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# 1 **Estimating agricultural ammonia volatilization over Europe** 2 **using satellite observations and simulation data**

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15

## 16 **Abstract**

17 Ammonia (NH<sub>3</sub>) is one of the most important gases emitted from agricultural practices. It affects air  
18 quality and the overall climate, and in turn influenced by long term climate trends as well as by short term  
19 fluctuations in local and regional meteorology. Previous studies have established the capability of the Infrared  
20 Atmospheric Sounding Interferometer (IASI) series of instruments aboard the Metop satellites to measure  
21 ammonia from space since 2007. In this study, we explore the interactions between atmospheric ammonia, land  
22 and meteorological variability, and long-term climate trends in Europe. We investigate the emission potential  
23 ( $\Gamma_{soil}$ ) of ammonia from the soil, which describes the soil – atmosphere ammonia exchange.  $\Gamma_{soil}$  is generally  
24 calculated in-field or in laboratory experiments; here, and for the first time, we investigate a method which assesses  
25 it remotely using satellite data, reanalysis data products, and model simulations.

26 We focus on ammonia emission potential during March 2011, which marks the start of growing season  
27 in Europe. Our results show that  $\Gamma_{soil}$  ranges from  $2 \times 10^3$  to  $9.5 \times 10^4$  (dimensionless) in a fertilized cropland,  
28 such as in the North European Plain, and is of the order of  $10 - 10^2$  in a non-fertilized soil (e.g. forest and grassland).  
29 These results agree with in-field measurements from the literature, suggesting that our method can be used in other  
30 seasons and regions in the world. However, some improvements are needed in the determination of mass transfer  
31 coefficient  $k$  (m s<sup>-1</sup>), which is a crucial parameter to derive  $\Gamma_{soil}$ .

32 Using a climate model, we estimate the expected increase in ammonia columns by the end of the century  
33 based on the increase in skin temperature (T<sub>skin</sub>), under two different climate scenarios. Ammonia columns are  
34 projected to increase by up to 50 %, particularly in Eastern Europe, under the SSP2-4.5 scenario, and might even  
35 double (increase of 100 %) under the SSP5-8.5 scenario. The increase in skin temperature is responsible for a  
36 formation of new hotspots of ammonia in Belarus, Ukraine, Hungary, Moldova, parts of Romania, and  
37 Switzerland.

38



## 39 1. Introduction

40 Ammonia ( $\text{NH}_3$ ) emissions have been increasing in a continuous manner from 1970 to 2017 (McDuffie et al.,  
41 2020). During the period 2008 – 2018 alone, the increase in ammonia columns in Western and Southern Europe  
42 accounted to  $20.8 \text{ \% yr}^{-1} (\pm 4.3 \text{ \%})$ , and to  $12.8 (\pm 1.3 \text{ \%})$  globally (Van Damme et al., 2021). Although ammonia  
43 alone is stable against heat and light, it is considered a very reactive base, whereas it constitutes the largest portion  
44 of the reactive nitrogen ( $\text{N}_r$ ) on Earth. The vast majority of atmospheric ammonia not deposited is transformed into  
45 fine particulate matter ( $\text{PM}_{2.5}$ ) composed of ammonium ( $\text{NH}_4^+$ ), through acid – base chemical reactions with  
46 available acids in the environment, namely sulfuric acid ( $\text{H}_2\text{SO}_4$ ), hydrochloric acid ( $\text{HCl}$ ), and nitric acid ( $\text{HNO}_3$ )  
47 (Yu et al., 2018), while only 10 % of the total ammonia gas are believed to be oxidized by hydroxyl radicals ( $\text{OH}^\cdot$ )  
48 (Roelle and Aneja, 2005).  $\text{PM}_{2.5}$  has degrading effects on human health, especially respiratory diseases (Bauer et  
49 al., 2016). In addition to agriculture, ammonia can be emitted from industrial processes, biomass burning (Van  
50 Damme et al., 2018), and natural activities such as from seal colonies (Theobald et al., 2006).

51  
52 Soils are known to be a source of atmospheric ammonia, especially in areas of intensive agricultural practices  
53 (Schlesinger and Hartley, 1992), and this is due to enriching the soil with the reactive nitrogen present in fertilizers.  
54 The increase in the application of synthetic fertilizers, and intensification of agricultural practices is believed to be  
55 the dominant factor of the global increase in ammonia emissions over the past century (Behera et al., 2013;  
56 McDuffie et al., 2020).

57  
58 Following the application of fertilizers, ammonium and ammonia are released in the soil. Prior to its volatilization,  
59 ammonia in the soil exists either in the gas phase ( $\text{NH}_{3(g)}$ ) or in the aqueous/liquid phase ( $\text{NH}_{3(aq)}$ ), the equilibrium  
60 between both states of ammonia is governed by Henry's law (Wentworth et al., 2014), as shown in A. The  
61 dissociation of ammonia in soil water is affected by soil acidity (pH) and temperature (Roelle and Aneja, 2005)  
62 (Eq. (A-1) and (A-2) in Appendix A); it is explained by the dissociation constant  $K_{\text{NH}_4^+}$ . Once released to the  
63 atmosphere, ammonia near the surface exists in the gas phase, hence Henry's law describes the equilibrium  
64 between ammonia in the soil (liquid phase), and near the surface (gas phase). This bi-directional exchange between  
65 the soil and the atmosphere will continue until the equilibrium is reached, and this occurs when ammonia  
66 concentration is equal to the compensation point  $\chi_{\text{NH}_3}$  (the concentration of ammonia at equilibrium). The flux of  
67 ammonia from the soil to the atmosphere (emission) occurs when the concentration of atmospheric ammonia is  
68 less than the compensation point  $\chi_{\text{NH}_3}$ , while ammonia deposition occurs when the concentration of ammonia is  
69 equal to or greater than  $\chi_{\text{NH}_3}$  (Flechard et al., 2011; Wichink Kruit, 2010). It is then crucial to quantify the  
70 compensation point in order to understand this bi-directional exchange. The main variables needed to calculate  
71  $\chi_{\text{NH}_3}$  are soil temperature ( $T_{\text{skin}}$ ) and  $\Gamma_{\text{soil}}$ , which is a dimensionless ratio between ammonium and pH ( $\text{NH}_4^+_{(aq)}$   
72 and  $\text{H}^+_{(aq)}$  concentrations, respectively, in the soil). All the equations are described in Appendix A (Eq. (A-1) to  
73 (A-15)).

74  
75 The soil emission potential ( $\Gamma_{\text{soil}}$ ) has been thoroughly investigated in field and controlled laboratory environments  
76 (e.g. David et al., 2009; Flechard et al., 2013; Massad et al., 2010; Mattsson et al., 2008; Nemitz et al., 2000;  
77 Wentworth et al., 2014, among others).  $\Gamma_{\text{soil}}$  is dimensionless and it can range from 20 (non-fertilized soil in a  
78 forest) to the order of  $10^6$  (mixture of slurry in a cropland). It is found to peak right after fertilizers application,  
79 due to the increase in ammonium content in the soil (a product of urea hydrolysis), reaching pre-fertilization levels  
80 10 days following the application (Flechard et al., 2010; Massad et al., 2010). Little information exists on regional  
81 or global scales to assess the large-scale spatial variability of ammonia emission potentials.

82  
83 In order to meet the needs for a growing population, agricultural practices have intensified during the period 2003  
84 – 2019 (more fertilizer use per surface area), resulting in an increase in the net primary production (NPP) per capita



85 (Potapov et al., 2022), subsequently increasing volatilized ammonia (increase in nitrogen soil content, and  
86 cultivated lands). In Europe alone, the area of croplands increased by 9 % from 2003 to 2019, and most of the  
87 expansion took place on lands that were abandoned for more than 4 years (Potapov et al., 2022). Between the year  
88 2008 and 2018, the increase in atmospheric ammonia columns accounted to 20.8 % ( $\pm 4.3$  %) in Western and  
89 Southern Europe (Van Damme et al., 2021). With the increase in croplands area and agricultural activities, climate  
90 change will have a significant effect on agricultural practices, with warmer climates enhancing the volatilization  
91 of ammonia from soils, especially in intensely fertilized lands (Shen et al., 2020).

92  
93 This study aims at exploring ammonia emission potential/volatilization in Europe, using infrared satellite data of  
94 ammonia columns, reanalysis temperature data, and chemical transport model simulations to provide information  
95 on chemical sources and sinks. We specifically study the relationship between satellite-derived ammonia  
96 concentration at the start of the growing season, soil emission potentials and their spatial variability over Europe  
97 during March of 2011. Section 2 provides the methods/datasets used. Simulation results are described in Sect. 3,  
98 and regional emission potentials are shown and discussed in Sect. 4. Using a climate model, future projections of  
99 ammonia columns are investigated under different climate scenarios in Sect. 5. Discussion and conclusions are  
100 listed in Sect. 6.

## 101 2. Methodology

### 102 2.1. Calculation of the emission potential

103  
104 In this study, we use IASI satellite data to calculate the ammonia emission potential  $\Gamma_{soil}$  instead of field soil  
105 measurements. In field studies,  $\Gamma_{soil}$  is calculated by measuring the concentration of ammonium ( $\text{NH}_4^+$ ) and  $\text{H}^+$   
106 ( $10^{-\text{pH}}$ ) in the soil; the ratio between both of these concentrations is  $\Gamma_{soil}$ . In this study, we use infrared satellite  
107 ammonia to have a regional coverage over Europe. With these, we cannot monitor soil content of ammonium nor  
108 its pH. This renders the remote  $\Gamma_{soil}$  calculation challenging, and less straight forward. The full derivation of the  
109 equation used to calculate the emission potential is explained in Appendix A, and was briefly described in the  
110 introduction. In short, upon its dissolution in the soil water, ammonia follows Henry's law. In steady state  
111 conditions between the soil and the near surface, the amount of the ammonia emitted and lost is considered equal.  
112 Based on this assumption, the soil emission potential (dimensionless) is calculated as follows Eq. (2-1) or Eq. (A-  
113 15) in Appendix A:

$$114 \Gamma_{soil} = \frac{[\text{NH}_3]^{col} \cdot T_{soil}}{\exp\left(\frac{-b}{T_{soil}}\right)} \frac{M_{\text{NH}_3}}{a \cdot N_a \cdot c'} \cdot \frac{1}{k\tau} \quad (2-1)$$

115  
116 Where  $[\text{NH}_3]^{col}$  is the total column concentration of ammonia (molecules  $\text{cm}^{-2}$ ), measured by satellite remote  
117 sensors,  $T_{soil}$  is the soil temperature at the surface, which can be expressed as the skin temperature,  $T_{skin}$  (Kelvin),  
118  $a$  and  $b$  are constants ( $a = 2.75 \times 10^3 \text{ g K cm}^{-3}$ ,  $b = 1.04 \times 10^4 \text{ K}$ ),  $M_{\text{NH}_3}$  is the molar mass of ammonia gas  
119 ( $M = 17.031 \text{ g mol}^{-1}$ ), and  $N_a$  is Avogadro's number ( $N_a = 6.0221409 \times 10^{23} \text{ molecules mol}^{-1}$ ),  $c'$  is  
120 equals to 100 and is added to convert  $k$  from  $\text{m s}^{-1}$  to  $\text{cm s}^{-1}$  (since  $[\text{NH}_3]^{col}$  is in molecules  $\text{cm}^{-1}$ ), and  $\tau$  the lifetime  
121 of ammonia (seconds).

