

## Quantification of eutrophic aerial compounds in Galicia (NW Spain): Part 1 - NH<sub>3</sub> inventory

A. GALLEGO, A. HOSPIDO, M. T. MOREIRA and G. FEIJOO

*Departamento de Ingeniería Química, Escuela Técnica Superior de Ingeniería*

*Universidad de Santiago de Compostela,*

*Rúa Lope Gómez de Marzoa, Campus Universitario Sur, 15782 - Santiago de Compostela, España*

Corresponding author: A. Gallego; e-mail: alejandro.gallego@usc.es

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### RESUMEN

En esta serie de artículos se han calculado por primera vez los inventarios de emisiones completos de NH<sub>3</sub> y NO<sub>x</sub> en Galicia (NW España), una región seriamente amenazada por la eutrofización provocada por estas emisiones. En esta primera parte se han estimado las emisiones de NH<sub>3</sub> de las principales fuentes (tanto agrícolas como no agrícolas) y se han cuantificado sus incertidumbres asociadas. Los resultados han demostrado que las emisiones asociadas al ganado (sobre todo por pollos broiler y ganado bovino) son especialmente relevantes en esta región representando prácticamente el 90% del total, siendo el uso de fertilizantes el segundo foco de emisión en importancia, mientras que el conjunto de fuentes no agrícolas representan menos de un 4%. El análisis de los valores de la incertidumbre ha demostrado que las investigaciones futuras en la región se deben centrar en establecer factores más específicos de emisión para las fuentes agrícolas (especialmente para ganado bovino, broilers y urea) para reducir dicha incertidumbre.

### ABSTRACT

In this series of articles, complete NH<sub>3</sub> and NO<sub>x</sub> emission inventories have been calculated for the first time for Galicia (NW Spain), a region in serious danger due to the eutrophication produced by these emissions. In this first part, NH<sub>3</sub> emissions corresponding to main sources (from agricultural and non-agricultural activities) have been estimated and the associated uncertainties have been quantified. The results have shown that livestock emissions (particularly by broilers and cattle) are especially relevant in this region, representing almost 90% of the total, the use of fertilizers being the second source in importance, while all the non-agricultural sources as a whole contribute less than 4%. The study of uncertainties has shown that future research in the region has to focus on the development of more specific emission factors for agricultural sources (especially for cattle, broilers and urea) to reduce this uncertainty.

**Keywords:** Eutrophication, inventory, NH<sub>3</sub>, Galicia, uncertainty, air emissions.

### 1. Introduction

Quantification of the emissions of atmospheric ammonia (NH<sub>3</sub>) and nitrogen oxides (NO<sub>x</sub>) is essential to guide strategies for emission control. Emission estimation is also important as input to atmospheric transport models used to estimate regional transport and deposition of pollutants. Data quality, together with associated uncertainty, will determine the value of the outputs of these models and the estimations derived.

Several inventories for NH<sub>3</sub> and NO<sub>x</sub> emissions have been published in different spatial scales including regional scales (e.g. Bouwman *et al.*, 1997; Koch *et al.*, 2001; Friedrich *et al.*, 2002;

Hellsten *et al.*, 2008). In Spain, national emission inventories, including  $\text{NH}_3$  and  $\text{NO}_x$ , have been calculated for the whole country as well as for several regions (Moreno and Lozano, 2006). In Galicia (NW Spain), Casares *et al.* (2005) only reported industrial emissions of  $\text{NH}_3$  and  $\text{NO}_x$ ; however, complete regional inventories of both pollutants are still lacking.

Emission of  $\text{NH}_3$  and  $\text{NO}_x$  and subsequent deposition represent nitrogen input with eutrophication effects on sensitive ecosystems, causing terrestrial and aquatic eutrophication (Bobbink *et al.*, 1992). Regional conditions (e.g. relative importance of agricultural activities) can highly affect this type of impacts (Finnveden and Potting, 1999). In this sense, Galicia is one of the regions in Europe with high soil and water eutrophication risk (EEA, 2001). The European Environment Agency reported that Galicia is one of the few European regions that present high values of N inputs from livestock activities ( $\geq 100$  kg N/ha) and, at the same time, medium values of nitrogen fertilizer use (50-100 kg N/ha) (EEA, 2000). In fact, an increase of nitrate concentration has been detected in many rural areas, rising in less than 10 years from values of  $<2$ -3 mg/L to values of 10-20 mg/L (Macías *et al.*, 2003). Even though the European Directive 91/270 establishes the limit of nitrate concentrations for human consumption at 50 mg/L, the high increase in a relatively short period of time is causing concern. In the specific case of aerial compounds, terrestrial eutrophication caused by atmospheric deposition of nitrogen compounds has been reported as a main threat to sustainability of terrestrial ecosystems in Galicia (Rodríguez and Macías, 2006), where the threshold for nitrogen deposition ensuring protection of forest ecosystems reportedly had been exceeded in 40% of the region. Rodríguez and Macías, 2006) have also identified the Atlantic heathlands, which are protected natural areas, as the Galician ecosystems most sensitive to the effects of eutrophication.

The above-mentioned facts justify carrying out regional inventories of eutrophying N emissions in Galicia. Therefore, the main objectives of these series are to update, or if not available, to calculate the emissions of these substances in Galicia. Furthermore, a quantitative estimation of uncertainties associated with these emissions is included. Part 1 presents the calculations done for  $\text{NH}_3$ . In a parallel study, Part 2 (pages 163-176, this issue) focuses on  $\text{NO}_x$  emissions.

## 2. Methods

### 2.1 Source selection

The 30 major emission sources obtained by CORINAIR from 28 European countries in September 1995 (EEA, 2007) were taken as basis for emission estimations. However, some considerations need to be made:

- No production of NPK fertilizers, ammonium, urea and ammonium nitrate takes place in Galicia, and therefore these sources were excluded.
- Use of latrines is insignificant, so this source was disregarded.
- Emissions from wastewater treatment, enteric fermentation from dairy cows and from other cattle contributed less than 1% to the  $\text{NH}_3$  emissions of the CORINAIR inventory (EEA, 2007) and, therefore, there is no methodology for calculation included in the EMEP/CORINAIR guidebook. Therefore, these sources were not estimated.
- Although excluded in the CORINAIR inventory, industrial and mobile sources have been considered here as they were regarded of importance by other authors (Sutton *et al.*, 2000; Klimont and Brink, 2004).

