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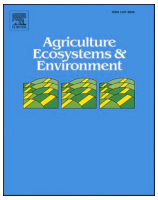

New N_2O emission factors for crop residues and fertiliser inputs to agricultural soils in Germany

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ABSTRACT

Direct agricultural N_2O emissions in Germany have so far been estimated using the default Tier 1 emission factor of 1% (0.3–3%) in accordance with the IPCC's default methodology. Since direct N_2O emissions is a "key category" in the German National Greenhouse Gas Inventory, the IPCC recommends the use of country-specific emission factors or models. With the aim of deriving country-specific and stratified N_2O emission factors, a meta-analysis was conducted using data collected from 71 individual studies comprising 676 separate emission measurements taken at 43 locations across Germany. A Bayesian generalised linear mixed-effects modelling approach was used to model N_2O fluxes and derive emission factors. In contrast to what is suggested by the 2019 Refinement to the 2006 IPCC Guidelines, the model results did not support a distinction being made between emission factors for synthetic and organic fertilisers. Instead, a model based on four environmental zones roughly representing the north-west, north-east, south-east and south-west parts of the country was developed. It was used to derive district-wise emission factors for direct N_2O emissions and revealed that northern districts had relatively lower emission factors than southern districts. The district-wise emission factors ranged from 0.38% to 0.92%. The national implied emission factor for direct N_2O emissions from managed agricultural soils was 0.62% (0.43–0.85%). Accordingly, the estimate of German national GHG emissions from agriculture in 2015 is 8.59% (calculated with global warming potentials from IPCC's fifth assessment report) lower than the estimate reported in the 2021 inventory submission to UNFCCC.

1. Introduction

Nitrous oxide (N_2O) is a strong greenhouse gas and catalyses the depletion of ozone in the stratosphere (IPCC, 2014a; Ravishankara et al., 2009). It is the third strongest contributor to global warming after carbon dioxide (CO_2) and methane (CH_4) (WMO, 2021). With an atmospheric lifetime of 131 years, it has a global warming potential (GWP) 265 times that of CO_2 over a 100-year time horizon (IPCC, 2014a). Approximately 40% of global N_2O emissions have anthropogenic sources with more than half of these being direct emissions from fertiliser nitrogen (N) inputs in agriculture (Tian et al., 2020; WMO, 2021).

As the basis of efforts to limit global warming (COP, 2015), accurate inventories of anthropogenic GHG emissions are essential. Signatories to the United Nations Framework Convention on Climate Change (United Nations, 1992) and in particular countries listed as Annex I parties therefore submit national emission inventories annually to the UNFCCC secretariat. Germany is one such country.

The Intergovernmental Panel on Climate Change (IPCC) provides guidelines for countries to estimate N_2O emissions along with other GHGs for reporting in their national inventories. The guidelines provide three "Tiers" based on the depth of data and scientific knowledge available to estimate emissions. Tier 1 uses global default emission factors (EFs), Tier 2 uses stratified or country-specific EFs, and Tier 3 uses measurements or complex modelling approaches requiring process-specific data at high resolutions (IPCC, 2006). An EF expresses the emission released by a specific and standard quantity of activity or input. In the case of direct N_2O emissions from N inputs to agricultural soils, the emission factor is the proportion of N input emitted as N_2O. Tier 1 (the lowest tier) uses a default emission factor (1% of N input) derived from global N_2O measurements (IPCC, 2019, 2006). The progression from lower to higher tiers increases the accuracy and also often reduces the uncertainty in estimating N_2O emissions and is therefore

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recommended by the IPCC guidelines.

The default Tier 1 EF for direct N_2O emissions from N inputs to soils (EF_1) has evolved over the years. The IPCC's revised 1996 guidelines (IPCC, 1997) had estimated an emission factor of 1.25% (0.25–2.25%) but related this value to the N input remaining in soil after ammonia volatilisation and nitric oxide emissions. With new N_2O measurement studies and data becoming available, the 2006 IPCC guidelines (IPCC, 2006) modified EF_1 to 1% (0.3–3%) of N input without deducting ammonia and nitric oxide losses. In 2019, the IPCC reduced the uncertainty range of EF_1 to 0.1–1.8% and published a disaggregated Tier 1 approach that stratifies by climate ("wet" and "dry") and within wet climates between synthetic fertilisers and other, i.e. organic or mixed N inputs (IPCC, 2019). This disaggregated Tier 1 approach increases EF_1 for synthetic fertilisers to 1.6% (1.3–1.9%) and decreases it for other N inputs to 0.6% (0.1–1.1%).

Germany is the second largest contributor of direct N_2O emissions from managed agricultural soils in Europe. For 2019, Germany reported direct N_2O emissions of 67.29 kt N_2O, which is 14.8% of Europe's N_2O emissions and is only surpassed by France's contribution of 19.1% (European Environment Agency, 2021). The direct N_2O emissions from agricultural soils in 2019 are 71.21% of the total N_2O emissions from German agriculture, which include N_2O emissions from livestock farming and manure management as well as indirect N_2O emissions from nitrate leaching and N volatilisation. N_2O emissions from agricultural soils are classified as a "key category" in the German national greenhouse gas inventory due to their significant contribution to overall emissions and the emission trend over time (Federal Environment Agency, 2021). Despite being part of a key category and the IPCC guidelines recommending the use of a higher tier approach for key categories, direct N_2O emissions in Germany are still being estimated using the default Tier 1 EF. While it may be suitable for estimating emissions at a global level, the Tier 1 approach does not accurately represent the actual direct N_2O emissions for many countries. Top-down estimates of N_2O emissions derived from inverse modelling based on atmospheric gas concentrations are not in good agreement with bottom-up estimates derived from inventory approaches at regional and sub-regional levels (Shcherbak et al., 2014; Tian et al., 2020). While there is good agreement at the global level and some regions such as South and South-East Asia, the top-down estimates are substantially lower than the bottom-up estimates for Europe (Tian et al., 2020), which could indicate an overestimation due to the Tier 1 EF_1 being too high for Europe. This disagreement therefore emphasises the need to derive country-specific Tier 2 EFs and investigate the appropriateness of the default Tier 1 EF for estimating country level N_2O emissions of European countries such as Germany.

Country-specific emission factors can only be derived when there is adequate availability of N_2O measurement data and sufficient stratified activity data, and are therefore difficult for many countries to achieve. As of 2021, only nine of the 43 UNFCCC Annex I countries have adopted Tier 2 or Tier 3 approaches to estimate N_2O emissions (UNFCCC, 2021). However, numerous N_2O measurement studies have been conducted over the past three decades throughout Germany, making it feasible to derive a national emission factor for the country.

The Tier 2 method requires stratified emission factors corresponding to variables influencing N_2O emissions. N_2O emissions from agricultural soils result primarily from microbial processes in the nitrogen cycle. Under aerobic conditions, ammonia is oxidised to nitrate via the intermediates hydroxylamine and nitrite, with N_2O produced as a by-product during hydroxylamine oxidation (Wrage et al., 2001). Even more important for N_2O emissions is the anaerobic process of denitrification, which is the reduction of nitrate to N_2. Since N_2O is an intermediate of this process and many microorganisms, in particular fungi, do not produce N_2O reductase, large quantities of N_2O can be emitted as a result of incomplete denitrification (Wrage et al., 2001). While nitrification and denitrification are the dominant N_2O forming processes, other microbial and chemical processes can be of relevance locally (Butterbach-Bahl et al., 2013). Due to the complex interaction of microbial processes, many factors are known to have an impact on N_2O emissions. These include substrate nitrogen species, soil pH, soil organic carbon (SOC), soil texture, soil moisture and temperature, and freeze-thawing events (Liu et al., 2010; Risk et al., 2013). Various drivers of N_2O emissions have been identified in different countries. The United Kingdom, for example, has identified fertiliser type (urea-based and other fertilisers) and annual rainfall to be primary drivers of N_2O emission whereas moisture regimes and topographic conditions are among the influential factors in Canada (Brown et al., 2021; Environment and Climate Change Canada, 2021). Previous meta-analyses of N_2O emissions in Germany have given preliminary knowledge about the magnitude and variability of N_2O emissions and about the main factors influencing them. Dechow and Freibauer (2011) identified climate and soil properties to be key influencers of N_2O emissions and found the average emission factor for N_2O emissions from mineral soils to be about 0.9%. Jungkunst et al. (2006) estimated a mean emission factor of 1.56% (median 0.63%), with the emission factor significantly varying according to the country's climatic zones.

