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Quantification of eutrophic aerial compounds in Galicia (NW Spain): Part 2 – NO_x inventory

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RESUMEN

Este estudio es la segunda parte de una serie en la que se han calculado los inventarios de emisiones de NH_3 y NO_x en Galicia (NW España), una región con un riesgo importante de eutrofización. En este artículo se han estimado las emisiones de NO_x de las principales fuentes así como sus incertidumbres asociadas. Los resultados demuestran que las fuentes móviles e industriales producen el 90% de las emisiones, principalmente debido al transporte por carretera, las plantas térmicas y las de cogeneración. La quema de combustibles para usos residenciales, el uso de fertilizantes, la quema de residuos agrícolas, los suelos, los incendios y los rayos son otras fuentes de emisión, aunque mucho menores. En el caso de los incendios, en regiones especialmente afectadas como Galicia, hay que prestar atención a las variaciones en número y tipo de hectáreas quemadas de un año a otro, porque pueden alterar significativamente el total de las emisiones de NO_x. Las emisiones provocadas por vehículos y maquinaria agrícola, pesquera y forestal, y por el transporte ferroviario y marítimo son los principales focos de incertidumbre debido a la ausencia de datos específicos para Galicia como tipo, edad y potencia de las máquinas. Otros focos de incertidumbre importantes son las emisiones de suelos e incendios debido a la ausencia de una metodología de cálculo más específica.

ABSTRACT

This study is the second part of a series where NH_3 and NO_x emission inventories for Galicia (NW Spain), a region with a great risk of eutrophication, have been developed. The principal sources of NO_x emissions in Galicia and their associated uncertainty have been calculated in this paper. The results prove that industrial and mobile sources produce 90% of the emissions, principally due to road transport, power and cogeneration plants. Use of fertilizers or fuels for residential purposes, burning of agricultural waste, soils, fires and lightning are minor sources of emissions. In the case of fires, in regions specially affected like Galicia, attention must be paid to variations in number and type of burnt areas from one year to another, because it can significantly change total NO_x emissions. The emissions produced by agricultural, fishery and forest vehicles and machines as well as by maritime and railway traffic are the main focus of uncertainty due to the lack of specific data for Galicia like type, age and power of the machines. Other important focuses of uncertainties are the emissions from soils and fires owing to the absence of a more specific methodology.

Keywords: Eutrophication, inventory, NO_x, Galicia, uncertainty, air emissions.

1. Introduction

As described in detail in Part 1 (pages 141-161, this issue) of this series, eutrophication is a major risk for both soils and watercourses in Galicia (NW Spain), being the emissions of aerial NH_3 and NO_x among the principal responsible causes (Rodríguez and Macías, 2006). In spite of these increasing environmental problems and although emission inventories are worldwide under development and in constant improvement (Parra *et al*., 2006), in Galicia, complete inventories of emissions of NO_x and NH_3 are still lacking and only a partial approximation for industrial emissions is available (Casares *et al*., 2005). For this reason, Part 1 of this series developed an inventory of NH_3 emissions in Galicia, and the main objectives of this Part 2 are the estimation of NO_x emissions in Galicia as well as the uncertainties associated therewith.

2. Methods

2.1 Source selection

As presented in Part 1, the 30 major focuses of emissions were obtained by CORINAIR for 28 European countries in September 1995 (EEA, 2007), these data being selected as basic criterion. However, some considerations need to be made:

- Nitric acid production was excluded, as no factories are located in the region of study.
- Regarding harbor emissions, only emissions related to arrival and departure of ships were calculated. Emissions from tug boats and harbor machinery could not be estimated due to lack of data.
- Some sources, such as fires and use of fertilizers, were included as they are expected to be significant contributors in Galicia.
- In addition, the main natural sources such as emissions derived from soils and lightning were also considered.

2.2 Methodologies for the calculation of emissions

As in Part 1, the methodologies proposed in the EMEP/CORINAIR Guidebook (EEA, 2007) were followed to calculate most of the source emissions. The reference year for this inventory was also 2001, unless otherwise properly specified in the text. NO_x emissions are always reported in tonnes (t) of NO_2.

2.3 Uncertainties

Uncertainty values were determined for every calculation. The methodology implemented is the one used for the NH_3 inventory (Part 1 of the series) and described there in detail. The basic factors used in this study are presented in Table I.

A summary of the uncertainty factors used along the inventory is included in the Annex.