- Emissions produced by residential consumption of biomass and carbon as well as use of sludge and diverse domestic products were taken into account because they were found of importance in the non-agricultural emission inventory of the UK (Sutton *et al.*, 2000) and they are also considered important in Galicia.
- Finally, emissions associated with fires were estimated, as they are believed to be significant sources in Galicia.

## 2.2 Methodologies for the calculation of emissions

The methodologies proposed by the EMEP/CORINAIR Guidebook were followed to calculate most of the emissions (EEA, 2007). This approach is international and fully recognized, as the parties of The Convention on Long Range Transboundary Air Pollution (including Spain) use this guidance as a reference to report annual emissions to the United Nations Economic Commission for Europe (UNECE). The reference year for the inventory is 2001. When data used is referred to a different period, it will be appropriately specified in the text. NH<sub>3</sub> emissions are always reported in tonnes (t).

## 2.3 Uncertainties

Uncertainty values were determined in each case. When available, the values estimated by the methodologies were considered and, if expressed in a range, the less favorable case was adopted. However, in most cases, uncertainty needed to be estimated, so the matrix described in Table I, adapted from Frischknecht and Jungbluth (2004), was followed.

Table I. Matrix used to assess the quality of data sources (adapted from Frischknecht and Jungbluth, 2004). Quality data decrease from left to right.

Realibility	Verified data <sup>b</sup> based on measurement	Verified data partly based on assumptions or non-verified <sup>c</sup> data based on measurements	Non-verified data partly based on qualified expert estimates	Qualified estimation (e.g. industrial)	Non-qualified estimate and unknown data origin
Uncertainty factor <sup>a</sup>	1.00	1.05	1.10	1.20	1.50
Temporal correlation	≤ 3 years of difference to reference year	≤ 6 years of difference to reference year	≤ 10 years of difference to reference year	≤ 15 years of difference to reference year	Unknown or ≥ 15 years of difference to reference year
Uncertainty factor <sup>a</sup>	1.00	1.03	1.10	1.20	1.50
Geographical correlation	Data from area under study	Average data from a larger area that includes the area of study	Data from a smaller area than area under study or from similar area <sup>d</sup>		Data from unknown or distinctly different area <sup>d</sup>
Uncertainty factor <sup>a</sup>	1.00	1.01	1.02		1.10

<sup>a</sup>Value defined by an expert panel; <sup>b</sup>verified data: published in public environmental reports of companies, official statistics, etc. <sup>c</sup>non-verified data: personal information by letter, fax or e-mail; <sup>d</sup>similarity expressed in terms of environmental legislation.

In addition, Frischknecht and Jungbluth (2004) established basic uncertainty factors (Table II) associated with energy and resources demand, infrastructures, transport and emissions of pollutants to water, air and soil. The obtained values will vary if emission, demand or consumption is produced by combustion, during a process or in agriculture. In all these cases it is considered that the calculation has an intrinsic or basic uncertainty value, independent of reliability, temporal or geographical correlation. The value of this factor will increase accordingly with the complexity of the calculation. For example,  $\text{NH}_3$  emissions from fertilizers are directly related with the type of fertilizer and the spring temperature of the region while  $\text{NO}_x$  release in the same case depends on nitrification and denitrification processes, which are influenced by factors that are much more complicated to estimate (presence of aerobic-anaerobic zones, etc.); therefore, the basic factor will be lower in this case for  $\text{NH}_3$  than for  $\text{NO}_x$ .

Table II. Basic uncertainty factors used (Frischknecht and Jungbluth, 2004).

	Combustion	Process	Agriculture
Air pollutants			
CH <sub>4</sub> , NH <sub>3</sub>	1.50	-	1.20
Resources			
Primary energy sources	1.05	1.05	1.05

A summary of the uncertainty factors used in the inventory is included in the Annex. Taking into account the four elements of uncertainty defined, geometric standard deviation (interval of 95%) is calculated as follows (Frischknecht and Jungbluth, 2004):

$$\sigma_g^2 = \exp \left[ [\ln(U_1)]^2 + [\ln(U_2)]^2 + [\ln(U_3)]^2 + [\ln(U_b)]^2 \right] \quad (1)$$

where  $U_1$  is the reliability uncertainty factor,  $U_2$  the temporal correlation uncertainty factor,  $U_3$  the geographic correlation uncertainty factor and  $U_b$  the basic uncertainty factor.

### 3. Emission calculation

#### 3.1 Livestock

Usually, livestock management is the major source of  $\text{NH}_3$  emissions (Klimont and Brink, 2004). In Spain, it is responsible for 78% of the total agricultural emissions (MAPA, 2001). This data does not have to match the results in Galicia because these can vary locally due to differences in animal type distribution (cattle, sheeps, pigs, poultry, etc.) and their respective nitrogen excretion and emission factors, which depend on agricultural practices, housing systems and climate (EEA, 2007).

Following the detailed method included in the EMEP/CORINAIR guidelines, and due to the importance of the emissions, 38 types of animals have been considered (Table III). The amount of these animals in Galicia was obtained from different sources (Bello *et al.*, 2004; CMR 2003a; MAPA, 2003; CMR, 2000)

Once the number of animals was known, the emission during grazing, in the stables (including storage of manure) and due to application of manure has been estimated (Table IV). As a first step, the amount of N produced per type of animal in Spain (considering the kind of feeding in this country) was obtained from different sources (Babot *et al.*, 2002; MAPA, 2001; Camaleño, 2008) except in the case of minks where, due to the lack of Spanish data, a generic value was used

Table III. Types of animals analyzed.

