Decreasing trends of ammonia emissions over Europe seen from remote sensing and inverse modelling

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16 **Abstract**

17   Ammonia (NH3), a significant precursor of particulate matter, not only affects

18 biodiversity, ecosystems, soil acidification, but also climate and human health. In addition, its

19 concentrations are constantly rising due to increasing feeding needs and the large use of

20 fertilization and animal farming. Despite the significance of ammonia, its emissions are

21 associated with large uncertainties, while its atmospheric abundance is difficult to measure.

22 Nowadays, satellite products can effectively measure ammonia with low uncertainty and a

23 global coverage. Here, we use satellite observations of column ammonia in combination with

24 an inversion algorithm to derive ammonia emissions with a high resolution over Europe for the

25 period 2013–2020. Ammonia emissions peak in Northern Europe, due to agricultural

26 application and livestock management, in Western Europe (industrial activity) and over Spain

27 (pig farming). Emissions have decreased by -26% since 2013 (from 5431 Gg in 2013 to 3994

28 Gg in 2020) showing that the abatement strategies adopted by the European Union have been

29 very efficient. The slight increase (+4.4%) in 2015 is also reproduced here and is attributed to

30 some European countries exceeding annual emission targets. Ammonia emissions are low in

31 winter (286 Gg) and peak in summer (563 Gg) and are dominated by the temperature dependent

32 volatilization of ammonia from the soil. The largest emission decreases were observed in

33 Central and Eastern Europe (-38%) and in Western Europe (-37%), while smaller decreases

34 were recorded in Northern (-17%) and Southern Europe (-7.6%). When complemented against

35 ground observations, modelled concentrations using the posterior emissions showed improved

36 statistics, also following the observed seasonal trends. The posterior emissions presented here

37 also agree well with respective estimates reported in the literature and inferred from bottom-up

38 and top-down methodologies. These results indicate that satellite measurements combined with

39 inverse algorithms constitute a robust tool for emission estimates and can infer the evolution of

40 ammonia emissions over large timescales.

41

1 Introduction

Ammonia (NH_3), the only alkaline gas in the atmosphere, constitutes one of the most reactive nitrogen species. It is produced from decomposition of urea, which is a rapid process when catalyzed by enzymes (Sigurdarson et al., 2018). The main sectors contributing to its production are livestock management and wild animals (Behera et al., 2013), biomass burning and domestic coal combustion (Fowler et al., 2004; Sutton et al., 2008), volcanic eruptions (Sutton et al., 2008), and agriculture (Erisman et al., 2007). Emissions from agricultural activity and livestock management represent over 80% of the total emissions (Crippa et al., 2020), while their regional contribution can reach 94% (Van Damme et al., 2018).

Once emitted, it is transported over short distances and deposited to water bodies, soil or vegetation with a typical atmospheric lifetime of a few hours (Evangeliou et al., 2021). It can then lead to eutrophication of water bodies (Stevens et al., 2010), modulate soil pH (Galloway et al., 2003) and «burn» vegetation by pulling water from the leaves (Krupa, 2003). It also reacts with the abundant atmospheric sulfuric and nitric acids (Malm, 2004) forming fine particulate matter (PM2.5) (Tsimpidi et al., 2007). While ammonia has a short atmospheric lifetime, PM2.5 resides significantly longer in the atmosphere, on the order of days to weeks (Seinfeld and Pandis, 2000), and hence is transported over longer distances. Accordingly, secondary PM2.5 can affect the Earth's radiative balance, both directly by scattering incoming radiation (Henze et al., 2012) and indirectly as cloud condensation nuclei (Abbatt et al., 2006). Its environmental effects include visibility problems and contribution to haze formation. Finally, PM2.5 affects human health, as it penetrates the human respiratory system and deposits in the lungs and alveolar regions (Pope and Dockery, 2006; Pope III et al., 2002) contributing to premature mortality (Lelieveld et al., 2015).

To combat secondary pollution, the European Union established a set of measures focusing on ammonia abatement, similar to the ones introduced by China (Giannakis et al., 2019). These measures aim at reducing ammonia emissions by 6% in 2020, relative to 2005. However, the lack of spatiotemporal measurements of ammonia over Europe makes any assessment of the efficiency of these measures difficult, as only bottom-up methods are used to calculate emission. These methods still show a slight increase (0.6% y^{-1}) up to 2018 mostly due to increasing agricultural activities (McDuffie et al., 2020). Such bottom-up approaches rely on uncertain land-use data and emission factors that are not always up to date, thus adding large errors to existing inventories.

74 During the last decade, satellite products have also become available to fill the gaps
75 created by spatially disconnected ground-based measurements. Data from satellite sounders
76 such as the Infrared Atmospheric Sounding Interferometer (IASI) (Van Damme et al., 2017),
77 the Atmospheric Infrared Sounder (AIRS) (Warner et al., 2017), the Cross-track Infrared
78 Sounder (CrIS) (Shephard and Cady-Pereira, 2015), the Tropospheric Emission Spectrometer
79 (TES) (Shephard et al., 2015), and Greenhouse Gases Observing Satellite (GOSAT) (Someya
80 et al., 2020) are publicly available. Most of them have been validated against ground-based
81 observations or complemented with other remote sensing products (Van Damme et al., 2015,
82 2018; Dammers et al., 2016, 2017, 2019; Kharol et al., 2018; Shephard et al., 2020; Whitburn
83 et al., 2016).

84 Accordingly, a few studies on ammonia emission calculations have been recently
85 published relying on 4D-Variational inversion schemes such as (Cao et al., 2022; Zhu et al.,
86 2013) or process based models (Beaudor et al., 2023; Vira et al., 2020). More recently, Sitwell
87 et al. (2022) proposed an inversion scheme for comparison between model profiles and satellite
88 retrievals using hybrid logarithmic and linear observation operator that attempts to choose the
89 best method according to the particular situation. In the present study, we use direct
90 comparisons between the CrIS ammonia retrievals and model profiles using the Least Squares
91 with Adaptive Prior Covariance (LS-APC) algorithm (Tichý et al., 2016), which reduces the
92 number of tuning parameters in the method significantly using variational Bayesian
93 approximation technique. We constrain ammonia emissions over Europe over the 2013–2020
94 period and validate the results against ground-based observations from EMEP (European
95 Monitoring and Evaluation Programme, https://emep.int/mscw/) (Torseth et al., 2012).

2 Methods

2.1 CrIS observations

98 To constrain ammonia emissions with inverse modelling, satellite measurements were
99 adopted from the Cross-Track Infrared Sounder (CrIS) onboard the NASA Suomi National
100 Polar-orbiting Partnership (S-NPP) satellite, which provides atmospheric soundings with a
101 spectral resolution of 0.625 cm^{-1} (Shephard et al., 2015). CrIS presents improved vertical
102 sensitivity for ammonia closer to the surface due to the low spectral noise in the ammonia
103 spectral region (Zavyalov et al., 2013) and the early afternoon overpass that typically coincides
104 with high thermal contrast, which is optimal for thermal infrared sensitivity. The CrIS Fast
105 Physical Retrieval (CFPR) (Shephard and Cady-Pereira, 2015) retrieves ammonia profiles at

106   14 levels using a physics-based optimal estimation retrieval, which also provides the vertical

107   sensitivity (averaging kernels) and an estimate of the retrieval errors (error covariance matrices)

108   for each measurement. As peak sensitivity typically occurs in the boundary layer between 900

109   and 700 hPa (~ 1 to 3 km) (Shephard et al., 2020) and the surface and total column

110   concentrations are both highly correlated with these boundary layer retrieved levels. The total

111   column random measurement error is estimated in the 10–15% range, with total errors to be

112   ~30% (Shephard et al., 2020). The individual profile retrieval levels show an estimated random

113   measurement error of 10–30 %, with total random errors estimates increasing to 60 to 100%

114   due to the limited vertical resolution (1 degree of freedom of signal for CrIS ammonia). These

115   vertical sensitivity and error output parameters are also useful for using CrIS observations in

116   applications (e.g. data fusion, data assimilation; model-based emission inversions; (Cao et al.,

117   2020; Li et al., 2019)), as a satellite observational operator can be generated in a robust manner.

118   The detection limit of CrIS measurements has been calculated down to 0.3–0.5 ppbv (Shephard

119   et al., 2020). CrIS ammonia has been evaluated against other observations over North America

120   with the Ammonia Monitoring Network (AMoN) (Kharol et al., 2018) and against ground-

121   based Fourier transform infrared (FTIR) spectroscopic observations (Dammers et al., 2017)

122   showing small bias and high correlations.