The objective of this study was to develop a new Tier 2 methodology to estimate German N_2O emissions associated with N input based on a meta-analysis of scientific results on N_2O emissions from agricultural soils. The meta-analysis was designed to identify the major factors influencing N_2O emissions in Germany and thereby provide a robust basis for determining country-specific Tier 2 emission factors. Particular attention was given to ensuring the methodology was in line with IPCC guidelines and tailoring the approach so that it could be part of the preparation of Germany's national greenhouse gas inventory.

2. Materials & methods

2.1. Data collection and organisation

Data collection started with the N_2O measurement data used in the 2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories consisting of 332 studies, of which 23 were from Germany. A literature review of N_2O measurement studies conducted in Germany was undertaken to increase the number of studies and expand the total number of measurements. A "measurement" hereinafter refers to the mean of all replicate N_2O measurements belonging to a treatment in a study. The scientific literature databases *Web of Science* and *Google Scholar* were searched for peer-reviewed literature using keywords 'N_2O' and 'Germany'. Apart from the exclusion criteria adopted in the IPCC's original database, such as the exclusion of studies conducted in the laboratory, organic soils, grazing studies, modelling studies and studies involving slow-release or inhibitor-stabilised fertilisers, three further exclusion criteria were included: studies pertaining to natural N_2O sources, studies whose measurement period was less than 150 days, and measurements that were taken from fallow land and legumes. Measurements with legumes were excluded because the additional N input from N fixation could generally not be quantified with the available data. Unlike the IPCC database, studies conducted on organic soils were included in the data collection.

In order to have a sufficient number of measurements in each location and make the modelling process more robust, measurement locations close to each other were grouped together to form location clusters. In view of the different emission dynamics between mineral and organic soils, the database was split into two different data sets for mineral and organic soils. Following the German national greenhouse gas inventory definition (Federal Environment Agency, 2021), locations with SOC content greater than 9% were classified as organic soils and the rest categorised as mineral soils. This national threshold is slightly lower than the IPCC default threshold of 12% (IPCC, 2014b).

Because cumulated emissions can be expected to correlate with measurement period, the measurement period was included in the database as a separate 'short-by' variable. By including this variable in

the models, it is possible to account for the confounding effect of measurement period and the transformation (short-by = 365 d – measurement period) allows extracting annual emission factors directly from the model fit.

Weather data from Germany's national meteorological service (DWD) were used to calculate long-term annual average precipitation and temperature (1990–2019) for studies lacking this information. Data from the German agricultural soil inventory (BZE-LW) were used to fill in missing SOC and pH values. Soil parameters for the depth 0–30 cm were included in the database and calculated from more detailed data where necessary. Locations were categorised according to their environmental conditions, using the environmental stratification zones from Metzger et al. (2005). A few of these zones were merged to ensure each of them contained a sufficient number of data points for modelling (Fig. 1a). There is no agricultural land use in the Alpine zone.

2.2. Modelling methods

Modelling was performed separately for mineral and organic soils. The modelling approach followed that of Bouwman (1996) who modelled N_2O emissions as a linear function of N application rates, making EF the slope of this model. The advantage of such a linear model is that it ensures that using the sum of N inputs to soil as the model input results in the sum of emissions, i.e. $\Sigma f(x) = f(\Sigma x)$, which is not the case for non-linear models. N_2O emissions generally exhibit a positively skewed pattern, with extreme values towards the higher tail. Previous studies, such as Stehfest and Bouwman (2006) and Walter et al. (2015), have handled this by log-transforming N_2O emissions, which, after back-transformation results in an exponential model. However, in the present study, N_2O emissions were not transformed, in order to preserve linearity. Instead, a generalised linear mixed model (GLMM) with the gamma distribution family and an identity link function was used to account for the skewness. A gamma distribution can approach varying levels of skewness (from symmetric to heavily skewed) depending on its shape parameter, which is estimated during model fitting. The mixed-effects model approach was used to account for correlation within location clusters and within the measurement years by including these as random effects.

The model fitting process was undertaken using the Bayesian approach. This allows prior knowledge about model parameters to be fed into the model as a prior probability distribution ('prior') which is then updated using the available data and results in a posterior probability distribution ('posterior'). Another advantage of this modelling method is that convergence can be difficult to achieve with complex generalised linear mixed-effects models and the underlying Markov chain Monte Carlo (MCMC) sampling algorithm of the Bayesian model helps avoid such difficulties by sampling over many chains, thereby increasing the likelihood of a successful fit. The Bayesian models were run in the R statistical computing platform (version 4.0.5, R Core Team, 2021) using the 'rstanarm' package (version 2.21.1 (Goodrich et al., 2020; Stan Development Team, 2020)).

Several models were constituted with a combination of potential predictors based on similar N_2O emission factor studies in Germany and other countries. 'Location' and 'measurement year' remained as random effects in all the models. The priors assigned to the fixed effects were primarily based on the 2019 refinement to the IPCC guidelines, where available, or otherwise on our own expert judgement.

Based on the availability of sufficient and necessary activity data, two types of models were formulated for the mineral soil subset: inventory-specific models that are in line with IPCC guidelines and are suitable for available national data, and a field-level model where the availability of activity data is not a constraint. A further separate model was formulated for the organic soil data subset to derive a separate N_2O EF for organic soils. An overview of the various generalised linear mixed-effects models derived for mineral and organic soils is given in Table 1.

For each model, the root mean squared error of the model fit ($RMSE_{fit}$) showing goodness of fit was calculated. In addition, the predictive performances of the models were evaluated using a k-fold cross-validation approach (with folds corresponding to location clusters) which resulted in the root mean squared error of cross-validation ($RMSE_{cv}$). Besides the root mean squared error of the model fit and cross-validation, a PSIS-LOO (Pareto smoothed importance sampling leave-one-out; Vehtari et al., 2017) cross-validation was also performed on the models to give a LOO information criterion (LOOIC) estimate. While the LOOIC values do not have any intrinsic meaning for individual models, they allow the models' performance to be compared.

2.3. Derivation and application of emission factors

The N_2O EFs for mineral and organic soils were averaged for each district (NUTS level 3 region; European Community, 2003), based on their respective agricultural area, to derive a unified N_2O EF for each district. The agricultural area (which includes cropland and grassland) of organic and mineral soils was taken from the database of the German LULUCF inventory (Federal Environment Agency, 2021).

The regional (district-wise) distribution of organic fertilisers and N inputs, i.e. biogas digestates, sewage sludge, manure and crop residues, was taken from the database of the German agricultural GHG inventory (Rösemann et al., 2021). Regional data on synthetic fertiliser applications are not available in Germany. There are statistics on synthetic fertiliser sales for NUTS level 2 regions, but due to interregional fertiliser trading these do not represent actual synthetic fertiliser use. Therefore, data on the regional distribution of synthetic fertiliser input for the period 2014–2016 were derived from regional agricultural nitrogen balances, which were modelled at municipality level. The Regional Agricultural and Environmental Information System (RAUMIS) was used as the model framework. RAUMIS is a regional agricultural supply model that depicts agricultural production and income at district level (NUTS level 3), ensuring consistency with regional and sectoral statistical information (Gömann et al., 2002; Henrichsmeyer et al., 1996). The model encompasses a series of environmental indicators such as spatial nutrient balances, that have been previously applied in nitrogen-related policy analyses in several studies (Ackermann et al., 2016; Henseler and Dechow, 2014; Kreins et al., 2007).

