE_EDGAR-ECMWF_2015 data are freely available https://doi.org/10.5281/zenodo.3967439, and consist of 11 grid-maps in NetCDF format and one Excel file with information on anthropogenic CO₂ emissions and their uncertainties. For detailed information on each file see Table 3.

420

Table 3: Detailed information on CHE_EDGAR-ECMWF_2015 data

File	General note	Field / Spreadsheet
	file has 2×8 fields with annual upper	"Lower" - lower uncertainty bound (2.5th percentile of log-normal distribution) for
	and lower uncertainty bounds in	yearly emissions, in percent
	percent per emission "group" and for	"Upper" - upper uncertainty bound (97.5th percentile of log-normal distribution) for
	all "groups" summed together on a	yearly emissions, in percent
ŝ	regular grid with 1800 pixels along the	"Sector" - emission "sector" numerical name. "0" represents emission "group"
ntie	latitude and 3600 pixels along the	ENERGY_S (with IPCC (2006) activity 1.A.1.a (subset)) standing for power industry
tair .	longitude, where values represent	emissions from super emitting power plants; "1" ENERGY A (1.A.1.a (rest), 4.C) -
<u>Uncertainties</u> 0.1.nc	centre of the grid-cell	power industry emissions from standard emitting power plants, & solid waste
<u>0.1</u>		incineration; "2" MANUFACTURING (1.A.2, 2.C.1, 2.C.2, 2.C.3, 2.C.4, 2.C.5, 2.C.6,
er).1		2.C.7, 2.D.1, 2.D.2, 2.D.4, 2.A.1, 2.A.2, 2.A.3, 2.A.4, 2.B.1, 2.B.2, 2.B.3, 2.B.4, 2.B.5,
à C		2.B.6, 2.B.8) – combustion for manufacturing (including autoproducers), & iron and
<u>Upper Lo</u> Percentage		steel production, & non-ferrous metals production, & non energy use of fuels, & non-
ent		metallic minerals production, & chemical processes; "3" SETTLEMENTS (1.A.4,
Uppe		1.A.5.a, 1.A.5.b.i, 1.A.5.b.i) – energy for buildings, residential heating; "4" AVIATION
		(1.A.3.a CRS, 1.A.3.a CDS, 1.A.3.a LTO) – aviation cruise, & climbing and descent,
Annual		<u>& landing and take off; "5" TRANSPORT (1.A.3.b, 1.A.3.d, 1.A.3.c, 1.A.3.e) – road</u>
An		transportation, & shipping, & railways, pipelines, off-road transport; "6" OTHER (1.A.1.b, 1.A.1.c, 1.A.5.b.iii, 1.B.1.c, 1.B.2.a.iii.4, 1.B.2.a.iii.6, 1.B.2.b.iii.3, 1.B.2.a.ii
		(1.A.1.b, 1.A.1.c, 1.A.5.b.m, 1.B.1.c, 1.B.2.a.m.4, 1.B.2.a.m.6, 1.B.2.b.m.3, 1.B.2.a.m, 1.B.2.a.jij.2, 1.B.2.a.jij.3, 1.B.2.b.jij, 1.B.2.b.jij.2, 1.B.2.b.jij.4, 1.B.2.b.jij.5, 1.C, 1.B.1.a.
		<u>1.B.2.a.III.2, 1.B.2.a.III.5, 1.B.2.0.II, 1.B.2.0.III.2, 1.B.2.0.III.4, 1.B.2.0.III.5, 1.C, 1.B.1.a,</u> 3.C.2, 3.C.3, 3.C.4, 3.C.7, 2.D.3, 2.B.9, 2.E, 2.F, 2.G) – oil refineries and transformation
		industry, & fuel exploitation, & coal production, & agricultural soils, & solvents and
		products use; "7" represents all "groups" summed together
		products use, / represents an groups summed together

>	0	file has 2×8×12 fields with monthly	file structure is identical to the file
Monthly Upper Low er Uncertainties	1.n		
r I	0.1	<u>upper and lower uncertainty bounds in</u> percent per emission "group" and for	Annual Upper Lower Uncertainties Percentage 0.1 0.1.nc, but per month (1, 2,, 12
onthly Upper L er Uncertainties	0.1		correspond to January, February,, December)
et:			
nc v	age	regular grid with 1800 pixels along the	
C [t]	ent	latitude and 3600 pixels along the	
er [0]	ICC	longitude, where values represent	
\geq	Pe	centre of the grid-cell	
		file has 3×8 fields with annual	"Sup_lower" - lower uncertainty bound (2.5th percentile of log-normal distribution) for
ties		emissions, and upper and lower	<u>yearly emissions of ENERGY_S, in kg·m⁻²·s⁻¹,</u>
ain		uncertainty bounds in kg·m ⁻² ·s ⁻¹ per	"Sup_upper" - upper uncertainty bound (97.5th percentile of log-normal distribution) for
ert		emission "group" and for all "groups"	yearly emissions of ENERGY_S, in $kg \cdot m^{-2} \cdot s^{-1}$,
nc		summed together on a regular grid	"Sup_flux" – yearly emissions of ENERGY_S, in kg·m ⁻² ·s ⁻¹
	2	with 1800 pixels along the latitude and	"Ene lower", "ene upper", "ene flux" – same, but for ENERGY_A, in kg·m ⁻² ·s ⁻¹
ver 1 +	-	3600 pixels along the longitude, where	"Man lower", "man upper", "man flux" - same, but for MANUFACTURING, in kg·m ⁻
õ c	Ď	values represent centre of the grid-cell	2.s ⁻¹
r Lower I	1.1		"Set lower", "set upper", "set flux" – same, but for SETTLEMENTS, in kg·m ⁻² ·s ⁻¹
Annual Upper Lower Uncertainties	-		"Avi lower", "avi upper", "avi flux" – same, but for AVIATION, in kg m ⁻² s ⁻¹
Up			"Tra lower", "tra upper", "tra flux" – same, but for TRANSPORT, in kg·m ⁻² ·s ⁻¹
al			"Oth lower", "oth upper", "oth flux" – same, but for OTHER, in kg·m ⁻² ·s ⁻¹
nu			"All lower", "all upper", "all flux" – same, but for all "groups" summed together, in
An			An lower, an upper, an nux – same, but for an groups summed together, in $kg \cdot m^{-2} \cdot s^{-1}$
les		file has 3×12 fields with monthly	<u>"Sup lower" – lower uncertainty bound (2.5th percentile of log-normal distribution) for</u>
Int D	0	emissions, and upper and lower	monthly emissions of ENERGY_S, in kg·m ⁻² ·s ⁻¹
rta	.nc	uncertainty bounds in kg·m ⁻² ·s ⁻¹ per	"Sup upper" – upper uncertainty bound (97.5 th percentile of log-normal distribution) for
Sul	0.1	ENERGY S emission "group" on a	monthly emissions of ENERGY_S, in kg·m ⁻² ·s ⁻¹
<u>Monthly Sup Upper</u> Lower Uncertainties	Ξ.	regular grid with 1800 pixels along the	"Sup_flux" – monthly emissions of ENERGY S, in kg·m ⁻² ·s ⁻¹
er Ith	0	latitude and 3600 pixels along the	"Month" - month numerical name, where 1, 2,, 12 correspond to January, February,
0 Mo		longitude, where values represent	, December
		centre of the grid-cell	
<u>Monthly Ene Upper</u> Lower Uncertainties		file has 3×12 fields with monthly	file structure is identical to the file
ii P	0	emissions, and upper and lower	Monthly Sup Upper Lower Uncertainties 0.1_0.1.nc, but with "ene_lower",
e l	.nc	<u>uncertainty bounds in kg·m⁻²·s⁻¹ per</u>	"ene_upper", "ene_flux" fields
DC En	0.1	ENERGY_A emission "group" on a	
75	-	regular grid with 1800 pixels along the	
er th	0		
<u>Month</u> Lower		longitude, where values represent	
		centre of the grid-cell	
<u>Man_Upper</u> incertainties		file has 3×12 fields with monthly	file structure is identical to the file
<u>u</u> U U	0	emissions, and upper and lower	Monthly Sup_Upper_Lower_Uncertainties_0.1_0.1.nc, but with "man_lower",
arts	ň.	uncertainty bounds in kg·m ⁻² ·s ⁻¹ per	"man_upper", "man_flux" fields
Mai	0.1	MANUFACTURING emission	
		"group" on a regular grid with 1800	
<u>Monthly</u> Lower	0	pixels along the latitude and 3600	
UO NO		pixels along the longitude, where	
M		values represent centre of the grid-cell	
H S		file has 3×12 fields with monthly	file structure is identical to the file
Monthly Set Upper Lower Uncertainties		emissions, and upper and lower	Monthly_Sup_Upper_Lower_Uncertainties_0.1_0.1.nc, but with "set_lower",
tai C	nc	uncertainty bounds in kg·m ⁻² ·s ⁻¹ per	"set_upper", "set_flux" fields
cer let	.1.	SETTLEMENTS emission "group" on	
Jnc	0.	a regular grid with 1800 pixels along	
r l	0.0	the latitude and 3600 pixels along the	
we		longitude, where values represent	
L K		centre of the grid-cell	
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file has 3×12 fields with monthly file structure is identical	to			file
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AVIATION emission "group" on a				
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a latitude and 3600 pixels along the				
longitude, where values represent				
integral grid with 1600 pixels along the integral latitude and 3600 pixels along the longitude, where values represent centre of the grid-cell				
	to		the	file
file has 3×12 fields with monthly emissions, and upper and lower uncertainty bounds in kg·m ⁻² ·s ⁻¹ per TRANSPORT emission "group" on a regular arid with 1800 pircle clong the		with		lower",
$\square = 2$ uncertainty bounds in kg·m ⁻² ·s ⁻¹ per "tra upper", "tra flux" fields	,			
표 한 TRANSPORT emission "group" on a				
Latitude and 3600 pixels along the longitude, where values represent centre of the grid-cell				
Iongitude, where values represent				
$\frac{1}{10}$ $\frac{1}{10}$ file has 3×12 fields with monthly file structure is identical	to		the	file
emissions, and upper and lower Monthly Sup Upper Lower Uncertainties 0.1_0.1.nc	e, but	with	"oth	lower",
file has 3×12 fields with monthly emissions, and upper and lower uncertainty bounds in kg·m ⁻² ·s ⁻¹ per oTHER emission "group" on a regular grid with 1800 pixels along the latitude and 3600 pixels along the longitude, where values represent centre of the grid-cell				
= <u>OTHER emission "group" on a regular</u>				
$\int \int \frac{\text{grid with } 1800 \text{ pixels along the}}{1000 \text{ pixels along the}}$				
intitude and 3600 pixels along the				
longitude, where values represent				
1 file has 2x12 fields with monthly file structure is identical	to		the	file
file has 3×12 fields with monthly emissions, and upper and lower uncertainty bounds in kg·m ⁻² ·s ⁻¹ for all "groups" summed together on a regular grid with 1800 pixels along the latitude and 3600 pixels along the longitude, where values represent centre of the grid-cell	to but	with	the "all	file lower".
\square	, Dui	witti	all	_lower ,
= an <u>uncertainty bounds in kg in s tot an</u> <u>an upper</u> , an <u>next nexts</u>				
regular grid with 1800 pixels along the				
\vec{z}				
E longitude, where values represent				
Image: Sector of the grid-cell				
file has 16 spreadsheets with listed "COUNTRY" – ISO Code (3-letter abbreviation of a g	geographi	cal entity	y), Geog	graphical
information per country (metadata, name (name of a geographical entity), Type (develop	ment leve	el of cou	untries s	statistical
emissions, uncertainties, statistical infrastructure, meaning with well-/less well-develope				
parameters) country (dependency, which country geographical en				
information (full name of a geographical entity, and wh	nat territor	ry it occu	upies on	the map
<u>of this study</u>)				
<u>"GROUP" – № (number of anthropogenic CO₂ em</u>				
name), IPCC (2006) activity (IPCC activities that are				
(short explanation of the "group"), Global emission				
- Emissions per group), this uncertainty bounds, // (
assumptions from free, lower and upper uncertainty	/ bounds	Tor cour	innes w	IIII well-
"YEARLY" – ISO Code (3-letter abbreviation of a get	ographica	1 optity)	group	("croup"
name), Budget, kt (yearly anthropogenic CO ₂ emission				
Yess well-developed statistical infrastructures) "YEARLY" – ISO Code (3-letter abbreviation of a gen name), Budget, kt (yearly anthropogenic CO ₂ emission geographical entity), Uncertainty bounds, % (calcul bounds and Budgets yearly uncertainties per "group" HO				
bounds and Budgets yearly uncertainties per "group"				
uncertainties lower/upper/average bounds), Contribution				
(share of each "group" in geographical entities total ye				
is always 100 %), Parameters of log-normal distribut				
distribution is assumed to be log-normal, so addition				
normal mean, log-normal standard deviation and log-normal				
"MONTHLY 01", "MONTHLY 02",, "MONTHL			<u>.</u>	
spreadsheet "YEARLY", but for a month (01, 02,	, 12	correspo	ond to	January,
February,, December)				