122  $k$  is the soil – atmosphere exchange coefficient or deposition velocity ( $\text{cm s}^{-1}$ ), also known as the mass transfer  
123 coefficient (this nomenclature will be used in this study). It is found to be affected by the roughness length of the  
124 surface, wind speed, the boundary layer height (Olesen and Sommer, 1993; Van Der Molen et al., 1990), and pH



125 (Lee et al., 2020). It can be explained by a resistance model often used to explain the exchange between the surface  
126 and the atmosphere (Wentworth et al., 2014). Different studies provide look up tables values of  $k$  for different land  
127 cover types and different seasons based on this resistance model (Aneja et al., 1986; Erisman et al., 1994; Phillips  
128 et al., 2004; Roelle and Aneja, 2005; Svensson and Ferm, 1993; Wesely, 1989).  
129 In general, the mass transfer coefficient  $k$  is in the order of  $10^{-3}$  to  $10^{-2}$   $\text{m s}^{-1}$  in a mixture of soil and manure, and  
130  $10^{-6}$  to  $10^{-5}$   $\text{m s}^{-1}$  in a mixture of manure alone (Roelle and Aneja, 2005). We discuss and provide more information  
131 on  $k$  in Sect. 4, and additional details on this calculation in general are provided in Appendix A.  
132

## 133 2.2. IASI ammonia, ERA5 T skin, and MODIS Land cover

134  
135 The Infrared Atmospheric Sounding Interferometer (IASI) is considered advanced the most innovative  
136 instrument onboard the polar-orbiting Metop satellites (Klaes, 2018). Three IASI instruments are onboard Metop-  
137 A, B and C, the series of satellites launched by the EUMETSAT (European Organization for the Exploitation of  
138 Meteorological Satellites) in 2006, 2012, and 2018, respectively. The Metop-A satellite was de-orbited in October  
139 2021 (Lentze, 2021), and as a result only two instruments (IASI-B and C onboard Metop-B and C) are operating  
140 today. The observations from IASI cover any location on Earth at 9:30 in the morning (AM) and in the evening  
141 (PM), local solar time. It can detect a variety of atmospheric species including trace gases (Clerbaux et al., 2009).  
142 The IASI Fourier-transform spectrometer monitors the atmosphere in the spectral range between 645 and 2760  
143  $\text{cm}^{-1}$  (thermal infrared), and is nadir-looking. IASI has a swath width that measures 2200 km, with a pixel size of  
144  $\sim 12$  km.  
145

146 Ammonia was first detected with IASI using the  $\nu_2$  vibrational band of ammonia ( $\sim 950$   $\text{cm}^{-1}$ ) (Clerbaux et al.,  
147 2009; Coheur et al., 2009). The ammonia total columns used in this study are the product of an Artificial Neural  
148 Network and re-analyzed temperature data from the European Centre for Medium-Range Weather Forecasts  
149 (ECMWF) product ERA5 ANNI-NH<sub>3</sub>-v3R-ERA5 (Van Damme et al., 2021). Several studies used ammonia data  
150 from IASI to study hotspots of ammonia of different source types including both natural and anthropogenic sources  
151 (Clarisse, Van Damme, Clerbaux, et al., 2019; Clarisse, Van Damme, Gardner, et al., 2019; Dammers et al., 2019;  
152 Van Damme et al., 2018, 2021; Viatte et al., 2021). Recently, IASI observations were used to study the effect of  
153 war and conflict on agricultural practices in Syria (Abeed et al., 2021).  
154

155 Fewer errors on the retrieval were observed during the day and over land (Van Damme et al., 2017), hence, we  
156 use only daytime ammonia measurements from IASI. Comparisons with ammonia measured using a ground-based  
157 instrument showed a good correlation of  $R=0.75$  (Viatte et al., 2021). Satellite ammonia data from CrIS (Crosstrack  
158 Infrared Sounder) (Shephard and Cady-Pereira, 2015) were compared with those from IASI, and were equally  
159 found to give similar results when looking at concentrations from a wildfire (Adams et al., 2019), showing  
160 consistency when studying seasonal and inter-annual variability (Viatte et al., 2020).  
161

162 In addition to ammonia, we look at skin temperature ( $T_{\text{skin}}$  or land surface temperature LST) data from the  
163 ECMWF reanalysis (ERA5) at a grid resolution of  $0.25 \times 0.25^\circ$  (Hersbach et al., 2020). ERA5 Temperatures are  
164 interpolated temporally and spatially to the IASI morning overpass ( $\sim 9:30$  A.M. local time), since we only consider  
165 daytime ammonia. ERA5 temperature data are also used in the retrieval process of the ammonia data we used in  
166 this study NH<sub>3</sub>-v3R-ERA5 (Van Damme et al., 2021).  $T_{\text{skin}}$  is defined as the temperature of the uppermost surface  
167 layer when radiative equilibrium is reached. It also represents the theoretical temperature required in order to reach  
168 the surface energy balance (ECMWF, 2016).  
169



170 In order to assign a mass transfer coefficient  $k$  to each land type, the moderate resolution imaging  
171 spectroradiometer (MODIS), a series of instruments orbiting the Earth aboard the Aqua and Terra satellites, is  
172 used. The data product MCD12Q1 (version 6) is a combined Aqua/Terra Land cover type product, with a spatial  
173 resolution of 500 m. This product provides maps of land cover type from 2001 through 2019 (Sulla-Menashe and  
174 Friedl, 2018). From the land use categories included in the MOD12Q1 product (Belward et al., 1999) we focus on  
175 croplands, forests, shrublands, and grasslands. We do not include barelands, snow cover, and urban areas in our  
176 analysis; we are not interested in studying these surfaces, since we focus on ammonia volatilization from the soil  
177 in areas amended with fertilizers. We show the emission potential in Forests and grasslands/shrublands for  
178 comparison with values in the literature. In an attempt to calculate an emission potential (Eq. (2-1)) that is relevant  
179 to the land cover/use, we therefore assign a mass transfer coefficient  $k$  to each land type based on literature values  
180 (Aneja et al., 1986; Erisman et al., 1994; Roelle and Aneja, 2005; Svensson and Ferm, 1993; Wesely, 1989) and  
181 we discuss it in Sect. 4.  
182

### 183 **2.3. Model simulations**

#### 184 **2.3.1. GEOS-Chem Chemistry Transport Model**

185  
186 In this study we use version 12.7.2 of the GEOS-Chem chemical transport model (Bey et al., 2001). The model is  
187 driven by the Modern-Era Retrospective Analysis for Research and Applications version 2 (MERRA-2) reanalysis  
188 product, including nested domains over Europe at a  $0.5^\circ \times 0.625^\circ$  horizontal resolution. MERRA-2 is the second  
189 version of the MERRA atmospheric reanalysis product by NASA Global Modulation Assimilation Office  
190 (NASA/GMAO) (Gelaro et al., 2017). Boundary conditions for the nested domains are created using a global  
191 simulation for the same months at  $2^\circ \times 2.5^\circ$  resolution. We generate model output for March of 2011, preceded by  
192 a one month of discarded model spin-up time for the nested run, and two months for the global simulation. March  
193 corresponds well to the month of fertilizer application in Europe, and as such to the beginning of the growing  
194 season (FAO, 2022; USDA, 2022).  
195

196 Output includes the monthly mean for selected diagnostics. Anthropogenic emissions are taken primarily from the  
197 global Community Emissions Data System (CEDS) inventory (Hoesly et al., 2018). Biogenic non-agricultural  
198 ammonia, as well as ocean ammonia sources, are taken from the Global Emission Inventories Activities database  
199 (GEIA, (Bouwman et al., 1997)). Open fire emissions are generated using the GFED 4.1s inventory (Randerson et  
200 al., 2015). We used the Harmonized Emissions Component module (HEMCO) to obtain the ammonia emissions  
201 over Europe (Keller et al., 2014).  
202

#### 203 **2.3.2. EC-Earth Climate model**

204 To analyze how future climate will affect ammonia concentration and emission potential, we use the ECMWF  
205 climate model the European Earth Consortium climate model (EC-Earth, <http://www.ec-earth.org/>). While other  
206 climate models exist, we choose this one because the ammonia product from IASI uses ERA5 for the retrievals  
207 and we calculate the emission potential from the T skin product of ERA5. The reanalysis uses the ECMWF  
208 Integrated Forecasting System for the atmosphere–land component (IFS). IFS is also used in EC-Earth and is  
209 complemented with other model components to simulate the full range of Earth system interactions that are  
210 relevant to climate (Döscher et al., 2021). We note that the versions of the IFS models used in ERA5 and in EC-  
211 Earth are not identical as the climate model product is not assimilated and is not initialized with observations