Municipality-level agricultural nitrogen flows were modelled using municipality-level statistical land use and livestock data from the Thünen Agraratlas,[1] regional yields, spatially explicit data on biogas plants, and data on inter-regional manure transportation as the core database. Nitrogen uptake from grazing and grassland management was modelled by a simplified balance of demand and supply of roughage as a feedstock to livestock and biogas plants.

Regional synthetic fertilisation was derived using linear, yield-depending nutrient requirement functions (Kreins et al., 2010). Site-specific characteristics affecting synthetic fertiliser requirements were included by deriving a local site factor (Henrichsmeyer et al., 1996; Krüll, 1988). RAUMIS calculates synthetic fertiliser inputs (S) as

$$S_m = \beta * R_m - \alpha * O_m,$$

where *m* indicates the municipality, β is a global adjustment factor, *R* is the aggregated crop-specific and site-specific nitrogen requirement derived from harvest data taking the preceding crop and nitrogen fixation into consideration, and *O* is the total amount of organic nitrogen applied (Henrichsmeyer et al., 1996). Only a part of this organic nitrogen is available for the crop. This available proportion *α* and the factor β were calibrated in such a way that aggregated regional fertiliser inputs

[1] Processed data. Original data from Statistische Ämter des Bundes und der Länder (2018) derived with the method from Gocht and Röder (2014). Version 2020.

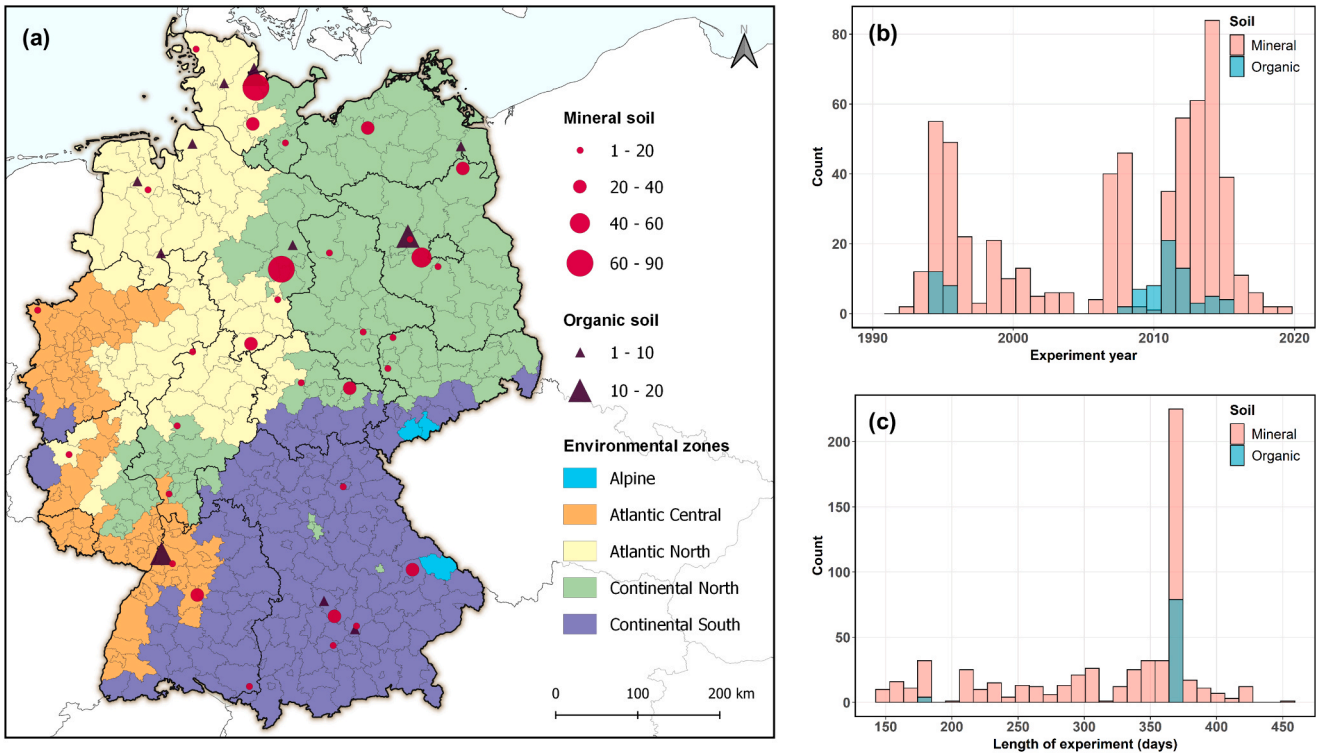

Fig. 1. (a) Map of Germany showing the five environmental zones and the location clusters of measurements. Symbol size indicates the number of measurements. Borders of federal states and districts are indicated. (b) Number of data points per year. (c) Histogram showing the measurement period.

equalled the averaged national statistics on synthetic nitrogen fertiliser sales (Statistisches Bundesamt, 2016) for the observed time period. The model ensures that the amount of mineral fertiliser applied is at least 10% of the nitrogen amount required, in every region. This model parameter, determined by experts from agricultural authorities based on documentation of previously mandatory farm-level "nutrient comparisons" (Gömann et al., 2020), reflects the minimum amount of mineral fertiliser assumed to be necessary for an effective crop management. Other model parameters (e.g. nitrogen contents of crops, nitrogen excretion per animal, share of crop residuals) were taken from (KTBL, 2018) and the German Fertilizer Ordinance (Düngeverordnung, 2021). The model does not allow for a distinction between different types of synthetic nitrogen fertiliser.

District-wise N inputs that were consistent with the emission inventory were derived by dividing the modelled synthetic fertiliser use in each district for the years 2014–16 by the modelled national total, and multiplying the resulting values by the total N input taken from the national GHG inventory for the year 2015. The district-wise sum of N inputs (synthetic fertilisers, animal manure, sewage sludge, biogas digestate and crop residue) were then multiplied by the corresponding district level unified EFs to derive total N_2O emissions for each district and thereby the whole country. The implied emission factors (IEF) for the individual and overall N inputs were calculated by dividing total emissions by the appropriate national sum of each N source. Uncertainties in N_2O emission values were derived using the Monte Carlo simulation based on Markov chain results from the model fit (i.e., Model 4 in Table 1). This approach only takes the uncertainty of the emission model into account and not the uncertainty of the spatial distribution of N inputs. The GWPs from the fifth assessment report of the IPCC (IPCC, 2014a) were used to convert emission units from N_2O and CH_4 to CO_2 equivalents.

3. Results

3.1. N_2O data

Of the 71 studies (676 measurements) in the database, most of the N_2O measurements had been performed on mineral soils (59 studies; 593 measurements). Although N_2O measurements on mineral soils had been taken throughout the country in 32 location clusters, about 75% of these measurements were from northern Germany in the Atlantic North and Continental North environmental zones (Fig. 1a). The measurements were taken over a wide temporal range between 1992 and 2019 (Fig. 1b), with most of the studies having measurement periods of more than 300 days (Fig. 1c). 22% of the measurements were from unfertilised control treatments and 63% of studies included such a control treatment. The pH of mineral soils ranged from 4.7 to 7.7 with a mean of 6.3 and the soil organic carbon (SOC) content was in the range of 0.5–4.7% with a mean of 1.57%.