3.1 Overview

The IPCC (2006) Guidelines for NIR for fossil CO2- uncertainty calculations and updated IPCC TFI (2019) provide vast

- 425 information about numerous human activities emitting CO₂ and how certain these values are. Use of the IPCC-TFI (2019) permitted to consider the 2019 emission factor and activity data uncertainties for petroleum refining, solid fuel manufacturing, transformation, processing and transport and oil and gas production, which differed significantly from the 2006 defaults. In order to use the same methodology globally and because CO₂ emissions are not technologically dependant, it was decided to omit regional (e.g. Europe) detailed information and use only information required for the most basic and
- 430 simplest (Tier 1) approach for emission reporting. The Tier 1 methodology to estimate CO₂ emissions from fossil fuel combustion follows the concept of carbon conservation (from the fuel combusted into CO₂). Uncertainties for all emission activities, sectors and groups can be derived following two different approaches of IPCC (2006): (Approach 1) propagation of error gives informative results even if the criterion "standard deviation divided by the mean value is less than 0.3" is not strictly met and data still have some correlation. The advantages are that it only needs uncertainty ranges for activity data
- 435 and emission factors, that are provided by IPCC and that it is relatively easy to improve in case of large and asymmetric uncertainties; (Approach 2) Monte Carlo simulation or similar techniques — suitable only if detailed category by category uncertainty information is available and complex calculations can be done. In order to use the same methodology for all world countries/geographical entities (i.e. not needing detailed information for each emission activity) it was decided to use the error propagation method (Approach 1).
- 440 To summarize, the final uncertainties per geographical entity per ECMWF fossil CO₂ emission group are based on: emission budgets calculated from CHE_EDGAR-ECMWF_2015 maps (upgraded combination of EDGARv4.3.2 and EDGARv4.3.2_FT2015), uncertainty default values from IPCC (2006) and IPCC TFI (2019), Tier 1 approach (error propagation method) and the definition of a log normal distribution (needed for non negative anthropogenic CO₂ emissions). It should be noted that all uncertainty calculations were done per country (geographical entity) and only then for comparison
- 445 purposes aggregated to Europe (28 members till end 2019) or global values assuming no correlation following IPCC (2006). Figure 1 shows a simplified scheme of the uncertainty calculation roadmap, followed by a detailed description below on how exactly yearly and monthly uncertainties are calculated.



3.2 Yearly uncertainties

3.2.1 Calculating uncertainty per each IPCC activity

Uncertainties in the emissions per IPCC activity from Table 2 Combined Uncertainties UC_{IPCCi} were calculated using
 uncertainties for emission factors EF_{IPCCi} and activity data AD_{IPCCi} in % provided in IPCC (2006) and IPCC TFI (2019)
 following Eq. (1):

$$UC_{IPCCi} = \sqrt{EF_{IPCCi}^2 + AD_{IPCCi}^2}.$$
(1)

It should be noted that IPCC (2006) and IPCC TFI (2019) provide upper and lower limits of emission factor and activity data, which are not always symmetrical. In order to preserve as much initial information as possible (and not to inflate artificially lower or upper limits of log normal emission distributions) all calculations were performed for upper and lower uncertainty limits separately although it is not required by the Approach 1 methodology. Moreover, IPCC (2006) provide default emission factor values for different fuels in transport-related activities (e.g. railways, aviation, etc.). Detailed fuel consumption information per each IPCC activity that result in long-cycle carbon was not available and it was decided to use

the most typical and consumed (common) fuel type (its emission factor value). Table 2 shows the most typical fuels for each

465 transport related sector.

3.2.2 Calculating uncertainty for each EDGAR sector

Uncertainties for each of the 70 IPCC activities from Table 2 are calculated with the error propagation method and combined into the 20 EDGAR sectors, following Eq. (2):

 $UC_{\underline{EUGARI}} = \sqrt{UC_{\underline{IPCC1}}^2 + UC_{\underline{IPCC2}}^2 + \dots + UC_{\underline{IPCCn}}^2},\tag{2}$

470 where *EDGARj* combined uncertainty per sector *j*, and *1*,2,...,*n* IPCC activities that are taken into account in a particular EDGAR sector; *UC*_{IPCC1}, *UC*_{IPCC2},..., *UC*_{IPCC2}, used in %.

3.2.3 Correction of EDGAR sector uncertainty due to underestimation by the chosen method

The EDGAR sector uncertainty had to be corrected, as the error propagation method of Approach 1 systematically underestimates the uncertainty unless the model is purely additive, which was not the case as EDGAR emissions are

- 475 estimated based on the sum of several product terms. To fix this underestimation IPCC (2006) advises using a correction factor. One example of a correction factor is proposed in Frey (2003), where the performance of an analytical approach for combining uncertainty in comparison to a Monte Carlo simulation with large sample sizes for many cases involving different ranges of uncertainty for additive, multiplicative, and quotient models are evaluated. Frey found that error propagation and Monte Carlo simulated estimates of the uncertainty half range of the model output agreed well for values of less than 100 %,
- 480 but with the increase of the uncertainty a systematic underestimation of uncertainty in the total inventory by the error propagation approach appeared. The relationship between the simulated and propagated error estimates was found to be well behaved, which led to a correction factor development for the large (i.e. greater than 100 %) total inventory uncertainties. This correction factor will not necessarily be reliable for very large uncertainties (i.e. greater than 230 %) because it was calibrated over the range of 10 to 230 %. As such, the correction factor *FC*, calculated following Eq. (3), was
- 485 applied if half range uncertainty estimated from the error propagation method was > 100 and < 230 % following Eq. (4):

FC -	-0.7200+1.0921·UC _{EDGARj} -1.63·10 ⁻³ ·UC _{EDGARj} +1.11·10 ⁻⁵ ·UC _{EDGARj}	$\left \right ^{2}$ (3)
- C _{EDGARJ} -	. UC _{EDGARj}],
(UC _{EDGARJ})	$= UC_{EDGARj} \cdot FC_{EDGARj},$	(4)

where *corr* corresponds to the corrected uncertainty; *UC_{EDGARy}* is given in %. In cases where *UC_{EDGARy}* was ≤ 100 and ≥ 230 %, *FC_{EDGARy}* was assumed to be equal to one. Only four sectors with non-energy use of fuels, chemical processes, fuel
exploitation and coal production emissions were corrected, Table 3 shows how these uncertainties were corrected. It should be noted that some uncertainty ranges for emission factors and/or activity data in IPCC (2006) and IPCC TFI (2019) are not symmetrical and have higher uncertainty values for the lower bound than for the upper bound, due to input from expert knowledge or available in-situ data, which lead to the same pattern in final prior uncertainty bounds.

495 Table 3: Sectors with corrected uncertainties (lower and upper bounds) for countries with well- (WDS) and less well-developed (LDS) statistical infrastructures

		IBCC (2000) and initian man		Constant	Prior uncertainty bounds, %						
No	ECMWF group	IPCC (2006) activities per EDGAR sector	Note	Country	Before co	orrection	After correction				
		EDGAK Sector		type	Low	Up	Low	Up			
	MANUFACTURING (part)	2.D.1, 2.D.2, 2.D.4	Non energy	WDS	112.0	112.0	121.7	121.7			
3		2.D.1, 2.D.2, 2.D.1	use of fuels	LDS	113.8	113.8	124.0	124.0			
-		2.B.1, 2.B.2, 2.B.3, 2.B.4, 2.B.5,	Chemical	WDS	100.9	<u>89.9</u>	107.8	89.9			
		2.B.6, 2.B.8	processes	LDS	100.9	89.9	107.8	89.9			
	1.B.2.a.ii, 1.B.2.a.iii.2, 1.B.2.a.iii.3,		Fuel	WDS	156.6	215.7	191.1	339.1			
7	OTHER (part)	H.B.Z.D.111.3. L.C.		LDS	166.8	223.2	210.9	364.5			
				WDS	107.4	300.5	115.8	300.5			
		1.D.1.u	production	LDS	107.4	300.5	115.8	300.5			

3.2.4 Forcing lognormal distribution on corrected EDGAR sector uncertainty

For models that are purely additive, and for which the half range of uncertainty is less than approximately 50 %, a normal 500 distribution is often an accurate assumption for the model output form. In this case, a symmetric probability distribution with respect to the mean can be assumed. But this is not the case for multiplicative (or mixed) models, or when the uncertainty is large for a non negative variable such as anthropogenic CO₂ emissions. A log normal distribution is typically an accurate assumption for the model output form, where the uncertainty range is not symmetric with respect to the mean, even though the variance for the total inventory may be correctly estimated from Approach 1. IPCC (2006) guidelines provide a practical 505 methodology based on Frey (2003) for approximate asymmetric uncertainty range calculations based on the error propagation method. According to this methodology key characteristics of the 95 % confidence intervals are: (i) approximately symmetric for small ranges of uncertainty, and (ii) positively skewed for large ranges of uncertainty. This methodology was applied if the corrected lower half-range uncertainty estimated from error propagation method was > 50 %. IPCC (2006) suggests to define parameters of the lognormal distribution in terms of the geometric mean μg (which can be 510 estimated based upon the arithmetic mean and the arithmetic standard deviation) following Eq. (5) and geometric standard deviation σg following Eq. (6):

$$\mu g_{EDGARj} = exp \left\{ ln(E_{EDGARj}) - \frac{1}{2} \cdot ln\left(1 + \left[\frac{(UC_{EDGARj})_{corr}}{200}\right]^2\right)\right\},$$
(5)
$$\sigma g_{EDGARj} = exp \left\{ \sqrt{ln\left(1 + \left[\frac{(UC_{EDGARj})_{corr}}{200}\right]^2\right)}\right\},$$
(6)

where E_{EDCARJ} is the anthropogenic CO₂ emissions per sector *j*; *corr* corresponds to the corrected uncertainty (i.e. corrected
 for the systematic underestimation of uncertainty calculated by the error propagation approach used in this study comparing to uncertainties calculated by using the Monte Carlo approach); UC_{EDGARJ} is in %.

Because calculations were performed for upper and lower uncertainty limits separately, there are two values of
$$(UG_{EUGARf})_{corr}$$
 : $[(UG_{EUGARf})_{corr}]_{low}$ — the absolute value of the lower uncertainty limit of sector *j*, and $[(UG_{EUGARf})_{corr}]_{high}$ the absolute value of the upper uncertainty limit of sector *j*. As it is preferred to preserve as much accuracy (extra knowledge) as possible in our calculations and not to inflate uncertainty upper or lower bounds artificially, lower $\{[(UG_{EUGARf})_{corr}]_{high}]_{tn}$ and upper $\{[(UG_{EUGARf})_{corr}]_{high}]_{tn}$ uncertainty half range from the error propagation method were calculated with a logarithmic transformation using $[\mu g_{EUGARf}]_{tow}$, $[\mu g_{EUGARf}]_{high}$ and $[\sigma g_{EUGARf}]_{tow}$; $[\sigma g_{EUGARf}]_{high}$ respectively according to the following Eq. (7) and Eq. (8) (see Figure 2 for visual representation of these equations):
525 $\{[(UG_{EUGARf})_{corr}]_{tow}\}_{ln} = \left(\frac{exp[in([\mu g_{EDGARf}]_{tow}]^{-1.96-Inf}[[\sigma g_{EUGARf}]_{tow}]^{-E_{EUGARf}}]_{high}}{E_{EUGARf}]_{tow}} + 100, \qquad (7)$
 $\{[(UG_{EUGARf})_{corr}]_{high}\}_{ln} = \left(\frac{exp[in([\mu g_{EDGARf}]_{tow}]^{-1.96-Inf}[[\sigma g_{EUGARf}]_{tow}]^{-E_{EUGARf}}]}{E_{EUGARf}} + 100, \qquad (7)$
 $\{[(UG_{EUGARf})_{corr}]_{high}\}_{ln} = \left(\frac{exp[in([\mu g_{EDGARf}]_{high}]^{+1.96-Inf}[[\sigma g_{EUGARf}]_{high}]^{-E_{EUGARf}}]}{E_{EDGARf}} + 100, \qquad (8)$
where *ln* corresponds to logarithmic transformation of the distribution; resulting values are not absolute.
It should be noted that according to this methodology (with constants for 2.5th and 97.5th percentiles, +1.96 and -1.96
respectively, from the Z table⁴) the lower uncertainty half range $\{[(UG_{EUGARf})_{corr}]_{high}\}_{ln}$ is approximately symmetric relative to the 0 (Gaussian distribution)
up to -20.0 %, then has rather rapid growth till -500.0 % (which with logarithmic transformation results in -486.0 %),
maxima at -1350.0 % (which with logarithmic transformation results in -582.6 %) and further gradual decrease.