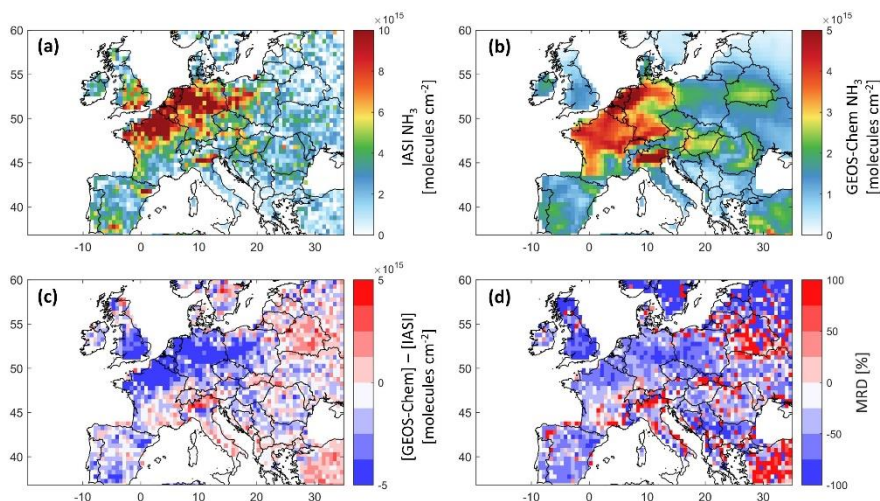


212 several times a day like ERA5. The EC-Earth simulations are included in the Climate model intercomparison  
213 project, phase 6 (Eyring et al., 2016), part of the Intergovernmental Panel on Climate Change (IPCC) report of  
214 2021 (Masson-Delmotte, et al., 2021). We use the so-called Scenario Model Intercomparison Project  
215 (ScenarioMIP), covering the period [2015 – 2100] for future projections under different shared socio-economic  
216 pathways (SSP) (Riahi et al., 2017). We analyze two scenarios, the SSP2-4.5 corresponding to “middle of the  
217 road” socio-economic family with a nominal  $4.5\text{W/m}^2$  radiative forcing level by 2100 - approximately  
218 corresponding to the RCP-4.5 scenario, and the SSP5-8.5 marks the upper edge of the SSP scenario spectrum with  
219 a high reference scenario in a high fossil-fuel development world throughout the 21st century.

### 220 3. GEOS-Chem model simulation: validation and analysis

#### 221 3.1. GEOS-Chem validation with IASI

222  
223 In order to analyse how well the model simulates atmospheric ammonia, we use the simulated GEOS-Chem  
224 monthly averaged (March 2011) ammonia total columns output (Sect. 2.3.1). We compare those to the IASI total  
225 columns of ammonia gridded on the same horizontal resolution ( $0.5^\circ \times 0.625^\circ$ ) and over the same month.



**Figure 1. Ammonia total column concentrations from IASI (panel a), and GEOS-Chem (panel b), the difference between both datasets (panel c) in molecules  $\text{cm}^{-2}$ , and the Mean Relative Difference (MRD) in % (panel d); all data are a monthly average of March 2011, and over Europe at a  $0.5^\circ \times 0.625^\circ$  grid resolution. Note that the colour bar limits are different between panels (a) and (b).**

226  
227 Figure 1 shows the IASI  $\text{NH}_3$  distribution (Figure 1a), and that from GEOS-Chem (Figure 1b), the bias between  
228 the two (Figure 1c), and the mean relative difference MRD (Figure 1d), all during March 2011. MRD is calculated  
229 as the mean of the ratio  $\frac{(\text{GeosChem } \text{NH}_3 - \text{IASI } \text{NH}_3) \times 100}{\text{IASI } \text{NH}_3}$  at each grid point.

230



231 Generally, both GEOS-Chem and IASI show coincident sources of ammonia, reflecting the good ability of the  
232 model to reproduce ammonia columns over major agricultural source regions in Europe. The bias between IASI  
233 and GEOS-Chem and the MRD are shown in Figure 1c and d. Ammonia columns from GEOS-Chem are  
234 underestimated by up to  $2 \times 10^{16}$  molecules/cm<sup>2</sup> in some source regions/over hotspots, especially in England, North  
235 Eastern France, the North European Plain (Netherlands, Belgium), and Spain (around Barcelona). Similar results  
236 were found in the study of Whitburn et al. (2016), in which they show that GEOS-Chem underestimates ammonia  
237 columns by up to  $1 \times 10^{16}$  molecules/cm<sup>2</sup> in Europe on a yearly average in 2009, notably in the North European  
238 Plain. It is important to note that, in our study, we compare only one month of data (March, 2011) that marks the  
239 start of the growing season in the majority of the countries of interest (FAO, 2022; USDA, 2022). The differences  
240 are mainly because of the time coincidence, and the fact that only cloud-free data are used to retrieve ammonia;  
241 IASI observes ammonia during the satellite overpass (~9:30 AM local time), whereas the GEOS-Chem simulation  
242 is averaged over the whole month including all hours of the day. In Western and Northern Europe, the MRD is  
243 mostly less than  $-50\%$ , for instance, in the North European Plain ( $-49\%$ ). If we look at the average MRD in  
244 regions of focus, we see that the Po Valley in Italy has the highest MRD value ( $+110\%$ ), whereas the best  
245 represented region is New Aquitaine in the southwest of France ( $-20\%$ ). The rest of the regions have mean MRDs  
246 that fluctuate between  $-64\%$  and  $-42\%$ . A summary of the results of this study, including the MRD over some  
247 source regions is listed in Table 1. Although the bias and MRD can be considered high, the spatial distribution is  
248 consistent between IASI and GEOS-Chem. Therefore, according to the steady state approximation, the  
249 meteorological and soil parameters affecting one dataset (e.g. IASI NH<sub>3</sub>) are applicable to the other (e.g. model  
250 simulation). It is worth noting that although we do not use the latest version of GEOS-Chem, the results we obtain  
251 reflects our current understanding of the regional chemistry at this horizontal and temporal resolution.

### 252 **3.2. Ammonia emissions, losses and lifetime in Europe**

253 In order to understand the NH<sub>3</sub> spatial variability in Europe during the application of fertilizers, a detailed analysis  
254 of the output of the GEOS-Chem simulation for the month of March 2011 is shown in Figure 2.

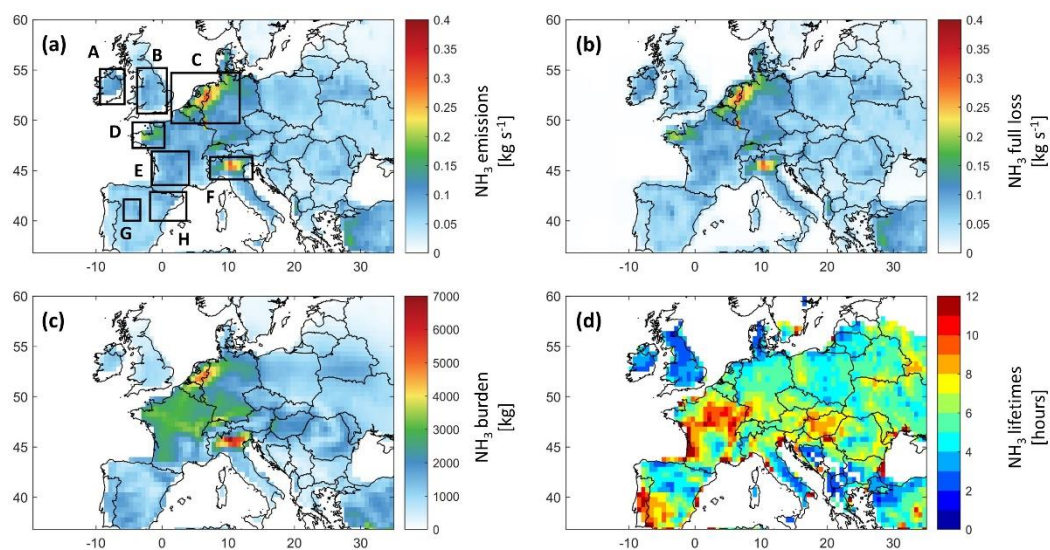
255 The anthropogenic sources (i.e. mainly agriculture) contribute 83 % of the total ammonia emissions during March  
256 2011 in Europe. The ammonia emissions from natural sources (i.e. soil of natural vegetation, oceans, and wild  
257 animals) follow representing 16 % of the total emissions, whereas the remaining 1 % correspond to the ammonia  
258 emissions from biomass burning and ships (not shown here).

259 Figure 2a shows ammonia monthly emissions. Most of them are due to agricultural activities (not shown here); we  
260 identify 8 source regions which we investigate thoroughly in this study shown as rectangles A to H. The highest  
261 agricultural sources over Europe include the North European Plain, Brittany, and the Po Valley (regions C, D, and  
262 F).

263 In the calculation of the total loss of ammonia (Figure 2b), we considered dry deposition, chemistry, transport, and  
264 wet deposition (in which we included ammonia loss to convection) from the GEOS-Chem model simulation, which  
265 are all possible loss processes for ammonia (David et al., 2009). Figure 2b shows that the largest losses occur  
266 logically where we have the highest sources detected (see Figure 2a).

267  
268





**Figure 2. Ammonia budget in Europe from GEOS-Chem: (a) Ammonia emissions from the Harmonized Emissions Component module (HEMCO) in  $\text{kg s}^{-1}$  with our regions of interest shown in rectangles, (b) ammonia full loss in  $\text{kg s}^{-1}$ , (c) ammonia total burden in kg, and (d) ammonia lifetime in hours. All plots refer to March 2011 and are presented at a  $0.5^\circ \times 0.625^\circ$  grid resolution.**

269 The total ammonia burden (Figure 2c) is calculated as the integrated sum of all ammonia columns in the model  
270 grid box. We can clearly detect ammonia hotspots over Europe, in particular the North European Plain, Brittany  
271 and the Po Valley, all regions characterized by intense agricultural activities, as the total emissions and deposition  
272 show (Figure 1 and Figure 2). We also see that the burden is generally the highest over France, Belgium, The  
273 Netherlands, and parts of Germany and Italy.

274  
275 We finally get the lifetime  $\tau_{ss}$  of ammonia (Figure 2d). In the case of a gas with a short lifetime, such as ammonia,  
276 the emissions are relatively well-balanced spatially by eventual sinks/losses (steady-state approximation).  
277 Therefore, we can calculate a steady-state lifetime as the ratio between the total burden  $B$  (Figure 2c) and the total  
278 emissions  $E$  or losses  $L$  (sum of all emitted / lost molecules, Figure 2a or b) using the following equation:  $\tau_{ss} =$   
279  $B/L$  (Plumb and Stolarski, 2013).