In addition to grasslands (16% of measurements), prominent crops grown during the measurement period were winter wheat/barley (22% of measurements), winter oilseed rape (15% of measurements) and maize (21% of measurements). Where measurements involved fertilisation, most of the nitrogen fertilisation was done through ammonium nitrate fertilisers and the rest split almost equally between cattle manure, biogas digestate and a mixture of organic and synthetic fertilisers. Ammonium nitrate fertilisers are the most common fertiliser category used in Germany. In 2015, the share of calcium ammonium nitrate (CAN) in total fertiliser nitrogen sales was 35.6% and that of urea ammonium nitrate solution (UAN) was 9.5% (Statistisches Bundesamt, 2016).

Eighty-three N_2O measurements from 12 studies were made on organic soils. Similar to the mineral soils, approximately 70% of these measurements were taken in northern Germany, where most of the organic soils are located. The measurements were conducted between 1994 and 2015 and most of them had a measurement period of 365 days except for four measurements that covered 184 days. Soil pH in the 11

Table 1
Linear mixed-effects models tested for direct N_2O-N emissions. Posterior means for the intercept and slope are in units of kg N_2O-N ha^{-1} yr^{-1} and kg N_2O-N/ kg N applied respectively. Some illustrative plots of the model components (priors and posteriors) can be found in the supplement.

Model No.	Linear predictor (fixed effects)	Random effects		Parameters	Priors (normal distribution with (mean, S.D.))	Posterior mean (credible interval)	$RMSE_{fit}$ (kg N_2O-N ha^{-1} yr^{-1})	LOOIC	$RMSE_{CV}$ (kg N_2O-N ha^{-1} yr^{-1})
		On intercept	On slope						
Mineral soils									
1	$\beta_0 + \beta_1 \bullet X_1 + \beta_2 \bullet X_2$	by location, by year	N input by location	β_0: Intercept (emission at zero fertiliser input)	N(1,0.5)	1.62 (1.11, 2.19)	2.16	1799	2.85
				β_1: Slope vs N input (emission factor)	N(0.01,0.0041)	0.0054 (0.0035, 0.0079)			
				β_2: Slope vs Experiment length ("short-by")	N(0,1)	−0.0022 (−0.0045, 0.0001)			
2	$\beta_0 + [\beta_{10} + \beta_{11} \bullet (X_2 == \text{“mixed fertiliser”}) + \beta_{12} \bullet (X_2 == \text{“organic fertiliser”})] \bullet X_1 + \beta_2 \bullet X_3$	by location, by year	N input by location	β_0: Intercept (emission at zero fertiliser input - synthetic fertiliser)	N(1,0.5)	1.80 (1.26, 2.43)	2.10	1808	3.25
				β_{10}: Slope vs N input (emission factor - synthetic fertiliser)	N(0.016,0.0015)	0.0125 (0.0094, 0.0156)			
				β_{11}: Slope vs N input (difference between synthetic and mixed fertiliser)	N(0.011,0.0051)	−0.0013 (−0.0035, 0.0009)			
				β_{12}: Slope vs N input (difference between synthetic and organic fertiliser)	N(0.006,0.0025)	−0.0009 (−0.0025, 0.0006)			
				β_2: Slope vs Experiment length ("short-by")	N(0,1)	−0.0018 (−0.0041, 0.0004)			
3	$\beta_0 + [\beta_{10} + \beta_{11} \bullet (X_2 == \text{“mixed fertiliser”}) + \beta_{12} \bullet (X_2 == \text{“organic fertiliser”})] \bullet X_1 + \beta_2 \bullet X_3$	by location, by year	N input by location	β_0: Intercept (emission at zero fertiliser input - synthetic fertiliser)	N(1,0.5)	1.62 (1.12, 2.18)	2.16	1804	2.85
				β_{10}: Slope vs N input (emission factor - synthetic fertiliser)	N(0.01,0.0041)	0.0055 (0.0035, 0.0080)			
				β_{11}: Slope vs N input (difference between synthetic and mixed fertiliser)	N(0,0.0041)	−0.0003 (−0.0023, 0.0019)			
				β_{12}: Slope vs N input (difference between synthetic and organic fertiliser)	N(0,0.0041)	0.0002 (−0.0013, 0.0017)			
				β_2: Slope vs Experiment length ("short-by")	N(0,1)	−0.0022 (−0.0045, 0.0001)			
4	$[\beta_{00} + \beta_{01} \bullet (X_2 == \text{“Atlantic North”}) + \beta_{02} \bullet (X_2 == \text{“Continental North”}) + \beta_{03} \bullet (X_2 == \text{“Continental South”})] + [\beta_{10} + \beta_{11} \bullet (X_2 == \text{“Atlantic North”}) + \beta_{12} \bullet (X_2 == \text{“Continental North”}) + \beta_{13} \bullet (X_2 == \text{“Continental South”})] \bullet X_1 + \beta_2 \bullet X_3$	by location, by year	N input by location	β_{00}: Intercept (emission at zero fertiliser input - Atlantic Central)	N(1,0.5)	1.78 (1.11, 2.49)	2.16	1800	2.78
				β_{01}: Intercept (difference between Atlantic Central and Atlantic North)	N(0,0.5)	−0.36 (−1.08, 0.37)			
				β_{02}: Intercept (difference between Atlantic Central and Continental North)	N(0,0.5)	−0.35 (−1.04 to 0.34)			
				β_{03}: Intercept (difference between Atlantic Central and Continental South)	N(0,0.5)	0.24 (−0.53, 1.01)			
				β_{10}: Slope vs N input (emission factor - Atlantic Central)	N(0.01,0.0041)				

(*continued on next page*)

Table 1 (*continued*)

Model No.	Linear predictor (fixed effects)	Random effects: On intercept	Random effects: On slope	Parameters	Priors (normal distribution with (mean, S.D.))	Posterior mean (credible interval)	$RMSE_{fit}$ (kg N_2O-N ha^{-1} yr^{-1})	LOOIC	$RMSE_{CV}$ (kg N_2O-N ha^{-1} yr^{-1})
						0.0072 (0.0037, 0.0108)			
				β_{11}: Slope vs N input (difference between Atlantic Central & Atlantic North)	N(0,0.0041)	−0.0023 (−0.0061, 0.0017)			
				β_{12}: Slope vs N input (difference between Atlantic Central & Continental North)	N(0,0.0041)	−0.0033 (−0.0071, 0.0006)			
				β_{13}: Slope vs N input (difference between Atlantic Central & Continental South)	N(0,0.0041)	0.0017 (−0.0036, 0.0071)			
				β_2: Slope vs Experiment length ("short-by")	N(0,1)	−0.0021 (−0.0044, 0.0003)			
5	$[\beta_{00} + \beta_{01} \bullet (X_2 == \text{“Atlantic North”}) + \beta_{02} \bullet (X_2 == \text{“Continental North”}) + \beta_{03} \bullet (X_2 == \text{“Continental South”}) + \beta_{04} \bullet (X_3 == \text{“grassland”}) + \beta_{05} \bullet (X_3 == \text{“other crops”}) + \beta_{06} \bullet (X_4 == \text{SOC} < 1) + \beta_{07} \bullet (X_5 == \text{pH} < 6) + \beta_{08} \bullet (X_6 == \text{"loam"}) + \beta_{09} \bullet (X_6 == \text{"sand"})] + [\beta_{10} + \beta_{11} \bullet (X_2 == \text{“Atlantic North”}) + \beta_{12} \bullet (X_2 == \text{“Continental North”}) + \beta_{13} \bullet (X_2 == \text{“Continental South”}) + \beta_{14} \bullet (X_3 == \text{“grassland”}) + \beta_{15} \bullet (X_3 == \text{“other crops”})] \bullet X_1 + \beta_2 \bullet X_7$	by location, by year	N input by location	β_{00}: Intercept (emission at zero fertiliser input - Atlantic Central; winter crops; SOC >1; pH >6; clay/silt)	N(1,0.5)	1.26 (0.57, 1.96)	2.03	1712	2.71
				β_{01}: Intercept (difference between Atlantic Central and Atlantic North)	N(0,0.5)	−0.45 (−1.12, 0.24)			
				β_{02}: Intercept (difference between Atlantic Central and Continental North)	N(0,0.5)	−0.27 (−0.91, 0.39)			
				β_{03}: Intercept (difference between Atlantic Central and Continental South)	N(0,0.5)	0.15 (−0.58, 0.90)			
				β_{04}: Intercept (difference between winter crops and grassland)	N(0,0.5)	0.51 (0.16, 0.84)			
				β_{05}: Intercept (difference between winter crops and other crops)	N(0,0.5)	0.51 (0.23, 0.79)			
				β_{06}: Intercept (difference between SOC >1 and SOC <1)	N(−0.5,0.5)	0.19 (−0.16, 0.53)			
				β_{07}: Intercept (difference between pH >6 and pH <6)	N(0.5,0.5)	0.40 (0.09, 0.73)			
				β_{08}: Intercept (difference between clay/silt and loam)	(0.2, 0.5)	0.39 (−0.03, 0.82)			
				β_{09}: Intercept (difference between clay/silt and sand)	(−0.2,0.5)	−0.46 (−0.84, −0.10)			
				β_{10}: Slope vs N input (emission factor - Atlantic Central)	N(0.01,0.0041)	0.0064 (0.0027, 0.0103)			
				β_{11}: Slope vs N input (difference Atlantic Central & Atlantic North)	N(0,0.0041)	−0.0015 (−0.0060, 0.0029)			
				β_{12}: Slope vs N input (difference Atlantic Central & Continental North)	N(0,0.0041)	−0.0028 (−0.0069, 0.0014)			
				β_{13}: Slope vs N input (difference Atlantic Central & Continental South)	N(0,0.0041)	0.0013 (−0.0039, 0.0067)			
					N(0,0.0041)				