123   Daily CrIS ammonia (version 1.6.3) was interpolated onto a 0.5°×0.5° grid covering all

124   of Europe (10°W–50°E, 25°N–75°N) for the period 2013–2020. Interpolation was chosen due

125   to the large number of observations (around 10,000 retrievals per day per vertical level), which

126   made the calculation of source-receptor matrices (SRMs) computationally inefficient. Through

127   interpolation we limited the number of observation (and thus the number of SRMs to be

128   calculated) to 2000 per day per vertical level. Sitwell et al. (2022) showed that the averaging

129   kernels of CrIS ammonia are significant only for the lowest six levels (the upper eight have no

130   influence onto the satellite observations) and therefore we considered only these six vertical

131   levels (~1018-619 hPa).

2.2   A priori emissions of ammonia

133   We used as a priori emissions for ammonia in the inversion algorithm the ones calculated

134   (i) from the most recent version of ECLIPSEv6 (Evaluating the CLimate and Air Quality

135   ImPacts of Short-livEd Pollutants) (Klimont, 2022; Klimont et al., 2017) combined with

136   biomass burning emissions from GFEDv4 (Global Fire Emission Dataset) (Giglio et al., 2013)

137   hereafter "EC6G4", (ii) a more traditional dataset from ECLIPSEv5, GFEDv4 and GEIA

138   (Global Emissions InitiAtive), hereafter "EGG" (Bouwman et al., 1997; Giglio et al., 2013;

Klimont et al., 2017), (iii) emissions calculated from IASI (Infrared Atmospheric Sounding Interferometer) and a 1-dimensional box-model and a modelled lifetime (Evangeliou et al., 2021), denoted as "NE" and (iv) from the high resolution dataset of Van Damme et al. (2018) after applying a simple 1-dimensional box-model (Evangeliou et al., 2021), hereafter denoted as "VD". Given the large uncertainty in ammonia emissions illustrated in Figure 1, we calculated the average of these four priors (hereafter "avgEENV") to establish the a priori emissions used in this study.

2.3 Lagrangian particle dispersion model for the calculation of source-receptor matrices (SRMs) of ammonia

SRMs were calculated for each $0.5°×0.5°$ grid-cell over Europe (10°W–50°E, 25°N–75°N) using the Lagrangian particle dispersion model FLEXPART version 10.4 (Pisso et al., 2019) adapted to simulate ammonia. The adaptation of the code includes treatment for the loss processes of ammonia adopted from the Eulerian model LMDZ-OR-INCA (horizontal resolution of $2.5°×1.3°$ and 39 hybrid vertical levels) that includes all atmospheric processes and a state-of-the-art chemical scheme (Hauglustaine et al., 2004). The model accounts for large-scale advection of tracers (Hourdin and Armengaud, 1999), deep convection (Emanuel, 1991), while turbulent mixing in the planetary boundary layer (PBL) is based on a local second-order closure formalism. The model simulates atmospheric transport of natural and anthropogenic aerosols and accounts for emissions, transport (resolved and sub-grid scale), and dry and wet deposition. LMDZ-OR-INCA includes a simple chemical scheme for the ammonia cycle and nitrate particle formation, as well as a state-of-the-art $CH_4/NO_x/CO/NMHC/O_3$ tropospheric photochemistry (Hauglustaine et al., 2014). To calculate chemical loss of ammonia to PM2.5, after a month of spin-up, global atmospheric transport of ammonia was simulated for 2013–2020 by nudging the winds of the 6-hourly ERA Interim Reanalysis data (Dee et al., 2011) with a relaxation time of 10 days (Hourdin et al., 2006). Using the EGG inventory, we calculated the e-folding lifetime of ammonia in the model, which was adopted in FLEXPART. We refer the reader to (Tichý et al., 2022) for a detailed description of the formalism. Atmospheric linearities of the system and a full validation against ground-based observation are also presented in the same paper.

FLEXPART releases computational particles that are tracked backward in time using ERA5 (Hersbach et al., 2020) assimilated meteorological analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF) with 137 vertical layers, a horizontal resolution of $0.5°×0.5°$ and one hour temporal resolution. FLEXPART simulates turbulence

172 (Cassiani et al., 2014), unresolved mesoscale motions (Stohl et al., 2005) and includes a deep
173 convection scheme (Forster et al., 2007). SRMs were calculated for 7 days backward in time,
174 at temporal intervals that matched satellite measurements and at spatial resolution of
175 0.25°×0.25°. This 7-day backward tracking is sufficiently long to include almost all ammonia
176 sources that contribute to surface concentrations at the receptors given a typical atmospheric
177 lifetime of about half a day (Van Damme et al., 2018; Evangeliou et al., 2021).

178 **2.4 Inverse modeling algorithm**

179   The inversion method used in the present study relies on optimization of the difference
180 between the CrIS satellite vertical profile observations, denoted as v^{sat}, and retrieved vertical
181 profile, v^{ret}. The latter are obtained by applying an instrument operator applied in logarithm
182 space (Rodgers, 2000) as follows:

$$\ln(v^{ret}) = \ln(v^a) + A(\ln(v^{true}) - \ln(v^a)) \qquad (1)$$

184 where v^{ret} is the retrieved profile concentration vector, v^a is a priori profile concentration
185 vector used in the satellite retrievals, v^{true} is the hypothetical true profile concentration vector
186 supplied by the model ($v^{true} = v^{mod}$), and A is the averaging kernel matrix (for each
187 0.5°×0.5° resolution grid-cell). Eq. (1) provides a useful basis for the calculation of the CrIS
188 retrievals if the retrieval algorithm is performing as designed, i.e., it is unbiased and the root
189 mean square error (RMSE) is within the expected variability. The v^{mod} term can be written as:

$$v^{mod} = Mx \qquad (2)$$

191 for each grid-cell of the spatial domain, where M is the grid-cell specific SRM calculated with
192 FLEXPART and x is the unknown grid-cell specific emission vector. The SRM matrix M is
193 calculated on circular surroundings around each grid-cell for computational efficiency. We
194 chose circles with a radius of approximately 445 km, equal to 4 degrees, which is shown to be
195 sufficient for reliable emission estimation and low sensitivity has been observed with this
196 choice. Since the vector x is unknown, we replace it by a prior emission x^a (see section 2.2) in
197 the initial step that is gradually refined iteratively based on the satellite observations.

198   The used inversion setup is based on iterative minimization of mismatch between v^{sat}
199 and v^{ret} updating (iteratively) the emission x such as below:

$$\arg\min_{x^a \to x} \left|\left| v^{sat} - v^{ret} \right|\right|_2^2 \qquad (3)$$

201 for each grid-cell of computational domain. The minimization problem is solved in two steps.
202   First, we construct the linear inverse problem for each year where v^{ret} from the given
203 surroundings, denoted here as S, forms the block-diagonal matrix v_S^{ret} while v^{sat} from the

204   given surroundings form an associated observation vector v_S^{sat}. This forms the linear inverse
205   problem:

$$v^{sat} = v_S^{ret} q_S \qquad (4)$$

207   where the vector q_S is a vector with coefficients denoting how x^a needs to be refined to obtain
208   emission estimate vector x. For one year, 6 vertical profiles, and 4 degrees radius, the size of
209   the the block-diagonal matrix v_S^{ret} is 13896 times 12, hence, the correction coefficient vector
210   q_S contain 12 values corresponding to each month. We solve Eq. 4 using the least squares with
211   adaptive prior covariance (LS-APC) algorithm (Tichý et al., 2016). The algorithm is based on
212   variational Bayesian methodology assuming non-negative solution and favoring solution
213   without abrupt changes and it minimizes the use of manual tuning (Tichý et al., 2020). The
214   method assumes the data model in the form of:

$$p(v^{sat}) = N(v_S^{ret} q_S, R) \qquad (5)$$

216   where N denotes the multivariate normal distribution and R the covariance matrix assumed in
217   the form $R = \omega^{-1} I_p$ with unknown precision parameter ω on its diagonal. Following Bayesian
218   methodology, we assign prior model to all unknown parameters, i.e. ω and q_S. Theirs prior
219   models are selected as:

$$p(\omega) = G(\vartheta_0, \rho_0) \qquad (6)$$

$$p(q_S) = tN(0, (LVL)^{-1}, [0, +\infty]) \qquad (7)$$

222   where $G(\vartheta_0, \rho_0)$ is the Gamma distribution (conjugate to the normal distribution) with prior
223   parameters ϑ_0, ρ_0 selected to 10^{-10} achive non-informative prior. The second term follows
224   truncated normal distribution with positive support and with specific form of a precision matrix.
225   We assume the precision matrix in the form of modified Cholesky decomposition which allows
226   for tractability of estimation of its parameters, matrices V and L. The matrix V is diagonal with
227   unknown diagonal parameters and the matrix L is lower bidiagonal with ones on the diagonal
228   and unknown parameters on its sub-diagonal, formalized as vectors v and l, respectively.
229   These parameters are estimated within the method, while purpose of vector v is to allow for
230   abrupt changes in q_S, and vector l to favor smooth estimates (see details in Tichý et al. (2016)).
231   All model parameters (ω, q_S, v, l) are estimated using the variational Bayes procedure where
232   we obtain not only point-estimates, but their full posterior distributions.