280  
281 We note that the  $\tau_{ss}$  is more or less the same whether we calculate it using the losses or the emissions. For instance,  
282 in selected source regions (rectangles in Figure 2a) the total emissions and losses are very close with very low  
283 biases that are less than 2% (not shown here). Our results show that  $\tau_{ss}$ , on a monthly average, can go up to 12  
284 hours, and it can reach 1 day (24 hours) in coastal regions such as region E in New Aquitaine in France. The latter  
285 can be related to the high probability of air stagnation is in that area in comparison to Northern Europe (Garrido-  
286 Perez et al., 2018), since higher  $\text{PM}_{2.5}$  pollution episodes were found under stagnant meteorological conditions  
287 (AQEG, 2012); and ammonium molecules carried on these  $\text{PM}_{2.5}$  can transform back into ammonia. Our results  
288 agree with the literature suggesting a residence time between a few hours to a few days (Behera et al., 2013; Pinder  
289 et al., 2008), and with those calculated by Evangeliou et al. (2021) over Europe, showing a monthly average of  
290 ammonia lifetime that ranges from 10 to 13 hours in Europe. The figure adapted from Evangeliou et al. (2021) is

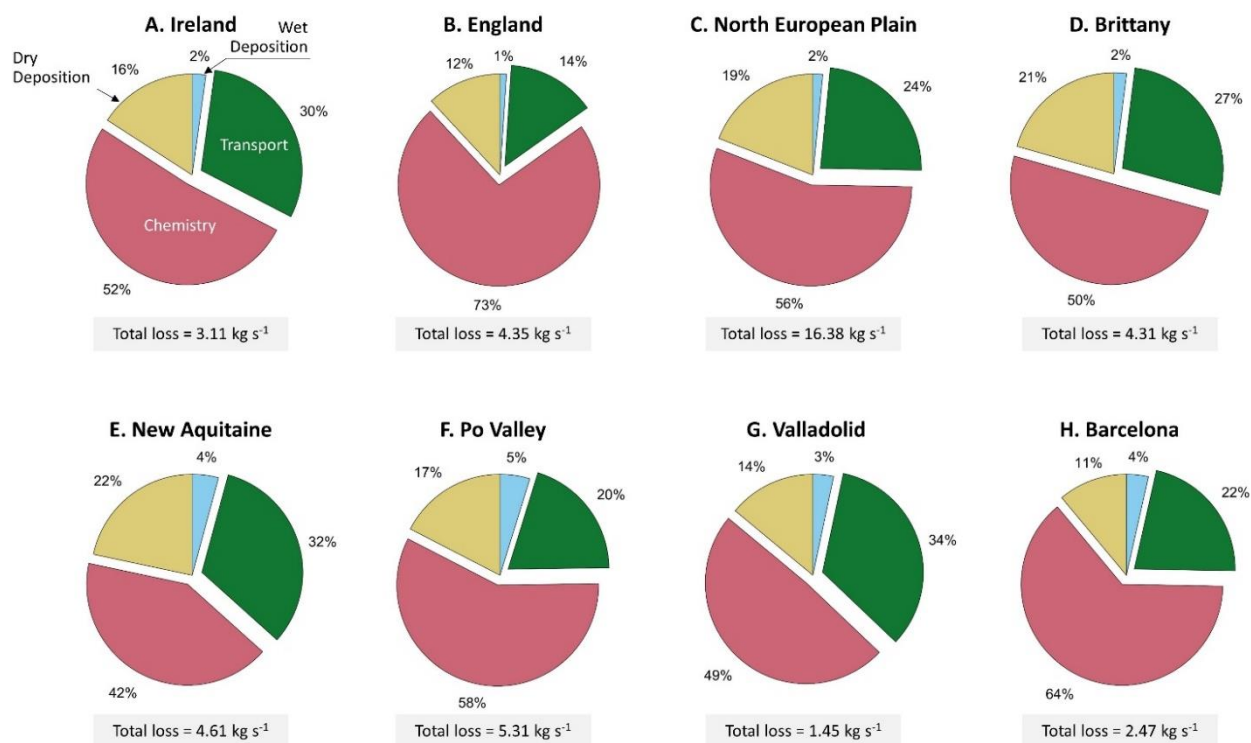


291 shown in supplementary material (Figure S1). Shorter lifetimes from industrial sources of ammonia were reported  
292 in Damers et al. (2019), with a mean lifetime of ammonia that is equal to 2.35 hours ( $\pm 1.16$ ). A recent study  
293 found lifetimes of ammonia that vary between 5 and 25 hours, roughly, in Europe (Luo et al., 2022); these values  
294 are higher since, in addition to ammonia loss, Luo et al. (2022) included the loss of ammonium, and thus  
295 considering the loss of ammonia only terminal when the ammonium is also lost/deposited. This approach is not  
296 considered here nor in Evangelidou et al. (2021).

297  
298 Notably, ammonia lifetime and burden (Figure 2c, and d) each have different spatial distribution compared to the  
299 other 2 panels (Figure 2a, and b). The ammonia residence time in the atmosphere varies depending on the sources  
300 and more importantly on the locally dominant loss mechanisms. For this reason, in Figure 3, we show the relative  
301 contribution of the ammonia loss mechanisms, presented as pie charts, for the agricultural source regions shown  
302 in black boxes in Figure 2a.

303  
304 The fastest loss mechanisms are either chemical (i.e. in the vast majority transformation to particulate matter) or  
305 through wet and dry deposition (Tournadre et al., 2020). Figure 3 shows that more than 50 % of the ammonia  
306 molecules in the atmosphere are lost to chemical reactions in most of the regions (A, B, C, H, and F). The shortest  
307 residence time of ammonia is observed in England, where the chemical removal was significantly higher than  
308 other sinks and represented up to 73 % of the total ammonia loss pathways, suggesting a rapid transformation into  
309 inorganic particulate matter ( $PM_{2.5}$ ). In the regions D, G and E the chemical loss makes up 50 %, 49 %, and 42 %,  
310 respectively. In fact, in March 2011, PM was found to be mostly composed of inorganic nitrate (41 %), and  
311 ammonium (20 %) (Viatte et al., 2022) over Europe, both of which are products of atmospheric ammonia. Nitrate-  
312 bearing  $PM_{2.5}$  are formed when nitric acid ( $HNO_3$ ) reacts with ammonia (Yang et al., 2022), and ammonium is a  
313 direct product of the hydrolysis of ammonia. 41% of the nitric acid formed in the atmosphere is produced from the  
314 reaction between nitrogen dioxide ( $NO_2$ ) and the hydroxyl radical (OH) (Alexander et al., 2020). These chemical  
315 pathways help explain the large chemical losses in most of the regions studied in Figure 3.

316  
317 Ammonia loss to transport is the highest in regions neighboring the Atlantic Ocean, accounting for 30 %, 27 %,  
318 32 %, and 34 % of total sinks in regions A, D, E, and G respectively. These regions are exposed to the North  
319 Atlantic Drift, also known as the Gulf Stream, that is associated with high wind speed and cyclonic activity (Barnes  
320 et al., 2022). In other regions, 14 % to 22 % of the total ammonia is lost to transport mechanisms, and in all regions,  
321 11 to 22 % is lost to dry deposition (Figure 3). During March, precipitation is relatively lower as compared to  
322 winter (December, January, February) in Europe. Furthermore, 2011 was a particular dry year compared to the  
323 1981 – 2010 average (Met Office, 2016). Drought was reported to be severe in areas such as France, Belgium and  
324 the Netherlands, and moderate in England and Ireland (EDO, 2011). This can help explain the low percentage of  
325 wet deposition during March 2011 (1 to 5 % out of the total loss of ammonia).



**Figure 3.** Repartition of the ammonia loss mechanisms for major agricultural areas in Europe, during March 2011, as retrieved from GEOS-Chem, with the total ammonia loss shown in a grey box under each pie chart (kg s<sup>-1</sup>). The regions are shown in black boxes in Figure 2a.

326

#### 327 4. Ammonia emission potential over Europe

328

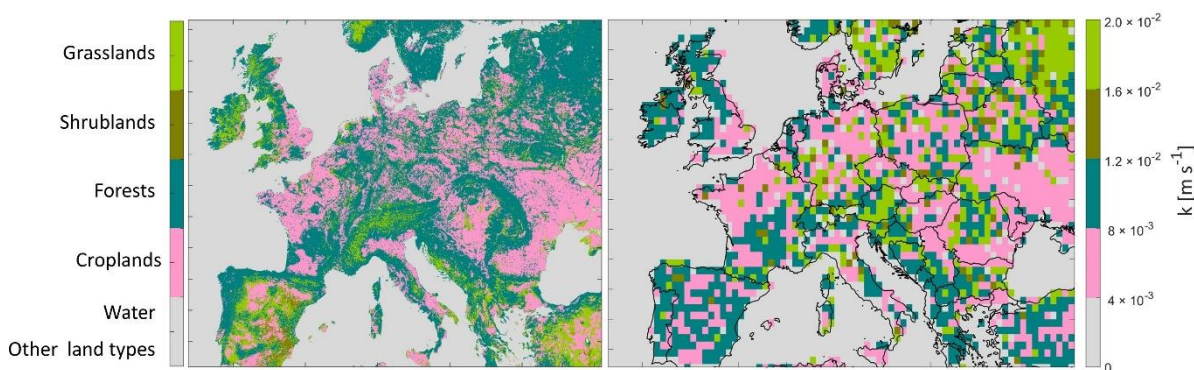
329 To calculate emission potential, a calculation of the mass transfer coefficient  $k$ , which relates to the land type, is  
 330 necessary. Figure 4 shows the land cover type from MODIS in Europe (left panel), and the corresponding assigned  
 331 mass transfer coefficient  $k$  (right panel) needed to calculate the emission potential (Eq. (2-1)). In order to choose  
 332 a mass transfer coefficient that is convenient for the different land types relevant to this study, we searched for  $k$   
 333 values in the literature. Not all land types have been studied for ammonia transfer coefficient.