Animal						i
Dairy cow	Holstein					1
	Others					2
	>24 months	Stud				3
Other cattle		Cows	Not calved	Sacrifice		4
				Milk production	Holstein	5
					Others	6
			Nurse cow		7	
			Calved	Nurse cow		8
	12-24 months	Bulls	Sacrifice			9
			Replacement			10
		Cows	Sacrifice			11
			Replacement			12
	<12 months (first months)	Sacrifice,		13		
		Others	Bulls	Sacrifice		14
				Replacement		15
			Cows	Sacrifice		16
				Replacement		17
Pigs	Suckling pig					18
	Fattening pigs	From 20 to 49 kg				19
		From 50 to 79 kg				20
		From 80 to 109 kg				21
		More than 109 kg				22
		Pigs for reproduction	Not calved	Not covered		23
	Covered			24		
	Calved		25			
Stud pigs					26	
Sheep	Sacrifice					27
	Reproduction	Ram				28
		Ewe	Not calved		29	
			Calved		30	
Goats	Reproduction					31
	Sacrifice					32
Horses, donkeys and mules						33
Poultry	Laying hen					34
	Broiler					35
	Turkeys, ducks and geese					36
Mother rabbits						37
Mother minks						38

(EEA, 2007). The next step is to calculate the ratio of N excreted in and outside the stables. For this calculation it must be taken into account that the amount of N excreted per type of animal is different in summer and winter due to feeding changes during these seasons. Therefore, ECOTEC (1994) takes into consideration the ratio summer/winter of N excretion and the time each type of animal stays inside the stable in winter and summer, in order to calculate the amount of N excreted in and outside the stables per type of animal in Spain. Finally, knowing the N excreted outside the stable and the  $\text{NH}_3$  emission factor during grazing per type of animal (ECOTEC, 1994; Bartrolí, 2003; EEA, 2007), we are able to calculate the total emissions during this stage. In order to calculate the emissions in the stables (including production and storage) the number of each type of animal has been multiplied by the animal-specific emission factors (kg N- $\text{NH}_3$  emitted in the stables/head and year) developed for Spain by Bartrolí (2003).

It has been assumed that N losses in stables in Spain are produced principally in form of N- $\text{NH}_3$ , therefore emissions in other forms are neglected (personal communication by Dr. Jordi Bartrolí, Universidad de Gerona, February 26, 2007, jordi\_bartrolí@yahoo.es). Having this in mind, N present in manure is the result of subtracting N excreted outside the stables and N- $\text{NH}_3$  emitted in the stables from total N excretion. Similar to other cases,  $\text{NH}_3$  specific emission factors (ECOTEC, 1994; EEA, 2007) have been applied in order to obtain  $\text{NH}_3$  emissions due to the application of manure.

### 3.2 Nitrogen fertilizer application

Volatilization of N-fertilizers has been estimated to contribute between 10 and 20% of agricultural  $\text{NH}_3$  emissions in Europe (ECOTEC, 1994). In Spain, this source is responsible for 22% of the total agricultural emissions (MAPA, 2001). Again, the detailed methodology of the EMEP/CORINAIR guidance was applied (EEA, 2007). In this methodology, different emission factors are established for three types of regions, defined as a function of spring temperature ( $t_s$ ): region A ( $t_s > 13^\circ\text{C}$ ), region B ( $6 < t_s < 13^\circ\text{C}$ ) and region C ( $t_s < 6^\circ\text{C}$ ). In 2001, 90% of the Galician meteorological stations measured values within the range of region B (CMA, 2002), hence that set of factors was used (Table V).  $\text{NH}_3$  losses from ammonium sulphate, ammonium nitrate and anhydrous nitrate have been found to increase markedly with increasing pH (Whitehead and Raistrick, 1990) and therefore a multiplication factor is needed if the soil is basic (EEA, 2007). Galician soils are predominantly acid and therefore this multiplication factor has been not applied (Álvarez *et al.*, 2005).

For quantification of the amount of each type of fertilizer used (Table V), national distribution of fertilizer types was applied to the total amount of fertilizers consumed in Galicia (CMR, 2003b; INE, 2008).

### 3.3 Waste treatment

Munday (1990) estimated that 0.073 t N (10% as N- $\text{NH}_3$ ) are emitted for each ton of methane produced in a landfill. Taking into account this data and methane emissions from controlled and uncontrolled landfills in Galicia of 9,514 t N,  $\text{NH}_3$  emissions were estimated as 84.3 t, within a range from 26.7 to 181 t (Bello *et al.*, 2004).

In 2003, 6,042 t of organic fraction waste was assigned to composting in Galicia (CMA, 2004), adding vegetation remains (1 part for each 2.5 parts of organic fraction, with an organic fraction density of 0.6 and a vegetation density of 0.3 (Bartrolí, 2003)) to improve the final quality of the compost. N contents of the organic and vegetal fraction have been estimated to be 0.84% and 1.36%, respectively, of their weight (Molina, 1997). During the composting process, 24.88% of this initial nitrogen content is lost (Soliva, 2001) and, in specific terms, an emission of 0.17 kg