(*continued on next page*)

Table 1 (*continued*)

Model No.	Linear predictor (fixed effects)	Random effects		Parameters	Priors (normal distribution with (mean, S.D.))	Posterior mean (credible interval)	$RMSE_{fit}$ (kg N_2O-N ha^{-1} yr^{-1})	LOOIC	$RMSE_{CV}$ (kg N_2O-N ha^{-1} yr^{-1})
		On intercept	On slope						
				β_{14}: Slope vs N input (difference winter crops & grassland)		0.0017 (−0.0009, 0.0044)			
				β_{15}: Slope vs N input (difference winter crops & other crops)	N(0,0.0041)	0.0033 (0.0011, 0.0055)			
				β_2: Slope vs Experiment length ("short-by")	N(0,1)	−0.0037 (−0.0056, −0.0018)			
				Organic soils					
6	$\beta_0 + \beta_1 \bullet X_1 + \beta_2 \bullet X_2$	by location, by year	N input by location	β_0: Intercept (emission at zero fertiliser input)	N(7.85,16.8)	4.58 (2.13, 7.45)	6.45	402	–
				β_1: Slope vs N input (emission factor)	N(0.01,0.0041)	0.0101 (0.0039, 0.0165)			
				β_2: Slope vs Experiment length ("short-by")	N(0,1)	−0.0111 (−0.0286, 0.0026)			

organic soil location clusters was in the range of 3.3–7.2 (mean 5.4) and SOC was in the range of 9.3–51.7% (mean 24.6%). Most of the N_2O measurements were taken on grasslands (82% of measurements) and a few on maize cropped lands (13% of measurements). Of these 83 measurements, 35 were taken under zero fertilisation conditions (83% of studies included such a control treatment) while the remainder involved ammonium nitrate fertilisers and cattle manure.

3.2. N_2O emission models for mineral soils

The IPCC Tier 1 method expresses N_2O emissions only as a function of N input. A corresponding model, only taking N inputs and the short-by parameter into account (Model 1; Table 1), yielded an emission factor of 0.54% (0.35–0.79%). The negative coefficient of the short-by parameter confirmed the expectation that cumulated N_2O emissions in measurements conducted for more than a year are higher, while shorter measurement periods correspond with lower emissions. The random intercepts grouped by measurement year appeared to be completely random which suggests the absence of a temporal trend that might be expected due to developments in measurement methodology or due to climate change. In Model 2 (Table 1), N input was then stratified by fertiliser type, i.e. synthetic and organic fertilisers as suggested in the IPCC (2019) refinement's disaggregated Tier 1 approach. Fertilisation for measurements that included both synthetic and organic fertilisers were categorised as mixed N inputs. The stratified EFs proposed by the 2019 refinement were taken as priors for synthetic fertilisers (1.6%) and organic (0.6%) N inputs. For mixed N inputs, the mean of the two EFs (1.1%) was used. The model contained the EF of synthetic fertilisers and the differences between EFs of other N inputs and synthetic fertilisers as slope parameters. The posterior emission factor for synthetic fertilisers was 1.25% (0.99–1.51%) and the emission factors for mixed and organic fertilisers were 1.12% (0.81–1.44%) and 1.16% (0.87–1.45%), respectively (Fig. 2, which shows the difference between EFs). Differences in EF between fertilisers were barely significant and the EFs were much higher than the overall EF derived from Model 1. This can be attributed to the high EF with narrow uncertainties, which was used as the prior for synthetic fertilisers, thus allowing only extremely small probabilities of a low EF.

The same model was fit with neutral priors, i.e. EF = 1% for synthetic, mixed and organic fertilisers. The neutral priors were chosen in such a way that they assign non-zero probabilities to the 2019 refinement result. Using the neutral priors resulted in a very different fit (Model 3; Table 1). The emission factor of synthetic fertilisers (0.55%; 0.37–0.75%) was almost the same as that of the EF from Model 1. Similar to Model 2 with strong priors for fertilisation, the emission factors for mixed (0.52%; 0.29–0.77%) and organic (0.56%; 0.37–0.78%) fertilisers were practically identical to that of synthetic fertiliser (Fig. 2). Both models involving fertiliser types did not have better performance metrics than the basic model, which indicates that the type of fertiliser had practically no impact on N_2O emissions.

Environmental zones, however, did have a strong effect on N_2O emissions (Model 4; Table 1). N input had the default 1% EF as prior and neutral priors were used for the differences in EF between the environmental zones. The EFs of the environmental zones were clearly distinguishable from a geographic perspective. Continental South had the highest EF (0.88%, 0.38–1.43%) followed by Atlantic Central (0.72%, 0.37–1.08%), Atlantic North (0.49%, 0.26–0.78%) and Continental North (0.39%, 0.17–0.66%). Thus, a clear divide was seen between the EFs of the environmental zones in northern and southern Germany. While having a slightly higher LOOIC value than the base model, the model with environmental zones had an identical RMSE of model fit and a marginally better predictive performance RMSE. Thus, although the performance was similar to Model 1, this model was preferable because it allowed the geographic distribution of N input to be taken into account, thereby increasing the accuracy of estimations of total national emissions.