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

	Combustion	Process	Agriculture
Air pollutants			
NO_x	1.50	-	1.40
Resources			
Primary energy sources	1.05	1.05	1.05

3. Emission calculation

3.1 Industrial sources

Industry is one of the major sources of NO_x emissions, principally due to combustion processes, where NO_x formation is mainly produced by the conversion of chemically bound nitrogen in the fuel and by fixation of atmospheric nitrogen stemming from combustion air (EEA, 2007). Casares *et al.*. (2005) calculated industrial NO_x emissions in 2001 by surveys of the 370 main installations responsible for air pollution in Galicia, showing a total emission of 53,400 t NO_x (value range between 50,857 and 56,070). Power and cogeneration plants were the main sources, accounting for 56 and 28% of the emissions, respectively.

3.2 Mobile sources

3.2.1 Road transport

NO_x in road transport is caused by the oxidation of N_2 from air in the combustion chamber (EEA, 2005). In 2005, transport (excluding international aviation and maritime transport) contributed 56% of total NO_x in EU-15 (EEA, 2007), road transport being the main source of this share with a contribution of 75%.

In the same way as NH_3 emissions in Part 1, NO_x emissions have been calculated using the program COPERT 4.5 (http://lat.eng.auth.gr/copert/) which is recommended by the EMEP/CORINAIR guidebook (EEA, 2007). The required data have been obtained from numerous bibliographic sources (André *et al*., 1999; Bello *et al*., 2004; Camaleño, 2008; CMA, 2002; DGT, 2002; INE, 2008; MMA, 2007). The calculation of emissions is very similar to NH_3 evaluation, and a detailed description can be found in Part 1 of these series.

Due to the great number of vehicle types and emission factors (96 different classes and 480 emission factors), Table II only presents the aggregated results per passenger cars, motorbikes, mopeds, buses, light and heavy-duty vehicles (< and > 3.5 t, respectively).

Table II. Emissions produced by road transport (program COPERT 4.5).

Type of vehicle	t NO_x/year
Passenger cars	25,006
Motorbikes	92
Mopeds	12
Buses	7,111
Light-duty vehicles (<3.5 t)	4,457
Heavy-duty vehicles (> 3.5 t)	5,355
Total (t NO_x)	42,033 (35,027-50,439)

The program COPERT 4.5 only allows calculating emissions produced exclusively by road transport, and therefore, emission factors for tractors are not included. As in the case of NH_3 in Part 1, the number of tractors, the amount of annually consumed combustible and the NO_x emission factor (taking into account if the tractor is gasoline or diesel-driven) have been considered in order to calculate these emissions (Table III).

Table III. Emissions produced by tractors.

	A) Number of vehicles[a]	B) Fuel consumed (t/vehicle year)[b]	C) NO_x factor (kg NO_x/t fuel)[b]	D) NO_x (t) $D = A\times B\times C\times 10^{-3}$
Gasoline tractors	144	2.016	7.56	2.20
Diesel tractors	9,056	2.016	50.21	917
Total (t NO_X)		919 (464-1.821)		

[a](INE, 2008); [b](Nemecek *et al.*, 2004).

3.2.2 Railways

The simple methodology suggested by EMEP/CORINAIR guidance was used. This methodology considers the multiplication of the tonnes of diesel consumed, namely 87,722 t/year in Galicia according to Bello *et al.* (2004), by the specific NO_X emission factor for railways (39.6 kg of NO_X/t) (EEA, 2007). As a result, railway transport in Galicia emitted 3,474 t NO_X (1840-6557).

3.2.3 Maritime traffic

The simple EMEP/CORINAIR methodology was used again to estimate emissions associated with maritime traffic. It consists in the multiplication of consumed fuel, for both international and national maritime traffic, by the NO_X emission factor for maritime activity (72 kg NO_X/t fuel) (EEA, 2007). Bello *et al.* (2004) estimated 44,361 tonnes of consumed fuel for the international maritime transport considering a 17 knots/hour velocity to cover the 200 nautical miles under Spanish jurisdiction. The result was an emission of 3,194 (2,090-4,670) t NO_X/year.

Concerning national maritime traffic, it can be divided into interior (canals and rivers) and coastal transport. The former can be considered practically negligible in Galicia compared with the latter, and was therefore excluded from the analysis. In 2000, the consumed fuel associated with national transport was estimated to be 54,375 t/year (Bello *et al.*, 2004), which means an emission of 3,915 (2,561-5,724) t NO_X.

3.2.4 Air traffic

The detailed methodology applied by EMEP/CORINAIR guidance was used for the estimation of these emissions (EEA, 2007).