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⁴ The Z-table is a mathematical table for the values of the cumulative distribution function of the normal distribution.



535 Figure 2: Visual representation of an empirical logarithmic transformation formula for upper and lower uncertainty bounds according IPCC (2006)

Table 4 shows the prior uncertainty values for each EDGAR sector and for two geographical entity types (i.e. with well and less well developed statistical infrastructure). These values are a combined IPCC activity uncertainty aggregated to EDGAR
540 sectors with the error propagation method and corrected for this method's underestimation. Also, as an example, Table 4 shows aggregated to ECMWF groups uncertainties with ensured log normal distribution for China (CHN), Europe (28 members till end 2019) and all world countries.

 Table 4: Prior uncertainties (lower and upper bounds) per each EDGAR emission sector and two geographical entity types (with well- (WDS) and less well-developed (LDS) statistical infrastructure) based on IPCC (2006) and IPCC-TFI (2019), and aggregated to the ECMWF group uncertainties for China (CHN), Europe (E28) and globe (GLB)

			Prior I	incertai	nty bour	ids, %	Uncertainty bounds, %					
No	ECMWF group	IPCC (2006) activities per EDGAR		WDS		LDS		IN,	E28,		GL	1
- 10	Lemmi group	sector	coun	tries	coun	tries	WDS		WDS		m	i x
				Up	Low	Up	Low	Up	Low	Up	Low	Up
4	ENERGY_S	1.A.1.a (subset)	8.6	3.0	12.2	3.0	8.6	3.0	5. 4	1.9	3.6	1.0
2	ENEDCV A	1.A.1.a (rest)	8.6	8.6	12.2	12.2	06	0 6	10	20	25	25
2	ENERGY_A	4 .C	40.3	40.3	4 1.2	<u>41.2</u>	8.6	8.6	2.8	2.8	3.5	3.5
	MANUFACTURING	1.A.2	8.6	8.6	12.2	12.2						
		2.C.1, 2.C.2	37.1	37.1	37.1	37.1						1
		2.C.3, 2.C.4, 2.C.5, 2.C.6, 2.C.7	73.2	73.2	73.2	73.2	12.8 1					1
3		2.D.1, 2.D.2, 2.D.4	121.7	121.7	124.0	124.0		19.4	3.9	5.8	5.7	8.6
		2.A.1, 2.A.2, 2.A.3, 2.A.4	70.9	70.9	93.0	93.0						1
		2.B.1, 2.B.2, 2.B.3, 2.B.4, 2.B.5, 2.B.6, 2.B.8	107.8	89.9	107.8	89.9						
4	SETTLEMENTS	1.A.4, 1.A.5.a, 1.A.5.b.i, 1.A.5.b.ii	<u>12.2</u>	<u>12.2</u>	26.0	26.0	12.2	12.2	4.2	4.2	3.9	3.9
		1.A.3.a_CRS	5.5	6.4	50.1	106.8						
5	AVIATION	1.A.3.a_CDS	5.5	6.4	50.1	106.8	3.5	4.1	1.4	1.6	17.3	58.1
		1.A.3.a_LTO	5.5	6.4	50.1	106.8						

		1.A.3.b	5.4	5.4	7.1	7.1						
6	TRANSPORT	1.A.3.d	5.4	5.1	50.0	50.0		8.2	1.6	1.8	4.3	6.4
		1.A.3.c, 1.A.3.e	50.3	106.9	50.5	107.0						
	OTHER	1.A.1.b, 1.A.1.c, 1.A.5.b.iii, 1.B.1.c, 1.B.2.a.iii.4, 1.B.2.a.iii.6, 1.B.2.b.iii.3	54.4	149.3	57.7	151.4						
7		1.B.2.a.ii, 1.B.2.a.iii.2, 1.B.2.a.iii.3, 1.B.2.b.ii, 1.B.2.b.iii.2, 1.B.2.b.iii.4, 1.B.2.b.iii.5, 1.C	191.1	339.1	210.9	364.5	39.7180.9	10.1	4 5.3	11.5	52.4	
		1.B.1.a	115.8	300.5	115.8	300.5						
		3.C.2, 3.C.3, 3.C.4, 3.C.7	70.7	0.0	70.7	0.0						
		2.D.3, 2.B.9, 2.E, 2.F, 2.G	25.0	25.0	50.0	50.0						

3.2.5 Calculating uncertainty for each ECMWF group

The next step is to combine these prior uncertainties for each EDGAR sector into ECMWF group uncertainties (see Table 4). Sector uncertainties are combined into group uncertainties by addition following Eq. (9) and Eq. (10):

$$UC_{ECMWFk} = \frac{\sqrt{(((UC_{EDGAR1})_{corr})_{in} \cdot E_{EDGAR1})^2 + (((UC_{EDGAR2})_{corr})_{in} \cdot E_{EDGAR2})^2 + \dots + (((UC_{EDGARn})_{corr})_{in} \cdot E_{EDGARn})^2}{|E_{EDGAR1} + E_{EDGAR2} + \dots + E_{EDGAR2}|},$$
(9)
$$E_{ECMWFk} = E_{EDEAR1} + E_{EDEAR2} + \dots + E_{EDGAR2},$$
(10)

where UC_{ECMWFk} and E_{ECMWFk} — combined uncertainty and total emissions per group k; 1,2,...,n — EDGAR emission sectors that are combined in a particular ECMWF group k; { $(UC_{EDGAR1})_{corr}$ }_{in}, { $(UC_{EDGAR2})_{corr}$ }_{in}, ..., { $(UC_{EDGARn})_{corr}$ }_{in} are in %. Combined group uncertainties are country specific, because they take into account sector budget and adjust

uncertainty values accordingly.

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3.2.6 Calculating mean and standard deviation of lognormally distributed ECMWF group uncertainty

Finally, we needed to ensure a log normal distribution of CO₂-emissions. Upper and lower uncertainty half range values per ECMWF group *k*-ECMWFk are descriptive, but not straight forward to use for emission perturbations in ensemble runs or flux inversions, where mean and standard deviation of the distribution are usually used. The lower and upper bounds of the 95 % probability range, which are the 2.5th and 97.5th percentiles respectively, calculated assuming a log normal distribution based on a corrected estimated uncertainty half range from an error propagation approach, are lower and upper uncertainty values. Taking this into account and using the Z-table for 2.5th and 97.5th percentiles *p*,-mean *p*th and standard deviation *p*th of log normal distribution can be calculated following Eq. (11):

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$$Z_{p} = \frac{ln([E_{ECMWFk}]_{p}) - \mu_{ECMWFk}^{ln}}{\sigma_{ECMWFk}^{ln}},$$
(11)
where the following variables are known:

$$p = 2.5 \Longrightarrow Z_{2.5} = -1.96, [E_{ECMWFk}]_{2.5} = E_{ECMWFk} \cdot \left(1 + \frac{[UC_{ECMWFk}]_{low}}{100}\right),$$
(12)

$$m = 0.75 \Longrightarrow Z_{m} = 1.06, [E_{m} = 1]_{m} = E_{m} = (1 + \frac{[UC_{ECMWFk}]_{low}}{100}),$$
(12)

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then simple system could be composed and solved accordingly following Eq. (14) and Eq. (15):



where $[UC_{ECMWFk}]_{tow}$ and $[UC_{ECMWFk}]_{high}$ are in %.

3.2.7<u>3.4</u> Example of uncertainty calculation

Table 54 shows a step-by-step example of how yearly uncertainties are calculated. Example, and Figure 2 shows plotted probability density functions based on computed log-normal parameters. The example shows calculations for the TRANSPORT "group,", that consists of several EDGAR-emission "sectors (one EDGAR sector consists even of several IPCC activities). Example". The example shows two countries with different statistical infrastructure development levels (country with well-developed statistical infrastructures is Germany, country with less well-developed statistical infrastructures is the Russian Federation) and significant differences in emission budgets.

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Table <u>5a4a</u>: Preparatory step for yearly uncertainty calculation – data collection, same values are applied for all countries of the same type, namely for countries with well- (WDS) and less well-developed (LDS) statistical infrastructures

(WDS)								 0 0
		1.A.3.d	1.A.3.d	Water-borne navigation	composition of 80 % diesel and 20 % residual fuel oil	2.1	1.1	55 00
			1.A.3.c	Railways	diesel	2.0	0.9	55 00
		1.A.3.c, 1.A.3.e		Other transportation – Pipeline	none (suggested to neglect)	0.0	0.0	0 C 0 C
		1.A.5.¢	1.A.3.e	Other transportation – Off-road	most typical emission factor	2.0	2.0	$5 \begin{array}{c} 1 \\ 0 \\ 0 \\ 0 \\ \cdot \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0$
	TRANSPORT <u>(6)</u>	1.A.3.b	1.A.3.b	Road transportation	most typical emission factor	5.0	5.0	55 00
Russian Federatio n (LDS)		1.A.3.d	1.A.3.d	Water-borne navigation	composition of 80 % diesel and 20 % residual fuel oil	2.1	1.1	55 00 00
			1.A.3.c	Railways	diesel	2.0	0.9	55 00
	TRAN	00 1.A.3.c, 1.A.3.e	Other transportation – Pipeline	none (suggested to neglect)	0.0	0.0	0 0 0 0	
		1.A.J.¢	1.A.3.e	Other transportation – Off-road	most typical emission factor	5.0	5.0	$5 \begin{array}{c} 1 \\ 0 \\ 0 \\ 0 \\ \cdot \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0$

Table <u>5b4b</u>: First part of yearly uncertainty calculation – same values are applied for all countries of the same type, namely for countries with well- (WDS) and less well-developed (LDS) statistical infrastructures

Country (Type)	IPCC (2006) activities per EDGAR	IPCC (2006) activity	Combined uncertainty per IPCC (2006) activity, <u>%</u> see Eq. (1)		Combined uncertainty per EDGAR_"sector,-", % see Eq. (2)		Corrected combined uncertainty per EDGAR-"sector ,", % see Eq. (3)-(4)	
	"sector"	activity	Low	Up	Low	Up	Low	Up
	1.A.3.b	1.A.3.b	5.4	5.4	5.4	5.4	5.4	5.4
C	1.A.3.d	1.A.3.d	5.4	5.1	5.4	5.1	5.4	5.1
Germany (WDS)	1.A.3.c, 1.A.3.e	1.A.3.c	5.4	5.1				
(WDS)		1.A.3.e	0.0	0.0	50.3	100.1	50.3	106.9
			50.0	100.0				
	1.A.3.b	1.A.3.b	7.1	7.1	7.1	7.1	7.1	7.1
Russian	1.A.3.d	1.A.3.d	50.0	50.0	50.0	50.0	50.0	50.0
Federation	1.4.2	1.A.3.c 1.A.3.e	5.4	5.1	50.5	100.3	50.5	
(LDS)	1.A.3.c,		0.0	0.0				107.0
	1.A.3.e		50.2	100.1				

Table <u>5e4c</u>: Second part of yearly uncertainty calculation – values are specific per <u>each</u>_geographical entity, <u>take into</u> <u>accountconsider</u> country type, namely if country has well- (WDS) or less well-developed (LDS) statistical infrastructure, and countries emission budget (values are from CHE_EDGAR-ECMWF_2015)

Country (Type)	IPCC (2006) activities per EDGAR <u>"sector"</u>	Emission budget 2015 per EDGAR "sector , <u>Mton",</u> . <u>10³ kt</u>	assumed budget Grouped uncertainty with per lognormallog- 2015 per normal distribution ECMWF assumed lognormallog- per per EDGAR "sector, see Eq. (5)- Mton", ·10 ³ kt see Eq. (9)-(105)		Immed lognormallog- nal distribution per MWF "group,", % see Eq. (9)-(105)uncertainty with ass lognormallog-norm distribution per ECP group," see Eq. (14)-(159)-c		neters of grouped ainty with assumed ormallog-normal ution per ECMWF <u>"group</u> ," q. (14) (159)-(10)			
		<u>10 Kt</u>	Low	Up	<u>see Eq. (6)</u>	Low	Up	mean	standard deviation	
	1.A.3.b	139.6	5.4	5.4						
Germany	1.A.3.d	1.0	5.4	5.1	143.0	5.3	5.7	11.9	0.0	
(WDS)	1.A.3.c, 1.A.3.e	2.3	40.3	135.5	145.0	143.0	145.0 5.5	5.7	11.9	0.0
Russian Federation (LDS)	1.A.3.b	131.7	7.1	7.1						
	1.A.3.d	7.4	40.1	57.2	206.9	14.1	44.8	12.3	0.1	
	1.A.3.c, 1.A.3.e	67.9	40.5	135.7	200.9	14.1	44.0	12.5	0.1	

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3.3 Monthly uncertainties

For Earth system modelling and data assimilation purposes a sub-yearly time scale is more appropriate. Monthly profiles of anthropogenic emissions are available and used in air quality models and are more certain than the sub-monthly profiles. The monthly profiles used in EDGARv4.3.2 are standardised to 12 monthly shares per EDGAR sector and per region (i.e.