334 For water bodies and other land types that are not considered here (see Sect. 2.2), the mass transfer values  $k$  were  
 335 set to zero and represented in grey colour in Figure 4. In a laboratory experiment, Svensson et al. (1993) reported  
 336  $k = 4.3 \times 10^{-3} \text{ m s}^{-1}$  for a mixture of soil and swine manure, as therefore, we assign this value to croplands. Due  
 337 to the lack of  $k$  values for non-fertilized forests, shrublands and grasslands in the literature, we used values  
 338 originally assigned for SO<sub>2</sub>, bearing in mind that these are approximate values and they reflect mostly the  
 339 conditions of the soil cover type (short, medium or tall grass). To assign a  $k$  value for forests, we used values  
 340 reported in Aneja (1986) ( $k = 2 \times 10^{-2} \text{ m s}^{-1}$ ), which originally represent deposition velocity (mass transfer) of



341 SO<sub>2</sub> in a forest (high crops). For shrublands and grasslands (the two land types have the same  $k$ ), we used the value  
342  $k = 8 \times 10^{-3} \text{ m s}^{-1}$  that has been reported in Aneja et al. (1986) as the deposition velocity (mass transfer) of SO<sub>2</sub>  
343 in a grassland (medium crops). These values are the best attempt to test the validity of using MODIS and lookup  
344 tables of  $k$  values to calculate a realistic soil emission potential. As a result, Figure 4 (left panel) includes 5 land  
345 types, while  $k$  values are reported for 4 land types (other land type/water, croplands, forests, and  
346 shrublands/grasslands).

347  
348 After choosing the  $k$  values, we assigned them for each land type on the (500 m × 500 m) grid. We then extrapolate  
349 the array with the  $k$  values from 500 m × 500 m to the resolution of GEOS-Chem (0.5° × 0.625° grid box). This  
350 leads to averaging different fine pixels with different land cover types into a coarser grid. The result is shown on  
351 the right panel of Figure 4.  
352



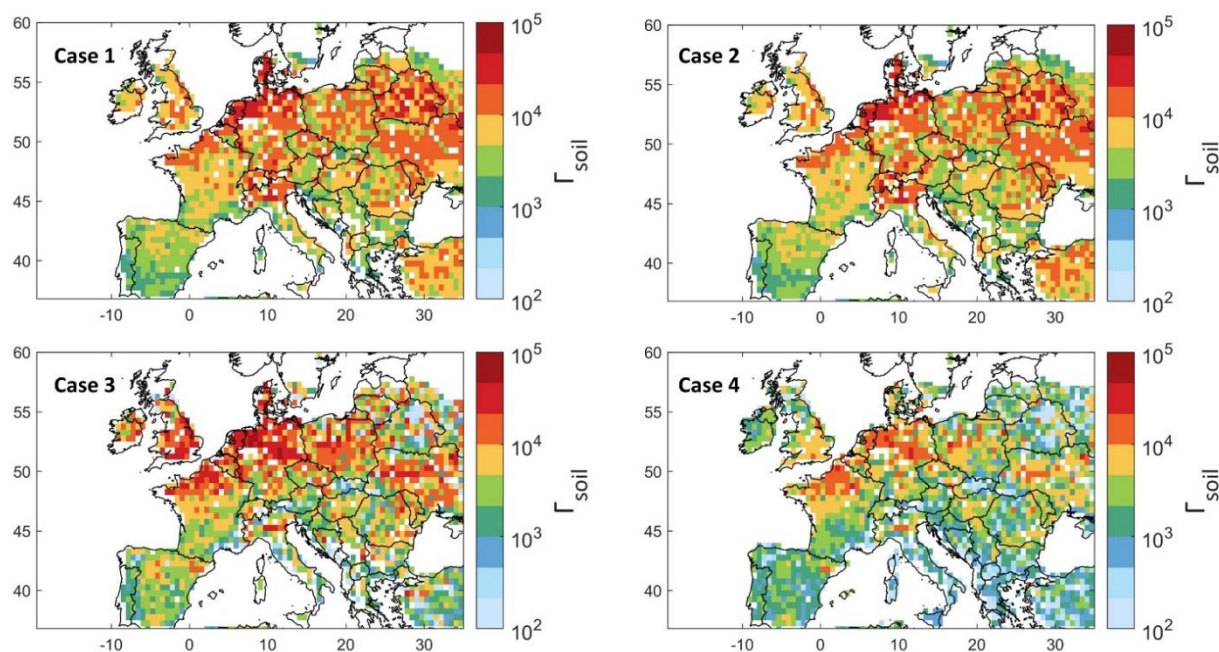
353 **Figure 4. MODIS Land Cover Type, at a 500 m × 500 m grid box (left panel), and interpolated mass transfer coefficient  $k$  on a horizontal resolution of 0.5° × 0.625° grid box (right panel).**

354 Uncertainties of this methodological approach can be summarized as follows:

- 355 (1) The  $k$  value assigned for croplands is approximate and therefore not the same in every cropland over  
356 Europe.
- 357 (2) The  $k$  value assigned for forests represents the SO<sub>2</sub> exchange in high croplands, and ammonia might  
358 change especially when the latter is highly affected by humidity; it can easily dissolve in the water film  
359 on leaves in high humid conditions.
- 360 (3) The extrapolation of a fine array (500 m × 500 m) will merge several grids together and average them in  
361 order to construct the coarser grid box (0.5° × 0.625°); the result is therefore an average that might mix  
362 croplands with neighboring forests/barelands/grasslands. This leads to a range of different  $k$  values that  
363 are shown on Figure 4.  
364

365 Using the land-type specific  $k$  value is necessary in order to reflect realistic emissions potential, as ammonia  
366 exchange in a forest is different from that of croplands or unfertilized grasslands, due to different barriers (long,  
367 medium or short crop / grass), and ammonium soil content in each land type.





**Figure 5. Ammonia soil emission potential ( $\Gamma_{soil}$ ) on a log10 scale from model simulation, observation and reanalysis for 4 different cases (see text for details).**

368  
369 We show in supplementary material Figure S2, the emission potential (similarly to what we show in Figure 5) but  
370 from a fixed and averaged  $k$  value for all land types. Figure S2 shows the importance of using a variable  $k$  that is  
371 adjusted to each land type is depicted in supplementary materials (Figure S2). In Figure S2, the fixed  $k$  used is  
372 calculated assuming 14 days of fertilization ( $k = 10^{-3} \text{ m s}^{-1}$ ), 7 days when  $k$  value reduces ( $k = 10^{-5} \text{ m s}^{-1}$ ), and  
373 10 days when  $k$  is low ( $k = 10^{-6} \text{ m s}^{-1}$ ) resulting in average of  $k = 4.5 \times 10^{-4} \text{ m s}^{-1}$ . The difference in the  
374 emission potential between fixed and spatially variable  $k$  is shown in supplementary material Figure S3, where we  
375 see that a fixed  $k$  might overestimate  $\Gamma_{soil}$  by 10 to  $10^3$  on a log10 scale (500 – 3000 %), in agricultural areas.

376  
377 Figure 5 illustrates the ammonia soil emission potential  $\Gamma_{soil}$  calculated using Eq. (2-1) and  $k$  values presented in  
378 Figure 4. After assigning the variable mass transfer coefficient, the remaining variables needed to calculate  $\Gamma_{soil}$   
379 in Eq. (2-1) are ammonia concentration and lifetime, as well as the skin temperature. For this reason, the emission  
380 potential  $\Gamma_{soil}$  shown in Figure 5 is calculated using different configurations.

- 381  
382 - Case 1: GEOS-Chem ammonia and lifetime with MERRA-2 T skin, i.e. simulated  $\Gamma_{soil}$ ,  
383 - Case 2: GEOS-Chem ammonia and lifetime and ERA5 Tskin, to check the effect of using ERA5 vs  
384 MERRA-2 for skin temperature,  
385 - Case 3: IASI ammonia, ERA5 T skin and GEOS-Chem ammonia lifetime,  
386 - Case 4: IASI ammonia, ERA5 T skin and ammonia lifetime from Evangeliou et al. (2021), that were  
387 calculated using LMDz-OR-INCA chemistry transport model. The latter couples three models: The  
388 general circulation model GCM (LMDz) (Hourdin et al., 2006), the INteraction with Chemistry and



389 Aerosols (INCA) (Folberth et al., 2006), and the land surface dynamical vegetation model (ORCHIDEE)  
390 (Krinner et al., 2005).

391  
392 Based upon the four cases, we calculate a range of emission potentials. When calculating  $\Gamma_{soil}$ , we filtered data  
393 points with ammonia total column concentration less than  $5 \times 10^{14}$  molecules  $\text{cm}^{-2}$ . The latter are mostly grid boxes  
394 concentrated above  $56^\circ$  North that we consider as noise (shown in white pixels on Figure 5).  
395

396 T skin from ERA5 and MERRA-2 agree very well, with a coefficient of determination  $r^2 = 0.97$  (Figure S4 in the  
397 supplementary material). This explains the excellent spatial correlation between cases 1 and 2. Since IASI-NH<sub>3</sub>  
398 retrievals use ERA5 T skin, this also suggests that using MERRA-2 or ERA5 does not affect our  $\Gamma_{soil}$  calculation.  
399 In case 3, the emission potential agrees spatially and in value with that of GEOS-Chem. However, we observe  
400 higher  $\Gamma_{soil}$  in regions such as Ireland, England, North France, Northeastern Spain, and Poland. This is due to the  
401 underestimation/overestimation of ammonia from GEOS-Chem as compared to IASI observations (Figure 1a). For  
402 instance,  $\Gamma_{soil}$  from IASI and ERA5 (case 3) differs with that from GEOS-chem and ERA5 (case 2) by up to -70  
403 % in the Po Valley (Italy) and +60 % in England. Looking at Table 1, this difference can be explained by the  
404 corresponding MRD for each of the regions, in which it is -64 % for England and +110 % for the Po Valley.  
405 Similarly, the differences between case 3 and 4 reach up to +66 % in England, and this is mostly due to the 10-  
406 hours difference between ammonia lifetime from GEOS-Chem and Evangeliou et al. (2021) (Figure S1 in the  
407 supplementary material). The lowest  $\Gamma_{soil}$  were obtained in case 4, due to the higher lifetimes than those calculated  
408 from GEOS-Chem (Figure S1); note that  $\Gamma_{soil}$  is inversely proportional to ammonia lifetime (Eq. (2-1). In fact, the  
409 longer ammonia stays in the atmosphere (longer lifetime), the less the flux will be directed from the soil to the  
410 atmosphere (less ammonia emission).