Aircraft operations were divided into two parts and associated emissions were calculated separately as they differ significantly (EEA, 2007):

- Landing/take-off (LTO) cycle, which includes all the activities (taxi-in and out, take-off, climb-out and approach-landing) that take place below an altitude of 1,000 m near the airport.
- Cruise, or the activities that take place at altitudes above 1,000 m. Cruise includes climb from the end of climb-out in the LTO cycle to cruise altitude, cruise and descent from cruise altitudes to the start of LTO operations of landing.
- The 86 different aircraft types that operated in 2001 in Galician airports (Bello *et al.*, 2004) were reclassified according to their characteristics in 29 generic classes. Each of these categories has their specific emission factor per LTO cycle and cruise (EEA, 2007). Therefore, depending on the type of airplane, the number of LTO cycles (Bello *et al.*, 2004) and the emission factor, the emissions associated with the landing and take-off were calculated (Table

IV). Regarding emissions associated with cruise activities, data from the principal routes that cover the Galician air space were used and an average distance per flight of 260 km (130 km for entering Galicia and 130 for leaving it) for all the planes with origin or destination in a Galician airport was estimated. Only emissions caused by aircraft landing or taking-off in Galicia were considered, as air traffic that crosses the region without stopping was assumed to be negligible.

Table IV. Emissions associated with air traffic.

Aircraft types	A) Number of LTO cycles[a]	B) Emission factor LTO (kg NO_x /cycle)[b]	C) Emission factor cruise (kg NO_x /260 km)[b]	D) NOx (t) $D= A\times(B+C)\times10^{-3}$
Airbus A310	19	23.2	33.8	1.08
Boeing 727-100	94	12.6	12.2	2.33
Boeing 727-200	3	12.6	12.2	0.07
Boeing 727-300	154	12.6	12.2	3.82
Boeing 737-200	2	8.3	11.1	0.04
Boeing 737-500	239	8.3	9.3	4.2
Boeing 737-400	155	8.3	9.3	2.73
Boeing 737-300	650	8.3	9.3	11.4
Boeing 737-700	335	8.3	9.3	5.9
Airbus A320	3,032	10.8	19.2	91.0
BAe 111	614	4.9	11	9.76
Boeing 747-100-300	20	55.9	80	2.72
Boeing 757	393	19.7	38	22.7
Boeing 767	1,120	26	30	62.7
McDonnell Douglas DC-9	1,374	7.3	11	25.1
McDonnell Douglas M81-88	9,172	12.3	21	305
McDonnell Douglas DC-10	22	41.7	62	2.3
McDonnell Douglas DC-8	2	7.3	10.6	0.04
Others[c]	1,354	2.94	1.9	6.55
Total (t NO_x)		560 (401-718)		

[a](Bello *et al*., 2004); [b](EEA, 2007); [c]Small aircrafts and helicopters.

3.2.5 Other mobile sources and machinery

In 2001, the annual consumption of diesel B for agricultural use (tractors and other machinery), fishery and forest machines was 7,531,000 GJ (Bello *et al*., 2004). The NO_X emission factor for the combustion of diesel in these sectors is 1.2 kg/GJ (IPCC, 1996). Therefore, total emission (subtracting the emissions associated with diesel tractors included already in subsection 3.2.1.) was 8,121 t NO_X (2,967-16,595).

3.3 Residential combustion

In 2001, a total of 2,721,550 GJ of energy for domestic purposes was produced from coal in Spain (López *et al*., 2005). An allocation of the Galician consumption was made on a population basis (data of Spanish and Galician population in 2001 from INE, 2008), the value obtained being 180,894 GJ. Using this figure together with the emission factor proposed by EEA (2007), the emission of NO_X associated with the use of coal for residential purposes was calculated (Table V). The same method was used for residential use of natural gas, butane, biomass, propane and diesel C (diesel specifically developed for heating), although in these cases the data referring to combustibles were obtained from Bello *et al*. (2004).

Table V. Emissions associated with residential combustion.

	A) Produced energy (GJ)	B) Emission factor ($g\ NO_x/GJ$)[c]	C) NO_x (t) $C = A \times B \times 10^{-6}$
Coal	180,894[a]	109.7	19,8
Natural gas	2,051,039[b]	57	117
Propane	2,538,684[b]	57	145
Butane	5,684,341[b]	57	324
Diesel C	9,416,869[b]	68	640
Biomass	3,138,000[b]	74.5	234
Total (t NO_x)		1,480 (491-2,868)	

[a](López *et al*., 2005) and (INE, 2008); [b](Bello *et al*., 2004); [c](EEA, 2007).

3.4 Fires

As in the case of NH_3 in Part 1, to compute the NO_x emissions produced by fires (Table VI), first the emitted carbon mass (M(C)) was calculated by equation (1) (Crutzen *et al*., 1979):

$$M(C) = 0.45 \times A \times B \times \alpha \times \beta \quad (1)$$

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^2), B the average biomass used as combustible referred to area (kg C/m^2), α the fraction of biomass in the surface related to total biomass of B, and β the efficiency of burnt biomass in the surface. Once M(C) is obtained, the emitted NO_x can be calculated using the factor of 8 g NO_x /kg of emitted C (Andreae, 1991).

