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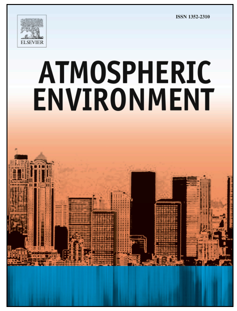

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AMMONIA EMISSIONS FROM THE AGRICULTURE SECTOR IN ARGENTINA; 2000–2012

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Abstract

Agriculture is one of the key economic sectors in Argentina and, in the last decades, the increase in prices and competitiveness of some grains has imposed important changes. In this process, crop cultivation occupied significant extensions of land areas previously dedicated to livestock farming, which in turn have experienced intensification in terms of production through an increasing share of feedlot systems. The agriculture sector is the main NH_3 emitter in Argentina, however no inventory developed locally has been thus far available. We estimated the time series 2000–2012 of NH_3 emissions, both at national and spatially disaggregated levels. National NH_3 emissions in 2012 amounted to 0.31±0.08 Tg, with the use of mineral fertilizers accounting for 43.0%, manure in pasture 32.5%, manure management 23.0% and agricultural waste burning 1.5%. Urea use was the major source of NH_3 emissions and its application on wheat and corn crops dominated the trend. Emissions from open biomass burning were estimated but not included in the national totals because of the difficulties in differentiating between agricultural (i.e., prescribed burning of savannas) and non-agricultural emission sources. Compared to this work, NH_3 emissions reported by EDGAR were 83% higher than our estimates. The time series of spatially distributed NH_3 emission estimates clearly showed the effect of the expansion of cropland, the displacement of planted areas of N-fertilizes crops by competing soybean cultivation and the relocation and intensification of beef cattle production. This new inventory constitutes a tool for policies concerning the impact of agricultural activities on air quality and contributes with more accurate and updated information useful for atmospheric chemical transport modeling. The accuracy and applicability of the inventory may be improved by local studies aimed at refining the spatial disaggregation by focusing in specific areas of fertilizer application, reflecting seasonal and monthly patterns in agricultural practices and climate conditions and addressing likely changes in diets, productivity and excretion rates over time.

Keywords

Ammonia emission inventory

Manure related activities

Nitrogen fertilizer use

Changes in agro-technology

South America

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1. Introduction

The growing concern about the global presence of ammonia (NH_3) in the atmosphere has led to introducing policies to reduce its emissions and proposing air quality standards in certain developed countries (Reis et al., 2012). European countries regularly report NH_3 emission estimates and have committed to achieve national emission ceilings according to a stipulated path (EEA, 2017; NEC Directive, 2016). In the United States there are some emissions reporting requirements (CERCLA, 2016; EPCRA, 2016) and the U.S. Environment Protection Agency has recently petitioned to include NH_3 in the air quality standards (AAQTF, 2014). For developing countries, an increasing number of studies on NH_3 emissions have been published in the last five years, particularly in China (Qiu et al., 2014; Xu et al., 2015; Yang et al., 2016; Zhou et al., 2015). Nevertheless, for South America, the only information available on NH_3 emissions is that reported in global databases (ECCAD, 2016). A recent study on short-lived climate pollutants in Latin America and the Caribbean, of which only the summary for decision makers is thus far available, estimated emissions of ammonia using the GAINS model and local information collected at country level (UNEP and CCAC, 2016).

Although ammonia is a short-lived species, it is the most abundant basic gas in the atmosphere that plays a key role in secondary aerosols formation, as it reacts with sulfuric acid and nitric acid to form ammonium sulfate and ammonium nitrate aerosols (Luo et al., 2015). These aerosols are constituents of fine particulate matter, which increases the morbidity and mortality levels of the population (Franck et al., 2011), and modifies the cloud condensation nuclei number thus changing the atmosphere radiative properties and climate (Forster et al., 2007). Agriculture activities, particularly livestock production and fertilizer application, constitute the main emission sources of NH_3 (Beusen et al., 2008). Taking into account that Argentina and Brazil belong to the top six agricultural exporters of the world (European Commission, 2013), the estimation of NH_3 emissions and their fate in the atmosphere is relevant for South America since it has been estimated that both countries contribute with about 7% of the NH_3 emissions worldwide (EDGAR, 2011). To the best of our knowledge this is the first study which estimates spatially disaggregated NH_3 emissions from agricultural activities in the entire territory of Argentina using activity data reported with high resolution by local information sources. It is worth noticing that previous work on NH_3 emissions from agricultural sources in the region are experimental studies focused on emission factors relative to the use of nitrogen (N) fertilizers and the corresponding NH_3 volatilization from soils (de Morais et al., 2013; Martins et al., 2015).

Argentina has a continental surface of 2,780,400 km^2 distributed in 24 provinces subdivided into a total of 512 districts. In agro-economic terms, the territory is usually divided into five regions with similar climatic and environmental conditions (see Figure 1): Northeast (NE), Northwest (NW), Semiarid (SA), Patagonia (Pat) and the Pampa Region, which due to its productive complexity can be divided into four sub-regions (Rearte, 2007): North (P-N); Southeast (P-SE), Southwest (P-SW) and West (P-W). Until the mid-twentieth century agricultural activities had concentrated in the fertile plains of the Pampa Region where the climate is temperate and humid with no dry season. Since 1960 agriculture had expanded to less fertile zones such as the NE and NW regions favored in part by a remarkable increase in precipitation over most subtropical Argentina (Barros et al., 2014). In conjunction with climate, planted cropland area has expanded under the influence of global and local economic conditions that favored the production of crops over livestock (Grau et al., 2005) and technological changes such as the introduction of new soybean genotypes which favored cultivation of this crop in lands previously dedicated to other crops (Viglizzo et al., 2011a). During this process, more than 15 million hectares historically devoted to livestock farming have been switched to crop cultivation (Rearte and Pordomingo, 2014; Viglizzo et al., 2011b). Soybean in particular has gained great importance because of its steady growth and the clear evolution towards its predominance over the rest of the crops, a process locally known as “soyzation” of the territory, which started by the end of 1990 and continued growing since then.