233   Second, the grid-cell specific coefficient vector q_S is propagated through Eq. 2 into Eq. 1
234   to refine a prior emission x^a and obtain estimated unknown emissions x. To maintain stability
235   of the method, we bound the ratio between prior and posterior emission elements to 0.01 and
236   100, respectively. This choice, motivated by Cao et al. (2020), omits unrealistically small or

high emissions, however, the bounds are large enough to allow for new sources, as well as for attenuation of old sources. To introduce these boundaries is necessary since the problem in Eq. 1 is ill-conditioned and the propagation through the equation may lead to unrealistic values due to numerical instability. For this reason, these boundaries are needed and the sensitivity to the choice of the prior emission are studied in Section 3.3.

Note that CrIS data for some spatiotemporal elements are missing in the dataset. In these cases, we interpolated the missing data following the method proposed by D'Errico (2023), which solves a direct linear system of equations for missing elements, while the extrapolation behavior of the method is linear. Another strategy recently adopted in the literature has been to tackle the missing data using total variation methodology (see details in Fang et al., 2023); however, the method has been limited so far to its use on point-source release, hence we did not use it in this work.

3 Results

3.1 Emissions of ammonia in Europe (2013–2020)

We analyze the CrIS ammonia satellite observations for Europe (10°W–50°E, 25°N–75°N) over the 2013-2020 period on monthly basis to derive ammonia emissions using the inverse modelling methodology described in Section 2.4. The inversion algorithm is applied to each year of CrIS observations separately with the use of the avgEENV prior emission.

The overall resulting spatial distribution of the posterior emissions of ammonia (denoted as posterior_avgEENV) averaged for the whole period are displayed in Figure 2 (top-left). The highest emissions occur in Northwestern Europe (including Northern Belgium, the Netherlands and northwestern Germany) and to a smaller extent in the Po Valley (Italy), and the Ebro Valley (Spain). Local maxima are also seen over Pulawy (Poland), South Romania and Kutina (Croatia) due to industrial applications (Clarisse et al., 2019; Van Damme et al., 2018). While ammonia emissions were not calculated high in the Po Valley (8 year average), it has been reported that in Lombardy, about 90% of the ammonia emissions there have been reported to originate from manure management (Lonati and Cernuschi, 2020). The Ebro Valley is characterized by intensive agricultural activities (Lassaletta et al., 2012; Lecina et al., 2010) and the Aragon and Catalonia regions by large pig farms (Van Damme et al., 2022). Finally, both Belgium and The Netherlands are countries in which intensive livestock activity is documented. It consists mostly of dairy cow, beef cattle, pig and chicken farming (Gilbert et al., 2018; Lesschen et al., 2011; Velthof et al., 2012).

269   Figure 2 (top-right) shows the annual posterior emissions discretized monthly for the
270 whole period (solid line) compared to prior ammonia emissions (dashed line), averaged for the
271 domain. Higher emissions than the prior ones were calculated, which is not necessarily
272 attributed to emission increases over Europe, but rather to miscalculation of emissions in the
273 prior bottom-up inventories that were used. A strong seasonal cycle is also observed peaking
274 in the middle of each year (summer) of the study period, but for several of these years, the
275 characteristic bimodal cycle also appears with another peak in spring (Beaudor et al., 2023).

276   To examine more closely the seasonal variability of ammonia emissions in Europe, we
277 present the monthly posterior emissions of ammonia averaged for the whole study period
278 (2013–2020) at the bottom-left panel of Figure 2 together with the prior ones. The total
279 emissions for each month based on the map element size and length of the respective month
280 were averaged for the whole study period. The same was done for each year in the bottom-right
281 panel. The interannual variability over the period between 2013 and 2020, is also apparent in
282 the monthly box and whisker plots of the posterior emissions. In addition, the spatial
283 distribution of monthly ammonia emissions averaged for the eight-year period is given in
284 Supplementary Figure S 1. It appears that ammonia emissions are very low in wintertime (DJF
285 average: 286 Gg) over Europe and increase towards summer (JJA average: 563 Gg), due to
286 temperature dependent volatilization of ammonia (Sutton et al., 2013), with the largest
287 emissions occurring in August (601 Gg). Although a clear peak of fertilization in early spring
288 is missing from the plot, emissions start to increase in early spring to peak in late-summer (Van
289 Damme et al., 2022) corresponding to the start and end of the fertilization periods in Europe
290 (Paulot et al., 2014). Fertilization is tightly regulated in Europe (Ge et al., 2020). It is only
291 allowed from February to mid-September in The Netherlands, while manure application is also
292 only allowed during the same period depending on the type of manure and the type of land (Van
293 Damme et al., 2022). In Belgium, nitrogen fertilizers are only allowed from mid-February to
294 the end of August (Van Damme et al., 2022), so as in Germany (restricted in winter months)
295 (Kuhn, 2017).

296   Finally, Figure 2 (bottom-right) shows the annual posterior emissions for the whole
297 period with the annual total emissions for each year. We observe a significant decrease in
298 ammonia posterior emissions over Europe during the 2013–2020 period. Emissions were
299 estimated as 5431 Gg for 2013 decreasing to 4890 Gg in 2014. A minor increase can be seen
300 in 2015 (5104 Gg), after which a significant decrease of 534 Gg (more than 10%) was estimated,
301 followed by the nearly constant plateau at the levels between 4383 Gg in 2017, 4323 Gg in
302 2019 and finally to 3994 Gg in 2020. The gradual decrease in ammonia emissions over Europe

303   since 2013 is also plotted spatially in Supplementary Figure S 2. It is evident that the restrictions
304   and measures adopted by the European Union to reduce secondary PM formation were
305   successful, as emissions in the hot-spot regions of Belgium, The Netherlands, Germany and
306   Poland declined drastically over time. However, an increase of +4.4% was observed in 2015. It
307   has been reported that ammonia emissions increased in 2015 and several European Union
308   Member States, as well as the EU as a whole, exceeded their respective ammonia emission
309   ceilings (EEA, 2017). The increase was reported to be +1.8% and was mainly caused by
310   increased emissions in Germany, Spain, France, and the United Kingdom. This was caused by
311   extensive use of inorganic nitrogen fertilizers (including urea application) in Germany, while
312   increased emissions in Spain were driven by an increase in the consumption of synthetic
313   nitrogen fertilizers and in the number of cattle and pigs (EEA, 2017). It should be mentioned
314   that a false decrease of ammonia in 2020 due to the COVID-19 pandemic is calculated by the
315   current methodology, mainly due to bias created by the decrease of NO_x and SO_2 that are
316   precursor species of the atmospheric acids, with which ammonia reacts (see Tichý et al., 2022).

317   **3.2   Country by country ammonia emissions**

318   Posterior annual emissions of ammonia for 2013–2020 are plotted for four European
319   regions (Western, Central and Eastern, Northern and Southern Europe), accompanied by
320   relative trends calculated as difference between year 2013 and 2020 divided by the average for
321   the whole period, in the left panel of Figure 3, while the estimated seasonal variation of each
322   region is shown on the right panels averaged over the whole eight-year period. Western Europe
323   includes Ireland, Austria, France, Germany, Belgium, Andorra, Luxembourg, The Netherlands,
324   Switzerland, and United Kingdom; Central and Eastern Europe include Albania, Bosnia and
325   Herzegovina, Bulgaria, Czechia, Croatia, Hungary, Belarus, Slovakia, North Macedonia,
326   Montenegro, Poland, Romania, Moldova, Slovenia, Ukraine, and Serbia; Northern Europe is
327   defined by Denmark, Estonia, Finland, Latvia, Lithuania, Faroe Islands, Norway, and Sweden;
328   finally, Southern Europe includes Cyprus, Greece, Italy, Portugal, Spain.