Table VI. Fires emissions.

Vegetation	A)[a]	B) α[b]	C) β[b]	D) B[b]	E) M(C) (t C)	F) Factor (kg NO_x/t C)[b]	G) NO_x (t) $G = F \times E \times 10^{-3}$
Bush	14,217	0.64	0.5	7.5	153,542	8	1,228
Trees	4,014	0.75	0.2	35	94,837	8	759
Grassland	122.5	0.36	0.5	2	198.4	8	1.59
Total (t NO_x)					1,989 (994-5,966)		

[a]Burnt hectares (MMA, 2008); [b](Crutzen *et al*., 1979).

3.5 Nitrogen fertilizer application

After nitrogen is applied to the soils, nitric oxide (NO) may be released during nitrification and denitrification. Estimates of NO emissions are very uncertain, but soils (including natural emissions) may contribute 4-8% of total NO_x emissions in Europe (EEA, 2007).

To calculate this source, both the amount of N in fertilizers used in 2001 (62,965 t; CMR, 2003a) and the emission factor of 0.007 t N-NO/t N in fertilizers (EEA, 2007) were considered, obtaining a value of NO_x (expressed as NO_2) emissions of 1,448 t (range 881-2,358).

3.6 Burning of agricultural waste

Similar to that observed in forest fires, burning of agricultural waste produces NO_x emissions, being a source of special importance in developing countries (IPCC, 1996). For this source, IPPC methodologies were used, which allow to calculate NO_x emissions from N emissions originated by burning of agricultural waste (IPCC, 1996). From the N_2O emission data associated with these burnings in Galicia in 2000 (Bello *et al.*, 2004) and using the IPCC emission factors (IPCC, 1996), N emissions were calculated first and, on the basis of these figures, NO_x emissions were obtained (Table VII).

Table VII. Emissions associated with burning of agricultural waste.

A) N_2O emissions during burning (t)[a]	B) N_2O emission factors (t N_2O/t N emitted)[b]	C) N emissions (t) C = A/B	D) NO_x emission factor (t NO_x/t N emitted)[b]	E) NO_x emissions (t) E = C × D
24	0.011	2,181.81	0.3975	867 (336 - 2,317)

[a](Bello *et al.*, 2004); [b](IPCC, 1996).

3.7 Lightning

Lightning and corona discharge during thunderstorm events cause atmospheric chemical reactions that take place at high voltages and temperatures. These reactions produce NO_x in the atmosphere (Sisterson and Liaw, 1990).

The majority of lightning falls on land while a marginal percentage hits the sea (Christian *et al.*, 2003). Taking into account that 0.33 lightning descends per km^2 and year (personal communication by Dr. Luis Rivas Soriano, Departmento de Física General y de la Atmósfera, Universidad de Salamanca, Spain), February 15, 2007; ljrs@usal.es) and that the Galician surface is 29,574 km^2 (INE, 2008), it was estimated that 9,759 lightning strokes occurred per year. However, and as EMEP inventories do, only the emissions of lightning produced within a range of 1 km above soil level were considered (20% of the total). EEA (2007) establishes an emission factor of 2.75 kg NO_x/lightning. Altogether, this results in a NO_x emission of 5.37 t (range 1.79-16.1).

3.8 Soil emissions

NO_x emissions, mainly in the form of NO, are produced by microorganisms in soil (EEA, 2007). Natural ecosystems tend to have modest fluxes, but soils that are nitrogen-enriched, especially agricultural regions, may have NO_x fluxes approaching those of anthropogenic sources (Williams *et al.*, 1992).

For this source, the detailed methodology of EMEP/CORINAIR guidance is used (EEA, 2007). The N-NO emission flux in ng/m^{2}s (F_{NO}) is calculated by the following equation (Williams *et al.*, 1992):

$$F_{NO} = A \exp(0.071 \times T_s) \tag{2}$$

where A is an experimental constant for grazing lands, forests and moist zones and T_s is the soil temperature in °C. These parameters were calculated by Novak and Pierce (1993) for different values of environmental temperature (T_a) (Table VIII).

Based on the data of the Climatologic Yearbook of Galicia for 2001 (CMA, 2002), T_a for Galicia was found to be 11.86°C. The data of N-NO flux calculated in equation (2) are multiplied by the area assigned for each use of the soil and thus NO_x emissions (expressed as NO_2) are obtained (Table IX).

Table VIII. Values of A and T_s (Novak and Pierce, 1993).

Type of soil	A	$T_s = f(T_a)$
Grassland	0.9	$T_s = 0.67\ T_a + 8.8$
Forest	0.07	$T_s = 0.84\ T_a + 3.6$
Moist zones	0.004	$T_s = 0.92\ T_a + 4.4$

Table IX. Emissions associated with soils.