Unlike the exponential growth in the production of cash crops, the production of beef cattle, the main livestock in the country, has experienced fluctuations in its stock without a clear trend. Beef cattle farming, which was traditionally based on pastoral production systems, has evolved over last decades towards (*i*) a geographical relocation of the cattle in lower-performance agricultural areas, and (*ii*) an intensification of beef cattle activity, through feeding modifications and the increased implementation of feedlot systems (Lence, 2010; Rearte and Pordomingo, 2014). Dairy cattle stock has also decreased while other livestock of lesser importance in the country, such as poultry or swine, has gained importance (SENASA, 2017).

The changes in climate, economic conditions and agro-technology mentioned before are expected to influence the magnitude and the distribution of NH_3 emissions in the country. More specifically, we hypothesized that the expansion of soybean, which have influenced both, the planted areas of the traditional crops of the country, such as wheat, corn and sunflower and the dynamics of livestock farming, has played a key role in the spatial and temporal pattern of NH_3 emissions. To contribute with update information aimed at addressing these issues we developed an inventory of NH_3 emissions from Argentina's agricultural sector for the 2000–2012 period at national and district levels. The manuscript is organized as follows: section 2 discusses the methodology and data used to estimate the emissions, results are presented and discussed in section 3 while concluding remarks are presented in section 4.

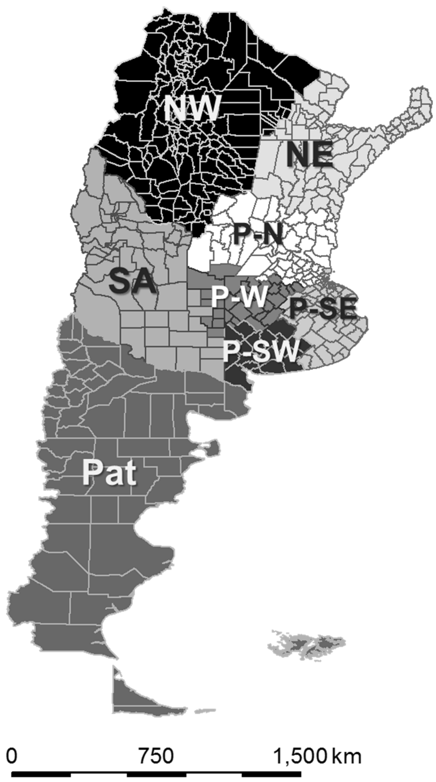

Figure 1: Agro-economic regions (large shaded areas) and districts (delimited by thin lines) of Argentina.

2. Methods and data

The general methodology applied, based on the European Monitoring and Evaluation Program (EMEP) approach (EMEP, 2013; Hutchings et al., 2013). The emission factors used are those suggested by the EMEP according to the level of detail adopted, which corresponds to Tier 2 in almost all cases.

The Third National Communication (TNC) of Argentina to the United Nations Convention on Climate Change (UNFCCC) constitutes the main basis for the AD. Additional data from other information sources were necessary (*i*) to complete gaps in the time series 2000–2012, (*ii*) to estimate emissions from poultry and swine manure, and (*iii*) for the spatial disaggregation by districts for the entire time series.

A description of the methodology is presented in sections 2.1–2.3. In addition, a summary of the data sources and the level of detail used for the estimation of the inventory (Table A.1), together with the activity data corresponding to each source (Table A.2), are presented as Supporting Material.

2.1. Animal husbandry and manure management

Simple and complex forms of N are present in animal waste derived from N-rich protein in feed that has not been completely converted into animal products. The ammonium ions (NH_4^+–N) are the main NH_3 source and the fraction of the N-compounds that decompose readily in these ions is denominated total ammoniacal-N (TAN). The emission sources of NH_3 arising from animal excreta are *(i)* manure deposited in buildings, in yards and during animal grazing, *(ii)* manure storage and *(iii)* land application of manure, where the term 'manure' includes both dung and urine. We used the tier 2 technology-specific approach of the unified methodology reported by EMEP (2013) for estimating NH_3 emissions from all types of livestock. This tier 2 approach is composed of 15 steps based on a mass balance, which considers the pathways for emission of N-compounds, by which emissions from manure management systems (E_{MMS}) and excreta deposited on pasture ($E_{grazing}$) are estimated (Eq. (1)).

(1) $E_{livestock} = E_{MMS} + E_{grazing} = (E_{MMS\_yard} + E_{MMS\_building}) + E_{grazing}$

In general, emissions from MMS occurred from manure managed in buildings housing livestock ($E_{MMS\_building}$) and outdoor yard areas (E_{MMS_yard}). These two components of E_{MMS} have three main contributors arising from (Eq. (2)): NH_3–N losses from the livestock building and yards, (E_{losses}), storage ($E_{storage}$) and land spreading of manure (E_{applic}).

(2) $E_{MMS\_i} = E_{losses\_i} + E_{storage\_i} + E_{applic\_i}$, with *i*=building, yard

Equations 1 and 2 summarize the main aspects of the 15-step tier 2 approach, for further details the reader is referred to EMEP (2013).

2.1.1.Livestock classes

To estimate $E_{livestock}$, livestock was organized in classes exhibiting similar characteristics in regard to feeding, excretion and weight. In this study, six classes were considered namely, beef cattle breeding, beef cattle fattening, dairy cattle, poultry, swine and other livestock. They were divided into subclasses as follows: beef cattle breeding (8 subclasses); beef cattle fattening (51), dairy cattle (2), poultry (2), swine (2) and other livestock (6), amounting to 71 subclasses, which are summarized below.

Beef cattle breeding, which includes cows for reproduction and male and female calves from birth to weaning, was disaggregated into eight sub-classes according to the different diets in the main agro-economic regions of the country (Vázquez Amabile et al., 2015).

Beef cattle fattening, which includes animals until they reach feat weight, was subdivided into 51 sub-classes. Fattening is carried out through three modalities: (*i*) sending animals to pastures without nutrition supplements, (*ii*) sending animals to pastures with nutrition supplements, and (*iii*) in feedlot systems, which imply intensive fattening through the incorporation of products formulated in the diets (rich in nitrogen) and reduced animal mobility through confinement in corrals. Sub-classes were defined regarding fattening modality, diet and fraction of manure that is managed, if any (Vázquez Amabile et al., 2015).