411  
412 In the four cases presented in Figure 5, we see similar spatial distribution of ammonia emission potential ranging  
413 from  $12 \times 10^{-1}$  in a forest to  $9.5 \times 10^4$  in a cropland (monthly average considering all the cases). In agricultural  
414 lands, our results show that  $\Gamma_{soil}$  ranges from  $2 \times 10^3$  to  $9.5 \times 10^4$ . Our values for croplands start at around  $10^3$ . In  
415 fact, most of the studies summarized in Zhang et al. (2010) reported  $\Gamma_{soil}$  that range mostly from  $10^3$  to  $10^4$  in  
416 fertilized croplands/grasslands; the minimum  $\Gamma_{soil}$  reported is in the order of  $10^2$  and the maximum is of the order  
417 of  $10^5$ . Therefore, our values fit within the range of  $\Gamma_{soil}$  calculated in the literature and summarized in Zhang et  
418 al. (2010) and the references within. Personne et al. (2015) focused on Grignon, an agricultural region near Paris,  
419 France ( $48^\circ 51'N$ ,  $1^\circ 58'E$ ). They obtained  $\Gamma_{soil}$  values between  $1.1 \times 10^4$  to  $5.8 \times 10^6$ . In the present study, the  
420 emission potential over this region is between  $5 \times 10^3$  (case 4) to  $2 \times 10^4$  (case 2). In this study, it is expected to  
421 obtain lower values than the ones measured over specific field. Therefore, we consider our results to be in good  
422 agreement with the obtained values in Personne et al. (2015), since ours reflect an average of a coarse patch of  
423 land of the size  $55 \times 70 \text{ km}^2$  approximately, with a 31-day mean.

424  
425 The mean emission potentials per ammonia source region in Europe (shown in rectangles in Figure 2 and Figure  
426 3) and per case are shown in Figure 6, and listed in Table 1. Table 1 shows the average lifetime from GEOS-Chem  
427 (hours), the average T skin from the three datasets that we used ( $^\circ\text{C}$ ), the average ammonia emission potential in  
428 all the cases examined (dimensionless), and the average ammonia columns from IASI and GEOS-Chem  
429 (molecules  $\text{cm}^{-2}$ ). The four cases show a similar pattern with the North European Plain exhibiting the highest  
430 emission potential. This has been shown in Figure 1, Figure 2, and Figure 5, as well as in Table 1, where  $\Gamma_{soil}$  is  
431 higher in regions with high ammonia columns. This is expected in fertilized lands (croplands), since  $\Gamma_{soil}$  is  
432 proportional to the concentration of ammonia near the surface. The latter increases when the soil content in  
433 ammonium ( $\text{NH}_4^+$ ) increases following the application of nitrogen-based fertilizers.  
434





435 Figure 6 also shows that for cases 1 and 2 (GEOS-Chem) the emission potential in the Po Valley is higher as  
 436 compared to case 3 (IASI), although it stays within the margin of error. This is due to the effect of temperature.  
 437 Table 1 shows that at the time of the IASI overpass,  $T_{skin}$  from ERA5 in the Po Valley is almost twice as large  
 438 (8.95 °C) as the monthly averaged temperature (4.46 °C). The effect of skin temperature through Eq. (2-1) makes  
 439 the emission potential highly dependent. In fact,  $\Gamma_{soil}$  is both directly and inversely proportional to  $T_{skin}$ , however,  
 440 the exponential in the denominator has ~10 times more effect on the value of  $\Gamma_{soil}$  than the  $T_{skin}$  in the numerator.  
 441 Therefore, through Eq. (2-1), we conclude that an increase in temperature by 1°C will reduce  $\Gamma_{soil}$  by around -8%.

442  
 443 The standard deviation (shaded area) is found to be the highest in the North European Plain, which is also the  
 444 largest region (hence higher variability is expected), especially when considering case 3 with IASI. IASI  
 445 distinguishes different source sub-regions, leading to higher spatial variability of ammonia, and therefore  $\Gamma_{soil}$ . As  
 446 Figure 5 has shown, case 4 has the lowest  $\Gamma_{soil}$ , with a factor of two lower than cases 1 to 3. This is due to the  
 447 longer lifetimes calculated by Evangeliou et al. (2021). However, we note that all the regions exhibit the same  
 448 inter-variability between each of the case, regardless of the lifetimes used.  
 449

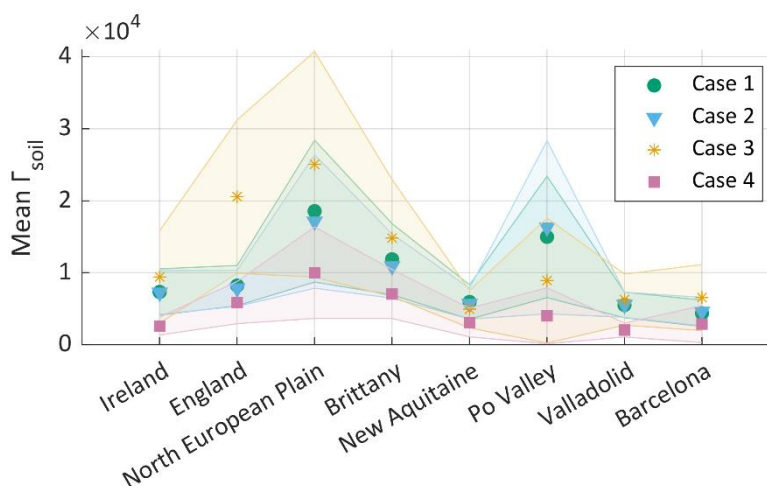


Figure 6. Mean ammonia emission potential  $\Gamma_{soil}$  per region and per case, with the error margin on the mean as the shaded area (95<sup>th</sup> percentile) for cases 1 to 4. The cases are explained in Figure 5 and its discussion.

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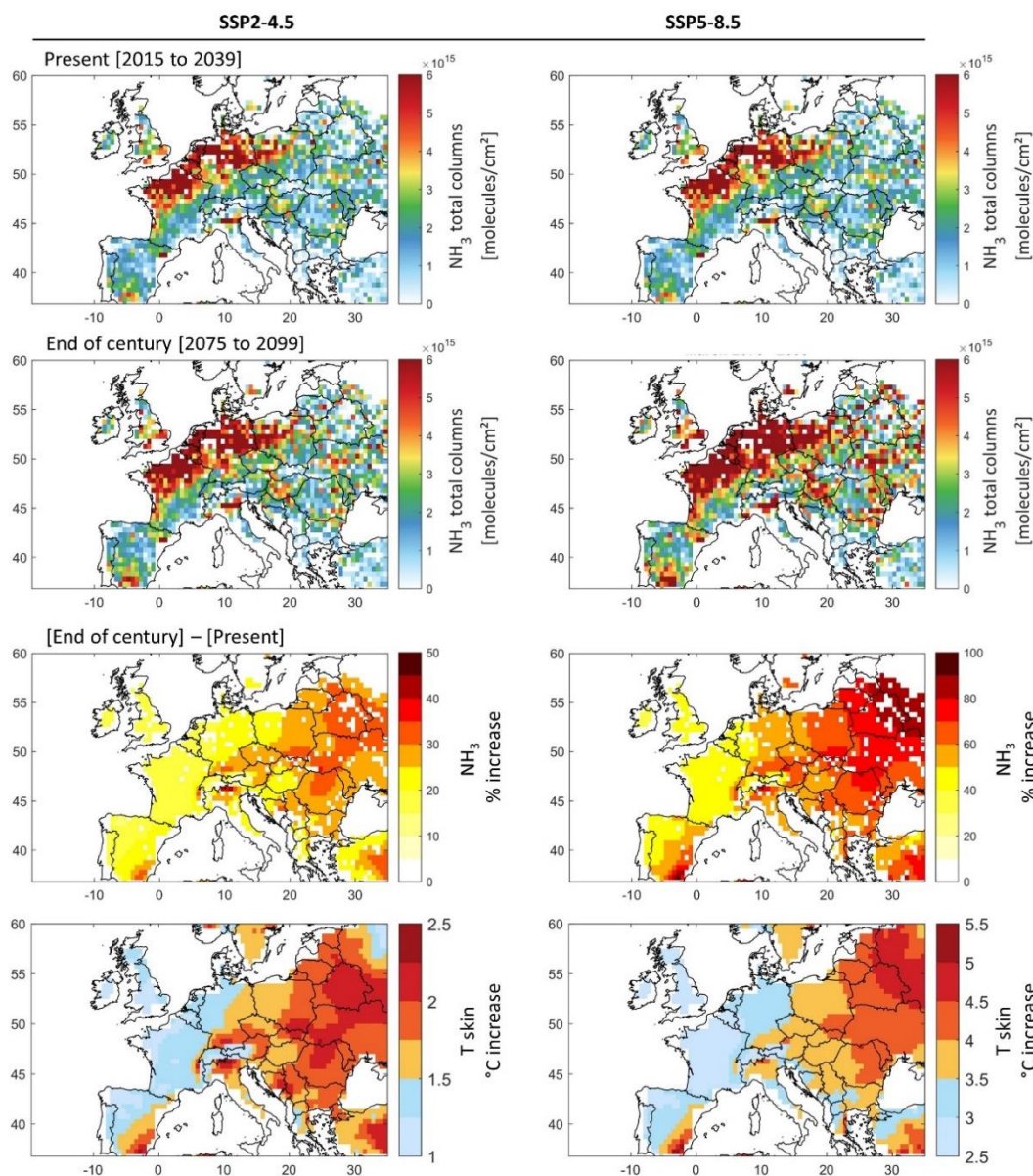
461 **Table 1.** Summary of NH<sub>3</sub> average lifetime, emission potential, concentrations and the T skin in selected regions  
 462 in Europe.