	A) Area (m^2)	B) A factor	C) T_s	D) F_{NO} (ng N-NO/sm^2)	E) NO_x (t) $E = A \times D \times 3153600 (46/14) 10^{-15}$
Forest	18,855,060,000[a]	0.9	16.75	2.955	5,774
Grassland	4,183,460,000[a]	0.07	13.56	0.183	79.5
Moist zones	706,773,600[b]	0.004	15.31	0.013	0.87
Total (t NO_x)			5,854 (1,951-17,563)		

[a](CMR, 2003b); [b](CMA, 2008).

4. Summary of results and discussion

4.1 Inventory of NO_x emissions

The main result of this study is the inventory of NO_x emissions for the region of Galicia (Table X). To the best of our knowledge, this is the first complete inventory for this specific region and therefore, no full comparison is possible with previous reports.

Table X. Summary of emissions.

Sources	New inventory	EMEP Galicia
Industry	53,400 (50,857-56,070)	55,051[a]
Combustible burning for non-industrial purposes	1,480 (491-2,868)	
Mobile sources	62,215 (45,350-86,526)	51,009[b]
Fertilizer use	1,448 (881-2,258)	
Agricultural waste burning	867 (336-2,317)	1487[c]
Fires	1,989 (994-5,966)	
Lightning	5.37 (1.79-16.1)	0[d]
Soils	5,854 (1,951-17,563)	
Total	127,258 (100,862-173,684)	107,547

Addition of emissions produced by the following sectors (SNAP-1997 codes): Combustion and energy and transformation industries, non-industrial combustion plants, combustion in manufacturing industry, production processes and waste treatment and disposal ([a]), road transport and other mobile sources and machinery ([b]), agricultural ([c]) and other sources and sinks ([d]) (UNECE, 1997).

However, some particular comments can be made with reference to other reported data, such as the values obtained by the EMEP program (EMEP, 2008). The EMEP researchers divide Europe in 50 × 50 km^2 grids and publish yearly values of emissions of NO_X (among others pollutants) per grid based on officially reported data by countries and expert criteria.

For frontier grids between countries, the percentage of the grid that belongs to each country is known, but these percentages are not available for internal regions of a country. Therefore, the percentage of each frontier grid that belongs to Galicia was calculated here. The total and source-based NO_X emissions and their comparison with the EMEP data for Galicia (year 2001) are summarized in Table X.

The major origins of NO_X emissions are mobile (road transport, railways, air traffic, etc.) and industrial sources, contributing both 90% of the total. Regarding the former, road transport causes 33% of all emissions, principally by passenger cars, followed by trucks and buses. Emissions associated with agricultural, fishery and forest machineries as well as maritime transport are also important (8,121 and 7,109 t, respectively), representing in both cases around 6% of the total.

To make these figures comparable to EMEP results, the following clarification is needed. When reporting their emissions to EMEP, countries are requested to report their national shipping emissions by grid cell, but international maritime data are separately reported and not allocated to member states. For this reason, the emission of 3,194 t NO_X associated with international maritime transport was subtracted in our inventory and the resulting comparison shows that the values are quite similar (59,021 versus 51,009 t).

Concerning industrial sources, power and cogeneration plants stand for 24 and 12%, respectively, of the total emissions of NO_X in Galicia. The sum of NO_X emissions from industrial activities and from heating for commercial and residential purposes given in our inventory (54,880 t NO_X) was almost the same as the equivalent emissions calculated by the EMEP (55,051 t NO_X).

After considering those two main streams, the remaining sources represent only 10% of the emissions. Agricultural sources (use of N-fertilizers and burning of agricultural wastes) account for 2,315 t NO_X (around 2% of total emissions), which is a value slightly higher than that reported by the EMEP, namely 1,487 t.

The EMEP program (EMEP, 2008) follows the classification of emission-generating activities listed in the current version of the Selected Nomenclature for Air Pollution (SNAP97) (EEA, 2007). This classification sets up 11 major sectors. Sector 11 (other sources and sinks) includes both natural sources (volcanoes, lightning, etc.) and sources related to human activity (forest and grassland conversion, abandonment of agricultural land, etc.). These emissions should also be reported, but only emissions from sources included in SNAP97 sectors 1 to 10 can be considered by the emission reduction protocols. This could be the reason why emissions produced by sources stated in sector 11 for the Galician selected grids are zero. In our inventory, the emission equivalents to the ones of sector 11 are 7,848 t NO_X, with soils being the major source. If we disregard emissions of sector 11 as well as emissions from international maritime transport, we will obtain a new figure for our inventory (116,216 t NO_X) that is closer to the figure of the EMEP (107,547 t NO_X).