Dairy cattle were disaggregated into two sub-classes: (*i*) dairy cows and (*ii*) other dairy cattle composed of animals for womb reposition and reproducers. Poultry was composed of two sub-classes: laying hens and broilers. Swine was also organized in two sub-classes: (*i*) sows and piglets, and (*ii*) rest of pig livestock. Within other livestock the following sub-classes have been included: sheep, buffalos, goats, camelids, horses and asses/mules.

2.1.2. Main variables

The calculation of emissions from animal excreta for the time series 2000–2012 involved the following variables:

- Annual average population (*AAP*) or the average number of animals of a particular sub-class in a specific year. *AAP* is a 71x13 matrix with rows for animal sub-classes and columns for years. Values were taken from the information compiled by Vázquez Amabile et al (2015). As $AAP \in Z^{71x13}$ contained gaps for certain sub-classes and years, we filled out this matrix using different strategies that are discussed below.
- Total annual excretion of N ($N_{ex} \in R^{71}$). The values in N_{ex} reflect the different diets and excretion rates for the different animal sub-classes; while as only one single value was adopted for each sub-class for the entire time period 2000–2012, it does not reflect N_{ex} sub-class temporal variability. However, there is a temporal variability in the average N_{ex} of each class composed of more than one sub-class, through the variation of the *AAP* in each one. Country-specific N_{ex} rates for beef cattle were adopted from Vázquez Amabile et al (2015) while those for the other 12 sub-classes were calculated using excreted nitrogen rates reported by IPCC (2006) taken into account animal weights representative of local conditions. The adoption of single values in each subclass for the mentioned parameters over the time series implies that no changes in diets, productivity and excretion rates were taken into account. Future studies may attempt to understand the potential influence of these likely changes on NH_3 emissions over time.
- The period of the year that the animals spent in buildings ($x_{building}$), on yards (x_{yard}) and/or during grazing ($x_{grazing}$), these three periods always total 1.0. For all animals in beef cattle breeding, other dairy cattle and other livestock, $x_{grazing}$=1.0, indicative that these animals spent the entire year on land ($x_{building}$=x_{yard}=0.0). Beef cattle fattening in Argentina is farmed either on pasture or feedlots, which normally have soil floor with excreta accumulating on the ground. Manure from feedlot was hence assumed as deposited on yards (x_{yard}=1.0). For dairy cows, $x_{grazing}$=0.92 and $x_{building}$=0.08; these values were estimated considering that these cows spend ten months in production (lactating cows) and two

months resting (dry cows). Lactating cows spend ~2 hours a day in milk rooms and the remaining time in the field together with dry cows; their excretions are deposited on land. Poultry is handled in farms, therefore $x_{building}$= 1.0. For swine, $x_{building}$=0.55 and $x_{grazing}$=0.45 for the entire time series. The estimation of these values for swine and those for beef cattle fattening are discussed later.

- The proportion of the N excreted as TAN (x_{TAN}). One value for dairy cattle and one value for non-dairy cattle were selected for all sub-classes for the entire time series. For the other animals, one value was selected for each sub-class for the entire time series. All values were adopted from EMEP (2013).
- The proportion of livestock manure deposited in buildings and handled as slurry or solid (x_{slurry}, x_{solid}), x_{solid}=(1-x_{slurry}). It goes without saying that this parameter is relevant for those sub-classes with $x_{building}$>0.0. For dairy cows and swine, x_{slurry}=1.0, as their excretions are treated as liquids in anaerobic lagoons. For poultry, x_{solid}=1.0, consistent with management practices.
- The amount of manure stored before spreading ($x_{store\ slurry}$, $x_{store\ solid}$ and $x_{store\ FYM}$, where FYM means litter-based farmyard manure). In this study we considered that manure has been always stored before being applied, $x_{store\ i}$=1.0.
- NH_3 emission factors (EFs): $EF_{grazing}$; EF_{yards}; EF_{build_solid} and EF_{build_slurry}; $EF_{applic\_solid/FYM}$ and EF_{applic_slurry}. For stored manure ($EF_{store\_solid/FYM}$, EF_{store_slurry}) in addition to NH_3, other N-species (N_2O, N_2 and NO) were considered. The EFs corresponding to each sub-class were taken from EMEP (2013).
- Addition of N in bedding for the animals ($m_{bedding}$). Broilers are the only animals farmed with beds of wheat or rice mulch, with approximately 3.5 kg/*AAP*.year of bed material aggregated (Chiappe, 2010).

2.1.3. Time series consistency

As was indicated in section 2.1.2, the matrix *AAP* contains gaps for different sub-classes and years, and this has consequences on the time series completeness of the remaining variables. In the TNC an outstanding work has been done on data collection and unification of the livestock farming systems on the estimation of beef cattle emissions, particularly for the period 2010–2012. However, for the previous years and for the remaining livestock classes, time series show significant inconsistencies. The following discussion focuses on the selection of key variables to consistently complete these series and on the inclusion of other animal classes relevant for NH_3 emissions.

Beef cattle, fattening

Information on beef cattle fattening class for the period 2010–2012 has been reported by Vázquez Amabile et al. (2015) based on 51 sub-classes distributed in the eight agro-economic regions of Argentina. For each sub-class, denominated fattening system in the original reference, the data include (*i*) number of animals (*AAP*), (*ii*) type of manure disposal (*TMD*), (*iii*) fraction of manure that is managed (*%MMS*) (if any) and (*iv*) N_{ex} associated with the diets. For each sub-class, the % of *AAP* with respect to the total number of animals in the class showed variability for 2010, 2011 and 2012 while the other three characteristics (*TMD*, *%MMS* and N_{ex}) remained constant. Values of *AAP* for the 51 subclasses for the period 2000–2009 were estimated in this study by extrapolating the information from the period 2010–2012, focusing on the intensification of feedlot farming occurred since 2000, and on the geographical relocation undergone by the beef cattle activity throughout the period.