329   The most significant decreases in ammonia emissions were estimated to be -38% in
330   Central and Eastern Europe and -37% in Western Europe, respectively. Quantitatively, Central
331   and Eastern Europe emissions were estimated to gradually drop from 2190 Gg in 2013 and to
332   1495 Gg in 2020 with a small increase in 2015 (2171 Gg) mainly because Germany, France
333   and the United Kingdom missed their emission targets (EEA, 2017). Western European
334   emissions of ammonia also declined constantly over time from 2041 Gg in 2013 to 1421 Gg in
335   2020. Smaller, yet significant, decreases were calculated over Northern Europe from 398 Gg in

2013 to 333 Gg in 2020 (-17%). Finally, Southern Europe exhibited a minor drop between years 2013 and 2014 (from 803 Gg in 2013 to 729 Gg in 2014) followed by a small increase until 2019 (from 729 to 803 Gg), and then decreased again in 2020 to 743 Gg. Overall, Southern European emissions decreased by -7.62%.

The seasonal cycle of ammonia was again characterized by the restrictions applied to the agricultural-related activities by the European Union member states (Figure 3, right panels). As such, emissions in Western, Central and Eastern and Southern Europe were very low in winter and started increasing when fertilization was allowed in early spring, whereas the increasing temperature towards summer increased volatilization and, thus, emissions of ammonia (Van Damme et al., 2022; Ge et al., 2020). Although much less marked than in other European regions due to lower prevailing temperatures and weaker agricultural applications, emissions in Northern Europe show the spring-summer temperature dependence. However, emissions were estimated to be double in winter rather following the cycle of SO2 (Tang et al., 2020). Emission may increase in Northern Europe in winter because OH and O3 concentrations are much lower, and the rate of converting SO2 to sulfate much slower. This means that less sulfate is produced and thus more NH3 stays in the gas form. Supplementary Figure S 3 shows prior emissions in Western, Central and Eastern, Northern and Southern Europe for EC6G4 and NE emission inventories. Both show the aforementioned increase in emissions during winter in Northeastern Europe. Specifically, the NE emissions that dominate the a priori emissions (avgEENV) as the highest inventory show an extreme winter peak in the north (emissions decline from 105 to 13 Gg). Therefore, there is a very strong dependence of the posterior seasonality of ammonia in Northern Europe, which may be also influenced by the used prior emissions, see uncertainty analysis in Section 3.3.

Country specific emissions of posterior ammonia on a monthly basis (eight-year average emissions) are shown for 20 countries in Supplementary Figure S 4. For countries such as Portugal, Spain, Italy, United Kingdom, The Netherlands, Belgium, Poland, Hungary, Denmark, Belarus and Romania two peaks can be clearly seen in late spring and end of summer. As discussed before, these peaks coincide with the two main fertilization periods in Europe (Paulot et al., 2014). However, it is expected that ammonia abundance is high throughout the entire spring–summer period (e.g., Greece, France, Germany, Czechia, Ukraine and Bulgaria) due to agricultural activity and temperature dependent volatilization (Sutton et al., 2013). Ammonia emissions in Finland, Sweden and Norway are smaller than in the rest of Europe and show a reverse seasonality.

3.3 Uncertainties in ammonia's posterior emissions

For the calculation of uncertainty of the estimated posterior emissions two different approaches were used. The first approach is based on uncertainty arising as a result of the inversion methodology. The standard deviation is calculated from posterior estimate which is in the form of Gaussian distribution such as

$$p_{\text{posterior}}(x_i) = N(\mu_i, \sigma_i^2) \qquad (8),$$

where N denotes normal (Gaussian) distribution and posterior parameters μ_i and σ_i are results of inversion for each element of the spatiotemporal domain. The uncertainty associated with any given spatial element is then a property of Gaussian distribution define with the square root of summed squared standard deviations:

$$\Sigma_{\text{location}} = \sqrt{\sum_t \sigma_{\text{location},t}^2} \qquad (9)$$

Here, we consider uncertainty calculated as 2σ standard deviations, i.e. 95% of the values lay inside the interval with the center in the reported emissions surrounded by the reported uncertainty.

The second approach is based on ensemble of the used prior emissions as an input for the inversion. The different ensemble members are built from five prior emissions (see Figure 1) while the uncertainty is calculated as the standard deviation of five resulting posterior emissions.

2.4*10*The calculated posterior uncertainty for our spatial domain and studied period (2013–2020) is shown in Figure 4Figure 4 (right). The uncertainty associated with Gaussian posterior for each year of the study period are depicted in Supplementary Figure S 5. The absolute uncertainty of Gaussian posterior ammonia emissions reaches a maximum of 23.3 ng m^{-2} s^{-1} or about 39% (relative value, calculated based on related maximum of posterior emissions). The uncertainty based on prior ensemble reaches a maximum of 60.2 ng m^{-2} s^{-1} which is equal to about 101% based on related maximum of posterior emissions. In general, the pattern of both posterior uncertainties, Gaussian posterior and prior ensemble respectively, are in agreement in theirs patterns and follow the one of the posterior emissions, with the highest values over (i) Belgium, the Netherlands, and Germany due to livestock, farming, and agricultural activity; (ii) Poland, South Romania and Croatia due to industrial applications; (iii) Catalonia due to pig farming; (iv) West France due to manure application. Nevertheless, the obtained posterior uncertainty remains low, and this depicts the robustness of the methodology used and the calculated posterior emissions of ammonia.

3.4 Validation of posterior emissions

As shown in Eq. 3 (Section 2.4), the inversion algorithm minimizes the distance between the satellite observations (v^{sat}) and the retrieved ammonia concentrations (v^{ret}). The latter is a function of different satellite parameters (e.g., averaging kernel sensitivities) and modelled ammonia concentrations using a prior dataset (v^{mod} or v^{true}) as seen in Eq. 1. The overall result is always propagated to v^{mod} iteratively, each time updating the prior emissions to obtain posterior ammonia. As specified in CrIS guidelines, modelled concentrations (v^{mod}) cannot be directly compared with satellite data (v^{sat}), while comparing v^{sat} with v^{ret} is not a proper validation method, because the comparison is performed for satellite observations that were included in the inversion (dependent observations), and the inversion algorithm has been designed to reduce the v^{sat}-v^{ret} mismatches. This means that the reduction of the posterior retrieved concentration (v^{ret}) mismatches to the observations (v^{sat}) is determined by the weighting that is given to the observations with respect to v^{ret}. A proper validation of the posterior emissions is performed against observations that were not included in the inversion (independent observations).

For these reasons, we compare modelled posterior concentrations of ammonia (v^{mod}) at the surface with ground-based observations over Europe from the EMEP (European Monitoring and Evaluation Programme, https://emep.int/mscw/) network (Torseth et al., 2012). The measurements are open in public and can be retrieved from https://ebas.nilu.no. We used measurements for all years between 2013 and 2020 from an average of 53 stations with 2928 observations for each station covering all Europe (Supplementary Figure S 6). The comparison is plotted for each of the 53 stations separately on a Taylor diagram in Figure 5. For all stations, the Pearson's correlation coefficient increased for the posterior ammonia (coloured circles) increased as compared to the prior one (coloured squares) reaching above 0.6 at several stations, while the normalized root mean square error (nRMSE) and standard deviation were kept below 2 (unitless) and 2 μg m-3, respectively, in almost all stations (except SI0008 in Slovenia).