4.2 Analysis of uncertainties

In order to estimate the uncertainty, where not established by the calculation methodology, the method developed by Frischknecht and Jungbluth (2004) was used (see section 2.3). As in the NH_3 inventory, the geographical and temporal factors (U_1 and U_2 in Table AI in the Annex) have low values because the used data are mostly elaborated for Galicia and reported for the reference year (2001). With regard to the reliability factor (U_3), all data are on level 2 or 4 (from a total of 5) because they are verified data partly based on assumptions or qualified estimation (according to expert criteria). For example, the amount of kg fuel consumed/tractor and year has been classified as level 4 because it is based on the criteria of experts, in this particular case, Nemecek *et al*. (2004). The basic factor (U_b) has been developed in accordance with the values proposed by Frischknecht and Jungbluth (2004) and for the categories established by these authors (energy and resources demand, infrastructures, transport, waste treatment and emissions of pollutants to air, soil and water), adopting the value of 1 in the remaining cases. Exceptionally, this basic factor has been considered 1 for the NO_x emissions of road transport and industry (instead of 1.5 as proposed by Frischknecht and Jungbluth, 2004) due to the high precision of the calculated emissions. As in Part 1, the uncertainty values proposed by the methodologies are shown in Table AII in the Annex.

The estimation of emissions produced by agricultural, fishery and forest vehicles and machines involves a great uncertainty. The application of an unspecific factor was the only option due to the lack of specific data for type, age and power of the machines in Galicia. Equally, the lack of specific data is responsible for the high uncertainty associated to maritime and railway traffic.

The other great focus of uncertainty is related to natural emissions from soils and fires. In both cases, the most detailed methodologies were used (EEA, 2007); however, uncertainty is associated with the methodologies as such, which require more specific future development of the calculations.

4.3 Sensitivity analysis of the importance of fires as a source

From all the sources analyzed in Galicia, only fires are likely to suffer high variability from year to year (Fig. 1). In fact, it can be argued that emissions from other sources (such as mobile, industrial, etc.) may also change within a short time, but they are controlled by regulation and those changes will be gradual. However, anthropogenic reasons (economic, number of fire-fighting squads, etc.) and climatic characteristics (temperature, wind and humidity) can severely affect the number of burnt hectares from year to year in a region like Galicia and, therefore, the associated annual emissions of NO_x.

The period of study was then expanded and calculations for several years were carried out (Fig. 1). The average emissions for the period 1997-2004 were 3,195±1,457 t NO_x, with the emission for 2001 (the year of reference for the whole inventory) being one of the lowest values for that period. However, the annual variation within those years seems not to have a great influence when compared with the absolute values of emission. Moving towards the years 2005 and 2006, when a high incidence of fires occurred, the level of NO_x emissions produced was 7,195 and 13,434 t NO_x, respectively. This can be really important when compared with other sources included in the global inventory (considering that the rest of the sources will remain in the same order of magnitude since 2001). Special attention should therefore be paid to the variability of fires when choosing the year of reference for the inventories as emissions can suffer a high variability.

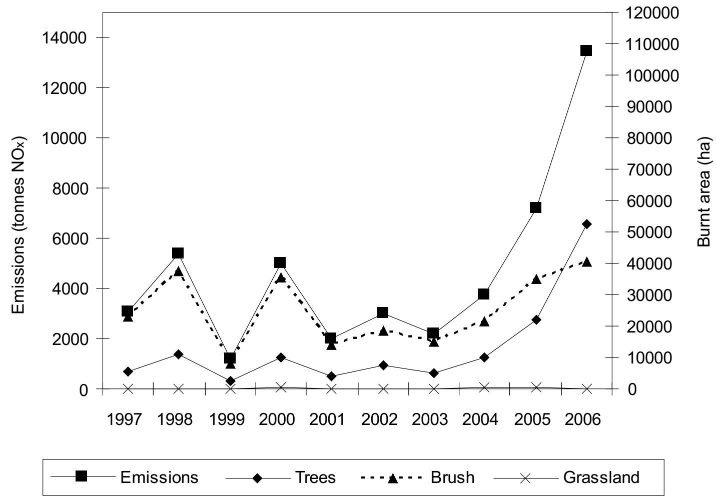

Fig. 1. NO_x emissions associated with fires (left axis) and areas of trees, brush and grassland burnt by fires (right axes) for the period 1997-2006 (MMA, 2008).

5. Conclusions

The main aim of this study was the calculation of a complete regional NO_X inventory for Galicia (NW Spain) together with the associated uncertainties. Although it was applied to a specific area, the procedure described in this paper can serve as a useful basis for the estimation of NO_X inventories and their uncertainties in other regions.