Table IV. Estimation of emissions from livestock production

i	Animals		Excretion		Emission during grazing			Emission in stables		Emission from manure applied to the soil	
	A) No. of animals	B) Total N factor (kg N/head year)	C) N in stables (kg N/head year)	D) N outside stables (kg N/head year)	E) N-NH <sub>3</sub> emission factor (kg N-NH <sub>3</sub> /kg N excret.)	F) N-NH <sub>3</sub> emission (t N-NH <sub>3</sub> )	G) N-NH <sub>3</sub> emission factor (kg N-NH <sub>3</sub> /head year)	H) N-NH <sub>3</sub> emission (t N-NH <sub>3</sub> )	I) N in manure (t N)	J) N-NH <sub>3</sub> emission factor (t N-NH <sub>3</sub> /t N in manure)	M) N-NH <sub>3</sub> emission (t N-NH <sub>3</sub> )
1	415,360	67.83	46.06	21.8	0.08	723	19,930	8,278	10,855	0.22	2,388
2	20,327	60.04	40.77	19.3	0.08	31	19,930	405	424	0.22	93
3	2,805	59.65	27.75	31.90	0.08	7	6,507	18	60	0.22	13
4	434	46.92	21.83	25.09	0.08	1	6,507	3	7	0.22	1
5	10,247	51.54	23.97	27.57	0.08	23	6,507	67	179	0.22	39
6	425	48.51	22.57	25.94	0.08	1	6,507	3	7	0.22	2
7	2,994	45.70	21.26	24.44	0.08	6	6,507	19	44	0.22	10
8	260,791	49.91	23.22	26.69	0.08	557	6,507	1,697	4,358	0.22	959
9	253	44.04	29.89	14.15	0.08	0.3	7,274	2	6	0.22	1
10	84	71.57	48.57	23.00	0.08	0.2	7,274	1	3	0.22	1
11	2,747	44.61	30.28	14.33	0.08	3	7,274	20	63	0.22	14
12	126,809	48.90	33.16	15.74	0.08	160	7,274	922	3,282	0.22	722
13	136,700	53.75	53.75	0	0	0	10,138	1,386	5,962	0.22	1,312
14	6,149	49.85	49.85	0	0	0	10,138	62	244	0.22	54
15	2,050	64.97	64.97	0	0	0	10,138	21	112	0.22	25
16	1,848	50.28	50.28	0	0	0	10,138	19	74	0.22	16
17	85,321	65.93	65.93	0	0	0	10,138	865	4,760	0.22	1,047
18	339,359	1.61	1.61	0	0	0	1,040	353	194	0.14	27
19	238,115	8.12	8.12	0	0	0	3,034	723	1,211	0.14	170
20	200,104	11.31	11.31	0	0	0	5,352	1,071	1,192	0.14	167
21	92,872	13.52	13.52	0	0	0	5,352	497	759	0.14	106
22	246	16.85	16.85	0	0	0	5,352	1	3	0.14	0.4
23	7,268	11.50	11.50	0	0	0	5,352	39	45	0.14	6
24	9,760	15.90	15.90	0	0	0	5,352	52	103	0.14	14
25	102,379	20.30	20.30	0	0	0	5,352	550	1,536	0.14	215
26	3,103	18.9	18.9	0	0	0	6,826	21	38	0.14	5
27	14,222	5.32	0.37	4.95	0.046	3	0.073	1	4	0.22	1
28	21,599	6.92	0.48	6.44	0.046	6	0.036	1	10	0.22	2
29	37,884	4.64	0.32	4.32	0.046	8	0.036	1	11	0.22	2
30	255,923	5.61	0.39	5.22	0.046	61	0.036	9	91	0.22	20
31	15,056	8.76	0.62	8.14	0.046	6	0.073	1	8	0.22	2
32	22,578	5.32 <sup>a</sup>	0.37	4.95	0.046	5	0.017	0.4	8	0.22	2
33	74,994	25.5	11.8	13.7	0.08	82	2,737	205	686	0.22	151
34	3,686 <sup>b</sup>	0.60	0.60	0	0	0	0.088	323	1,889	0.376	710
35	101,196 <sup>b</sup>	0.30	0.30	0	0	0	0.066	6,649	23,710	0.72	17,071
36	48,890	0.60	0.60	0	0	0	0.066	3	26	0.20	5
37	141,945	4.33	4.33	0	0	0	0.471	67	548	0.20	110
38	53,300	4.10	4.10	0	0	0	0.491	26	192	0.20	38
Total (t NH <sub>3</sub> ) <sup>c</sup>					1,684		62,643 (35,156-99,045)		24,382		25,522

<sup>a</sup> The same excretion factor as for sheep for sacrifice is considered; <sup>b</sup> Multiplied by 1000; <sup>c</sup> Total NH<sub>3</sub> (t NH<sub>3</sub>) = (Total F + Total H + Total M)(17/14). The results from uncertainty calculations are expressed within parentheses. References used in the calculations: Camaleño (2008), EEA (2007); Bello *et al.* (2004); Bartoli (2003); CMR (2003a); MAPA (2003); Babot *et al.* (2002); MAPA (2001); CMR (2000); ECOTEC (1994)

Table V. Emissions from fertilizer application.

	A) Amount of N in the fertilizers (t N) <sup>a</sup>	B) Emission factor (t N-NH <sub>3</sub> /t N) <sup>b</sup>	C) Emissions (t NH <sub>3</sub> ) C=A×B×17/14
Complex nitrogen fertilizers	17,737	0.015	323
Ammonium nitrosulphate	4,366	0.02	106
Ammonium sulphate	811	0.015	14.8
Calcium ammonium nitrate	12,470	0.015	227
Ammonium nitrate	6,353	0.015	116
Urea	17,299	0.17	3,571
Calcium nitrate	864	0.015	15.7
Saltpeter	64	0.015	1.16
Agricultural ammonium	177	0.03	6.44
Nitrogen solutions	2,824	0.09	309
Total (t NH <sub>3</sub> )		4,690 (2,345-7,035)	

<sup>a</sup>(INE, 2008;CMR, 2003b); <sup>b</sup>Values for region B (EEA, 2007).

NH<sub>3</sub>/t waste is produced, considering as waste the sum of organic and vegetal fractions (Pagans *et al.*, 2006). In compost application to soil, the NH<sub>3</sub> volatilization factor is assumed to be half of the factor used when sewage sludge is applied (0.258 t N-NH<sub>3</sub>/ t N in sludge (Doka, 2003)), because the remaining nitrogen compounds are poorly volatile (Bartolí, 2003). Calculations of NH<sub>3</sub> emissions associated with composting are summarized in Table VI.

Only emissions from sludge applied to agricultural land are included here (Table VII), because NH<sub>3</sub> emitted due to landfill disposal was already quantified (see above) and emission associated with sludge incineration will be included in subsection 3.5.

Emissions associated with urban waste and dangerous waste management are also included in subsection 3.5.

Table VI. Emission associated with composting.

A) Waste (t)	B) N (t)	C) Inputs (t N) C=B <sub>1</sub> +B <sub>2</sub>	D) Emissions in composting process (t NH <sub>3</sub> ) D= (A <sub>1</sub> +A <sub>2</sub> ) × 0.17/1000	E) N. remaining (t) E=C– C × 0.2488	F) Total emissions compost (t NH <sub>3</sub> ) F=D+E × (0.258/2) × (17/14)
Organic (6,042)	50.75 <sup>a</sup>	111.14	1.78	83.49	14.9 (5.43-47.6)
Veget. (4,441)	60.39 <sup>a</sup>				

<sup>a</sup>The N contents of the organic and vegetal fraction have been estimated to be 0.84 and 1.36%, respectively, of their weight (Molina, 1997).

Table VII. Emission produced by the use of sludge for agricultural purposes.

A) Agricultural sludge (t dry matter)	B) N contents (g N/t dry matter)	C) Amount of N in sludge (t N) C= (A×B)/106	D) Volatilization factor (t N-NH <sub>3</sub> /t N)	E) Emission (t NH <sub>3</sub> ) E=C×D×(17/14)
14,472 <sup>a</sup>	1,820 <sup>a</sup>	26.34	0.258 <sup>b</sup>	8.25 (2.48-27.5)

<sup>a</sup>Personal communication by Vicente Amores Torrijos, Ministerio de Agricultura y Pesca (6, December 2006; vamorest@mapya.es); <sup>b</sup>(Doka, 2003).