To further show how posterior emissions of ammonia affect modelled concentrations, we chose six stations (DE0002 in Germany, NO0056 in South Norway, ES0009 in Spain, NL0091 in the Netherlands, HU0002 in Hungary and PL0005 in Poland) from the EMEP network (highlighted in red in Supplementary Figure S 6), and we plot prior and posterior concentrations against ground-based ammonia over time for the whole study period (2013–2020) in Supplementary Figure S 7. Given the long period of plotting, we average observations every week and modelled concentrations every month for a more visible representation of the

comparison. To evaluate the comparison, we calculate a number of statistic measures, namely nRMSE, the normalized mean absolute error (nMAE) and the root mean squared logarithmic error (RMSLE) as defined below:

$$nRMSE = \frac{\sqrt{\sum_{i=1}^{n} \frac{1}{n}(m_i - o_i)^2}}{\frac{1}{n}\sum_{i=1}^{n} o_i} \qquad nMAE = \frac{\sum_{i=1}^{n}|m_i - o_i|}{\sum_{i=1}^{n} o_i}$$

$$RMSLE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(\log m_i - \log o_i)^2} \qquad (11)$$

where n is sample size, m and o the individual sample points for model concentrations and observations of ammonia indexed with i. As one can see in Supplementary Figure S 7, all statistics were improved in all six stations and posterior concentrations were closer to the observations. However, individual peaks were in many cases misrepresented in the model. Whether this is a result of the measurement technique or the fact that local sources cannot be resolved at the spatiotemporal resolution of CTM and FLEXPART (given the short lifetime of atmospheric ammonia) needs further research. The best results were obtained at station ES0009 (Spain), where model captures the seasonal variation of the observations during the whole study period (2013–2020). In all other stations, the seasonality is maintained albeit steep peaks in the observations are lost.

4 Discussion

4.1 Comparison with emissions inferred from satellite observations

We compared our posterior estimates with two recently published studies on ammonia emission in Europe (Cao et al., 2022; Luo et al., 2022). Luo et al. (2022) used IASI observations for the period 2008 to 2018 to estimate ammonia emissions in a global domain. Their method was based on updating prior emissions with correction term computed using differences between observed and simulated ammonia columns combined with calculated ammonia lifetimes. The key indicators calculated for the European domain in Luo et al. (2022) are a linear trend for the 2008–2018 period, average annual emissions, and relative trends. Note that we compare our eight-year period with a decade in Luo et al. (2022). The comparison is depicted in Figure 6. Our estimates (Figure 6, left panel) are in good agreement with those calculated by Luo et al. (2022). The linear trend was estimated as -1.27 Tg for the period by Luo et al. (2022), while our estimate is -1.44 Tg. The spatial distribution of the trend is also given in Figure 6 (left panel). The key decrease is observed mainly in France, Germany, and middle Europe, while the increasing trend is observed mostly in Spain, parts of Italy, and

Greece. The average annual ammonia emission for the European domain in Luo et al. (2022) was estimated to be 5.05 Tg while our estimate is 4.63 Tg. Our lower estimate (by approximately 8%) may be attributed to use of more recent period considered in our study, but both methods agree that the trend in Europe is negative. The relative decrease estimated by Luo et al. (2022) is -25.1%, while we calculate -31.02%, which is again in very good agreement.

Cao et al. (2022) used CrIS observations for the year 2016 in order to estimate ammonia emissions for 25 European Union members (EU25), namely Austria, Belgium, Bulgaria, Croatia, Republic of Cyprus, Czech Republic, Denmark, Estonia, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, Netherlands, Poland, Portugal, Romania, Slovakia, Slovenia, and Spain. The method was tested with uni-directional and bi-directional flux schemes. The uni-directional dry deposition scheme assumes only air to surface exchange of ammonia ignoring changes in environmental conditions, while the bi-directional scheme captures dynamics in measured ammonia fluxes. Total estimated ammonia emissions for the EU25 region by the uni-directional scheme (posterior_uni) and the bi-directional scheme (posterior_bi) were reported as 3534 Gg N y−1 and 2850 Gg N y−1, respectively. The posterior_bi estimate is very close to our estimate for EU25 for the year 2016, which is 2712 Gg N y−1, while the posterior_uni is approximately 30% higher. A uni-directional dry deposition scheme ignores the impacts of changes in environmental conditions (e.g., soil temperature, soil wetness, soil pH, fertilized condition, and vegetation type) on ammonia emissions from fertilized soil and crops (volatilization), which likely lead to high biases in top-down estimates. Ammonia in LMDz-OR-INCA model, that was used to capture ammonia's losses, resembles a partially bi-directional treatment, where emissions and deposition are both possible at the same time without any use of a compensation point; this may explain the 30% difference.

The detailed EU25 emissions for the year 2016 are displayed in Figure 6 (right panel) for posterior_uni (red), posterior_bi (yellow), our post_avgEENV (blue), and priors used by Cao et al. (2022) and in our study (dashed red and blue, respectively). As seen from Figure 6, our posterior estimates (post_avgEENV) have more similar characteristics with posterior_bi, with monthly difference to be less than factor of 2 positive or negative from Cao et al. (2022). Note that the posterior_uni estimates are always a factor of 3 higher than our posterior estimates for ammonia emissions. The main differences can be observed during February-March and October-November periods where our estimates are generally lower than those from Cao et al. (2022).

4.2   Assessment of ammonia's atmospheric linearities

Ammonia is a particularly interesting substance due to its affinity to react with atmospheric acids producing secondary aerosols. In most cases, it is depleted by sulfuric and nitric acids. In acidic atmospheres where total ammonia (TA=gas, aqueous and solid) is less than twice the total sulfate ([TA]<2[TS]), all the available ammonia is taken up by the aerosol phase. In ammonia-rich environments ([TA]>2[TS]), the excess ammonia reacts with nitric acid forming ammonium nitrate. If RH is too high, ammonium nitrate is aqueous (Seinfeld and Pandis, 2000). As ammonia reacts with sulfate, it neutralizes sulfuric acid decreasing its concentration. Part of the sulfate may be replaced by nitric acid increasing ammonium nitrate content in the aerosol. If RH is high and particles are aqueous, the sulfate that reacts with ammonia and decreases, increases the equilibrium vapor pressure of ammonia with nitric acid shifting the reaction towards production of free ammonia (Seinfeld and Pandis, 2000). However, production of ammonia is a rare event and lots of prerequisites must be fulfilled in order to take place.

The latter is illustrated in Supplementary Figure S 8a that shows the frequency distribution of gain (negative numbers) or loss (positive numbers) due to all chemical processes affecting ammonia into the inversion domain (10°W–50°E, 25°N–75°N), for the study period (2013 – 2020) and the lowest sigma-p vertical levels (~1018-619 hPa) that were significant in satellite observations (see averaging kernels in section 2.1) (Sitwell and Shephard, 2021). The figure shows mostly positive numbers indicating that atmospheric ammonia reacts towards secondary aerosol formation. The spatial distribution of gain/loss of ammonia is shown in Supplementary Figure S 8b. The pixels indicating production of gaseous ammonia are located in marine regions, where we chose to not perform inversions, as they are an order of magnitude lower (Bouwman et al., 1997), thus less significant. No continental pixels showing gain of ammonia were were detected, which would cause simulated backwards in time to fail with our Lagrangian model (see next paragraph). Our approximation, although simplistic, provides computational efficiency when simulating SRMs in backward mode using FLEXPART (Pisso et al., 2019).

Seibert and Frank (2004) reported that standard Lagrangian particle dispersion models cannot simulate non-linear chemical reactions. However, all the other processes occurring during the atmospheric transport of trace substances are linear, i.e., advection, diffusion, convective mixing, dry and wet deposition, and radioactive decay. First-order chemical reactions, where the reaction rates can be prescribed, are also linear. Non-linear chemistry cannot be calculated because neither the background chemistry is modeled nor is the coupling

531 of the tracked plume (forward or backward) to this background. Technically, the SRM in

532 FLEXPART is calculated for a receptor with a certain mean mixing ratio (χ) and an emitting

533 source ($q_{i,n}$) in a certain discretization of the space (index i) and time (index n), as:

534 $$\frac{\chi}{q_{i,n}} = \frac{1}{J}\sum_{j=1}^{J} \Delta t_{i,j,n} \frac{p_{j,n}}{\rho_{i,n}} \qquad (12)$$

535 where J is the total number of backward trajectories (particles index j) originating from the

536 position of the receptor χ and ending at a certain discretized time (index n) in certain

537 discretized space position (index i) for a time interval $\Delta t_{i,j,n}$, and where the air density is $\rho_{i,n}$.

538 The further function $p_{j,n}$ ($p_{j,n} \leq 1$) represents the relative (to the initial receptor state) decay

539 of the mass value in the particle in its travel from the receptor to the discretized space time

540 interval (j,n) due to any linear decay process (e.g. deposition, linear chemical decay) for a

541 perfectly conserved scalar $p_{j,n} = 1$. So, for linear decaying species a direct SRM can be

542 calculated explicitly among all relevant receptor points and all positions in space and time. The

543 existence of the SRM (**H**), linking directly mixing ratios at the receptor points with emissions,

544 is the pre-requisite to apply simple inversion algorithms such as the one we use in the present

545 study.