595 Northern temperate zone, Equator, Southern temperate zone). They do not take into account the specificity of a single year and are not varying within a geographical entity (country). We used these global yearly and monthly emission maps for 2010 to calculate for each month a multiplication factor per 0.1°×0.1° grid cell of the sector specific maps. Then multiplication factors were combined with CHE_EDGAR ECMWF_2015 maps and monthly country and sector specific CO₂ emission budgets are calculated.

600 Uncertainties for monthly budgets are obviously larger than yearly ones and instead of one standard deviation σ (Quilcaille et al., 2018) two or three standard deviations, 2σ or 3σ respectively are commonly used (Oda et al., 2018; Andres et al., 2014; Andres et al., 2011). We decided to be more analytical:

1) to use the same procedure as for annual uncertainty calculation but base it on monthly emission budgets (i.e. uncertainties for IPCC activities are combined to EDGAR sectors with error propagation method, corrected for systematic

605 underestimation by error propagation method, and adapted to have log normal distribution; see Eq. (1) (8)). Obtained monthly uncertainties are the same or even smaller than the yearly ones, because empirical equations applied use emission budgets, which are smaller for individual months compared to the yearly values;

2) to calculate the correlation α (an uncertainty boosting parameter) between yearly and monthly uncertainties based on an analysis of the variations over the different months following Eq. (16):



 $((E_{MONTH1} \cdot UC_{MONTH1})^2 + (E_{MONTH2} \cdot UC_{MONTH2})^2 + \dots + (E_{MONTH12} \cdot UC_{MONTH12})^2)_{+}$ where E and UC correspond to sectoral emission budget and uncertainty in kton and % respectively, YEAR, MONTH1, MONTH12 yearly and monthly (January, February, ..., December) values. Eq. (16) is based on the rule for combining uncorrelated uncertainties under addition of the error propagation equation (see Eq. (9)) and 615

assumption that each month's uncertainty should be enhanced (boosted) by the same value;

3) to multiply the prior yearly uncertainties from Table 4 by the boosting parameter (specific per country and emission sector) and use the result as a first guess of monthly prior uncertainties;

to iterate calculation steps 1) to 3) in order to find the best boosting parameter (to have the best fit between yearly 4) 620 and combined 12 month uncertainties) from Eq. (16) for each country and emission sector. Once the best boosting parameter was found (i.e. maximum difference between α from previous iteration and the current one over all countries and emission sectors became less than acceptable threshold) calculated monthly uncertainties per each EDGAR sector were grouped into 7 ECMWF groups and log normal distribution of CO₂ emissions was ensured, see Eq. (9) (15).

Figure 3 has simplified roadmaps for yearly and monthly uncertainty calculations.

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6x10⁻⁶ 4x10⁻⁶ 2x10⁻⁶ n

Yearly uncertainties

-100000 150000 200000 250000 300000 350000 400000 450000 550000 CO₂ anthropogenic emissions, ktons

Monthly uncertainties



Figure 3: Simplified roadmaps for yearly2: Probability density functions (for Germany (left) and monthlythe Russian Federation (right) uncertainty calculation and their relation (bottom)

The prior error covariance matrix of the emission inventory is required as an input to the inversion system. According to the 630
 IPCC (2006) all anthropogenic CO₂-emissions are assumed to be fully uncorrelated, hence the prior error correlations between grid-cell emissions from the same sector should be assumed negligible if country- and/or sector-specific information is lacking. For the first-implementation, ECMWF-)) based on computed log-normal mean and standard deviation for TRANSPORT "group covariance matrices per each geographical entity have the same representation – emission group is fully correlated with itself and fully uncorrelated with any other group. For an example of ECMWF group covariance matrices see Table S4 from the Supplementary Information, section S.3. Due to the lack of information available to properly characterize the error correlations and error variances in the inventory, a refinement of those prior statistics will be carried out in a follow-on paper (Bousserez, 2019) using atmospheric CO₂-observations. For this, the maximum likelihood of the prior error standard deviations and error correlations lengths will be estimated following approaches described in Wu et al. (2013)."

3.4 Gridding uncertainties

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Calculated yearly and monthly uncertainties per country and <u>sectoremission "group"</u> were assigned to each grid-box on the global map. National uncertainties were applied uniformly across each country. Figure 4<u>3</u> shows an example of the upper and lower uncertainty limits of anthropogenic CO₂ emission flux for <u>the</u>_TRANSPORT "group". It should be noted that uncertainties related to the spatial distribution (representativeness of the proxy data and their uncertainty) should be much

645 higher than the ones presented in this study. This research does not address uncertainties related to the spatial distribution. In the future we planit is planned to address these uncertainties too. For example, by following Oda et al. (2019) to characterize spatial patterns of the disaggregation errors in ourthe emission maps.



Figure 43: CO₂ emission flux uncertainties (lower (left) and upper (right) bounds) for TRANSPORT <u>"group"</u> in kg·m⁻²·s⁻¹

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4 Generated dataset

CHE_EDGAR_ECMWF_2015 data (Choulga et al., 2020) are freely available <u>https://doi.org/10.5281/zenodo.3967439</u>, and consist of 11 grid maps in NetCDF format and one Excel file with information on anthropogenic CO₂ emissions and their uncertainties. For detailed information on each file see Table 6.

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Table 6: Detailed information on CHE_EDGAR-ECMWF_2015 data

File	General note	Field / Spreadsheet
ver He	file has 2×8 fields with annual upper	"Lower" lower uncertainty bound (2.5th percentile of log normal distribution) for
ן ל יו ל	and lower uncertainty bounds in % per	yearly emissions, in %-
1.1.1	each emission group and for all groups	"Upper" upper uncertainty bound (97.5th percentile of log-normal distribution) for
<u>∦</u> ·≣ ợ	summed together on a regular grid	yearly emissions, in %
15 \$ \$	with 1800 pixels along the latitude and	"Sector" emission sector numerical name. "0" represents emission group ENERGY_S
∣ <u> </u>	3600 pixels along the longitude, where	(with IPCC (2006) activity 1.A.1.a (subset)) standing for power industry emissions from
∰ Ti ĝ	values represent centre of the grid-cell	super emitting power plants; "1" group ENERGY_A (1.A.1.a (rest), 4.C) power
l⊈ aª		industry emissions from standard emitting power plants, & solid waste incineration; "2"

-		
hintiesMonthly_Upper_Low er_Uncertainties	file has 2×8×12 fields with monthly upper and lower uncertainty bounds in % per each emission group and for all groups summed together on a regular grid with 1800 pixels along the latitude and 3600 pixels along the longitude, where values represent centre of the grid cell file has 3×8 fields with annual emissions, and upper and lower uncertainty bounds in kg m ² -s ¹ - per each emission group and for all groups	group MANUFACTURING (1.A.2, 2.C.1, 2.C.2, 2.C.3, 2.C.4, 2.C.5, 2.C.6, 2.C.7, 2.D.1, 2.D.2, 2.D.4, 2.A.1, 2.A.2, 2.A.3, 2.A.4, 2.B.1, 2.B.2, 2.B.3, 2.B.4, 2.B.5, 2.B.6, 2.B.8) combustion for manufacturing (including autoproducers), & iron and steel production, & non ferrous metals production, & non energy use of fuels, & non metallic minerals production, & chemical processes; "3" group SETTLEMENTS (1.A.4, 1.A.5.a, 1.A.5.b.i, 1.A.5.b.ii) energy for buildings, residential heating; "4" group AVIATION (1.A.3.a_CRS, 1.A.3.a_CDS, 1.A.3.a_LTO) aviation cruise, & climbing and descent, & landing and take off; "5" group TRANSPORT (1.A.3.b, 1.A.3.d, 1.A.3.c, 1.A.3.c) road transportation, & shipping, & railways, pipelines, off road transport; "6" group OTHER (1.A.1.b, 1.A.1.c, 1.A.5.b.iii, 1.B.1.c, 1.B.2.a.iii, 4, 1.B.2.a.iii, 6, 1.B.2.b.iii.3, 1.B.2.a.iii, 1.B.2.a.iii.3, 1.B.2.b.ii, 1.B.2.b.iii.2, 1.B.2.b.iii.4, 1.B.2.b.iii.5, 1.C, 1.B.1.a, 3.C.2, 3.C.4, 3.C.7, 2.D.3, 2.B.9, 2.E, 2.F, 2.G) oil refineries and transformation industry, & fuel exploitation, & coal production, & agricultural soils, & solvents and products use; "7" represents all groups summed together file structure is identical to the file Annual_Upper_Lower_Uncertainty bound (2.5 th percentile of log normal distribution) for yearly emissions of ENERGY_S group, in kg m ⁻² s ⁻¹ , "Sup_upper" upper uncertainty bound (07.5 th percentile of log normal distribution) for yearly emissions of ENERGY_S group, in kg m ⁻² s ⁻¹ ,
Amnual_Upper_Lower_Uncertainties_ 0.1_0.1.ne	each emission group and for all groups summed together on a regular grid with 1800 pixels along the latitude and 3600 pixels along the longitude, where values represent centre of the grid cell	yearly emissions of ENERGY_S group, in kg·m ² ·s ⁻¹ , "Sup_flux" yearly emissions of ENERGY_S group, in kg·m ² ·s ⁻¹ "Ene_lower", "ene_upper", "ene_flux" same, but for ENERGY_A group, in kg·m ⁻² ·s ⁻¹ "Man_lower", "man_upper", "man_flux" same, but for MANUFACTURING group, in kg·m ⁻² ·s ⁻¹ "Set_lower", "set_upper", "set_flux" same, but for SETTLEMENTS group, in kg·m ⁻² ·s ⁻¹ "Avi_lower", "avi_upper", "avi_flux" same, but for AVIATION group, in kg·m ⁻² ·s ⁻¹ "Tra_lower", "tra_upper", "tra_flux" same, but for TRANSPORT group, in kg·m ⁻² ·s ⁻¹ "Oth_lower", "oth_upper", "oth_flux" same, but for OTHER group, in kg·m ⁻² ·s ⁻¹ "All_lower", "all_upper", "all_flux" same, but for all_groups summed together, in kg·m ⁻² ·s ⁻¹
Monthly_Sup_Upper_ Lower_Uncertainties_ 0.1_0.1_me	file has 3×12 fields with monthly emissions, and upper and lower uncertainty bounds in kg m ⁻² -s ⁻¹ per ENERGY_S emission group on a regular grid with 1800 pixels along the latitude and 3600 pixels along the longitude, where values represent centre of the grid-cell	 "Sup_lower" lower uncertainty bound (2.5th percentile of log normal distribution) for monthly emissions of ENERGY_S group, in kg m²-s⁻¹ "Sup_upper" upper uncertainty bound (97.5th percentile of log normal distribution) for monthly emissions of ENERGY_S group, in kg m²-s⁻¹ "Sup_flux" monthly emissions of ENERGY_S group, in kg m²-s⁻¹ "Month" month numerical name, where 1, 2,, 12 correspond to January, February,
Monthly_Ene_Upper_1 Lower_Uncertainties_1 0.1_01_me	file has 3×12 fields with monthly emissions, and upper and lower uncertainty bounds in kg m ² s ⁴ per ENERGY_A emission group on a regular grid with 1800 pixels along the	file structure is identical to the file Monthly_Sup_Upper_Lower_Uncertainties_0.1_0.1.nc, but with "ene_lower", "ene_upper", "ene_flux" fields