The principal results concerning the values obtained can be summarized as follows:

- Industrial activities and mobile sources are the major sources of NO_X emissions, accounting for 90% of the total inventory. In the case of industry, power and cogeneration plants are the main focus, with a share of 24 and 12% of the total emissions, respectively. Road transport stands for 33% of the emissions, principally due to passenger cars, followed by trucks and buses.
- Agricultural sources (use of fertilizers and burning of agricultural waste), soil emissions and fires represent minor sources of emissions (2, 1.6 and 5%, respectively).
- In regions that have especially been affected by fires, as in the case of Galicia, attention has to be paid to the variability over the years when defining the year of reference as this source can significantly alter the total NO_X emissions.
- The emissions produced by agricultural, fishery and forest vehicles and machines as well as by maritime and railway traffic are the main focus of uncertainty due to the lack of specific data for Galicia like type, age and power of the machines. In addition, the absence of a more specific methodology is also an important source of uncertainty for the emissions originated by soils and fires.

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Annex

Uncertainty factors used along the inventory

Table AI. Uncertainty factors calculated by the authors.

	A) Reliability (U_1)	B) Temperature (U_2)	C) Geographical (U_3)	D) Basic (U_b)	E) Results $E = \exp\sqrt{[\ln(A)]^2 + [\ln(B)]^2 + [\ln(C)]^2 + [\ln(D)]^2}$
Industrial sources					
NO_x emission	1.05	1	1	1	1.05
Mobile sources					
NO_x emission road vehicles	1.2	1	1	1	1.2
No. of tractors	1.05	1	1	1	1.05
kg cons. fuel/tractor year	1.2	1	1.02	1.05	1.21
NO_x emission factor (tractors)	1.2	1	1.02	1.5	1.56
t consumed diesel/railw. year	1.2	1	1	1.05	1.21
NO_x factor (railways)	1.2	1.03	1.02	1.5	1.56
t diesel consumption (maritime traffic)	1.2	1	1	1.05	1.21
t diesel consumption (other mobile sources)	1.2	1	1	1.05	1.21
NO_x emission (other mobile sources)	1.2	1.03	1.10	1.5	1.58
Residential combustion					
Galician population	1.05	1	1	1	1.05
Spanish population	1.05	1	1	1	1.05
Coal consumption	1.2	1	1.01	1.05	1.21
Biomass, propane… consumption	1.2	1	1	1.05	1.21
Burning of agricultural waste					
N_2O emission	1.2	1.03	1.10	1.5	1.58
Nitrogen fertilizers application					
N-NO factor (fertilizer)	1.2	1.03	1.02	1.4	1.47

[a] Default value = 1 for items not included in the categories defined by Frischknecht and Jungbluth (2004) (energy and resources demand, infrastructure, transport, waste treatment and emissions of pollutants to air, soil and water). Exceptionally, this basic factor has been considered to be 1 for the emissions of road transport and industry due to the high precision of the calculated emissions.

Table AII. Uncertainty factors proposed by different methodologies.

	Value	Reference
Mobile sources		
Emission factor (maritime traffic)	57-87	(EEA, 2007)
NO_x emission air traffic (LTO cycles)	± 10%	(EEA, 2007)
NO_x emission air traffic (cruise)	± 40%	(EEA, 2007)
Residential combustion		
Emission factor for all combustibles	± 60%	(EEA, 2007)
Fires		
NO_x emissions	3	(EEA, 2007)
Burning of agricultural waste		
N_2O emission factor	0.008-0.014	(IPCC, 1996)
NO_x emission factor	0.308-0.486	(IPCC, 1996)
Nitrogen fertilizers application		
t fertilizers consumed	± 10%	(EEA, 2007)
Lightning		
NO_x emissions	3	(EEA, 2007)
Soil emissions		
NO_x emissions	3	(EEA, 2007)

References

André M., U. Hammarström and I. Reynaud, 1999. Driving statistics for the assessment of pollutant emission from road transport. INRETS Report LTE 9906. Paris, France, 194 pp.

Andreae M. O., 1991. Biomass burning. Its history, use, and distribution and its impact on environmental quality and global climate. In: *Global biomass burning: Atmospheric, climatic, and biospheric implications* (J. S. Levine, Ed.). MIT Press, Massachusetts, USA, 3-21.

Bello P., G. Grueiro, F. Macías, D. Rodríguez, R. Rodríguez, L. Rodríguez and Y. Romero, 2004. Inventario de emisións de gases de efecto invernadoiro en Galicia. Colección Técnica de Medio Ambiente. Consellería de Medio Ambiente. Santiago de Compostela, Spain, 168 pp.

Camaleño, M., 2008. Inventario de emisiones procedentes de los sectores primario y terciario en Castilla y León, 2002-2005. Propuestas de mejora de la calidad ambiental. Doctoral Thesis. Universidad de Valladolid, Valladolid, Spain, 1074 pp.