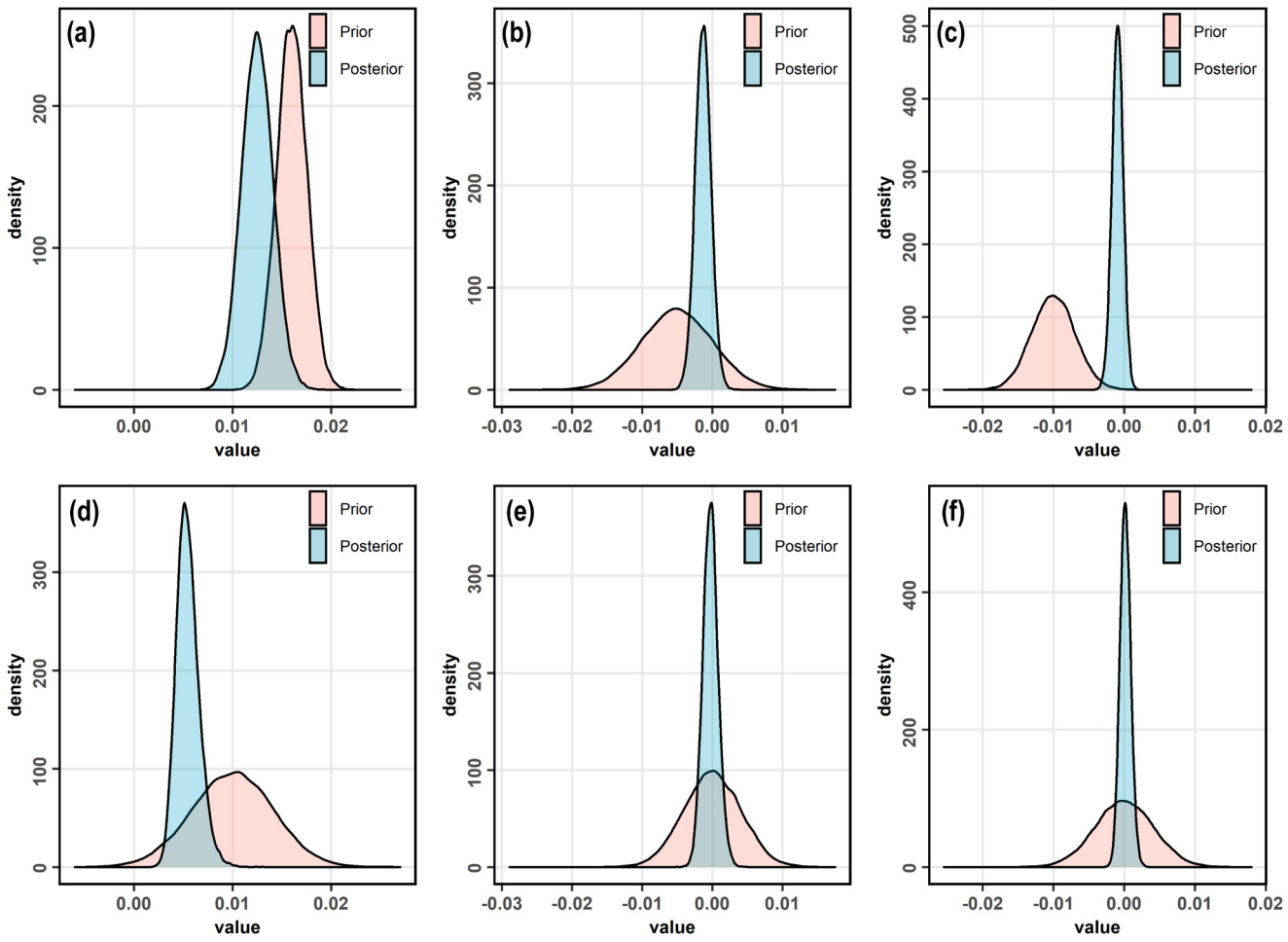

Fig. 2. Comparison of prior vs posterior distributions of Model 2 (a,b,c) with strong priors for fertiliser types and Model 3 (d,e,f) with neutral priors. Distributions in a and d represent the emission factor for synthetic fertilisers; b and e represent the difference between emission factors of synthetic and mixed fertilisers; c and f represent the difference between emission factors of synthetic and organic fertilisers.

A more complex model included the variables crop categories, SOC, pH and texture in addition to environmental zones (Model 5; Table 1). The crop types were consolidated and categorised as 'winter crops', 'grassland' and 'other crops'. Clay and silt texture classes were grouped together to form 3 texture classes: 'sand', 'loam' and 'clay/silt'. SOC and pH were grouped into two discrete classes, [<1; >1] and [<6; >6] respectively, so that measurements were split uniformly in each class. This model, whose predictors were similar to that of Stehfest and Bouwman (2006) except for fertiliser types, performed marginally better than all the other models. The EFs for winter crops in this model were similar to those of Model 4 whereas the EFs for grasslands and other crops were higher in magnitude.

Emissions at zero fertilisation were consistently higher than the assigned prior of 1 kg N_2O-N ha^{-1} yr^{-1} in all the models except for the complex model 5, which had emission at zero fertilisation lower than the assigned prior in the Atlantic North environmental zone (0.81 kg N_2O-N ha^{-1} yr^{-1}). They were in a similar pattern to that of the EFs for the four environmental zones, i.e. the higher the EF, the higher the emission at zero fertilisation for each zone.

3.3. N_2O emission model for organic soils

The N_2O fluxes measured from organic soils were generally higher than those from mineral soils. Extreme N_2O fluxes were common in several locations, with an anomalous value of 56.4 kg N_2O-N ha^{-1} reported by Flessa et al. (1998). N_2O emissions from organic soils were modelled separately since the factors influencing N_2O emission in these soils were expected to be different from those of mineral soils, e.g. groundwater levels have a strong impact and strong mineralisation of soil organic matter occurs in drained organic soils. Since the number of N_2O measurements was too small to explore the influence of possible predictors, a simple mixed-effects model was formulated using N input and short-by with location clusters and experiment year as random effects (Model 6; Table 1).

Following the guidance of IPCC (2014b), the default emission factor of 1% (0.1–1.8%) from the IPCC (2019) was also used as the prior for N input to organic soils and a neutral prior was given for the short-by parameter. The prior for emissions at zero fertilisation was derived from the German Tier 2 methodology for estimation of N_2O emissions resulting from mineralisation of soil organic matter in drained organic soils (Tiemeyer et al., 2020). The mean of the emission factors for cropland and grassland in drained organic soils was used as the prior, i.e., the expected value of the prior was 7.9 kg N_2O-N ha^{-1} yr^{-1} with a large standard deviation of 16.8 kg N_2O-N ha^{-1} yr^{-1}. The model yielded an emission factor of 1.01% (0.39–1.65%) for annual N_2O emissions and annual emission at zero fertilisation of 4.58 (2.13 – 7.45) kg N_2O-N ha^{-1} yr^{-1}. The effect of the short-by parameter on emission rates was close to zero as most of the measurements in organic soils had a measurement period of one year.

The RMSE of the model fit ($RMSE_{fit}$) was 6.45 kg N_2O-N ha^{-1} yr^{-1}, which is relatively poor but expected given the small number of measurements.

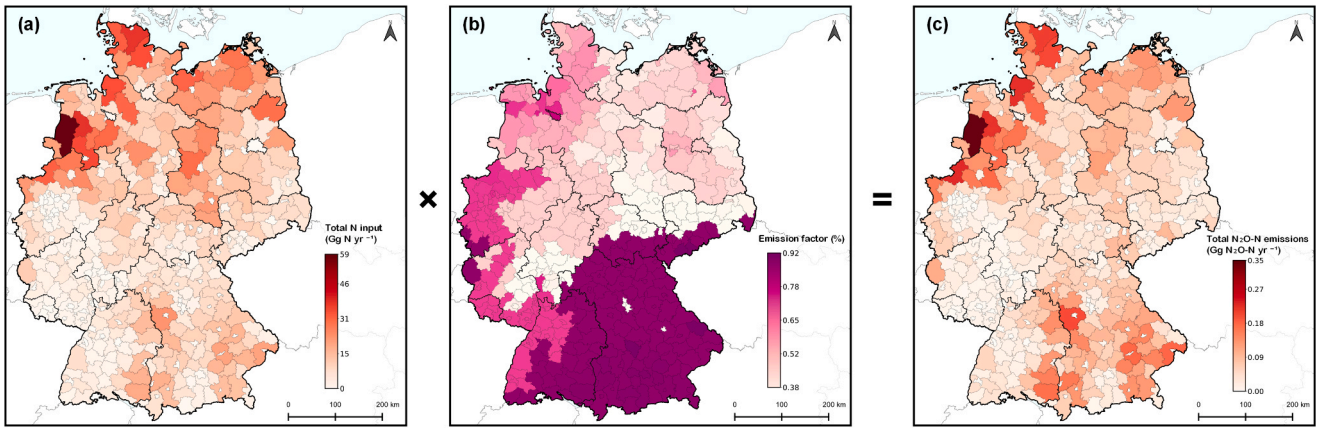

Fig. 3. Maps illustrating the steps of district-wise total N_2O-N estimation: (a) Total annual N inputs including synthetic fertilisers, animal manure, sewage sludge, biogas digestate and crop residues (Gg N yr^{-1}), (b) Unified emission factors (%) representative of mineral and organic soils, (c) Total annual N_2O-N emissions (Gg N_2O-N yr^{-1}).