In Argentina, while manure from feedlot is always managed, the excreta from cattle fattening on land is handled in a variety of ways, ranging from totally unmanaged (grazing) to totally managed. However, the emissions from excreta disposed on pasture, whether managed (reported under MMS) or not (reported under grazing), were estimated using the same methodology; for this reason, feedlot animals have been differentiated from the rest. Data sources of number of animals in feedlot at country level include (*i*) official statistics on average annual stock for the period 2007–2012 (SENASA, 2017) and a (*ii*) compilation of the livestock outputs from feedlots for 2001, 2007 and 2009 (Lence, 2010). The methodology applied for completing the time series is detailed in the Supporting Material (section A.I.), together with the description of the distribution of fattening stock (51x13 matrix), the N_{ex}, the type of manure disposal and manure fraction that is managed, corresponding to each sub-class by agro-economic regions and by year (Table A.3).

Beef cattle, breeding

The number of animals in breeding activity (AAP_{breed} 8x13 matrix) was determined from the difference between the total number of beef cattle and the fattening stock of each agro-economic region and by year. Regarding N_{ex}, we adopted country specific data reported for each agro-economic region for 2010–2012 (Vázquez Amabile et al., 2015) for the entire time period 2000–2012. In rigor, although diet variations for beef cattle have occurred there is no accurate data to reflect this situation. However, the AAP_{breed} matrix describes the geographical relocation of the animals occurred throughout the period. Given the fact that the diet and the resultant N excretion vary from region to region, values for N_{ex} were assigned accordingly and in consequence the time series of annual country-level N_{ex} exhibited a temporal variability related to the relocation of cattle, as happened with the beef cattle fattening class.

Other dairy cattle

Other dairy cattle are composed of animals for womb reposition and reproducers. The time series corresponding to these animals' stock was performed using the values reported by in Vázquez Amabile et al. (2015) for the period 2010–2012, those reported in the 2002 national agricultural census (CNA), and considering a linear behavior to complete the missing years. The average weights of the animals of the sector, necessaries for the N_{ex} determination, were estimated keeping the stock proportion corresponding to 2010.

Poultry

The *AAP* data for the entire period 2000–2012 is only available for the entire class. Figures for the two poultry sub-classes (broilers and laying hens) were obtained by interpolation of the values reported by Finster (2014) for 2000 (60/40 broilers/hens) and those for 2010–2012 (68/32broilers/hens) reported in the TNC.

Swine

The disaggregation of swine sub-classes (sows and piglets, rest of pig livestock) was made using swine total stock data (SAGyP, 2016; SENASA, 2017) and considering that the swine stock composition registered for 2002 (INDEC, 2002) has not varied significantly during the period analyzed.

There are three modes of swine farming in Argentina: (*1*) *intensive*, with all animals in confinement; (*2*) *extensive*, with all animals in the field; and (*3*) *mixed*, sows and piglets in the field, and the rest of the animals in confinement. It was estimated that 40% of the sows, together with their piglets, are grown under mode (*1*), other 40% under (*2*), and the remaining 20% under (*3*). For the rest of animals, based on expert judgment, we estimated that every 1 animal in system (*3*) there are 3 in system (*1*) and 1.4 in system (*2*). With this information and the stock composition, we estimated the following values $x_{building}$=0.55 and $x_{grazing}$=0.45 for the entire time series and for the entire swine class. The resulting distribution between confinement types by animal sub-classes was 41/59 sows and piglets/rest of animals for confined animals (whose excreta is removed with water and disposed in anaerobic lagoons), and 74/26 sows and piglets/rest of animals for those farming in the field (grazing).

2.2. N-containing fertilizers

Direct soil emissions of NH_3 arise from the use of three types of N-containing fertilizers in the form of (*i*) ammoniacal nitrogen, (*ii*) other compounds that decompose quickly in ammonium, such as urea ($(NH_2)_2OC$) and (*iii*) nitrates. Although nitrates do not emit NH_3 directly, they can contribute to its emissions through crop foliage due to the increase in the concentration of nitrogen in the leaves.

According to official information, in Argentina the use of manure as a fertilizer for crop production is not a frequent practice (Vázquez Amabile et al., 2015). Therefore, the entire emissions from manure spreading on land are reported under MMS, and in this section only mineral fertilizers are considered.

The emissions from the use of N-fertilizers were estimated according to a tier 2 (EMEP, 2013) as described below.

(3) $E_{fert} = \sum_{i=1}^{n} \sum_{j=1}^{m} \{m\_fert_{i,j} \cdot EF_{i,j} \cdot [1 - p\_alk_j \cdot (1 - c_i)]\}$

In the Eq. (3) E_{fert} denotes NH_3 emissions in kg/year, $m\_fert_{i,j}$ is the nitrogen mass provided by the fertilizer *i* applied in the region *j* in kg N/year, $EF_{i,j}$ is the emission factor of the fertilizer *i* in region *j* in kg NH_3/kg N. The term p_alk_j is used to account for the proportion of region *j* where the soil pH>7, and for the whole Argentinean agricultural territory p_alk = 0 since the corresponding pH has been reported as lower than 7 (Saint Rozas et al. (2011)).

The consumption data of fertilizers differentiated by product were taken from the Chamber of the Argentine Industry of Fertilizers and Agrochemicals (CIAFA, 2017) for the entire time series considered. The annual consumption of N-fertilizers varied within the range 400–900 Gg N/year in the period 2000–2012 (Figure 2). Urea was the main N-fertilizer used in the country, accounting for ~60% of the total N-consumption. The use of urea ammonium nitrate solution (UAN), which contains 32% N, has gained relevance over the years, with consumption increasing from <10% between 2000–2004 up to ~20% for 2005–2012. Simple phosphates (monoammonium phosphate: MAP, and diammonium phosphate: DAP) have slightly reduced their participation while N-contribution of the remaining fertilizers has oscillated between 8% and 15%.