### 3.4 Humans and pets

Direct human emission of NH<sub>3</sub> originates from breath, sweat, cigarette smoking and infant excretion (under 3 years of age) (Sutton *et al.*, 2000) (Table VIII). In case of perspiration, emissions are produced due to hydrolysis and volatilization of urea from sweat (Lee and Dollar, 1994). Several studies have demonstrated NH<sub>3</sub> emissions among the substances emitted by breathing and smoking (Lee and Dollar, 1994; Davies *et al.*, 1997). In the case of infant excretion, urea in diapers does not enter the sewage system and may hydrolyze producing NH<sub>3</sub> emissions (Lee and Dollar, 1994; Sutton *et al.*, 2000). In the same way, emissions from pets (dogs and cats) are mainly associated with excretion (Sutton *et al.*, 2000).

Table VIII. Emissions produced by persons and pets.

	A) Amount (animal or person)	B) Emission factor (g N-NH <sub>3</sub> /animal-person year) <sup>a</sup>	C) NH <sub>3</sub> (t) $C = A \times B \times 10^{-6} \times 17/14$
Persons	2,695,880 <sup>b</sup>	3.0 <sup>c</sup> 14.0 <sup>d</sup>	9.82 45.8
Minors (<3 years)	66,332 <sup>b</sup>	13.7	1.10
Smokers	663,186 <sup>b</sup>	17.8	14.3
Cats	76,808 <sup>e</sup>	100	10.3
Dogs	265,335 <sup>e</sup>	610	196.5
Total (t NH <sub>3</sub> )		278 (117-827)	

<sup>a</sup>(Sutton *et al.*, 2000); <sup>b</sup>(INE, 2008); <sup>c</sup>breathing emission factor; <sup>d</sup>sweating emission factor; <sup>e</sup>Data for year 2004 (INE, 2008; Refugio, 2004).

### 3.5. Industrial sources

Casares *et al.* (2005) calculated NH<sub>3</sub> emissions in 2001 performing surveys in the 370 main enterprises responsible for air pollution in Galicia. Total emission was 525 t NH<sub>3</sub> (within a range of 437.5 to 630 t), power and cogeneration plants being the main sources with contributions of 58 and 15%, respectively.

### 3.6 Mobile sources

#### 3.6.1 Road transport

The EMEP/CORINAIR methodology for the calculation of emissions produced by road transport has been computerized by the European Environment Agency in a program called COPERT (Computer Programme to Calculate Emissions from Road Transport). It was developed by Ntziachristos and Samaras (1999) and has suffered successive updates (EEA, 2007). The present paper uses the last released version 4.5 (<http://lat.eng.auth.gr/copert/>).

NH<sub>3</sub> emissions in road transport are calculated by adding emissions from two different sources, namely the thermally stabilized engine operation (“hot emissions”) and the warming-up phase (“cold emissions”). In the first case, the emissions here calculated are based on the application of specific hot emission factors developed for 96 types of vehicles (passenger cars, buses, light and heavy-duty vehicles –< 3.5 t and >3.5 t–, motorbikes and mopeds classified by registration year, cubic capacity and fuel type), taking also into account mileage and driving mode (urban, rural or highway). Emissions in the warming-up phase further depend on ambient temperature

and average trip distance. The majority of these cold emissions are produced in urban driving by passenger cars and light-duty vehicles and therefore, COPERT only considers these sources for these specific emissions. For buses and heavy-duty vehicles a load factor is included, because the fact of being more or less loaded influences the amount of emissions. Table IX shows the used data and references.

The number of vehicles of each class is multiplied by the corresponding factor in order to estimate NH<sub>3</sub> emissions and this data is added to obtain the global values. Table X presents the global values of NH<sub>3</sub> emissions for the different types of vehicles in Galicia.

The program COPERT 4.5 does not allow to calculate emissions associated with tractors. In this particular case, the number of tractors, the amount of annually consumed fuel and the NH<sub>3</sub> emission factor (taking into account if the tractor is gasoline or diesel-driven) have been considered (Table XI). Data refers to a standard 80 CV tractor that consumes 3.36 kg fuel/h and is used 600 h/year (Nemecek *et al.*, 2004).

Table IX. Used parameters to calculate NH<sub>3</sub> emissions (program COPERT 4.5).

Parameters	References
Factor $\beta^a$	(CMA, 2002) (Camaleño, 2008)
Load factor	(André <i>et al.</i> , 1999)
No. of vehicles <sup>b</sup> (registration year, cubic capacity and fuel)	(Camaleño, 2008; DGT, 2002; INE, 2008)
Average speed <sup>b</sup> (urban, rural, highway driving)	(MMA, 2007)
Average annual mileage <sup>b</sup>	(Bello <i>et al.</i> , 2004)
%km covered/year according to driving (urban, rural, highway)	(Camaleño, 2008)
Fuel consumption	(INE, 2008)
Fuel characteristics (concentrations of sulphur, cadmium, etc.)	(Camaleño, 2008)

<sup>a</sup>It indicates % of annual mileage covered before the engine reaches its normal operation temperature; <sup>b</sup>types of vehicles considered: Passenger cars, motorbikes, mopeds, buses, light and heavy-duty vehicles (< > 3.5 t).

Table X. NH<sub>3</sub> emissions produced by road transport (program COPERT 4.5).

Type of vehicles	NH <sub>3</sub> /year
Passenger cars	393
Motorbikes	1.57
Mopeds	0.66
Buses	1.71
Light-duty vehicles (< 3.5 t)	17.1
Heavy-duty vehicles (> 3.5 t)	2.08
Total (t NH <sub>3</sub> )	416 (347 - 499)

Table XI. NH<sub>3</sub> emissions produced by tractors.

	A) Number of vehicles <sup>a</sup>	B) Fuel consumed (t/vehicles year) <sup>b</sup>	C) NH <sub>3</sub> factor (kg NH <sub>3</sub> /t fuel) <sup>b</sup>	D) NH <sub>3</sub> (t) D = A×B×C×10 <sup>-3</sup>
Gasoline tractors	144	2.016	0.04	0.012
Diesel tractors	9.056	2.016	0.02	0.37
Total (t NH <sub>3</sub> )			0.38 (0.19 - 0.73)	

<sup>a</sup>(INE, 2008); <sup>b</sup>(Nemecek *et al.*, 2004).