546   Inversion of observation to obtain emission for non-linear chemically reactive species

547 entails the need to run a chemistry transport model (CTM) forward (and its adjoint backward)

548 in time from time t_0 to time $t_{\square}$ evolving the full state of the atmosphere in relation to the

549 emissions and boundary conditions. Subsequently, a cost function is evaluated by an iterative

550 descent gradient method that implies running the adjoint of the forward model (Fortems-

551 Cheiney et al., 2021). Note that an iterative algorithm means that the forward and adjoint

552 models run several times in sequence until the estimated minimum of the cost function is

553 reached.

554   To overcome these complexities, we examine the linearities of our method and show that

555 FLEXPART simulates ammonia efficiently, we evaluate modelled ammonia against ground-

556 based measurements of ammonia from the EMEP network (https://emep.int/mscw/) in Europe,

557 EANET (East Asia acid deposition NETwork) in Southeastern Asia (https://www.eanet.asia/)

558 and AMoN (Ammonia Monitoring Network in the US, AMoN-US; National Air Pollution

559 Surveillance Program (NAPS) sites in Canada) in North America

560 (http://nadp.slh.wisc.edu/data/AMoN/). The SRMs for ammonia calculated with the backward

561 mode express the emission sensitivity (in seconds), which yields a modelled concentration in

562 the receptor point (station) when coupled with gridded emissions (in kg m-2 s-1) at the lowest

563 model level (100 m). Here, we used the ECLIPSEv5, GFED4 and GEIA, (EGG) emissions

564 (Bouwman et al., 1997; Giglio et al., 2013; Klimont et al., 2017) to get concentrations. To check

565 the consistency of the proxy used in the SRMs of ammonia, we also simulated surface

566 concentrations of ammonia with FLEXPART in forward mode using the same emissions. We

567 have chosen two random ground-based stations from each of the three measuring networks

568 (EMEP, EANET, AMoN) to compare modelled concentrations. For consistency, we also plot

569 the resulting surface concentrations from the LMDz-OR-INCA model (Supplementary Figure

570 S 9).

571   Modelled concentrations (forward and backward FLEXPART and the CTM LMDz-OR-

572 INCA) at each station have been averaged to the temporal resolution of the observations.

573 Supplementary Figure S 10 shows Taylor diagrams of the comparison between FLEXPART

574 simulated concentration in forward and backward mode. Plotting backward versus forward

575 results is a common procedure to infer whether a Lagrangian model produces reasonable results

576 (Eckhardt et al., 2017; Pisso et al., 2019). In general, the forward and backward simulations

577 show very good agreement for the depicted receptor points. For example, ammonia

578 concentration at stations AL99, CA83, and VNA001 (Supplementary Figure S 9) are simulated

579 similarly, and the mean concentrations are almost identical in the forward and backward modes.

580 However, during some episodes there can be notable differences (e.g., at DE0002R) as seen

581 before (Eckhardt et al., 2017). The Taylor diagram for the respective comparison

582 (Supplementary Figure S 10) show high Pearson's correlation coefficients (>0.7), low standard

583 deviations (<1 μg N m-3) and root means square errors (RMSEs <0.7 μg N m-3).

5 Conclusions

585   Today, a large debate takes place about ammonia abatement strategies for Europe, but

586 also for Southeastern Asia, in an effort to reduce secondary formation and, thus, mitigate

587 climate crisis (van Vuuren et al., 2015). These strategies include (a) low nitrogen feed by

588 reducing ammonia emissions at many stages of manure management, from excretion in

589 housing, through storage of manure to application on land, also having positive effects on

590 animal health and indoor climate (Montalvo et al., 2015); (b) low emission livestock housing,

591 which focuses on reducing the surface and time manure is exposed to air by adopting rules and

592 regulations regarding new livestock houses (Poteko et al., 2019); (c) air purification by

593 adopting technologies to clean exhaust air from livestock buildings (Cao et al., 2023) and

594 others. Here we used satellite observations from CrIS and a novel inverse modelling algorithm

595 to study the spatial variability and seasonality of ammonia emissions over Europe. We then

evaluated the overall impact of such strategies on the emissions of ammonia for the period 2013–2020. The main key messages can be summarized below:

- The highest emissions over the 2013–2020 study period occur in North Europe (Belgium, the Netherlands and northwestern Germany). At a regional scale, peaks are seen in Western Europe (Poland, South Romania and Croatia) due to industrial activities, in Spain (Ebro Valley, Aragon, Catalonia) due to agricultural activities and farming, in Belgium and The Netherlands due to livestock activity (dairy cow, beef cattle, pig and chicken farming).

- Ammonia emissions are low in winter (average: 286 Gg) and peak in summer (average: 563 Gg), due to temperature dependent volatilization of ammonia, while a notable peak attributed to fertilization can be seen in early spring during some years.

- Over the 2013–2020 period, European emissions of ammonia decreased from 5431 Gg in 2013 to 3994 Gg in 2020 or about -26%. Hence, the restrictions adopted by the European Union members were effective in reducing secondary PM formation.

- A slight emission increase of +4.4% in 2015 appears for several European Union Member States (Germany, Spain, France, and the United Kingdom) who exceeded the respective ammonia emission targets. Part of the 2020 ammonia decrease might be attributable to the COVID-19 pandemic restrictions.

- The largest decreases in ammonia emissions were observed in Central and Eastern Europe (-38%, 2190 Gg in 2013 to 1495 Gg in 2020) and in Western Europe (-37%, 2041 Gg in 2013 to 1421 Gg in 2020). Smaller decreases were calculated in Northern Europe (-17%, 398 Gg in 2013 to 333 Gg in 2020) and, Southern Europe (-7.6%, from 803 Gg in 2013 to to 743 Gg in 2020).

- The maximum calculated absolute uncertainty of Gaussian posterior model was 23.3 ng m^{-2} s^{-1}, or about 39% (relative value) and calculated maximum based ensemble of prior emissions was 60.2 ng m^{-2} s^{-1}, or about 101% following the spatial distribution of the posterior emissions.

- Comparison of the concentrations calculated with prior and posterior ammonia emissions against independent (not used in the inversion algorithm) observations showed improved correlation coefficients and low nRMSEs and standard deviations. Looking at timeseries of six randomly selected stations in Europe, we also found that posterior surface concentrations of ammonia were in accordance with the ground-based measurement, also following the observed seasonal trends.

- Our results agree very well with those from Luo et al. (2022) (decreasing trend: -1.44 versus -1.27 Tg, annual European emissions: 4.63 versus 5.05 Tg) and those from Cao et al. (2022) following their methodology (their posterior_bi estimate for EU25 and year 2016 was 2850 Gg N y−1, while we calculate 2712 Gg N y−1).
- The relatively low posterior uncertainty and improved statistics in the validation of the posterior surface concentrations denote the robustness of the posterior emissions of ammonia calculated with satellite measurements and our adapted inverse framework.

Data availability. The data generated for the present paper can be downloaded from ZENODO (https://doi.org/10.5281/zenodo.7646462). FLEXPARTv10.4 is open access and can be downloaded from https://www.flexpart.eu/downloads, while use of ERA5 data is free of charge, worldwide, non-exclusive, royalty-free and perpetual. The inversion algorithm LS-APC is open access from https://www.utia.cas.cz/linear_inversion_methods. CrIS ammonia can be obtained by request to Dr. M. Shephard (Mark.Shephard@ec.gc.ca). EMEP measurements are open in https://ebas.nilu.no. FLEXPART SRMs for 2013–2020 can be obtained from the corresponding author upon request.