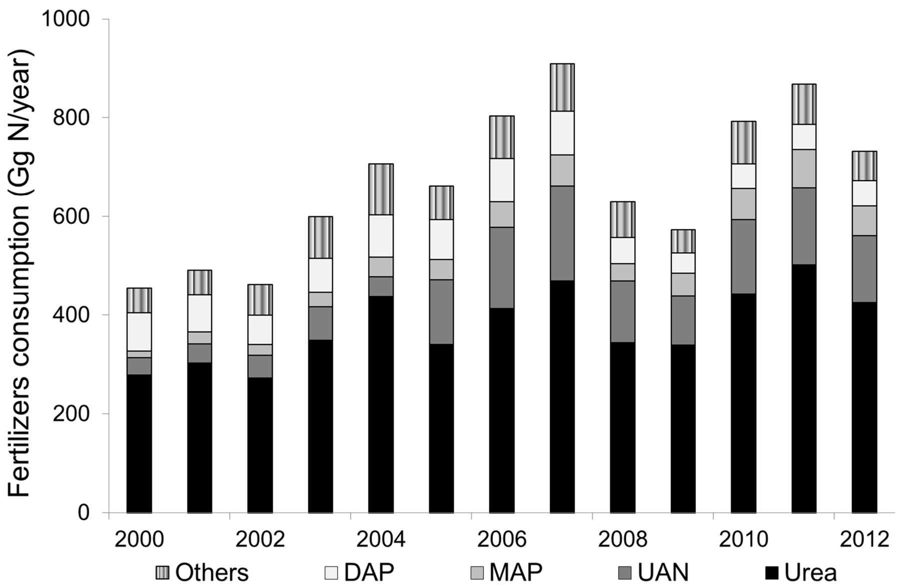

Figure 2: Annual consumption of fertilizers for the period 2000–2012, in Gg of N. (DAP: diammonium phosphate, MAP: monoammonium phosphate, UAN: urea ammonium nitrate solution, others include: ammonium nitrate, sulfate and thiosulfate, potassium nitrate, nitrogen-phosphorus-potassium mixtures, and other N-compounds).

For each crop type i, the emissions from total N-fertilizer used (N_i) were spatially disaggregated on the basis of planted area ($area_i$) and the ratio of N-fertilizer per area ($N_i/area_i$). To this end, crops were organized into *major*, *minor* and *other*. The set of major N-fertilized crops include: wheat and corn, which together represented ~60% of N-consumption, sunflower, malting barley, sorghum, and pastures that accounted for ~22% for 2006, 2011 and 2012 (years with available data on the proportion of the total amount of N-fertilizers consumed by crops) (Fertilizar, 2017). Minor crops consist of fruit trees, citruses, vine and grape, potato, sugar cane, tobacco and rice. Major and minor crops accounted for ~96% of N-consumption in the country for 2006, 2011 and 2012, assigning the remaining 4% to the hereinafter called *other crops*. For each of the six major crops, we obtained data on (*i*) annual cultivated area in each district (2000–2012) (PDA, 2017) and (*ii*) the proportion of the total amount of N-fertilizers consumed (N_i/N_T) only for 2006, 2011 and 2012 (Fertilizar, 2017). Based on these data, we noted that although the ratios N_i/N_T varied, the sum of N_i was practically constant at a level of 0.82 N_T. This value (0.82) was adopted for the ratio $(1/N_T)\sum_{i=1}^{6} N_i$ for all years with missing information. The consumption per crop i in each year j in the periods 2000–2005 and 2007–2010 was estimated by linear correlation of $N_{i,2006}/area_{i,2006}$, $N_{i,2011}/area_{i,2011}$ and $N_{i,2012}/area_{i,2012}$ subject to $(1/N_{T,j})\sum_{i=1}^{6} N_{i,j} = 0.82$. Then, for each district, the amount of total N per crop and year is trivially calculated by the product of the estimated $N_{i,j}/area_{i,j}$ and the corresponded planted area ($area_{i,j}$) from the national statistics.

For minor and other crops, we used information on (*i*) planted area per district, which was available only for 2002 (PDA, 2017) and (*ii*) N_i/N_T for 2006, 2011 and 2012 (Fertilizar, 2017). In the absence of further information, the value of planted area per district in each year was assumed equal to that in 2002. As the values of N_i/N_T did not show significant variations in the years with available data, the corresponding averages were taken as representative for the whole period. To the best of our knowledge, this study reports for the first time activity data relative to N-consumption by minor and other crops.

2.3. Biomass burning

Emissions from biomass burning during agricultural activities include those arising from (*i*) agricultural waste burning and (*ii*) prescribed burning of savannahs. In Argentina, agricultural waste burning (AWB) practices occur only under sugar cane and flax cultivation.

2.3.1. Agricultural waste burning

Emissions corresponding to AWB were calculated on the basis of the following expression:

$$(4) \quad E_{rb} = \sum_{i=1}^{n} A_i \cdot Y_i \cdot s_i \cdot d_i \cdot p_{b_i} \cdot C_{f_i} \cdot EF_{rb_i}$$

where E_{rb} denotes NH_3 emissions, i is each crop (sugar cane or flax), A_i (ha) is the area occupied by crop i, Y (kg ha^{-1}) is the average yield, s is the waste/crop rate, d is the dry matter content, p_b is the proportion of waste burnt in the fields, C_f is the burning factor, and EF_{rb} is the NH_3 emission factor (kg NH_3/kg dry matter).

Data on the amount of cultivated crops (kg), equivalent to the product $A_i.Y_i$, were taken from national statistics sources (PDA, 2017) for flax, and from producer association reports (CAA, 2016) for sugar cane. The proportion of flax area burnt in the fields (2000–2012) was estimated as 50% while that of sugar cane exhibited a reduction factor of 1% per year from 82% in 2002 to 70% in 2012 (Vázquez Amabile et al., 2015). In the absence of crop-specific EFs, the same EMEP tier 1 default value was used for both crops. For the remaining parameters (*s*, *d* and *Cf*), we adopted the values in the IPCC 1996 guidelines (Houghton, 1997).

2.3.2. Other biomass burning

In Argentina there are comprehensive data on the surface affected by fires from the burning of the following types of vegetation: pastures, shrublands, native forests and cultivated forests (MAyDS, 2016). However, this source does not report whether these fires were associated with land use change or agricultural practices (prescribed burning). Therefore, we had no choice other than estimating the emissions from other biomass burning (OBB) in an integrated manner although in total they have not arisen from agricultural activities. These emissions were calculated on the basis of the EMEP guidelines from the carbon emitted by each type of vegetation, weighed by the NH_3 proportion by the emitted carbon (this methodology is described in the Supporting Material, section A.II.).

2.4. Spatial distribution of emissions

Emissions from beef cattle (MMS and grazing) were directly estimated in a disaggregated manner at district level. For all other livestock classes, emissions were estimated at national level and then disaggregated using the 2010 stock (SENASA, 2017) as reference, assuming that for each animal type, the annual share of the stock in each district with respect to the national stock was practically constant throughout the period analyzed.