<u>_Man_Upper</u> Uncertainties 0.1.ne	file has 3×12 fields with monthly	file structure is identical to the file
E E E	emissions, and upper and lower	Monthly_Sup_Upper_Lower_Uncertainties_0.1_0.1.nc, but with "man_lower",
日常も	uncertainty bounds in kg m ⁻² s ⁻¹ per	"man_upper", "man_flux" fields
4 3 1	MANUFACTURING emission group	
チモル	on a regular grid with 1800 pixels	
Monthly_ _Lower_1 _0.1	along the latitude and 3600 pixels	
onthly ower: _0_	along the longitude, where values	
l ¥ ∔ ∣	represent centre of the grid-cell	
	file has 3×12 fields with monthly	file structure is identical to the file
<u>Set_Upper_</u> Jncertainties L_0.1.nc		
よう しょう しょう しょう しょう しょう しょう しょう しょう しょう しょ	emissions, and upper and lower	
日書業	uncertainty bounds in kg m ⁻² s ⁻¹ per	<u>"set_upper", "set_flux" fields</u>
3 8 7	SETTLEMENTS emission group on a	
L B J	regular grid with 1800 pixels along the	
Monthly Lower_U 0.1	latitude and 3600 pixels along the	
Month Lower 6	longitude, where values represent	
ĽĽ	centre of the grid-cell	
	file has 3×12 fields with monthly	file structure is identical to the file
Upper tainties ne		Monthly_Sup_Upper_Lower_Uncertainties_0.1_0.1.nc, but with "avi_lower",
<u>Uppe</u> Jncertaintie: 1_0.1.nc	emissions, and upper and lower uncertainty bounds in $\log m^2$ of nor	"avi_upper", "avi_flux" fields
1 # 1	uncertainty bounds in kg·m ⁻² ·s ⁻¹ per	-avi_upper, avi_nux_nerus
Avi- nceri	AVIATION emission group on a	
1111	regular grid with 1800 pixels along the	
Monthly. Lower_L 0.1	latitude and 3600 pixels along the	
Monthl Lower_ 0	longitude, where values represent	
ĽŻ	centre of the grid-cell	
TI TI	file has 3×12 fields with monthly	file structure is identical to the file
Tra_Upper_ Jncertainties_ L_0.1.ne	emissions, and upper and lower	Monthly_Sup_Upper_Lower_Uncertainties_0.1_0.1.nc, but with "tra_lower",
e ≣∵∄	uncertainty bounds in kg·m ⁻² ·s ⁻¹ per	"tra upper", "tra flux" fields
];∦,¶]	TRANSPORT emission group on a	au_upper, au_nux netus
H, ∰, di	require and with 1900 nivels along the	
N - H - H	regular grid with 1800 pixels along the	
450	latitude and 3600 pixels along the	
Monthly Lower_L 0.1	longitude, where values represent	
	centre of the grid-cell	
Oth_Upper_ ncertainties_ _0.1.ne	file has 3×12 fields with monthly	file structure is identical to the file
± <u></u> <u></u>	emissions, and upper and lower	Monthly_Sup_Upper_Lower_Uncertainties_0.1_0.1.nc, but with "oth_lower",
P 3 8	uncertainty bounds in kg·m ⁻² ·s ⁻¹ per	"oth_upper", "oth_flux" fields
⊈ ∰ +i	OTHER emission group on a regular	
	grid with 1800 pixels along the	
≜∄∄ま	latitude and 3600 pixels along the	
17 17	longitude, where values represent	
Monthly. Lower_L 0.1	centre of the grid-cell	
	file has 3×12 fields with monthly	file structure is identical to the file
J pper_ inties e	me mas 3×12 metas with monthly	file structure is identical to the file Monthly Sup Upper Lower Uncertainties 0.1 0.1.nc, but with "all lower",
Monthly_All_Upper Lower_Uncertainties 0.1_0.1.nc	emissions, and upper and lower	
11 ∄ ⋕	uncertainty bounds in kg in 5 101 an	"all_upper", "all_flux" fields
L Jncerta 	groups summed together on a regular	
L≞⊥	grid with 1800 pixels along the	
Monthly. Lower_U 0.1	latitude and 3600 pixels along the	
≣₿≱	longitude, where values represent	
12 ≴ _	centre of the grid-cell	
	file has 16 spreadsheets with listed	"COUNTRY" ISO Code (3-letter abbreviation of a geographical entity), Geographical
₩\$	information per country (metadata,	name (name of a geographical entity), Type (development level of countries statistical
ਪ੍ਰੀ 🖞	emissions, uncertainties, statistical	infrastructure, meaning with well-/less well developed statistical infrastructure), Main
	parameters)	country (dependency, which country geographical entity in question belongs to), Full
	parameters)	country (dependency, which country geographical clutty in question belongs to), Pun
_ <u></u> <u></u> , <u>₹</u>		intermetion (till nome of a geographical entity, and what torritory it occupies on the man
CHE_EDGAR_ 2015.xlsx		information (full name of a geographical entity, and what territory it occupies on the map of this study)

	"GROUP" № (number of anthropogenic CO2 emission group), ECMWF group (group
	name), IPCC (2006) activity (IPCC activities that are included in each group), Note
	(short explanation of the group), Global emission budget 2015, Mton (total global
	emissions per group), Prior uncertainty bounds, % (initial, calculated purely based on
	assumptions from IPCC, lower and upper uncertainty bounds for countries with well-
	Aess well-developed statistical infrastructures)
	"YEARLY" ISO Code (3 letter abbreviation of a geographical entity), ECMWF group
	(group name), Budget, kton (yearly anthropogenic CO2 emission budget per group and
	total per geographical entity), Uncertainty bounds, % (calculated based on Prior
	uncertainty bounds and Budgets yearly uncertainties per group and total per geographical
	entity, uncertainties lower/upper/symmetrical bounds), Contribution to total countries
	uncertainty, % (share of each group in geographical entities total yearly uncertainty, total
	contribution is always 100 %), Parameters of log-normal distribution (anthropogenic
	CO2 emission distribution is assumed to be log normal, so additionally for modelling
	purposes log normal mean, log normal standard deviation and log normal variance were
	calculated)
	"MONTHLY 01", "MONTHLY 02",, "MONTHLY 12" - same explanation as for
	spreadsheet "VEARLY", but for a month (01, 02,, 12 correspond to January,
	February,, December)

54 Comparison and sensitivity

4.1 Comparison of total uncertainty of global CO2 emission datasets

660 Calculated emissions and uncertainties of fossil CO₂ have been compared to other <u>global_data</u> sets based on the country-specific data reported to UNFCCC₂ and on fuel-specific data reported in the energy statistics of IEA. The global values and their uncertainty at a 2σ range for the CHE_EDGAR-ECMWF_2015 dataset show the lowest value of -4.7/+9.6 % or ±7.1 % range, see Table 7<u>5</u>. This result might be attributed to the methodology, in particular considering that (i) all calculations were done at the country level and then aggregated to global level assuming no correlation following IPCC (2006), (ii) all calculations were done separately for upper and lower uncertainty bounds to preserve original information with asymmetric confidence intervals for large uncertainties (not required for the Approach 1 described in IPCC (2006), according to Approach 1 from IPCC (2006)in which only the higher uncertainty value of the asymmetric interval should be used – leads to artificial inflation of uncertainty upper or lower limit), and (iii) might be also because in this study we were not taking into account proxy grid-map uncertainties are not considered.

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Table 75: Comparison of global anthropogenic CO₂ emission uncertainty at 2σ associated with certain emission datasets

Name	Global uncertainty at 2σ , %	References
BP	no quantitative assessment of uncertainty associated with its emissions dataset	Andrew (, 2020)
CDIAC	±8.4 %	Andres et al. (., 2016)
CEDS	no quantitative assessment of uncertainty associated with its emissions dataset, limited information in	Hoesly et al . (<u>.,</u>2018)
CHE_EDGAR-ECMWF_2015	±7.1 % (-4.7/+9.6 %)	Andrew-(, 2020)
EDGAR	±9.0 %	Janssens-Maenhout et al. (., 2019)

EIA	no quantitative assessment of uncertainty associated with its emissions dataset	Andrew-(, 2020)
Global Carbon Project (GCP)	±10.0 %	Friedlingstein et al . (2019)
IEA	no quantitative assessment of uncertainty associated with its emissions dataset	Andrew-(, 2020)
ODIAC	$\pm 8.4 \%^{5}$	Oda et al . (<u>.,</u> 2018)

In this paper we decided to focus on some specific geographical areas — chosen to be among most emitting in total or per emission group, The contribution of each emission "group" to the total uncertainty per grid-cell is assessed. Figure 4 shows
675 which "group" contributes the most to the total uncertainty per grid-cell. TRANSPORT "group" contributes most to the grid-cell uncertainty over the Unites States of America (due to road and off-road transport) and over the ocean (due to shipping). AVIATION "group" contributes most over main flight routes all over the Globe. OTHER "group" contributes the most over agricultural areas, and regions with oil refineries and transformation industry and fuel exploitation. MANUFACTURING "group" contributes most over areas in e.g. Vietnam and Bangladesh. ENERGY_A (and ENERGY_S) "group" contributes the most to the grid-cells (e.g. South Africa). SETTLEMENT "group" contributes the most to the grid-cell uncertainty over either very dense, either very sparce populated areas.



Figure 4a: Main emission "group" that contributes to the total uncertainty per grid-cell - Global region

⁵ The difference between ODIAC and CDIAC gridded data is 3.3-5.7 % (Oda et al., 2018).



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Figure 4b: Main emission "group" that contributes to the total uncertainty per grid-cell – European (left) and China (right) regions



690 <u>Figure 4c: Main emission "group" that contributes to the total uncertainty per grid-cell – the Russian Federation (top) and the</u> <u>United States of America (bottom) regions</u>


Figure 4d: Main emission "group" that contributes to the total uncertainty per grid-cell – Brazil (top left), India(top right), Indonesia (bottom left), and Japan (bottom right) regions

4.2 Dependence of the country-specific statistical infrastructure

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<u>Also, some specific geographical areas are analysed: chosen to be among the most emitting in total or per emission "group",</u> and the most typical or most influential for a certain region. A list of these geographical entities and development levels of their statistical infrastructures are presented in Table <u>86</u>.

Table 86: List of selected geographical entities with their statistical in	ifrastructure's development levels
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ISO Code	Geographical name	Туре
GLB	All World Countries	mixed-developed statistical infrastructure
E28	Europe (28 members till <u>until</u> end 2019)	well-developed statistical infrastructure
DEU	Germany	well-developed statistical infrastructure
ESP	Spain	well-developed statistical infrastructure
FRA	France	well-developed statistical infrastructure
GBR	United Kingdom	well-developed statistical infrastructure
POL	Poland	well-developed statistical infrastructure
BRA	Brazil	less well-developed statistical infrastructure
CHN	China	well-developed statistical infrastructure

IDN	Indonesia	less well-developed statistical infrastructure
IND	India	well-developed statistical infrastructure
JPN	Japan	well-developed statistical infrastructure
RUS	Russian Federation	less well-developed statistical infrastructure
USA	United States of America	well-developed statistical infrastructure

5.1 Global versus country-specific results

705 In order to see how <u>the_development level</u> of country's or geographical entity's statistical infrastructure <u>is influencinginfluences the</u> emission uncertainty of that country or geographical entity itself and (possibly) the globe, uncertainty calculations for selected entities were performed twice – with their original and inverse types (i.e. <u>a</u> country with <u>a</u> well-developed statistical infrastructure becomes <u>a</u> country with <u>a</u> less well-developed statistical infrastructure and vice <u>a</u> versa). More details on <u>a</u> geographical entity's statistical infrastructure development level (e.g. how it was determined) are given in the Supplementary Information, section S.4<u>5</u>. Figure 5 shows sectoral emission budgets, uncertainties and contributions in percentage, to the total uncertainty of <u>a</u> country or geographical entity with its original and inverse statistical infrastructure development levels. The biggest impact of development level change <u>can be noticedoccurs</u> for countries with larger emission budgets. On average, total uncertainties of selected countries (see Table <u>86</u>) changed by 1-2 %; "group" uncertainties changed in line with prior uncertainties from Table <u>4</u>-and countries emission budgets, as reported in Table <u>97</u>.