### 3.6.2 Other mobile sources and machinery

Diesel B (i.e. diesel subsidized by the Spanish Government) is used for railway transport, cogeneration, agricultural consumption (tractors and other machinery), fishery and forest machines. In 2001, a total of 456,214 t of Diesel B was consumed in Galicia (personal communication by Carmen Togores, Instituto Energético de Galicia, February 27, 2007; carmentogores@inega.es), without taking into account the amount consumed by either cogeneration or diesel tractors. Both emissions are specifically considered in subsections 3.5 and 3.6.1, respectively. The emission factor for diesel consumption by other mobile sources is 0.007 g NH<sub>3</sub>/kg diesel (EEA, 2007) and, as a result, the annual emission is 2.95 t NH<sub>3</sub>, within a value range of 1.71 - 5.98.

### 3.7 Residential combustion of coal and biomass

In Spain, the energy produced from coal in 2001 for domestic purposes was 2,721,550 GJ (López *et al.*, 2005). Allocation of consumptions on a population basis (data of Spanish and Galician population in 2001 from INE, 2008) allows estimating energy production from this source in Galicia (180,894 GJ). According to EEA (2007), the emission factor is 0.3 g NH<sub>3</sub>/GJ and therefore Galician emission amounts to 0.054 (0-0.181) t of NH<sub>3</sub>.

Residential consumption of biomass is difficult to estimate, because it is mainly consumed in rural areas, where consumption is difficult to register. Data from Bello *et al.* (2004) considered an amount of 3,138,000 GJ/year. An emission factor of 3.8 g NH<sub>3</sub>/GJ was used (EEA, 2007), thus obtaining an emission in Galicia of 11.9 (0-36.1) t of NH<sub>3</sub>.

### 3.8 Household sources

NH<sub>3</sub> is a known component of many domestic products but generally estimations of emissions from these sources are not elaborated (Sutton *et al.*, 2000). Some of these sources are house-cleaning products, nitrogen fertilizers for domestic use, ammonia refrigerators, and solvents used in building construction. Emissions per house were calculated combining the emissions reported by Sutton *et al.* (2000) for the UK and the UK census of houses (National Statistics, 2001), and these factors were then applied to the number of houses in Galicia (INE, 2008) (Table XII).

Table XII. Emissions from household sources

Source	A) Number of houses <sup>a</sup>	B) NH <sub>3</sub> emission factor (g NH <sub>3</sub> /house) <sup>b</sup>	C) NH <sub>3</sub> (t) C = A×B×10 <sup>-6</sup>
Fridges	900,376	5.74	5.17
Solvents		39.3	35.4
Fertilizers		11.0	9.91
Cleaning products		18.75	16.9
Total (t NH <sub>3</sub> )	67.3 (17.0 - 224)		

<sup>a</sup>(INE, 2008); <sup>b</sup>(Sutton *et al.*, 2000; National Statistics, 2001).

### 3.9 Fires

Nitrogen containing compounds have been measured in biomass burning emissions in several studies (e.g. Lee and Dollar, 1994; Ward and Hardy, 1991). The dominant nitrogenous species are NO<sub>x</sub> in the flaming phase and NH<sub>3</sub> in the smoldering phase of combustion (Dennis *et al.*, 2002). To

compute these emissions (Table XIII), the detailed EMEP/CORINAIR methodology was applied (EEA, 2007) and the emitted carbon mass (M(C)) was calculated by equation (2) (Crutzen *et al.*, 1979):

$$M(C) = 0.45 \times A \times B \times \alpha \times \beta \quad (2)$$

where M(C) is the carbon mass emitted (kg C), 0.45 is the average fraction of carbon in wood, A the burnt area (m<sup>2</sup>), B the average biomass which acts as combustible per unit of area (kg C/m<sup>2</sup>),  $\alpha$  the fraction of biomass in the surface with respect to total biomass of B and  $\beta$  the efficiency of burnt biomass in the surface.

Once M(C) is obtained, the emitted NH<sub>3</sub> can be calculated using the factor 1.8 g NH<sub>3</sub>/kg of emitted C (Andreae, 1991).

Table XIII. Fire emissions.

Vegetation	A) <sup>a</sup>	B) $\alpha^b$	C) $\beta^b$	D) B <sup>b</sup>	E) M(C) (t C)	F) Factor (kg NH <sub>3</sub> /t C) <sup>b</sup>	G) NH <sub>3</sub> (t) G = F×E×10 <sup>-3</sup>
Brush	14.217	0.64	0.5	7.5	153.542	1.8	276
Trees	4.014	0.75	0.2	35	94.837	1.8	171
Grassland	122.5	0.36	0.5	2	198.4	1.8	0.36
Total (t NH <sub>3</sub> )					447 (149-1,342)		

<sup>a</sup>Burnt hectares (MMA, 2008); <sup>b</sup>(Crutzen *et al.*, 1979).

### 3.10 Wild animals, forests and natural grassland

Although data availability is very limited for these emissions, they were calculated due to their importance within natural emissions. According to the CORINAIR inventory for 28 European countries (EEA, 2007), contribution of forests and natural grassland was, on average, 0.8 and 0.3%, respectively. By applying these percentages to the total NH<sub>3</sub> emissions inventoried in Galicia, emissions of 561 (range 317-993) t from forests and 210 (range 119-372) t from natural grassland were obtained.

Other inventories reported a contribution of 0.2% due to wild animals (Sutton *et al.*, 2000; Bouwman *et al.*, 1997). Assuming here the same share, an emission of 140 (range 89.9-219) t of NH<sub>3</sub> could be derived.

## 4. Summary of results and discussion

### 4.1 Inventory of NH<sub>3</sub> emissions

The main result of this study is the inventory of NH<sub>3</sub> emissions for the region of Galicia (Table XIV). To the best of our knowledge, this is the first complete inventory for this specific region and therefore no comparison is possible with previous reports. The comparison with other inventories is also difficult as they are usually focused only on emissions produced by livestock and use of fertilizers. However, some inventories have been calculated for the UK with a set of sources very similar to those of our study, which allows a certain level of global comparison (Misselbrook *et al.*, 2000; Sutton *et al.*, 2000). Therefore, comparison is only viable by tendency analysis rather than by exhaustive comparative studies of specific results. The latter would have no sense due to the high level of uncertainty associated with some emission sources.