Emissions associated with fertilizers were estimated at a national level and assigned to the different N-consuming crops, as detailed in section 2.2. The spatial disaggregation from *major crops* was carried out on the basis of those crops distribution in the national territory from the information available at district level for the whole period (PDA, 2017). For *minor crops* surface planted data by district for 2002 (INDEC, 2002) has been used, neglecting the possible displacements during the period analyzed. Because of lack of information the *other crops* group, that accounts only for ~4% of the fertilizer emissions, has not been spatially disaggregated. For reporting purposes, these emissions were equally distributed throughout the country's territory.

Emissions from *other biomass burning* were estimated in a disaggregated manner at district level by type of vegetation burnt. Emissions from AWB were estimated at national level and then disaggregated at district level according to the location of sugar plants with active production, weighing the value of emissions with the annual production of each plant. Emissions from the burning of flax residues, which accounted for <2% of emissions from AWB, were distributed equally.

2.5. Uncertainty analysis

Uncertainties associated with the calculated emissions were estimated using the tier 1 approach of the IPCC 2006 guidelines, based on error propagation from the uncertainties in the activity data, emission factors and other estimation parameters. Most of the activity data used in this study was collected by national statistics agencies; however, the uncertainties associated with these data were not published. Thus, estimation of the uncertainty associated with activity data was mostly based on expert judgment, much of it based on the considerations done in the TNC. Given the diversity of emission factors and other parameters used in our study several sources of information were used to estimate the associated uncertainties. Although the complete list of criteria and information sources used to estimate the uncertainties is reported in Supporting Material, we briefly indicate below the main sources of information other than expert judgment that we used to estimate the uncertainties values adopted for EFs and parameters.

For livestock, an uncertainty of 50% was adopted for N_{ex} (IPCC's Expert Group Report suggested value), and values in the range 9–14% for x_{TAN} depending on the type of animals, based on the maximum difference among the modeling results reported in the bibliography (Reidy et al., 2009, 2008). For cattle under feedlot systems, the uncertainty assigned to the EF was based on the results obtained through the NARSES model (Webb and Misselbrook, 2004) while for the other livestock categories, the EF uncertainties were taken from the EMEP. EF uncertainties of the different types of fertilizers were based on the N loss data reported in EMEP for micrometeorological measurements, wind tunnel experiments and other type of tests. The values adopted for biomass burning EFs and associated parameters were those included in the EMEP guidelines.

3. Results and Discussion

3.1. National emissions

Table 1 reports NH_3 annual emissions in Argentina for 2000–2012, disaggregated according to four main sources: (*i*) manure management, (*ii*) manure in pasture, (*iii*) fertilizers and (*iv*) agricultural waste burning; the corresponding uncertainty ranges are also indicated. Regarding excreta from animal husbandry, the emissions estimated in the disaggregated manner described in section 2.1 were grouped in Table 1 as follows: the emissions under manure management are those arising from excreta handled in feedlot and in pasture reported by our information sources as “managed”, while those under manure in pasture are those arising from animal “grazing”. Since the use of manure as a fertilizer for crop production is not a common agricultural practice in Argentina, emissions from land application of manure were reported under "manure management", and attributed to animal husbandry in line with the TNC.

Emissions from all four sources were higher in 2012 than in 2000, increases were in the order: manure management (82.5%) > direct soil emissions (60.6%) >> agricultural waste burning (8.0%) > manure in pasture increased (3.0%). Total emissions have been reported excluding emissions from OBB because, as calculated, they include emissions from fires not related to agricultural activities (see section 2.3.2). In 2012, NH_3 emissions in Argentina, excluding OBB, amounted to 313 Gg, estimated with an uncertainty level of 26%. Manure in pasture was the largest contributor to the uncertainty, followed by direct soil emissions with a much lesser contribution by manure management. The time series of estimates exhibited decreasing uncertainty values from 31% in the early years to 26% in the late years. This decrease in uncertainty was mainly associated with the decreasing participation of beef cattle under grazing (manure in pasture) in the total NH_3 emissions.

Table 1: Annual ammonia emissions from the four categories of the Argentinean agriculture sector.

NH_3 (Gg)	**Manure management[1]**	**Manure in pasture**	**Fertilizers**	**Agricultural waste burning**	**Total inventory**	**Other biomass burning[2]**
2000	39 (29–50)	99 (37–161)	84 (51–116)	4 (1–7)	**226 (155–297)**	69
2001	42 (30–53)	104 (41–167)	91 (56–126)	4 (1–8)	**241 (168–315)**	100
2002	44 (32–56)	107 (40–173)	83 (51–115)	4 (1–7)	**238 (164–313)**	56
2003	48 (35–61)	109 (41–177)	107 (66–148)	5 (1–8)	**269 (188–349)**	65
2004	51 (37–65)	110 (42–179)	129 (78–180)	5 (1–8)	**295 (209–381)**	8
2005	54 (40–69)	111 (42–181)	115 (73–157)	5 (1–9)	**285 (203–368)**	16
2006	57 (42–73)	113 (42–185)	139 (88–190)	5 (1–9)	**315 (226–404)**	11
2007	60 (45–76)	114 (42–185)	158 (99–216)	5 (1–9)	**337 (243–431)**	3
2008	65 (48–82)	110 (41–179)	111 (69–153)	5 (1–10)	**291 (209–374)**	9
2009	71 (52–90)	106 (41–171)	106 (65–146)	5 (1–9)	**288 (209–367)**	14

2010	66 (49–83)	99 (41–159)	142 (88–196)	5 (1–8)	**312 (231–394)**	3
2011	69 (52–87)	99 (41–157)	158 (98–219)	5 (1–8)	**331 (245–416)**	4
2012	72 (54–90)	101 (41–162)	135 (83–186)	5 (1–8)	**313 (231–394)**	3

[1] The entire emissions from manure spreading on land are included under manure management (manure is not used as a fertilizer for crop production in Argentina).

[2] Emissions from open biomass burning other than that of agricultural wastes were reported but not included in the national totals because they include emissions arising from land use and land use change.