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1012   **FIGURES & LEGENDS**

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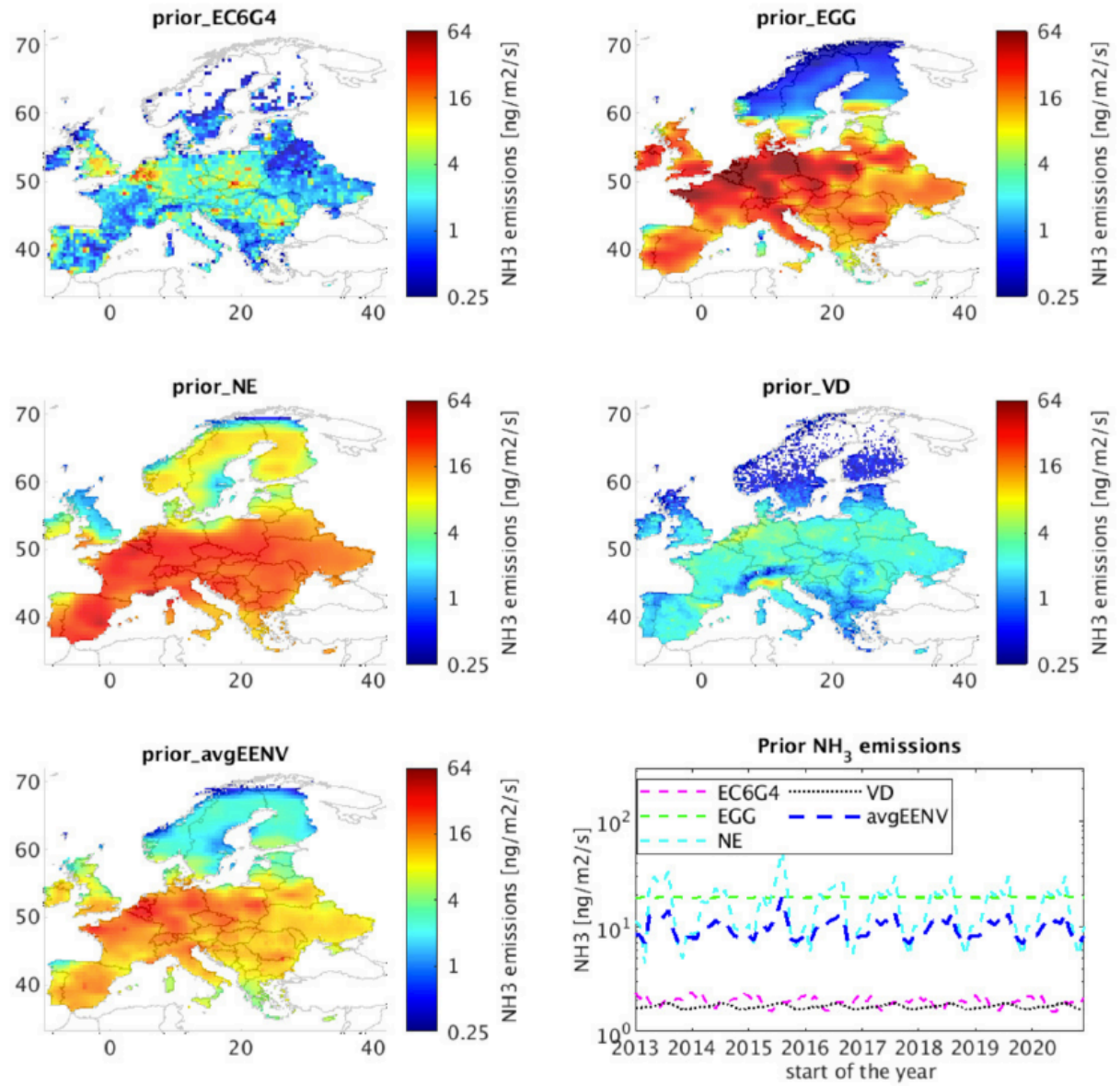
1014   **Figure 1.** Four ammonia prior emissions (EC6G4, EGG, NE, VD) are displayed in the first two
1015   rows. The combined prior (avgEENV) is displayed in the bottom left. The temporal variability
1016   of all five prior emissions is given in bottom right.

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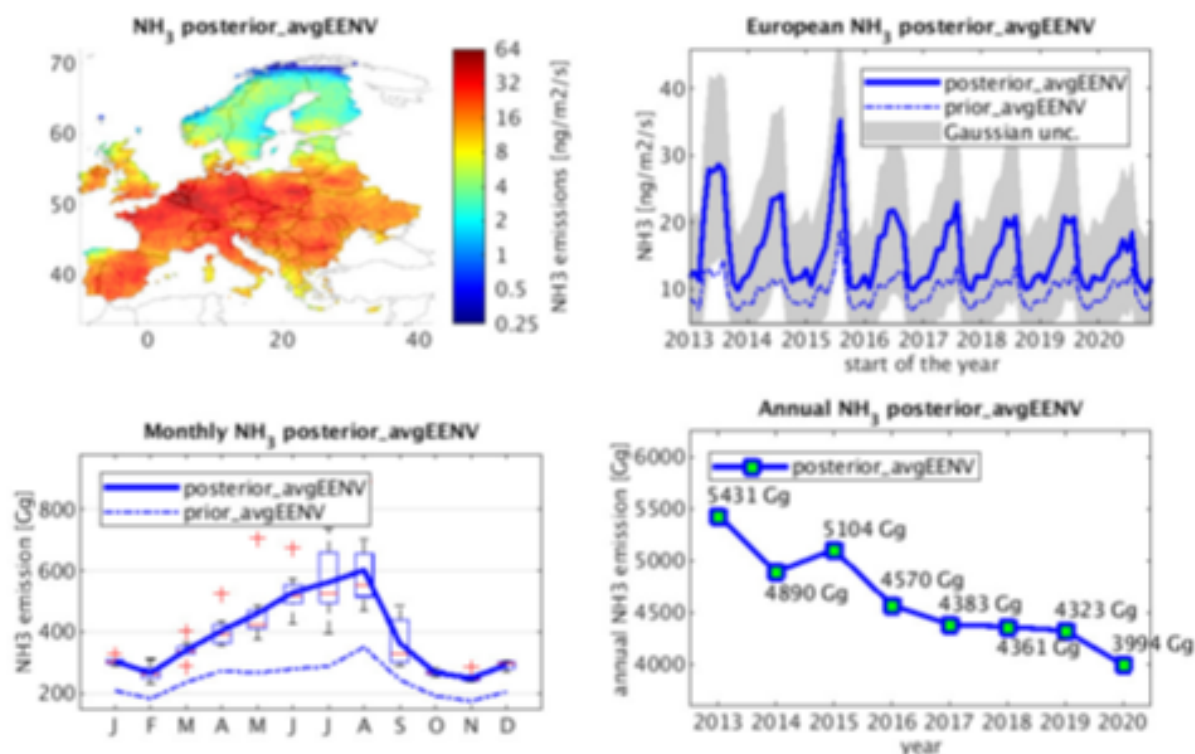
Figure 2. The spatial distribution of posterior ammonia emissions (posterior_EENV, top-left) together with its temporal distribution (top-right). The Gaussian uncertainty of the posterior emissions is also plotted. Monthly average (bottom-left) and annually average (bottom-right) estimates are also plotted. The monthly average posterior emissions over the studied period are accompanied by the box plot where the red line indicates the median, the bottom and top edges of the Boxes indicating the 25th and 75th percentiles, respectively, and the whiskers extend to the most extreme data points not considered as outliers, which are denoted using red crosses. Solid blue lines refer to the posterior ammonia emissions, while dashed ones to the prior emissions (avgEENV).

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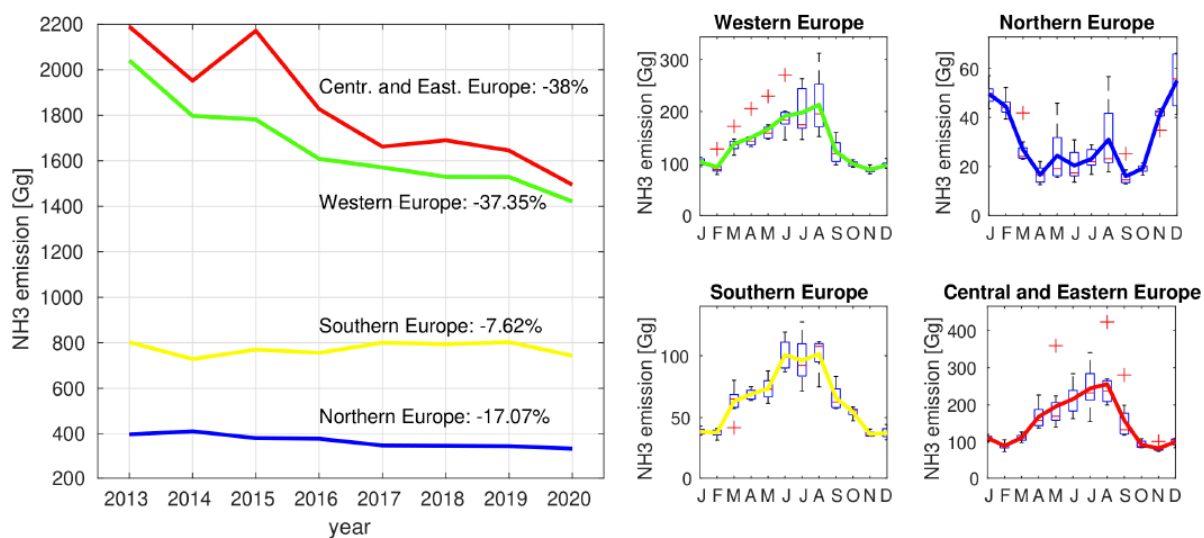
1030 **Figure 3.** Left: Annual posterior emissions of ammonia in Southern (yellow), Western (green),
1031 Northern (blue), and Central and Eastern (red) Europe. Right: Monthly average posterior
1032 emissions of ammonia accompanied by box plots, where the red line indicates the median, the
1033 bottom and top edges of the box indicate the 25th and 75th percentiles, respectively, and the
1034 whiskers extend to the most extreme data points (not considered outliers), which are represented
1035 using red crosses.