Table XIV. Summary of Galician NH<sub>3</sub> inventory.

Sources	Emission (t NH <sub>3</sub> )
Livestock	62,643 (35,156-99,045)
Nitrogen fertilizer application	4,690 (2,345-7,035)
Waste treatment	107.5 (34.6-256)
Humans and pets	278 (117-827)
Industry	525 (438-630)
Mobile sources	420 (349-507)
Residential combustion of coal and biomass	12.0 (0-3-6.3)
Household	67.3 (17.0-224)
Fires	447 (149-1,342)
Wild animals	140 (89.9-219)
Forests	561 (317-993)
Natural grassland	210 (119-372)
Total (t NH <sub>3</sub> )	70,101 (39,131-111,486)

As expected, livestock management was found to be the major source of NH<sub>3</sub> with a contribution of 89.4%, a value near the top limit of the range of 70-90% reported by Klimont and Brink (2004) for Europe. In UK, a country with an important livestock production, the contribution of livestock management accounts for 75% of the emissions (Sutton *et al.*, 2004), value comparatively lower than that for Galicia. If only the main sources (livestock management and fertilizers use) are considered, livestock management accounts for 96.1% of emissions in Galicia, similar to the figure of 92% reported for a high livestock production country such as the Netherlands (Koch *et al.*, 2001). Nevertheless, for Spain or Germany, this value is lower, namely 78 and 84%, respectively (MAPA, 2001; Döhler *et al.*, 2002). Taking into account all these facts, it can be concluded that livestock contribution to the NH<sub>3</sub> emissions has a great relevance in Galicia, especially compared with the rest of Spain.

Going a bit further and analyzing the activities included within livestock management, emissions from stables and from manure after application are the main sources, accounting for 49 and 48% of the total livestock emissions, respectively, while emissions during grazing are insignificant (3%). Looking at the contributions by animal types, broilers stand for almost half of the amount (46%) of livestock emission, followed by cattle (43%) and pigs (8%). This important role of broilers has not been reported by other national inventories (contributions of 6-15% according to Döhler *et al.*, 2002; MMA, 2002; Sutton *et al.*, 2004), but this is understandable in view of their large number in Galicia, namely 101,196,000 animals.

The second major source is the use of agricultural fertilizers that represents 6.7% of the total emissions, being urea the responsible for three quarters of these emissions. The emissions for the UK are very similar in importance accounting for 8% of the total (Sutton *et al.*, 2004).

The non-agricultural sources, a heterogeneous group, account for 3.9% of emissions, the principal items here are comprised of mobile sources, forestry, industry and fires. Other emissions which are closely related to agricultural activity such as use of tractors, use of sludge for agricultural purposes, production and use of compost, and use of other mobile sources and machinery (it has been estimated that 25% of the use is for agricultural purposes), not usually considered in other inventories as agricultural sources, are negligible, representing only 0.03% of the emissions.

#### 4.2. Analysis of uncertainties

The objective of this work was dual, the second goal being the inclusion of uncertainties associated with each emission. For this purpose a semi-quantitative method has been used. As can be observed in Table AI, the uncertainty due to geographical and temporal correlation of the data used is low because most of the data are obtained for Galicia and reported for the reference year (2001). Regarding reliability, only in the case of emissions associated with forests and natural grassland the worst value (level 5) has been adopted because the approximate estimation established lacks precision. In most of the remaining cases, the figures used are based on verified data (published in public environmental reports of companies, official statistics, etc.) partly relying on assumptions (level 2) or on qualified estimation according to expert criteria (level 4). As an example, the number of smokers in Galicia has been obtained from verified data (official statistics) partly based on assumptions (among others, the estimated percentage of smokers only takes into account people older than 16 years) and is therefore considered as level 2. Finally, the basic factor has been established in accordance with the values proposed by Frischknecht and Jungbluth (2004) (Table II), adopting in the remaining cases a value of 1. For the emissions of road transport, this basic factor has also been considered 1 (instead of 1.5 proposed by Frischknecht and Jungbluth, 2004) due to the high precision of the calculated emissions. On the contrary, in the case of emissions associated with wild animals, forests and natural grassland, the maximum value has been adopted for the basic factor (1.5) due to the high intrinsic uncertainty of these results. As mentioned in section 2.3, when the methodologies establish an uncertainty value, the latter has been selected by default, those values being collected in Table AII of the Annex.

In the case of the two major sources, livestock and agricultural fertilizers, reduction of the uncertainties is difficult because the most detailed methodology and the best local emission factors available (on Spanish level for livestock and Region B level for fertilizers) were used. Even so, the EEA methodology proposes an uncertainty factor of  $\pm 30\%$  for livestock and  $\pm 50\%$  for fertilizers. To reduce these uncertainties, efforts have to focus on the definition of more specific emission factors (especially for cattle, broilers and urea) that provide a better description of the activities at least at Spanish, and when available, at Galician level. For example, with regard to livestock emissions it would be very interesting to obtain, in the first place, two new emission factors, on the one hand, for the production of manure in the stables and on the other hand, for the storage of this manure. In the course of time, these factors should become more and more sophisticated, considering facts like the type of manure (liquid, solid or mixed), the way the manure is stored (e.g. utilization of covered or uncovered tanks, storage time) or applied (use of special equipment to reduce emissions, e.g., shallow injection). In order to obtain these specific factors, intensive field measurement campaigns must be made, as performed for industrial emissions in Galicia. These are expensive and time-consuming campaigns but they can be justified by the fact that the sources in question represent 96.1% of  $\text{NH}_3$  emissions. In the case of non-agricultural sources, research has to concentrate on the development of more specific factors for fires, forest, wild animals, grassland, pets (primarily, emissions from dogs) and humans (primarily, emissions from sweat and smokers).

#### 5. Conclusions

The results of this study allowed raising a complete  $\text{NH}_3$  inventory and its uncertainties for Galicia

(NW Spain). Although it has been applied to a specific region, this work can become a useful basis for the estimation of NH<sub>3</sub> inventories and its uncertainties in other parts of the world.