Emissions from manure-related activities constituted the main source of NH_3 emissions, accounting for ~60% of the total. Analyzing each component, it can be observed that the shares in emissions from manure in pasture decreased from 44% in 2000 to 32% in 2012 while those from manure management increased from 17% in 2000 to 23% in 2012. This is mainly a consequence of the greater relevance in feedlot practices and poultry farming; e.g., the annual average livestock on feedlot increased from 0.45 million head in 2000 to 1.31 million head in 2012 (i.e., 191%) while that of poultry increased in 165%. Argentina´s beef cattle was historically based on pastoral production systems, however, the increment in the proportion of the animals farmed in feedlots (from ~1% of the total beef cattle population in 2000 to ~3% in 2012) was reflected in a relevant increase in the proportion of beef cattle emissions from manure management category: from 14% of the total emissions from beef cattle in 2000 to 26% in 2012 (Figure 3). The maximum value of emissions from manure management in 2009 was associated with a substantial increase in the number of registered feedlot animals. This increase may be attributed to a counter measure taken by the government in the period 2007–2009, which provided economic compensation to feedlots for the use of grains to feed animals aimed at ensuring meat supply to the market under high price levels of grains (ONCCA, 2010, 2007).

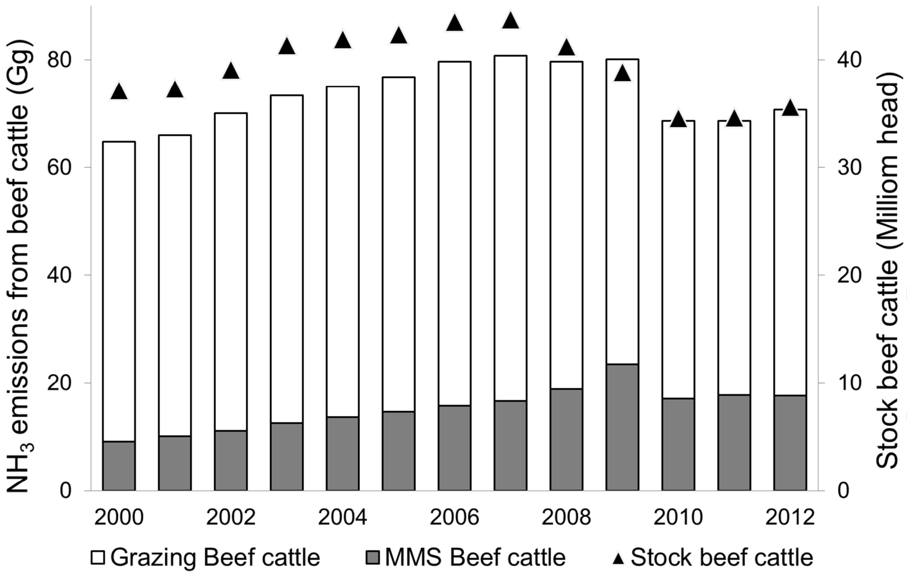

Figure 3: Beef cattle emissions (Gg of NH_3) from grazing and manure management systems (left axis) and average annual stock (million head) (right axis). Note that the entire emissions from manure spreading on land are included under MMS.

The use of N-fertilizers is the other main source, being in general the most significant single source since 2004. In general, the raise in emissions was associated with higher levels of N-fertilizer use associated with the displacement of cultivation areas to less fertile soils, while the variability was largely associated with market conditions, severe drought events occurred in 2008 and 2009, and lingering floods in 2002/2003 and 2012 (Barros et al., 2014).

With regard to the burning of agricultural waste, although it accounted for < 2% of NH_3 emissions, its relevance resides on the fact that it is customarily carried out during specific periods (early spring in the southern hemisphere) and in small areas in the country (sugar cane plantations). This situation, coupled with the presence of compounds co-emitted with NH_3 deserves attention in studies of atmospheric chemistry dynamics.

Emissions from OBB whether they are related to agricultural practice or not exhibited a unique behavior with levels in the range ~56–100 Gg NH_3 in 2000–2003 and in the range ~3–16 Gg NH_3 in 2004–2012. The large differences in emissions between the early years and the last years of the period considered may not be related to an actual decrease in fires but most likely to a methodological evolution in data collection, which was indicated by the source of AD (MAyDS, 2016). Considering that (*i*) the reported values include emissions from land use and agricultural activities and (*ii*) the large uncertainty associated with the AD, we reiterate that these emissions are reported as an information item and note that they should be considered in a cautious manner for alternative purposes. In addition, this situation underlines the need to collect adequate information that would enable to disaggregate AD by type of fire and to obtain a consistent time series.

Excreta from six livestock types (beef cattle, dairy cattle, horses, poultry and swine) and the use of three types of fertilizers (urea, UAN and simple phosphates) contributed to >95% of total NH_3 emissions in Argentina, excluding OBB, in the period 2000–2012. Crop fertilization with urea was the main source of NH_3, contributing 30.0% in 2000 and 33.1% in 2012; it was followed by beef cattle (breeding and fattening) with shares decreasing from 27.7% in 2000 to 21.8% in 2012. Dairy cattle were the third source in order of importance in 2000, representing 11.4% of the total, with emissions maintained at a relatively constant level throughout the period exhibiting a decrease in shares to 7.5% in 2012 as a consequence of the increasing importance of other emission sources. Poultry, became the third contributor in 2012 (11.5% of the total) as a consequence of the relevant increase in the poultry population; it was fifth in 2000 (6.3%) after horses whose contribution was in the range 5.9% (2011)–8.5% (2002) throughout the whole period. The combined contribution of simple phosphates and UAN was in the range 6.2–12.2%, the participation of UAN in the total NH_3 emissions raised from 2.0% in 2000 to 5.4% in 2012 reaching a maximum of 7.1% in 2007 while that of simple phosphates was at relatively stable levels between 3.8% (2002) and 5.0% (2006–2007), reflecting the consumption pattern discussed in section 2.2.