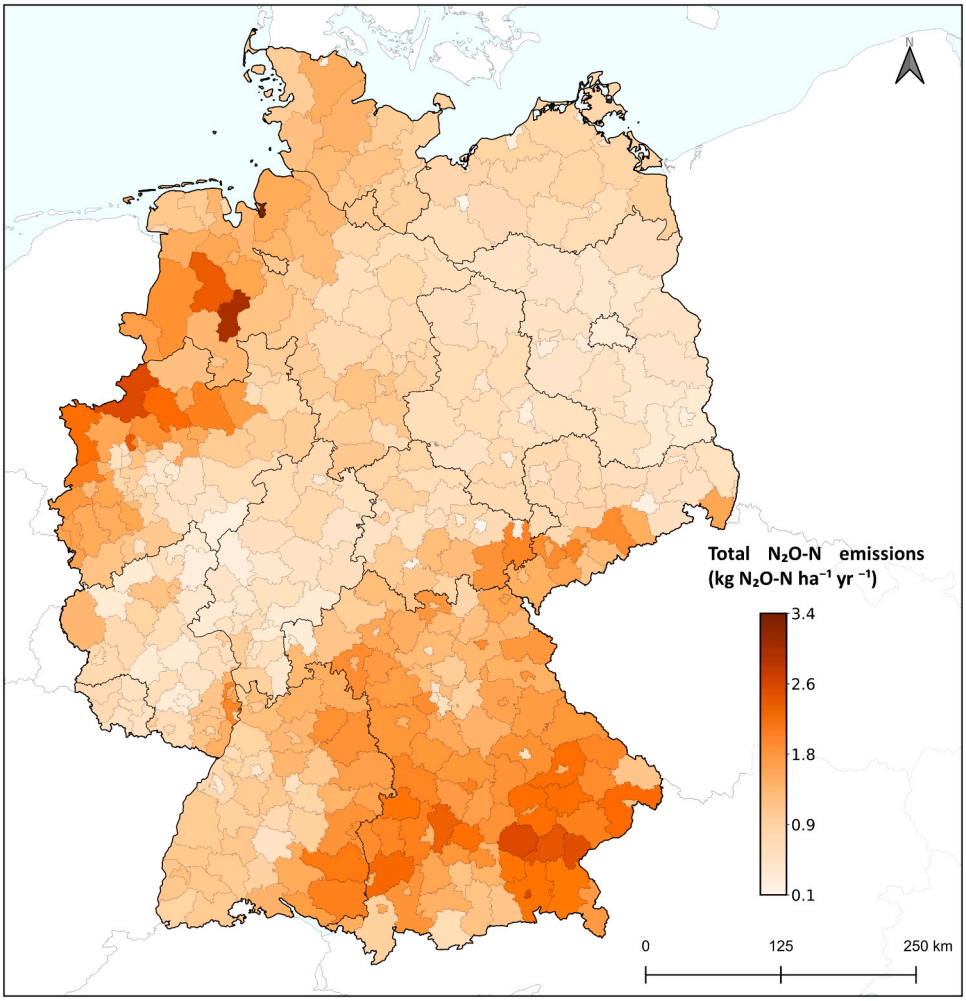

Fig. 4. District-wise N_2O-N emissions related to agricultural area (kg N_2O-N ha^{-1}). Borders of federal states and districts are indicated.

3.4. *Unified emissions from mineral & organic soils*

The district-wise unified emission factor resulting from the weighted average of EFs associated with mineral (Model 4) and organic (Model 6) soils is shown in Fig. 3b. The EFs ranged from 0.38% to 0.92% across the country, with EFs for the south-west and south-east environmental zones generally higher than those of the two northern zones. Organic soils are prevalent in the north as well as close to the Alps, i.e. Germany's southern border, and can be clearly seen as increased emission factors within the climatic zones. District-wise N inputs, i.e. the sum of synthetic fertiliser, animal manure, sewage sludge, biogas digestate and crop residue inputs, in 2015 showed a pattern almost opposite to that of the

Table 2
Comparison of Tier 1 (IPCC, 2006, 2019, aggregated Tier 1 value) and Tier 2 (this study) implied emission factors and corresponding N_2O emissions for 2015 (submission 2021).

GHG sources	Tier 1		Tier 2	
	Implied emission factors (kg N_2O-N/kg N)	N_2O emissions (kt)	Implied emission factors (kg N_2O-N/kg N)	N_2O emissions (kt)
Inorganic N fertilisers	0.01	27.28	0.0061 (0.0042 – 0.0084)	16.69
Animal manure applied to soils	0.01	15.38	0.0066 (0.0046 – 0.0090)	10.13
Sewage sludge applied to soils	0.01	0.29	0.0057 (0.0041 – 0.0077)	0.17
Other organic fertilisers applied to soils (digestates)	0.01	4.77	0.0065 (0.0045 – 0.0087)	3.08
Crop residues	0.01	9.57	0.0059 (0.0041 – 0.0081)	5.67
Overall (Uncertainty ranges: IPCC, 2006; IPCC, 2019)	0.01 (0.003 – 0.03; 0.001 – 0.018)	57.29	0.0062 (0.0043 – 0.0085)	35.74

EFs, with the districts to the north of Germany and a few districts in the south having the highest N fertiliser application rates up to 59.2 Gg N yr^{-1} (Fig. 3a). Unlike the EFs, where most of the high figures were concentrated in the south, the total N_2O emissions from all N sources displayed a divergent pattern, with the district of "Emsland" in north-west Germany having the highest emission of 0.35 Gg N_2O-N yr^{-1} (Fig. 3c) due to high N inputs combined with a large proportion of organic soils. Lower N_2O emissions were predominantly estimated for the west-central districts, which are mostly urban areas and have lower N fertiliser application rates. Overall, the national total direct N_2O emissions from the five N sources in 2015 were estimated to be 22.75 Gg N_2O-N yr^{-1}, of which synthetic fertilisers contributed 10.62 Gg N_2O-N yr^{-1} followed by animal manure at 6.45 Gg N_2O-N yr^{-1}. Emissions from other N sources, i.e. sewage sludge, biogas digestates and crop residues were 0.11, 1.96 and 3.61 Gg N_2O-N yr^{-1} respectively.

Generally, crop production and thus N inputs are higher in the northern zones than the south (Fig. 3a). However, the southern environmental zones had higher emission factors than the north (Fig. 3b). As a result, relatively high district-wise total N_2O emissions are estimated for the north and the south-east regions (Fig. 3c). When relating N_2O emissions to agricultural area, high values occur in the north-west, which is a region of concentrated livestock farming (Statistisches Bundesamt (Destatis), 2020), and in the south-east region (Fig. 4).