Casares J. J., R. Rodríguez, P. Maceira, J. A. Souto, S. Ramos, M. A. Costoya, A. Sáez and J. M. Vellón, 2005. Inventario, análisis y proyección de las emisiones atmosféricas industriales en Galicia. Universidade de Santiago de Compostela. Servizo de Publicaciones e Intercambio Científico, Santiago de Compostela, Spain, 281pp.

Christian H. J., R. J. Blakeslee, D. J. Boccippio, W.L. Boeck, D. E. Buechler, K. T. Driscoll, S. J. Goodman, J. M. Hall, W. J. Koshak, D. M. Mach and M. F. Stewart, 2003. Global frequency and distribution of lightning as observed from space by the Optical Transient Detector. *J Geophys. Res*. **107** (D1), 4005.

CMA, 2002. Anuario climatolóxico de Galicia. Centro de Desenvolvemento Sostible. Consellería de Medio Ambiente, Santiago de Compostela, Spain, 108 pp. CMA, 2008. http://medioambiente.xunta.es/. Consellería de Medio Ambiente. July 2008.

CMR, 2003a. *Anuario de estatística agraria 2001*. Consellería do Medio Rural, Santiago de Compostela, Spain, 260 pp.

CMR, 2003b. Macromagnitudes agrarias 2001. Consellería do Medio Rural, Santiago de Compostela, Spain, 153 pp.

Crutzen P. J., L. E. Heidt, J. P. Krasnec, W. H. Pollock and W. Seiler, 1979. Biomass burning as a source of atmospheric gases CO, H_2, N_2O, NO, CH_3C and COS. *Nature* **282**, 253-256.

DGT, 2002. Anuario general estadístico, 2001. Dirección General de Tráfico, Madrid, Spain, 48 pp.

EEA, 2005. ETC-ACC Air emissions spreadsheet for indicators 2004. Environmental European Agency, Copenhagen, Denmark, 33 pp.

EEA, 2007. EMEP/CORINAIR Emission inventory guidebook - 3rd edition. 2007 Update. Technical report No. 30. Environmental European Agency, Copenhague, Denmark, 2734 pp.

EMEP, 2008. www.emep.int. July 2008.

Frischknecht R. and N. Jungbluth, 2004. Overview and methodology. Ecoinvent Database v1.1. Swiss Centre for Life Cycle Inventories, Dübendorf, Switzerland, 75 pp.

INE, 2008. www.ine.es. Instituto Nacional de Estadística. July 2008.

IPCC, 1996. Revised 1996 IPCC Guidelines for national greenhouse gas inventories: Reference manual. Workbook (Volumen 2). UK Meteorological Office, Bracknell, United Kingdom, 366 pp.

López J. M., L. J. Sánchez and V. Meneu, 2005. Impactos sobre el sector energético. In: *Evaluación preliminar de los impactos en España por efecto del cambio climático* (J. M. Moreno, Ed.). Madrid, España, 617-653.

MMA, 2007. Inventario de emisiones de gases de efecto invernadero de España. Años 1990-2005. Ministerio de Medio Ambiente, Madrid, Spain 396 pp.

MMA, 2008. http://www.incendiosforestales.org/estadisticas.asp. Ministerio de Medio Ambiente. July 2008.

Nemecek T., A. Heil, O. Huguenin, S. Meier, S. Erzinger, S. Blaser, D. Dux and A. Zimmermann, 2004. Life cycle inventories of agricultural production systems. Ecoinvent Database v. 1.1. Report No. 15. The Swiss Centre for Life Cycle Inventories, Zurich, Switzerland, 289 pp.

Novak J. and T. Pierce, 1993. Natural emissions of oxidant precursors. *Water Air Soil Poll.* **67**, 57-77.

Parra R., P. Jiménez and J. M. Baldaseno, 2006. Development of the high spatial resolution EMI-CAT2000 emission model for air pollutants from the north-eastern Iberian Peninsula (Catalonia, Spain). *Environ. Pollut.* **140**, 200-219.

Rodríguez L. and F. Macías, 2006. Eutrophication trends in forest soils in Galicia (NW Spain) caused by the atmospheric deposition of nitrogen compounds. *Chemosphere* **63**, 1598-1609.

Sisterson D. L. and Y. P. Liaw, 1990. An evaluation of lightning and corona discharge on thunderstorm air and precipitation chemistry. *J. Atmos. Chem.* **10**, 83-96.

UNECE, 1997. Procedures for estimating and reporting emission data under the convention on long-range transboundary air pollution. Steering Body to the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP). Twenty-first session, Geneva, Switzerland, September 1997.

Williams E., A. Guenther and F. Fehsenfeld, 1992. An inventory of nitric oxide emissions from soils in the United States. *J. Geophys. Res.* **97**, 7511-7519.