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Table <u>97</u>: Country's statistical infrastructure (countries with well- (WDS) and less well-developed (LDS) statistical infrastructures) influence on emission uncertainty

Impact on the uncertainty	ECMWF group <u>"Group"</u> <u>name</u>	Cause description
most substantial	SETTLEMENTS	 consists only from residential heating emissions; high differences in prior uncertainties for WDS and LDS, ±12.2 % and ±26.0 % respectively
	MANUFACTURING	 budget usually makes a significant part of country's total emission budget; globally mainly composed from combustion for manufacturing with rather low prior uncertainty (±8.6 % and ±12.2 % for WDS and LDS respectively) and non-metallic minerals production with much higher uncertainties (±70.9 % and ±93.0 % for WDS and LDS respectively); also contains emissions from very uncertain non-energy use of fuels (±121.7 % and ±124.0 % for WDS and LDS respectively) and chemical processes (-107.8/+89.9 % both for WDS and LDS) emissions, though their global share in this "group" is only ~7.0 %
strong	ENERGY_A	 budget usually makes a significant part of country's total emission budget; composed of emissions from standard power plants with rather low uncertainties (±8.6 % and ±12.2 % for WDS and LDS respectively) and solid waste incineration with much higher uncertainties (±40.3 % and ±41.2 % for WDS and LDS respectively); for the Globe the ratio of solid waste incineration to energy emissions is ~1/100, which keeps the total "group" prior uncertainty quite low ±3.5 %; NB!Note: geographical entities with higher ratios will have higher uncertainties
	ENERGY_S	• composed of emissions from super power plants only with rather low prior uncertainties (-8.6/+3.0 % and -12.2/+3.0 % for WDS and LDS respectively) for all geographical entities

mild	TRANSPORT	 globally mainly composed of road transportation with rather low uncertainty (±5.4 % and ±7.1 % for WDS and LDS respectively) and shipping emissions with low uncertainties - 5.4/+5.1 % for WDS and high uncertainties ±50.0 % for LDS; also contains rather uncertain railways, pipelines and off-road transport emissions (~ - 50.4/+107.0 % for both WDS and LDS), though their global share in this "group" is ~16.0 % only; <u>NB!Note:</u> all international shipping is included in All World Countries geographical entity
small	AVIATION	 extremely high differences in prior uncertainties for WDS and LDS (-5.5/+6.4 % and -50.1/+106.8 % respectively), though this "groups" share in global emissions is only 2.3 %; <u>NB!Note:</u> all international aviation is included in All World Countries geographical entity
negligible	OTHER	 composed of very uncertain components with usually almost the same prior uncertainties for WDS and LDS; main composite globally (~78.0 %) are emissions from oil refineries and transformation industry with prior uncertainties -54.4/+149.3 % and -57.7/+151.4 % for WDS and LDS respectively; also usually has the highest contribution to the country's total uncertainty







Figure 5: Emission budgets, uncertainties and contributions in percentage to the total uncertainty of the country with their original and inverse types (countries with well- (WDS) and less well-developed (LDS) statistical infrastructures): impacting mainly country itself, e.g. the Russian Federation (RUS), India (IND), impacting also Europe (E28), e.g. Germany (DEU), impacting even global values, e.g. China (CHN)

Alterations in some countries' (e.g. Germany, France) statistical infrastructure's development levels lead to changes in Europe (28 members tilluntil end 2019) uncertainties, with the most substantial change for the SETTLEMENTS "group" (e.g. 2.5 and 1.0 % respectively). Huge changes (> 10.0 %) in Europe's (28 members tilluntil end 2019) AVIATION "group" uncertainty %-percentage value can be due to the variation of statistical infrastructure development level for Germany, United Kingdom, France or Spain; though this groups "group's" contribution to the Europe's (28 members till end 2019) total uncertainty remains negligible. Alterations in statistical infrastructure development levels for China or the United States of

America, modify even global uncertainties because these countries substantially contribute to the <u>total</u> global emission budget, e.g. China emits $\sim 1/3$ of the global anthropogenic CO₂ budget and can change global total uncertainty up to 0.5 %.

5.2 Yearly and monthly uncertainties

In order to 4.3 Effect of increasing temporal resolution from yearly to monthly

- To increase the emission temporal resolution, monthly emissions and their uncertainties were calculated combining yearly 735 emissions, monthly multiplication factors, and adapted uncertainty calculation methodology (see Section $\frac{3.32.2}{3.32.2}$). Prior yearly uncertainties were multiplied by a dimensionless uncertainty boosting parameter α (same value for each month) to compute prior monthly uncertainties, which were further used together with monthly emission budgets for countries countries monthly uncertainty calculation. Monthly uncertainties (just like yearly uncertainties) are determined by empirical formulas from IPCC (2006), hence their values depend on) with monthly emission budgets, which relate to 740 (weighted with the total number of days in a month-(e.g. even with a flat yearly cycle months with more days have higher emission budgets, i.e. month emissions are sum of daily values). To eliminate this dependency, we looked straight away at). The dimensionless uncertainty boosting parameter α is applied, see Table 108 for most common values for countries with well- and less well-developed statistical infrastructures per EDGAR sectors. "sector". Boosting parameters become active (α \neq 1)), when absolute uncertainty values are \geq 25.0 %, and α increases with the increase of absolute uncertainty following a 745 third order polynomial. For lower bound uncertainties, α has biggerlarger values and steeper growth than for upper bound uncertainties (e.g. -25.0 % $\triangleq \alpha = 1.5$ and -124.0 % $\triangleq \alpha = 2.6$; +25.0 % $\triangleq \alpha = 0.8$ and +124.0 % $\triangleq \alpha = 1.2$), and α behaves in the same way for countries with well- and less well-developed statistical infrastructures. Discrepancies in a different geographical entity's (country's) boosting parameters might be for several reasons, main ones are: (i) "sector" emissions were zero (e.g. super power plant emissions of the energy "sector" had no emissions); (ii) "sector" uncertainties were ≥ 50.0 750 % and needed to be adapted accordingly byto log-normal distribution technique (e.g. agriculture this is the case for the agricultural soils "sector" with prior uncertainties -70.7/+0.0 % both for countries with well- and less well-developed statistical infrastructures). Most significant; discrepancies in α are for agriculture soils sector (e.g. instead of lower/upper values from Table 108 for countries with well developed statistical infrastructures France has agricultural soils are France α = 1.8/3.1, United Kingdom - 1.8/7.2, China - 1.8/8.4, Japan - 1.8/10.8; instead of lower/upper values from Table 10 for countries with less well developed statistical infrastructures, Brazil has $\alpha = 1.8/0.0$, the Russian Federation – 1.8/5.6).
- 755

Table 108: Dimensionless (DN) boosting parameter uncertainties (lower and upper bounds) for countries with well- (WDS) and less well-developed (LDS) statistical infrastructures

		MWF IPCC (2006) activities per EDGAR-"sector"		Uncertainty boosting parameter, DN					
N₂				WDS countries		untries			
	group Group name			Up	Low	Up			
1	ENERGY_S	1.A.1.a (subset)	1.0	1.0	1.0	1.0			
2		1.A.1.a (rest)	1.0	1.0	1.0	1.0			
2	ENERGY_A	4.C	1.8	0.8	1.9	0.8			
	3 MANUFACTURING	1.A.2	1.0	1.0	1.0	1.0			
3		2.C.1, 2.C.2	1.7	0.8	1.7	0.8			
		2.C.3, 2.C.4, 2.C.5, 2.C.6, 2.C.7	2.0	0.9	2.0	0.9			

		2.D.1, 2.D.2, 2.D.4	2.6	1.2	2.6	1.2
		2.A.1, 2.A.2, 2.A.3, 2.A.4	2.0	0.9	2.3	1.0
		2.B.1, 2.B.2, 2.B.3, 2.B.4, 2.B.5, 2.B.6, 2.B.8	2.4	1.0	2.4	1.0
4	SETTLEMENTS	1.A.4, 1.A.5.a, 1.A.5.b.i, 1.A.5.b.ii	1.0	1.0	1.5	0.9
		1.A.3.a_CRS	1.0	1.0	1.7	1.1
5	AVIATION	1.A.3.a_CDS	1.0	1.0	1.7	1.1
		1.A.3.a_LTO	1.0	1.0	1.7	1.1
		1.A.3.b	1.0	1.0	1.0	1.0
6	TRANSPORT	1.A.3.d	1.0	1.0	1.7	0.9
		1.A.3.c, 1.A.3.e	1.7	1.1	1.7	1.1
		1.A.1.b, 1.A.1.c, 1.A.5.b.iii, 1.B.1.c, 1.B.2.a.iii.4, 1.B.2.a.iii.6, 1.B.2.b.iii.3	1.7	1.4	1.8	1.4
7	OTHER	1.B.2.a.ii, 1.B.2.a.iii.2, 1.B.2.a.iii.3, 1.B.2.b.ii, 1.B.2.b.iii.2, 1.B.2.b.iii.4, 1.B.2.b.iii.5, 1.C	3.0	2.4	3.1	2.5
		1.B.1.a	2.5	2.2	2.5	2.2
		3.C.2, 3.C.3, 3.C.4, 3.C.7	1.8	0.0	2.0	0.0
		2.D.3, 2.B.9, 2.E, 2.F, 2.G	1.5	0.8	1.7	0.9

760 In general, Brazil, Indonesia and India have a very weak yearly cycle with quite high monthly uncertainties throughout the year. Globe, Europe (28 members tilluntil end 2019), Germany, Spain, France, United Kingdom, Poland, China, Japan, the Russian Federation, and the United States of America have more pronounced yearly cycles, most significant for SETTLEMENTS and ENERGY_A (and ENERGY_S where present) "groups;", and less significant for AVIATION, TRANSPORT and MANUFACTURING "groups;". This is in line with the monthly profiles applied in EDGARv4.3.2 for Northern and Southern temperate zones, and Equator, see Janssens-Maenhout et al. (2019). In the summer months for the Northern temperate zone, a strong decrease in SETTLEMENT and ENERGY_A (and ENERGY_S where present) "groups" emissions was observed, with a light decrease in MANUFACTURING "group" emissions, and a light increase in AVIATION and TRANSPORT "groups" emissions. This corresponds rather well with the assumption that most of the population in the Northern hemisphere must-heat their houses during winter, and the work with the ability and travel more

5.34.4 Comparison for selected European countries with UNFCCC, and TNO and other data

The CHE_EDGAR-ECMWF_2015 dataset containing 7 global gridded fossil CO₂ emission flux maps, and country- and ECMWF-"group-"-specific emission budgets and uncertainties have been assessed with independent data. Global emission budget values from different datasets are almost never the same, therefore it is important to first identify why estimates differ

775 between datasets—<u>. First</u>, datasets might use <u>the</u> same country-level information as primary input, <u>neverthelessthough</u> differences in inclusion, interpretation, and treatment of that data lead to diverse results in emissions; <u>second</u>_. <u>Second</u>, try to harmonise <u>e.g.</u> data inclusion or omission across datasets to have more clarity in the discrepancies. For Europe (28 members <u>tilluntil</u> end 2019), Germany, Spain, France, United Kingdom, Poland, Japan, the Russian Federation and the United States of America, emission and uncertainty data was collected from UNFCCC NIR. The

- 780 aggregation of the IPCC (2006) activity-specific emissions and uncertainties into 7 ECMWF "groups" was done assuming no correlation, following IPCC (2006). Although IPCC (2006) has a standard table to report GHG emissions, uncertainties can be reported in less detail by a more general category (e.g. 2.D only instead of 2.D.1, 2.D.2, 2.D.3, 2.D.4), meaning information "harmonization" required lots of careful time-consuming country-specific technical work by the authors of this paper.
- The Netherlands Organisation for Applied Scientific Research (TNO) has-recently prepared the first version of their GHG and co-emitted species emission database (TNO_GHGco_v1.1) that covers the entire European domain (at 0.1°×0.05° resolution)- also-for), including CO₂ (distinguishing between fossil fuel and biofuel). Initial emission data is from the UNFCCC (Common reporting format-(,_CRF), tables) and the European Monitoring and Evaluation Programme/Centre on Emission Inventories and Projections for air pollutants (EMEP/CEIP). These data were harmonized, checked for gaps, errors
- 790 and inconsistencies, and (where needed) replaced or completed using emission data from the Greenhouse gas-Air pollution Interactions and Synergies (GAINS) model (Amann et al., 2011). Moreover, inland shipping emissions were replaced with TNO's own estimates and sea shipping is based on automatic identification system (AIS) based tracks. Expert judgement is used to assess the quality of each data source and to make choices on which source to use. The resulting emissions were checked in detail with regard toregarding their absolute value and trends (Kuenen et al., 2014). In this study we used
- 795 emission budgets from 30 TNO sectors provided by TNO (Super et al., February 2020, personal communication), and prior uncertainties calculated from IPCC (2006) and IPCC TFI (2019) see Table 11 (NB! all uncertainty calculations were done per country and only then for comparison purposes aggregated to Europe (28 members till end 2019) values assuming no correlation following IPCC (2006)).its refinements (IPCC, 2019) are used. In addition, TNO has provided Tier 2 (Monte Carlo approach) uncertainties based on the same budgets and uncertainties from submitted NIR reports based on <u>a</u> Tier 1
- 800 approach. The Monte Carlo simulations were done at the highest detail level (nomenclature for reporting (NFR) sector/fuel type) assuming correlations between certain sectors (for more information see Super et al. (2020)), and then emissions were aggregated to ECMWF "groups" assuming no correlation.