The national implied EFs were similar for all N sources, with 0.7% for animal manure and biogas digestates and 0.6% for synthetic fertilisers, sewage sludge and crop residues (Table 2). The difference between EFs for synthetic fertilisers and animal manure is a function of the spatial distribution of the N sources between organic and mineral soils. For instance, the north-west region with high concentration of livestock is also a region with higher concentration of organic soils (Fig. S1). The overall implied emission factor was 0.62% (0.43–0.85%) which is significantly lower and has a narrower uncertainty range than both the default Tier 1 EF of 1% (0.3−3%) currently used and the IPCC (2019) Tier 1 EF of 1% (0.1–1.8%). The implied emission factor for synthetic fertilisers (0.61%) was also considerably lower than the proposed IPCC (2019) stratified EF for synthetic fertilisers 1.6% (1.3–1.9%).

4. Discussion

The new N_2O EFs for Germany confirmed the initial hypothesis of this study that N_2O emissions are overestimated by the IPCC's default 1% EF. The lower magnitude of N_2O emissions is in agreement with Tian et al. (2020), where estimates from the bottom-up approach used for national GHG inventories in Europe are higher overall than top-down estimates. In comparison with Tier 2 EFs for other countries, Germany's new direct N_2O IEF for inorganic fertilisers is similar to that of Japan, New Zealand and the United Kingdom, while countries such as Ireland, the Netherlands, the Russian Federation and the USA report IEFs above 1% (UNFCCC, 2021). The IEF for organic fertilisers was almost the same as that for inorganic fertilisers, and was comparable with those of Japan and the United Kingdom.

This variability of EFs with respect to geographical locations not only exists between countries but also within Germany, with measured N_2O emissions generally being low in the north-east and substantially higher in the south-east. The differing emission factors in different regions have previously been explained by the effect of climatic conditions, soil properties such as soil aeration, pH, and agricultural land use (Dechow and Freibauer, 2011; Jungkunst et al., 2006). Consequently, environmental stratification, which integrates climate and geomorphology, is a good predictor in the derived empirical models.

In contrast to what is reported by other countries in their inventories and what is suggested in the 2019 IPCC Refinement, the extensive data from Germany do not support different EFs for synthetic and organic fertilisers. There are reasons for expecting lower EFs for organic N input. Larger ammonia losses from organic fertilisers than from synthetic fertilisers result in less nitrogen available for N_2O production. Furthermore, extensive agriculture with low N surplus (such as organic farming systems) use organic fertilisation but not synthetic fertilisers. However, there are also arguments against lower EFs for organic N inputs. Slower N uptake by crops after organic fertiliser application and the longer time span over which organic nitrogen is mineralised in soils provide opportunities for N_2O production (Petersen, 1999; Velthof et al., 2003). Unfortunately, studies in the database that compared synthetic and organic fertilisers were usually not optimally set up for EFs to be compared. Several treatments of varying N input would be needed for both fertiliser types. It is therefore possible that the effect of fertiliser type also exists in Germany and would become evident if more dedicated experiments were conducted. However, the present study showed that emissions from synthetic fertilisers are not as high as the levels suggested by the 2019 IPCC refinement (IPCC, 2019).

There has been considerable debate over whether the N_2O response to N input is non-linear. Kim et al. (2013b) and Shcherbak et al. (2014) conducted meta-analyses and found a trend of N_2O emissions increasing exponentially if N input exceeds crop needs. Screening of the data collected from Germany did not reveal this non-linearity, but most of the studies were focused on recommended fertilisation practices, thus over-fertilisation is likely to be rare in the dataset. A linear relationship was therefore assumed for modelling purposes. There are substantial advantages of a linear relationship for an inventory because the models can be upscaled simply by using aggregated values for N input. A non-linear model that is calibrated on field-scale data cannot be upscaled to district-scale as easily as a linear model.

Jungkunst et al. (2006) highlight that the correlation between soil properties and N_2O emissions, which commonly exists on a site-specific scale, decreases at a national level. Although the inclusion of soil properties and crop types, the latter being a proxy for timing of fertilisation and N uptake as well as the quality of crop residues, slightly improved the model's predictive performance, generally the effects were rather uncertain. The effect of soil properties on EF could not be included in the empirical model because the model became over-parameterised and impossible to fit successfully. It would be preferable to use a process model to quantify these effects, but sufficient fine-scaled data are not available for Germany's emission inventory and

thus fitting a process model was beyond the scope of this study. An empirical model was not fitted on soil data at a district level because soil properties are difficult to aggregate for districts and can vary considerably, e.g. pH within a district varies due to pH management. That kind of a model can therefore be expected to have a worse predictive performance than the simpler model that only stratifies by environmental zones. A model adding only crop type as predictor was not considered for the inventory because the annual spatial distribution of crop types in Germany is not available for the whole time series from 1990. Furthermore, winter crops are the main crops in Germany and therefore also had the largest proportion of data points in the N_2O database.

According to the model results, expected N_2O emissions at zero fertilisation were between 1.4 kg N ha^{-1} yr^{-1} in the north and 2.0 kg N ha^{-1} yr^{-1} in the south of Germany on mineral soils and about 4.6 kg N ha^{-1} yr^{-1} on organic soils. In their global meta-analysis, Kim et al. (2013a) observed a mean background emission of 1.5 kg N ha^{-1}, but reported a higher value of 2.4 kg N ha^{-1} for Germany. Unfortunately, no distinction was made between mineral and organic soils. However, background emissions from agricultural land are more of an experimental artefact because unfertilised controls are not representative of good agricultural practice. Kim et al. (2013a) list several drivers of background emissions: fertilisation and soil management in previous years, atmospheric N deposition and plant residues. Furthermore, N_2O emissions can result from mineralisation of soil organic matter, which also interacts with fertilisation (Craine et al., 2007). A strong correlation was observed between the background emissions from the environmental zones and their emission factors ($R^2 = 0.96$). This suggests that environmental controls of N_2O production differ more between these regions than the availability of nitrogen in control treatments, particularly in newly established unfertilised plots.

Since N_2O emissions were only modelled as a function of exogenous nitrogen (excluding N deposition) and endogenous sources and legacy effects cannot be disentangled, the new EF was only applied to inputs of synthetic fertilisers, organic fertilisers, biogas digestates, sewage sludge and crop residues. Although crop residues were not included in the N input of the calibration data, the example set by IPCC was followed and it was assumed that the EF was also valid for this N input. This was supported by the correlation between background emissions and EFs, as crop residues are likely to be a large part of N input not accounted for in the dataset. However, we are not confident that the new EFs are applicable to nitrogen from a very slow release process such as mineralisation of soil organic matter or to non-agricultural areas. It is therefore recommended that, until further research findings are available, the IPCC default EF for direct N_2O emissions from the mineralisation of soil organic matter should be continued to be used in Germany's emission inventory.

5. Conclusions

According to the new methodology in this study, the new best estimate of GHG emissions from German agriculture in 2015 is 5.71 Tg CO_2-eq. (8.59%) lower than that reported in 2021 submission (66.51 Mt CO_2-eq, converted with GWPs according to (IPCC, 2014a)). Inventory compilers can easily apply this new method to the whole time series from 1990 to the present day, which would change the estimate of mitigation achieved in that period. As reported mitigation of N_2O emissions from soils between 1990 and 2019 (15.27%) was lower than GHG mitigation in the agricultural sector (19.63%, converted with GWPs according to (IPCC, 2014a)), the smaller contribution of N_2O emissions from soils to total agricultural emissions results in a larger estimated sectoral mitigation. With the new EFs, the N_2O mitigation from agricultural soils was marginally lower than the 2021 submission at 15.08%, while the sectoral N_2O mitigation increased to 19.94%. Furthermore, due to the smaller share of N_2O emissions from soils, mitigation measures focusing on methane emissions from livestock farming will have a stronger relative effect than measures for reducing N_2O emissions. However, all emission sources in agriculture, including N_2O emissions from soils, will need to be mitigated if the ambitious goals of climate protection policy (Deutscher Bundestag, 2019; European Commission, 2020) are to be achieved.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.agee.2021.107640.

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