463

Region	Country	$\tau_{NH_3}$ [hours]	T skin [°C]			$\Gamma_{soil} \times 10^4$ [dimensionless]				NH <sub>3</sub> concentrations [molecules $\times 10^{15}$ cm <sup>-2</sup> ]		
			ERA5 IASI Overpass	ERA5	MERRA- 2	Case 1	Case 2	Case 3	Case 4	IASI	GEOS- Chem	Mean MRD [%]
Ireland	Ireland	3.34	8.74	5.78	6.23	0.73	0.72	0.94	0.26	2.5	1.5	- 46
England	England	3.15	8.54	5.87	5.73	0.82	0.78	2.06	0.58	4.8	1.2	- 64
North European Plains	Belgium, Netherlands	5.16	7.46	4.93	4.57	1.86	1.71	2.51	1.00	7.7	3.9	- 49
Brittany	France	6.93	10.48	8.13	8.16	1.19	1.09	1.48	0.70	5.8	3.7	- 60
New Aquitaine	France	8.05	11.25	7.72	7.47	0.59	0.57	0.49	0.30	4.0	2.9	- 20
Po Valley	Italy	7.10	8.95	4.46	5.46	1.50	1.63	0.89	0.40	3.8	4.0	+ 110
Valladolid	Spain	4.53	11.64	6.87	6.93	0.55	0.55	0.62	0.20	2.5	1.3	- 42
Barcelona	Spain	4.94	12.61	7.05	9.44	0.43	0.46	0.65	0.28	3.2	1.5	- 49



464 5. Ammonia under future scenarios



**Figure 7.** First and second rows: Ammonia total column concentrations during March (monthly averages) under the present climate [2015 to 2039] (first row), and in the end of century climate [2075 to 2099] (second row), under the socio-economic scenarios SSP2-4.5 (left) and SSP5-8.5 (right). Third and fourth rows: The percentage increase in ammonia concentration (third row), and the change in T skin in °C (fourth row) by the end of the century [2075 to 2099] with respect to present climate [2015 to 2039] under SSP2-4.5 (left) and SSP5-8.5 (right). Ammonia columns were calculated using ammonia emission potential  $\Gamma_{soil}$  derived from IASI and ERA5 for March 2011 (case 3), and EC-Earth T skin simulations for SSP2-4.5 and SSP5-8.5 extending from 2015 till 2099.



465 As seen in Eq. (2-1), higher skin temperatures favour volatilization of ammonia from the soil. In an attempt to  
466 understand how our simplified emission potential model behaves under changing climate, as well as under future  
467 scenarios, we adopt the future T skin simulations from EC-Earth climate model, into Eq. (2-1). The two climate  
468 socio-economic scenarios that we consider are SSP2-4.5 (“middle of the road” scenario where trends broadly  
469 follow their historical patterns), and SSP5-8.5 (a world of rapid and unconstrained growth in economic output and  
470 energy use) (Riahi et al., 2017). The same Figure constructed using  $\Gamma_{soil}$  from GEOS-Chem (case 1) is shown in  
471 the supplementary material as Figure S5.

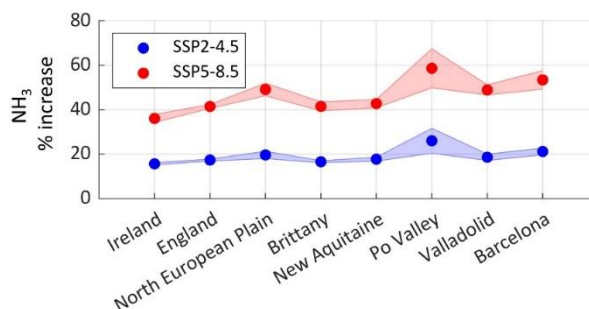
472 We calculate current and future ammonia columns assuming that the emission potential  $\Gamma_{soil}$  remains unchanged.  
473 In other words, we assume that the same amount of fertilizers and manure is used until 2100 in the agricultural  
474 fields and farms (unchanged ammonium soil content).

475 Figure 7 shows ammonia columns during the 25-year [2015 – 2039] representing the present climate (upper  
476 panels), and the end of the century [2075 – 2099] (middle panels). The ammonia columns in the 25-year average  
477 climate of the end of century with respect to present day climate (lower panels) are also shown.

478  
479 Spatially, the present climate ammonia columns calculated from the T skin of the climate model and our emission  
480 potential from IASI (case 3 in Figure 5), agree very well with those shown in Figure 1. We do not aim at validating  
481 or directly comparing the two, as we are only interested in the climate response on ammonia concentration, i.e. by  
482 the difference due to skin temperature increase (lower panels).

483  
484 From Figure 7 (lower panels) it can be seen that the increase in ammonia columns by the end of the century is  
485 more severe on the east side of Europe. Under the most likely scenario (SSP2-4.5), ammonia columns vary between  
486 +15 % in France, to around +20 % in the North European Plain (Figure 7). The largest increase is detected in  
487 Eastern Europe, where ammonia columns show an increase of up to a +50 % (Figure 7, lower left panels), creating  
488 new potential hotspots/sources of ammonia in Belarus, Ukraine, Hungary, Moldova, parts of Romania and  
489 Switzerland. Under the SSP5-8.5 scenario, the results show an increase in ammonia columns of up to +100 % in  
490 Eastern Europe (Figure 7, right lower panel). This is directly related to the higher projected increase in skin  
491 temperature over these regions. Other studies have equally reported Eastern Europe to be more affected by climate  
492 change under future scenarios, as compared to western Europe (European Environment Agency, 2022; Jacob et al.,  
493 2018). Spatially, the increase in ammonia coincides with the increase in T skin.

494 Figure 8 depicts the change in ammonia columns under the SSP2-4.5 and SSP5-8.5 scenarios, for our source  
495 regions (shown as rectangles in Figure 2). Ammonia columns increase is foreseen to be the highest in the Po Valley  
496 (Italy) with +26 % and +59 % under SSP2-4.5 and SSP5-8.5 respectively. It is then followed by the agricultural



**Figure 8.** The percentage increase in ammonia concentration by the end of the century [2075 to 2099] with respect to the present climate [2015 to 2039] under the two climate scenarios SSP2-4.5 (blue) and SSP5-8.5 (red), in the source regions investigated in this study. The shades around each line represent the standard deviation from the mean.



497 areas around Barcelona (Spain), and the North European Plain (Belgium, Netherlands) with an increase of +21 %  
498 (+49 %) and +20 % (+53 %) respectively, under the SSP2-4.5 (SSP5-8.5) scenario. Under the SSP5-8.5, the  
499 increase in ammonia columns in percentage is more than twice the change under SSP2-4.5 (+127 % in the case of  
500 the Po Valley for instance). The Po Valley is adjacent to the Alps mountains, and due to global warming, this  
501 region is expected to experience increased evapotranspiration (Donnelly et al., 2017), which is a major factor that  
502 leads to the volatilization of ammonia.

503 The local and regional effect of volatilization of ammonia under different climate scenarios remains difficult to be  
504 properly assessed. Even under the “middle of the road” scenario 2-4.5, and without climate extremes (e.g.  
505 heatwaves), Europe might be facing big challenges in air or downwind agricultural regions, since chemistry and  
506 atmospheric transport (Figure 3) drive the loss of ammonia during the growing season in this part of the world.

507  
508 An increase in ammonia concentration poses a significant and yet poorly understood effect on local and regional  
509 air quality through the increase in PM<sub>2.5</sub> concentration. We note, however, that ammonia columns in the soil are  
510 governed by a threshold. Higher temperatures will increase the rate of volatilization of ammonia from the soil, but  
511 only up to a certain point where no dissolved ammonium is left. Plants, however, can also be a source of ammonia  
512 when exposed to stressful conditions. For example, under heat stress and in instances where there are no ammonia  
513 in the air, increase in air temperature results in exponential increase in ammonia emission from plants’ leaves  
514 (Husted and Schjoerring, 1996).

## 515 6. Discussion and conclusions

516  
517 Agriculture worldwide has fed the human race for thousands of years, and will continue to do so, as mankind  
518 highly relies on it. Emissions from agricultural activities will inevitably increase, in order to meet the expected  
519 yield. In this study, we use a variety of state-of-the-art datasets (satellite, reanalysis and model simulation) to  
520 calculate the first regional map of ammonia emission potential during the start of the growing season in Europe.  
521 The emission potential can be used as a proxy to calculate ammonia columns in the atmosphere, and as such to  
522 assess its deposition, atmospheric transport, and contribution to PM formation. First, we show that the GEOS-  
523 Chem chemistry transport model is able to reproduce key spatio-temporal patterns of ammonia levels over Europe.  
524 The ammonia budget is governed by the emissions over source regions (North European Plain, Brittany and the  
525 Po valley), as well as by key loss processes. We find that chemical loss pathway is responsible of 50 % or more of  
526 the total ammonia loss over Europe. From the GEOS-Chem simulation, we calculate the average ammonia lifetime  
527 in the atmosphere which ranges between 4 and 12 hours in agricultural source regions of Europe. From this, and  
528 using the mass transfer coefficient for different land cover types, we calculate a range of emission potentials  
529  $\Gamma_{soil}$  from IASI and GEOS-Chem. We find that  $\Gamma_{soil}$  ranges between from  $2 \times 10^3$  to  $9.5 \times 10^4$  in fertilized lands  
530 (croplands). Choosing a variable  $k$  from the literature, and based on different land cover types from MODIS, we  
531 calculate  $\Gamma_{soil}$  values that are consistent with those found in the literature. The increase in T skin is expected to  
532 have an effect on the emission of ammonia from the soil. Using T skin from the EC-Earth climate model, we  
533 estimate ammonia columns by the end of the century [2075 – 2099], and compare it to columns of the present  
534 climate [2015 – 2039]. Our results show that ammonia columns will double under the SSP5-8.5 scenario, and will  
535 increase by up to 50 % under the most likely SSP2-4.5 scenario. The eastern part of Europe is the most affected  
536 by the change in temperatures, and it is where we find the highest ammonia columns increase. Among the regions  
537 of focus, Italy, Spain, Belgium and the Netherlands are the most affected, as compared to France, England and  
538 Ireland. The highest increase in ammonia columns is observed in the Po Valley in Italy (+59 % under the SSP5-  
539 8.5).