 Table 11: Prior uncertainties (lower and upper bounds) per each TNO emission sector based on IPCC (2006) and IPCC-TFI

 805
 (2019), and aggregated to the ECMWF group uncertainties for Germany (DEU) and Europe (E28)

				Prior uncertainty bounds, %			Uncertainty bounds, %			
₽	ECMWF group	IPCC (2006) activities per TNO sector	WDS co	ountries	DEU		E28			
			Low	Up	Low	Up	Low	Up		
1	ENERGY_S	1.A.1.a (subset)	8.6	3.0	0.0	0.0	0.0	0.0		
2	ENEDCY	1.A.1.a (rest)	8.6	8.6	8.6	8.6	5 3.1	2 1		
ź	ENERGY_A	4 .C	4 0.3	4 0.3	ð.0			3.1		
		1.A.2	8.6	8.6	8.3					
		2.C.1, 2.C.2	37.1	37.1						
2		2.C.3	10.2	10.2		0.0		20		
3	MANUFACTURING	2.C.4, 2.C.5, 2.C.6, 2.C.7	72.5	72.5		9.0	3.0	3.6		
		<u>2.D.2</u>	106.8	106.8						
		2.D.1, 2.D.4	50.3	50.3						

						T		
		<u>2.A.1</u>	36.7	36.7				
		<u>2.A.2, 2.A.3, 2.A.4</u>	60.7	60.7				
		2.B.1, 2.B.2, 2.B.3, 2.B.4, 2.B.5, 2.B.6, 2.B.8	107.8	89.9				
4	SETTLEMENTS	<u>1.A.4</u>	12.2	12.2	12.1	12.1	4.2	4.0
+	BETTLEMENTS	1.A.5.a, 1.A.5.b.i, 1.A.5.b.ii	0.0	0.0	12.1	12.1	4.2	4.2
		1.A.3.a_CRS	5.5	6.4				
5	AVIATION	1.A.3.a_CDS	5.5	6.4	5.5	6.4	1.9	2.2
		1.A.3.a_LTO	5.5	6.4				
		1.A.3.b	5.4	5.4				
	TDANGDODT	1.A.3.d	5.4	5.1	E 1	7.4	1.0	2.1
6	TRANSPORT	1.A.3.c	5.4	5.1	5. 4	7.4	1.8	3.1
		1.A.3.e	50.0	106.7				
		1.A.1.b	8.6	8.6				
		1.A.1.c	12.2	12.2	8.1			
		1.A.5.b.iii, 1.B.1.c, 1.B.2.a.iii.4, 1.B.2.a.iii.6, 1.B.2.b.iii.3	0.0	0.0				
7	OTHER	1.B.2.a.ii, 1.B.2.a.iii.2, 1.B.2.a.iii.3, 1.B.2.b.ii, 1.B.2.b.iii.2, 1.B.2.b.iii.4, 1.B.2.b.iii.5	176.3	267.2		19.6	3.7	12.4
		1.C	50.0	100.0				
		1.B.1.a	115.8	300.5				
		3.C.2	50.0	0.0				
		3.C.3, 3.C.4, 3.C.7	50.0	0.0				
		2.D.3, 2.B.9, 2.E, 2.F, 2.G	25.0	25.0				

Figure 6 shows emission budgets and uncertainties in <u>MtonsMegatonne</u>, and contributions in <u>%percent</u> to the total geographical entity's uncertainty for Europe (28 members tilluntil end 2019), Germany, France and United Kingdom with their original statistical infrastructure development types based on data from CHE EDGAR-ECMWF 2015 (in pink), 810 UNFCCC (in yellow), and TNO_GHGco_v1.1 Tier 1 (in blue) and Tier 2 (in green); plots for Spain and Poland are not shown here. Out of the four different sources, usually UNFCCC and TNO_GHGco_v1.1 Tier 2 uncertainties are the lowest ones and CHE EDGAR-ECMWF 2015 - the highest one. It should be noted that: (i) UNFCCC uncertainties were aggregated to ECMWF "groups" individually per each country as uncertainties are reported in a rather free form, and thus could be aggregated from different levels of precision, (ii) uncertainties for Europe (28 members tilluntil end 2019) from 815 CHE EDGAR-ECMWF 2015 are rather low as they were calculated by aggregating information of 28 countries, rather than assuming it to be a one geographical entity from the beginning as it is done in UNFCCC from 28 countries, and (iii) differences in uncertainties of CHE_EDGAR-ECMWF_2015 with other sources, especially in fuel dependent emission groups, might be due to biofuels, as CHE EDGAR ECMWF 2015 is not taking them into account (NB! other datasets do take biofuels into account), and other sources (e.g. according to UNFCCC SETTLEMENT group uncertainties for United 820 Kingdom are ±24.5 % (contributes 95 % of United Kingdom's total uncertainty), which is twice higher according to other

sources it might be explained by use of other fuels, e.g. or other fuels (e.g. wood and/or coal for residential heating). Differences in uncertainties between CHE_EDGAR-ECMWF_2015 and TNO_GHGco_v1.1 Tier 1 show additional value in

more detailed emission budget knowledge, (i.e. if we know for certain that country has no-where absence of the uncertain glass production then this rather uncertain activity can be excluded fromin the non-metallic minerals production "sector" decreases overall uncertainty calculation.). Differences in uncertainties between TNO_GHGco_v1.1 Tier 1 and TNO_GHGco_v1.1 Tier 2 show additional value in an advanced calculation technique, using a more sophisticated, data demanding Monte Carlo approach instead of simple error propagation. Overall, there is quite good agreement in emission budgets and uncertainties from different sources of emission data.

825



Germany, country with well-developed statistical infrastructure



Data	Emission budget	Uncertainty bound, %			
Data	2015, <u>MtonMt</u>	Low/Up	Range		
CHE_EDGAR- ECMWF_2015	783.7	-4.7/+8.7	±6.7		
UNFCCC _{Tier1}	794.0	-2.7/+2.7	±2.7		
TNO_GHGco _v1.1 _{Tier1}	791.2	-4.3/+4.6	±4.5		
TNO_GHGco _v1.1 _{Tier2}	791.2	-3.8/+3.6	±3.7		

France, country with well-developed statistical infrastructure



Contribution to total uncertainty,	Data	Emission budget		inty , %
	Data	2015, Mton<u>Mt</u>	Low/Up	Range
n to tot	CHE_EDGAR- ECMWF_2015	333.8	-5.1/+10.7	±7.9
Itior	UNFCCC _{Tier1}	334.1	-1.6/+1.6	±1.6
ontribu	TNO_GHGco _v1.1 _{Tier1}	340.6	-4.3/+6.4	±5.3
ŏ	TNO_GHGco _v1.1 _{Tier2}	340.6	-3.8/+3.8	±3.8



830 Figure 6: Emission budgets, uncertainties and contributions in percentage to the total uncertainty for Europe (E28), Germany (DEU), France (FRA) and United Kingdom (GBR) with their original statistical infrastructure development types

Emission budgets, Tier 1 uncertainties, and contributions in percentage to the total geographical entity's uncertainty for Japan, the Russian Federation and the United States of America from CHE_EDGAR-ECMWF_2015 could be compared

only with UNFCCC data (plots not shown here). UNFCCC uncertainties are usually lower than the ones calculated in this 835 study. Main The main reason for that is the use of country-specific emission data and activity data uncertainties, which are lower than default values suggested by IPCC (2006) and its refinements (IPCC TFI (, 2019). Only for the fuel dependent "groups" (e.g. AVIATION) might UNFCCC uncertainties might be higher than in this study, as rather uncertain biofuels might be taken into account (NB!Note: CHE EDGAR-ECMWF 2015 does not take biofuels into account). Also, emission 840 budgets reported to UNFCCC show some differences from the ones from CHE EDGAR-ECMWF 2015. For Japan-, "group" budgets agree rather well, and the total budget difference is ~1.0 %. For the Russian Federation, major differences are in the ENERGY A (and ENERGY S) and MANUFACTURING "groups,", which results in ~6.0 % higher total budget of CHE EDGAR-ECMWF 2015. For the United States of America, major differences are ~200 MtonMt and ~100 MtonMt for the SETTLEMENTS and OTHER "groups" respectively, which results in ~4.0 % higher total budget than based on 845 UNFCCC data. Recent comparison of different gridded global datasets by Andrew (2020) pointed out that only a few of these datasets provide quantitative uncertainty assessment, see the summary in Table 7. Comparing5. Compared to other global emission uncertainty values, CHE EDGAR-ECMWF 2015 shows the lowest values mainly due to the aggregation technique.

4.5.4 Sensitivity to the fuel specificity

850 As mentioned above, for transport related emission uncertainty calculations only the most typical fuel type (for aviation, railways, shipping) and emission factor uncertainty (for road and off-road transport) were used, because detailed fuel consumption information per IPCC activity was not available for this study. The EDGAR dataset development team do have specific fuel information globally, which could be used for uncertainty calculation. The EDGAR dataset with incorporated fuel-specific activity data and emission factor uncertainties and Tier 1 approach for uncertainty calculation (see 855 Supplementary Information, section S. $\frac{5}{6}$, is hereinafter referred to as EDGAR-JRC. Country budget uncertainties were calculated by considering "full fuel" splitting and by taking into consideration the assumption that the emission factor factors, from sectors sharing the same fuel, are fully correlated. This latter assumption transformed the sum in guadrature of Eq. (2) into a linear summation (Bond et al., 2004; Bergamaschi et al., 2015). The uncertainty of activity data werewas set in accordance with IPCC (2006) guidelines, in the range 5.0 to 10.0 % for combustion activities, 10.0 to 20.0 % for combustion 860 in the residential sector, 25.0 % for bunker fuels in the marine transport, 35.0 % for industrial processes of cement, lime, glass, ammonia (the range of uncertainty values refers to the 95 % confidence interval of the mean, assigned separately to countries with well- and less well-developed statistical infrastructures). Uncertainties from EDGAR-JRC dataset aggregated to the ECMWF-"group" level were compared with the ones from CHE_EDGAR-ECMWF_2015, see Table 129 for Europe (28 members tilluntil end 2019) and all world countries, and Table S8 from the Supplementary Information, section S.56, for all the rest geographical entities from Table 8. NB! Group contribution to the geographical entity's (country's) total 865 uncertainty is zero when group has no emissions⁶. Emission uncertainties from EDGAR-JRC reflect the share of fuel composing the emission of each country and are in line with the estimates by CHE_EDGAR-ECMWF_2015 for those countries where the fuel-composite uncertainty is closer to the average value assigned (see Table 4)... Uncertainties calculated with fuel-specific data are usually smaller; when prevailing fuel coincides with typical fuel type from