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1038 **Figure 4.** Absolute uncertainty of posterior emissions of ammonia calculated as 2σ (left
1039 panel) and from a member ensemble (right panel) comprising posterior emissions calculated
1040 with five different priors (**Figure 1**) averaged for the whole study period 2013–2020.

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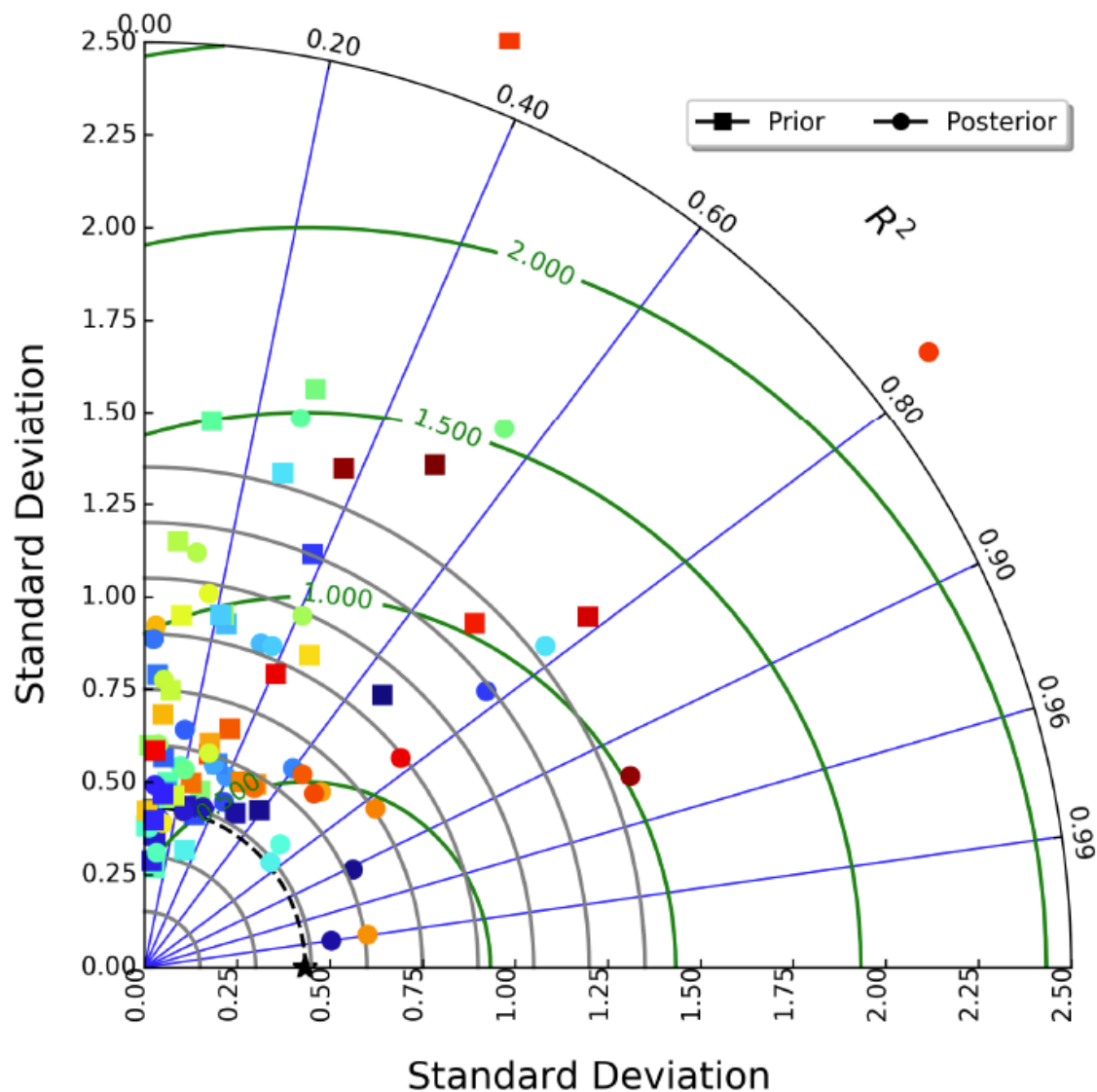
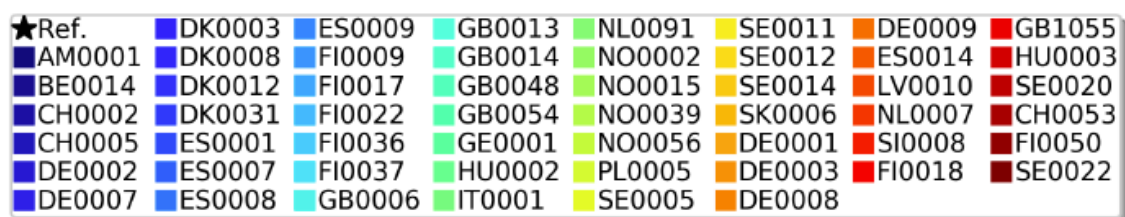
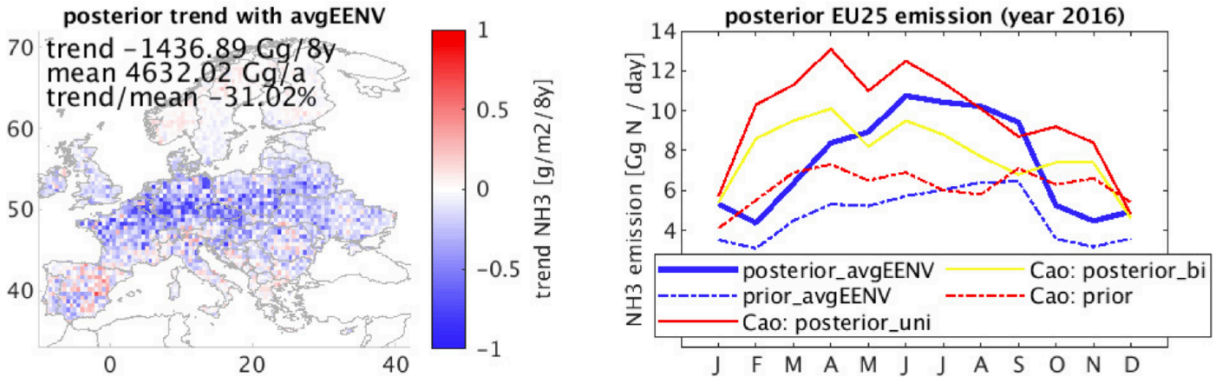


Figure 5. Modelled concentrations of ammonia with prior and posterior emissions against ground-based observations from 53 EMEP stations for 2013–2020 presented in a Taylor diagram. The diagram shows the Pearson's correlation coefficient (gauging similarity in pattern between the modelled and observed concentrations) that is related to the azimuthal angle (blue contours); the standard deviation of modelled concentrations of ammonia is proportional to the radial distance from the origin (black contours) and the centered normalized RMSE of modelled concentrations is proportional to the distance from the reference standard deviation (green

 contours).

 Figure 6. Left: spatial distribution of ammonia emission trends computed for the studied period
2013–2020 in the same way as in (Luo et al., 2022), where also trend, mean, and trend/mean
are defined/computed in the same way. Right: comparison of ammonia emissions from the
EU25 countries for the year 2016 from our posterior calculations (posterior_avgEENV, blue)
and results from Cao et al. (2022) (posterior_uni in red and posterior_bi, in yellow).

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