- Regarding the quantified emissions, the main conclusions are:
- Livestock is the major source of NH<sub>3</sub> emissions, representing 89.4% of these emissions, mainly produced by cattle and broilers.
- Application of N-fertilizers, principally urea, also plays an important role in the generation of NH<sub>3</sub> emission.
- Non-agricultural sources are of less importance, although attention should be paid to mobile sources, forestry, industry and fires.

Based on the uncertainties calculated, future research should be focused, in the first place, on the computation of precise emission factors better adapted to local conditions for livestock (cattle and broilers) as well as fertilizer application, and secondly, on the development of a more specific emission factor for fires, forest, pets (primarily, emissions from dogs) and humans (principally emissions from sweat and smokers).

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### Annex

#### *Uncertainty factors used along the inventory*

Table AI. Uncertainty factors calculated by the authors.

	A) Reliab. (U <sub>1</sub> )	B) Temp. (U <sub>2</sub> )	C) Geogr. (U <sub>3</sub> )	D) Basic (U <sub>b</sub> )	E) Results $E = \exp \sqrt{[\ln(A)]^2 + [\ln(B)]^2 + [\ln(C)]^2 + [\ln(D)]^2}$
<i>Livestock</i>					
Number of animals	1.05	1	1	1	1.05
<i>Waste treatment</i>					
Methane emissions	1.20	1	1	1.20	1.30
Vegetal and organic fraction	1.05	1	1	1	1.05
N contents (veget. and org.)	1.05	1.03	1.02	1	1.06
NH <sub>3</sub> emission in composting	1.05	1	1.02	1	1.05
N losses	1.05	1	1.02	1	1.05
Amount of sludge	1.10	1	1.01	1	1.11
Amount of N in the sludge	1.10	1	1.01	1	1.11

Continues in next page.

	A.Reliab. (U1)	B.Temp. (U2)	C. Geogr. (U3)	D. Basic (Ub)	E. Results $E = \exp\sqrt{[\ln(A)]^2 + [\ln(B)]^2 + [\ln(C)]^2 + [\ln(D)]^2}$
<i>Humans and pets</i>					
No. of persons	1.05	1	1	1	1.05
< 3 years	1.05	1	1	1	1.05
Smokers	1.05	1	1	1	1.05
No. of cats and dogs	1.20	1.03	1.01	1	1.20
<i>Industrial sources</i>					
NH <sub>3</sub> emissions	1.20	1	1	1	1.20
<i>Mobile sources</i>					
NH <sub>3</sub> emission road vehicles	1.20	1	1	1	1.20
No. of tractors	1.05	1	1	1	1.05
NH <sub>3</sub> emissions factor (tractors)	1.20	1	1.02	1.50	1.56
Kg consum. fuel/ tractor year	1.20	1	1.02	1.05	1.21
t diesel B other machinery	1.10	1	1	1.05	1.11
NH <sub>3</sub> emissions factor (diesel B)	1.20	1.03	1.02	1.50	1.56
<i>Residential combustion of coal and biomass</i>					
Spanish population	1.05	1	1	1	1.05
Coal consumption	1.20	1	1.01	1.05	1.21
Biomass consumption	1.20	1	1	1.05	1.21
<i>Emissions from household sources</i>					
No. of houses in Galicia	1.05	1	1	1	1.05
No. of houses in UK	1.05	1	1	1	1.05
<i>Wild animals, forest and natural grassland</i>					
NH <sub>3</sub> emissions from forest	1.50	1.03	1.02	1.50	1.77
NH <sub>3</sub> emissions grassland	1.50	1.03	1.02	1.50	1.77
NH <sub>3</sub> emissions wild animals	1.20	1	1.02	1.50	1.56

<sup>a</sup>Default value = 1 for items not included in the categories defined by Frischknecht and Jungbluth (2004): energy and resources demand, infrastructures, transport, waste treatment and emissions of pollutants to air, soil and water. For the emissions of road transport, the basic factor has been considered to be 1 due to the high precision of the calculated emissions. For emissions associated with wild animals, forests and natural grassland, the maximum value of the basic factor (1.5) has been adopted due to the high intrinsic uncertainty of these results.



Table AII. Uncertainty factors proposed by different methodologies.

	Value	Reference
<i>Livestock</i>		
Emission factors	± 30%	(EEA, 2007)
<i>Nitrogen fertilizer application</i>		
Amount of fertilizer and emission factor	± 50%	(EEA, 2007)
<i>Waste treatment</i>		
N-NH <sub>3</sub> emission per t CH <sub>4</sub>	2	(Sutton <i>et al.</i> , 2000)
N-NH <sub>3</sub> emission factor when sludge/compost is applied	3	(Doka, 2003)
<i>Humans and pets</i>		
g N-NH <sub>3</sub> /year emitted per person by breathing (Sutton <i>et al.</i> , 2000)	1.0-7.7	
g N-NH <sub>3</sub> /year emitted per person by sweating	2.1 - 74.9	(Sutton <i>et al.</i> , 2000)
g N-NH <sub>3</sub> / year emitted per minors of 3 years	2.8 - 63.2	(Sutton <i>et al.</i> , 2000)
g N-NH <sub>3</sub> /year per smoker	8.9 - 39.1	(Sutton <i>et al.</i> , 2000)
g N-NH <sub>3</sub> /year per cat	50 - 160	(Sutton <i>et al.</i> , 2000)
g N-NH <sub>3</sub> /year per dog	300 - 930	(Sutton <i>et al.</i> , 2000)
<i>Residential combustion of coal and biomass</i>		
Emission factor for coal	±150%	(EEA, 2007)
Emission factor for biomass	±150%	(EEA, 2007)
<i>Households sources</i>		
NH <sub>3</sub> contents in cleaning products	2.5 - 10%	(Sutton <i>et al.</i> , 2000)
% of NH <sub>3</sub> present in cleaning products emitted	10 - 50%	(Sutton <i>et al.</i> , 2000)
g NH <sub>3</sub> produced by fridges per house	2.28 - 13.1	(Sutton <i>et al.</i> , 2000)
g NH <sub>3</sub> produced by solvents per house	11.4 - 149.0	(Sutton <i>et al.</i> , 2000)
g NH <sub>3</sub> produced by fertilizers per house	3.65 - 24.1	(Sutton <i>et al.</i> , 2000)
<i>Fires</i>		
Global factor of uncertainty	3	(EEA, 2007)

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