- 870 CHE_EDGAR-ECMWF_2015 emission "group" uncertainties from both sources are quite similar. It should be noted here that: (i) countries total uncertainty is higher in EDGAR-JRC due to the aggregation technique (full correlation is assumed), (ii) AVIATION "group" uncertainties are higher in EDGAR-JRC due to prior aggregation of all three aviation connected sectors (cruise, climbing & descent, and landing & take off).
- 875 Table 129: Aggregated to the ECMWF-"group" level uncertainties (lower and upper bounds) in <u>%percent</u> and contributions in <u>%percent</u> to the total uncertainty (CV) for Europe (E28) and globe (GLB) from EDGAR-JRC (with extra fuel type knowledge) and CHE_EDGAR-ECMWF_2015 (with typical fuel only)

Constant	ECMWF group"Group" name	EDGAR-JRC			CHE_EDGAR-ECMWF_2015			
Country		Low, %	Up, %	CV, %	Low, %	Up, %	CV, %	
	ENERGY_S	0.0	0.0	0.0	-3.6	1.0	0.0	
	ENERGY_A	-2.9	2.7	42.4	-3.5	3.5	11.0	
	MANUFACTURING	-4.3	4.3	41.3	-5.7	8.6	34.0	
GLB	SETTLEMENTS	-2.5	2.5	1.9	-3.9	3.9	1.1	
GLD	AVIATION	-4.2	5.8	0.5	-17.3	58.1	6.1	
	TRANSPORT	-2.5	2.6	7.7	-4.3	6.4	8.1	
	OTHER	-5.9	6.2	6.2	-11.5	52.4	39.7	
	TOTAL	-4.8	4.8	100.0	-2.3	4.8	100.0	
	ENERGY_S	0.0	0.0	0.0	-5.4	1.9	0.2	
	ENERGY_A	-2.0	2.4	56.4	-2.8	2.8	13.3	
	MANUFACTURING	-2.2	2.2	12.6	-3.9	5.8	20.0	
E28	SETTLEMENTS	-2.5	2.5	15.1	-4.2	4.2	8.8	
E28	AVIATION	-2.4	2.8	0.0	-1.4	1.6	0.0	
	TRANSPORT	-1.3	1.3	7.2	-1.6	1.8	2.8	
	OTHER	-5.0	5.0	8.7	-10.1	45.3	54.9	
	TOTAL	-3.3	3.6	100.0	-1.6	3.3	100.0	

The uncertainties derived in this study are an upper bound of the uncertainty estimation compared to the uncertainties calculated with more detailed information, as done by the countries and reported to UNFCCC or to the uncertainties calculated with fuel-specific data. Even though sometimes differences might be quite high in %,percentage values, they are usually quite small in Mtons. Taking into account that fuel data is not publicly available, requires a lot of time to collect and implement, and is not available globally it was decided not to use it in this study for Tier 1 uncertainty calculationsMegatonne.

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5.54.6 Atmospheric sensitivity to nationally disaggregated emissions

The gridded emissions are a required input to the ECMWF<u>IFS</u> model used to simulate atmospheric CO_2 globally (Agusti-Panareda et al., 2014; Agusti-Panareda et al., 2019). Ideally, uncertainties at a grid-cell level would be preferred by the models in general, which is a difficult time-consuming task. In order to To check if these calculations are necessary the

890 <u>usefulness of the information-intensive derivation of uncertainties at a grid-cell level,</u> it was decided to run some experiments. High-resolution (~25 km horizontal resolution, 137 vertical levels) simulations with <u>the ECMWF Integrated</u> Forecasting System (IFS) model have been performed to assess the atmospheric sensitivity to fully resolved emissions compared to nationally smoothed (global emission budget is conserved), see Figure 7.



Figure 7: Anthropogenic CO₂ flux source distribution <u>at ~25 km resolution</u> – fully resolved (left), country aggregated (right)

Model simulations were performed for January 2015 with <u>3three</u> hourly output. Anthropogenic, fire, ocean and biogenic fluxes (large-scale model BIAS mitigated by biogenic CO₂ flux adjustment scheme BFAS) were considered. For the full model configuration description see McNorton et al. (2020). The atmospheric response to using either fully or partially resolved emissions compared with nationally smoothed emissions after a 10-day period are shown in Figure 8. It was noted that point sources (e.g. power plants, factories) can be easily detected if they comprise <u>a</u> substantial part of countries total emission budget (e.g. in South Africa). If point sources are distributed homogeneously over the country and other areal sources are rather high as well, it becomes really difficult to detect one extra/missing emitting hotspot (e.g. in Germany). China is a very good example for both cases, as its western part has very <u>littlefew</u> hotspots and they are easy to detect over the low emitting background, and its. Its eastern part, however, has lots of hotspots and high emitting areal sources which makemaking it almost impossible to disentangle emissions from <u>a</u> single power plant or factory from the high emitting

background. In general, even by resolving a single sector, in this case the energy sector (see Figure 8), a difference in the atmospheric response is evident. Differences of several ppm are detected over multiple regions, highlighting the importance of using high resolution spatially resolved emissions. With increase of both flux and transport model resolutions these differences are expected to increase further with steeper atmospheric CO₂ gradients.

Fully resolved emissions (global domain) sector resolved emissions (European domain) Only energy 5 Difference in Total Column CO2, in ppm 0.1 -0.2 -0.1 -0.3 -0.1 0.3 0.1 0.2

Figure 8: Difference-5 Recommendations and conclusion

A pre-processor has been created that allows derivation of the upper and lower band uncertainty grid-maps, while making use of an appropriate classification of more certain and uncertain sectors. These grid-maps allow assessment of the error propagation of 915 country emission budgets following the IPCC 2006 Guidelines for National Greenhouse Gas Inventories. It is a first step in evaluating where to provide more effort in reducing the propagated error budget that can be taken up in any global or regional atmospheric response tomodel as a first step. The method has been applied using resolved and country aggregated emissions for January 2015 with IFS model at ~25 km resolution after 10-day simulation; the difference is calculated using both fully resolved emissions (left) or by only resolving the energy sector emissions (right)

0.3

920

910

6 Conclusions and discussion

The new CHE EDGAR-ECMWF 2015 dataset with anthropogenic fossil CO₂ emissions and their uncertaintiesEDGARv4.3.2 FT2015 and with a new 7×7 covariance matrix for the atmospheric transport model was compiled and tested. The fossil CO2 emissions include all long-cycle carbon emissions from human activities, such as fossil 925 fuel combustion, industrial processes (e.g. cement) and products use, but excludes emissions from land-use change and forestry, Human CO₂ emission inventories were processed into gridded maps to provide an estimate of prior CO₂ emissions, aggregated in 7 main emissions groups: 1) energy production super emitters, 2) energy production standard emitters, 3) manufacturing, 4) settlements, 5) aviation, 6) other transport at ground level and 7) others, with estimation of their uncertainty and covariance. For the first implementation it is assumed that each emission group is fully correlated with itself

- 930 and fully uncorrelated with any other group (only diagonal values are non zero and equal to log normal variance).
- The CHE_EDGAR ECMWF_2015 represents the 2015 global fossil CO₂ emissions prior at 0.1°×0.1° resolution that has been for the first time to our knowledge bridging the inventory community and the atmospheric modelling community. In fact, the uncertainty calculations fully respect the detailed error propagation approach recommended by IPCC (2006) guidelines for GHG inventories while these datasets as prior-input were processed such that the uncertainty information
- 935 could be fully taken up by the ECMWF model IFS. Estimation of emission uncertainties is purely based on IPCC (2006) and IPCC TFI (2019) emission factor and activity data uncertainty values and assumptions — mainly that emissions are fully uncorrelated. Uncertainties related to the spatial distribution (representativeness of the proxy data and their uncertainty) were not assessed in this study, but they can be included by the user on top of the calculated emission uncertainties. All calculations, performed for the year 2015, are documented so that the methodology and algorithms used can be easily
- 940 adapted for any other year. The dataset can be directly used in inverse modelling, and ensemble data assimilation applications, such as those envisaged within the Copernicus Atmosphere Monitoring Service (CAMS) system. CHE_EDGAR_ECMWF_2015 consists of 11 global NetCDF files with gridded yearly and monthly upper and lower bounds of uncertainties in % and kg·m⁻²·s⁻¹ per each ECMWF group and their sum, and 1 Excel file with 16 spreadsheets with the same information listed per country (metadata, emissions, uncertainties, statistical parameters).
- 945 Calculated emissions and uncertainties of fossil CO₂ have been compared to other data sets based on the country specific data reported to UNFCCC and on fuel specific data reported in the energy statistics of IEA. The global values and their uncertainty at a 2σ range for the CHE_EDGAR ECMWF_2015 dataset show the lowest value of 4.7/+9.6 % or ± 7.1 % range due to the methodology used. to the ECMWF IFS ensemble spread to characterise the carbon dioxide (CO₂) atmospheric concentrations' uncertainties in the prototype of the Copernicus CO₂ Monitoring and Verification Support
- 950 <u>Capacity.</u> At country level the CHE_EDGAR-ECMWF_2015 dataset provides generally larger uncertainty ranges, that are reduced when more detailed information is available to reduce the uncertainties; in.<u>In</u> summary, using the information that is-uniformly available for all countries, a coherent uncertainty representation is obtained.

The CHE_EDGAR ECMWF_2015 dataset has been tested to provide application in the ECMWF IFS Earth system ensemble spread to characterise the CO₂ atmospheric concentrations' uncertainties in the prototype of the Copernicus CO₂. Monitoring

- 955 and Verification Support Capacity. Annual and monthly uncertainties have been evaluated in the ECMWF's atmospheric transport model IFS ensemble simulations as well as the sensitivity tosheds light on the spatial distributionrepresentativeness of anthropogenic CO₂the emissions (McNorton et al., 2020). Results show to be rather sensitive to the spatial distribution proxies, and most updated proxies and prior uncertainties. While the emission-intensive point sources were checked w.r.t. their spatial location, the diffuse emission sources are better adapted forgridded using spatial proxy data assimilation 960 applications. This needs to be studied in a future research project, the Prototype system for a Copernicus CO₂ service
- (CoCO2), With CHE_EDGAR-ECMWF_2015 implemented in the IFS model it was demonstrated that follows the choice of

the spatial proxy data has a strong influence on the model results. As such, it is proposed that this is analysed in comparison to other datasets, going beyond the current CHE research project.

evaluation of the probability density of the spatial proxy itself. Contribution of representativeness errors to uncertainties and
 time correlation are neglected in CHE_EDGAR-ECMWF_2015 and will need to be assessed in successive future studies.
 foreseen under the Prototype system for a Copernicus CO₂ service (CoCO₂) project, following up on the CO₂ Human Emissions (CHE) project.

The estimation of global gridded emissions with their spatially and temporally distributed uncertainties constitute the backbone for atmospheric inversions to estimate anthropogenic emissions from atmospheric concentrations (Pinty et al., 2017). Dedicated satellite missions (e.g. Copernicus anthropogenic CO₂ monitoring mission CO₂M described in Janssens-Maenhout et al. (2020)) are being planned to monitor anthropogenic emissions from space and substantially reduce emission uncertainties. The developments in the emission uncertainty, based on prior knowledge computation presented in this paper is, are an important preparatory step for an ensemble-based CO₂ Monitoring and Verification System prototype, such as the one developed within the CHE project.

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Data availability. EDGARv4.3.2 data are open access and available at 29 June 20202021, http://edgar.jrc.ec.europa.eu/overview.php?v=432&SECURE=123, last access: doi:https://data.europa.eu/doi/10.2904/JRC DATASET EDGAR, documented in Janssens-Maenhout et al. (2019). CHE EDGAR-ECMWF 2015 data (Choulga et al., 2020) are freely available https://doi.org/10.5281/zenodo.3967439, and 980 documented in this paper. CHE_UNC_APP anthropogenic CO2 emission uncertainty calculation tool (Choulga et al., 2021) is freely available https://doi.org/10.5281/zenodo.5196190, and documented in this paper.

Author contribution. All the authors participated in the <u>uncertainty calculation tool CHE_UNC_APP design and</u>
 <u>CHE_EDGAR_CHE-ECMWF 2015</u> maps generation (methodology, data generation), model experiment set-up, and
 analysis of the result. Margarita Choulga and Greet Janssens-Maenhout wrote the manuscript with contributions from all the other authors.

Competing interests. The authors declare that they have no conflict of interest.

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