540



541 We calculate ammonia concentration under future climate and during the start of the growing season (March) in  
542 Europe. However, in order to grasp the yearly budget of ammonia, it is crucial to apply this method to all seasons  
543 of the year; especially in regions with extensive agricultural activities, such as the United States, India, and China.  
544 In addition to this, more field measurements of ammonia emission potential ( $I_{soil}$ ) in different land use / cover  
545 types are required, this can help us perform better comparison with emission potentials calculated from model and  
546 satellite data. Finally, having ammonia columns at different times of the day, from field observations or satellite  
547 measurements will allow quantification of daily emission potentials, that will in turn help us understand its diurnal  
548 variability. This will be ensured with the launch of the Infrared Sounder (IRS) on the Meteosat Third Generation  
549 (MTG) geostationary satellites scheduled in 2025.





550  
551

## A. Appendix A

### 552 1. Ammonia-Ammonium equilibrium

553 Ammonia ( $NH_3$ ) is a water-soluble gas, it undergoes protonation with  $H^+$  from the hydronium ion  $H_3O^+$  in an  
554 aqueous solution in order to give ammonium ( $NH_4^+$  cation), the dissociation equation is expressed as follows:



556 Or



558 With  $K_{NH_4^+}$  as the ammonium-ammonia dissociation equilibrium constant that can be expressed as:

$$559 K_{NH_4^+} = \frac{[NH_3(aq)][H^+]}{[NH_4^+(aq)]} \quad (A-3)$$

560  
561 The solubility of ammonia in water is affected by the temperature and the acidity (pH) of the solvent (water). The  
562 equilibrium constant can be expressed as follows:

$$563 K_{NH_4^+} = 5.67 \cdot 10^{-10} \exp\left[-6286 \left(\frac{1}{T} - \frac{1}{298.15}\right)\right] \quad (A-4)$$

### 564 2. Henry's equilibrium

565 Upon its dissolution in water,  $NH_3$  obeys the Henry's law. Ammonia gas ( $NH_3(g)$ ) near the surface of the solvent  
566 is in equilibrium with the dissolved ammonia in the aqueous phase  $NH_3(aq)$  (in water). Henry's equilibrium is  
567 expressed as follows:



569 With  $H_{NH_3}$  as the Henry's constant, it can be expressed as follows (Wichink Kruit, 2010):

$$570 H_{NH_3} = \frac{[NH_3(aq)]}{[NH_3(g)]} = 5.527 \cdot 10^{-4} \cdot \exp\left[4092 \left(\frac{1}{T} - \frac{1}{298.15}\right)\right] \quad (A-6)$$

571 The partial pressure of ammonia near the surface of the soil can be calculated using Henry's constant and the  
572 dissociation equilibrium (Wichink Kruit, 2010):

$$573 P_{NH_3} = \frac{K_{NH_4^+} [NH_4^+]}{H_{NH_3} [H^+]} = \frac{5.67 \cdot 10^{-10} \cdot \exp\left[-6286 \left(\frac{1}{T} - \frac{1}{298.15}\right)\right]}{5.527 \cdot 10^{-4} \cdot \exp\left[4092 \left(\frac{1}{T} - \frac{1}{298.15}\right)\right]} \times \frac{[NH_4^+]}{[H^+]} \quad (A-7)$$

574 If we use the ideal gas law ( $PV=nRT$ ), we can draw the link between the mass density of ammonia ( $NH_3(g)$ ) and  
575 the partial pressure:



$$\chi_{NH_3} = \frac{P_{NH_3} \cdot M_{NH_3}}{R \cdot T} \quad (\text{A-8})$$

576  
 577 Where  $\chi_{NH_3}$  is the concentration of  $NH_3$  at the soil surface ( $kg\ m^{-3}$ ),  $P_{NH_3}$  is the partial pressure of  $NH_3$  near the  
 578 surface (atm),  $M_{NH_3}$  is the molar mass of  $NH_3$  ( $kg\ mol^{-1}$ ),  $R$  is the gas constant ( $0.082\ atm\ L\ mol^{-1}\ K^{-1}$ ), and  $T$  is  
 579 the temperature in Kelvin.

580 Substituting Eq. (A-5) in (A-6) we get:

$$\chi_{NH_3} = \frac{2.75 \cdot 10^9 \left(\frac{gK}{m^3}\right)}{T_{soil}} \exp\left[\frac{-1.04 \cdot 10^4}{T_{soil}}\right] \Gamma_{soil} \quad (\text{A-9})$$

581  
 582 Where  $\chi_{NH_3}$  is the concentration of ammonia at the soil surface at equilibrium ( $g\ m^{-3}$ ), and is referred to as the  
 583 compensation point,  $T_{soil}$  is the temperature of the soil (Kelvin),  $\Gamma_{NH_3}$  is the  $NH_3$  emission potential from the soil  
 584 and is a dimensionless ratio between  $[NH_4^+]$  and  $[H^+]$ .

### 585 3. Ammonia total columns from IASI

586 In this study we use the total columns of ammonia from IASI (molecules  $m^{-2}$ ) in order to calculate the emission  
 587 potential  $\Gamma_{soil}$ , we should draw the link between these columns and this parameter. The bi-directional exchange of  
 588  $NH_3$  between the surface and the atmosphere can be expressed by the flux (assuming a flux independent of time)  
 589 (Roelle and Aneja, 2005; Zhang et al., 2010):

$$Flux_{NH_3} = k ([NH_3]^{soil} - [NH_3]^{atm}) \quad (\text{A-10})$$

590  
 591 Where  $Flux_{NH_3}$  is the bidirectional flux between the soil and the atmosphere (molecules ( $m^2\ s^{-1}$ )),  $k$  is the soil –  
 592 atmosphere exchange velocity ( $m\ s^{-1}$ ), also known as the mass transfer coefficient;  $[NH_3]^{soil}$  is the concentration  
 593 of  $NH_{3(g)}$  in the soil, and  $[NH_3]^{atm}$  is the concentration of  $NH_{3(g)}$  in the atmosphere (molecules  $m^{-3}$ ).

594 Assuming a first order dissociation of  $NH_3$ , we can express the change in the  $[NH_3]^{col}$  total columns as follows:

$$\frac{d [NH_3]^{col}}{dt} = Flux_{NH_3} - k' [NH_3]^{col} \quad (\text{A-11})$$

595  
 596 Where  $k'$  is the rate of dissociation of first order  $k' = 1/\tau$  ( $m\ s^{-1}$ ), with  $\tau$  the lifetime of  $NH_3$  in the atmosphere.  
 597 Assuming steady state, and considering the  $[NH_3]^{atm}$  as the  $[NH_3]^{col}$ , and  $[NH_3]^{soil}$  as  $\chi_{NH_3}$ , Eq. (A-9) can be  
 598 written as:

$$k \left( \frac{N_a \cdot \chi_{NH_3}}{M_{NH_3}} - \frac{1}{c} [NH_3]^{col} \right) = \frac{[NH_3]^{col}}{\tau} \quad (\text{A-12})$$

599  
 600 Where  $c$  is the column height and is equal to 6 km. It is important to note that we neglect the effect of transport by  
 601 wind since we only look at large regions. Finally, the total column of ammonia  $[NH_3]^{col}$  can be written as:

$$[NH_3]^{col} = \frac{N_a \cdot \chi_{NH_3}}{M_{NH_3} \cdot \left(c + \frac{1}{k\tau}\right)} \quad (\text{A-13})$$

602  
 603 The column height is not considered anymore because it is negligible compared to  $1/k\tau$ , using Eq. (A-6) in (A-  
 604 11) we get:



$$[NH_3]^{col} = \frac{2.75 \cdot 10^{27} \left(\frac{gK}{cm^3}\right)}{T_{soil}} \exp\left[\frac{-1.04 \cdot 10^4}{T_{soil}}\right] \Gamma_{NH_3} \cdot k\tau \quad \left(\frac{molecules}{cm^2}\right) \quad (A-14)$$

605

606 Note that  $2.75 \cdot 10^{27} = \frac{a \cdot N_a \cdot c'}{M_{NH_3}} \left(\frac{K \text{ molecules}}{cm^3}\right)$ , where  $a = 2.75 \cdot 10^3 \text{ (g K cm}^{-3}\text{)}$ ,  $N_a$  Avogadro's number  
607 ( $6.0221409 \times 10^{23} \text{ molecules mol}^{-1}$ ),  $10^{-2}$  is added to convert  $k$  from  $m \text{ s}^{-1}$  to  $cm \text{ s}^{-1}$ , and  $M_{NH_3}$  the molar mass of  
608  $NH_3$  ( $17.031 \text{ g mol}^{-1}$ ). The emission potential of  $NH_3$  from the soil can we written as:

$$\Gamma_{soil} = \frac{[NH_3]^{col} \cdot T_{soil}}{\exp\left(\frac{-b}{T_{soil}}\right)} \frac{M_{NH_3}}{a \cdot N_a \cdot 10^{-2}} \cdot \frac{1}{k\tau} \quad (A-15)$$

609

610 Where  $b = 1.04 \times 10^4 \text{ K}$ .



## 611 Author contribution

612 RA contributed to the conception and design of the article, developed the code, wrote the manuscript, analysed and  
613 interpreted of the data, and approved the version for submission; CV, CC, and PFC revised the manuscript; WCP provided  
614 the GEOS-Chem simulation data, and revised the manuscript; NE provided ammonia lifetime calculation using the LMDz-  
615 OR-INCA chemistry transport model and commented on the manuscript; MVD and LC contributed to the acquisition of the  
616 IASI ammonia data (NH<sub>3</sub>-v3R-ERA5), and revised the manuscript; SS contributed to the conception and design of the article,  
617 provided the EC-Earth temperature data, and revised the manuscript, and approved the version for submission.

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## 629 Competing interests

630 The authors are aware of no competing interests.

## 631 Data accessibility statement

632 The IASI-NH<sub>3</sub> used in this study are retrieved from the AERIS data infrastructure (<https://iasi.aeris-data.fr/nh3r-era5/>). ERA5  
633 skin temperature from 1979 to present are available for download in the following DOI: [10.24381/cds.adbb2d47](https://doi.org/10.24381/cds.adbb2d47). The GEOS-  
634 Chem outputs used in this study are only available upon request. EC-Earth3 model output prepared for CMIP6 ScenarioMIP  
635 are retrieved here: <https://doi.org/10.22033/ESGF/CMIP6.727>. The MODIS land cover data are available for download in the  
636 following link: <https://doi.org/10.5067/MODIS/MCD12Q1.006>.

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