Regarding emissions from the use of N-fertilizers, in addition to considering the contribution of the different fertilizer types, it is worth considering their disaggregation by crops. Corn and wheat combined accounted for 70% of the NH_3 emissions from fertilizer use in 2000 and 58% in 2012. The relative contributions of these crops interchanged throughout the period, in 2000 wheat was responsible for 43% of the emissions and corn for 27%, while in 2012 corn contributed with 34% of the emissions and wheat with 24%. The combined share of corn and wheat decreased as a consequence of the higher contribution of other crops such as malting barley or sorghum, which jointly accounted for 4% (3.4 Gg) of the emissions in 2000, and 15% (19.8 Gg) in 2012. The contribution of the remaining crops to NH_3 emissions did not show relevant variations in the period.

For further details, a complete set of the emissions estimated is presented as Supporting Material (Table A.4).

3.2. Spatial disaggregation

On the basis of the estimated national NH_3 emissions, we estimated the emissions by agro-economic regions and by district. Although we carried out this spatial distribution considering all relevant NH_3-emitting activities, the following discussion focused only on the three components that most contributed to both, levels and spatial distribution: fertilizers, manure management and manure in pasture for beef cattle. Regarding animal husbandry, the discussion below considers beef cattle only because of its relevant contribution to emission levels. Poultry also contributed significantly to the level and especially to the trend but not to variability in spatial distribution since the population of laying hens and broilers has been located roughly in the same area throughout the period. This was the reason for not having explicitly addressed these animals in this section, which focuses on the differences on spatial distribution of NH_3 emissions in the period 2000–2012, which according to our results were significant across the country's agro-economic regions (Table 2).

Table 2: Distribution of NH_3emissions (Gg) arising from fertilizers and beef cattle per agro-economic region.

NH_3 (Gg) by region	Fertilizers		Beef cattle			
			Manure management		Manure in pasture	
	2000	2012	2000	2012	2000	2012
National level (Gg)	83.8	134.6	9.1	17.6	53.5	50.4
Pampa Region	61.7	86.8	7.0	13.4	35.0	28.2
Southeast (P-SE)	9.1	12.2	1.3	1.6	8.8	8.5
Southwest (P-SW)	13.0	16.2	0.3	0.2	6.1	5.1
West (P-W)	12.8	20.4	2.6	2.9	8.4	6.1
North (P-N)	26.9	38.0	2.8	8.7	11.7	8.5
Northeast (NE)	5.3	10.2	0.5	0.6	11.6	13.5
Northwest (NW)	10.0	25.8	0.6	1.7	3.0	4.1
Semiarid (SA)	5.2	10.3	0.9	1.6	3.2	3.5
Patagonia (Pat)	1.5	1.5	0.1	0.4	0.7	1.1

Emissions from crop fertilization increased in all regions excepting Patagonia; however, the shares showed a decrease in the Pampas (from 73.7% in 2000 to 64.5% in 2012) and a sharp increase in the NW (11.9% in

2000 and 19.2% in 2012) and, to a lesser degree, in the NE and the SA regions (from ~6% to ~8%). This spatial variation was related to the relative increase in cultivated areas of soybean. Figure 4 shows that the increase in the total area planted in the country was practically driven by the growth in soybean cultivation while the area for N-consuming crops remained relatively constant. Soybean is mainly produced in the Pampas (Figure 4), therefore the increase in soybean area planted did not imply a decrease in total area planted for other crops but led to the displacement of cultivation areas of N-fertilized crops from the Pampas to NE, NW and SA. It is somehow paradoxical that the cultivation of a crop that does not consume N as nutrient has had such a significant influence on the spatial pattern of NH_3 emissions associated with the use of N-fertilizers.

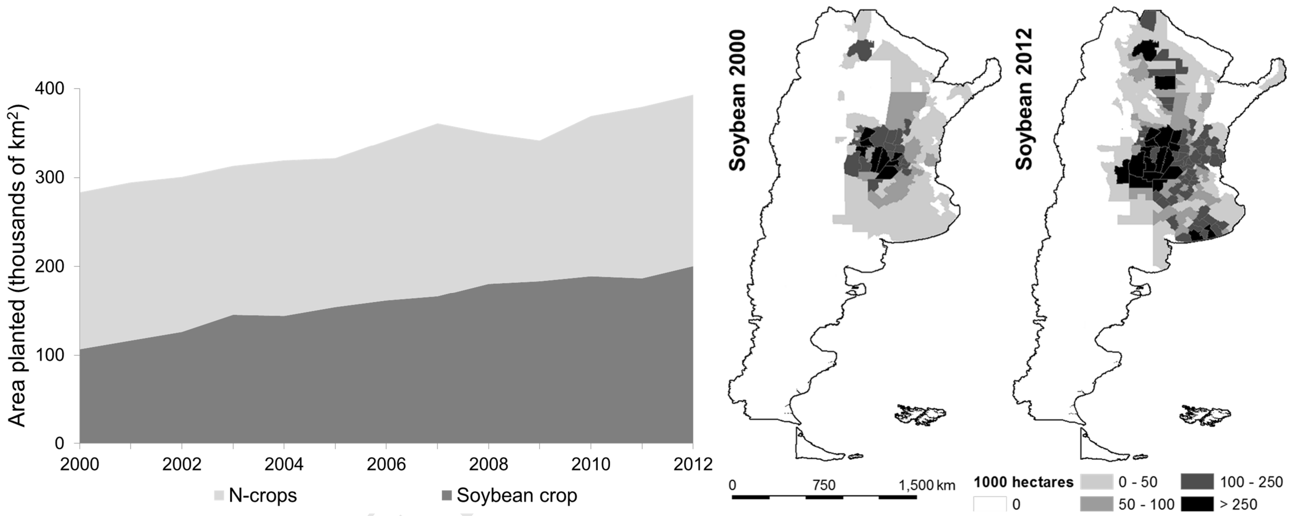

Figure 4: Planted area (thousand km^2) of soybean and N-fertilizer crops in Argentina (left), and spatial distribution of soybean crops (thousand hectares) in 2000 and 2012 (right).

Emissions from beef cattle manure management exhibited increases across the country, most noticeably in the northern Pampas, as a result of the intensification of cattle production systems in feedlots. Emissions from beef cattle manure in pasture, which decreased at national level, exhibited significant variations across regions; they decreased in the Pampas while they increased in the other regions, indicating that the observed trend at national level was clearly driven by the diminishing of beef cattle stock in the Pampas. This is consequence of market conditions coupled with the displacement of grazing beef cattle to other regions.

A finer resolution at district level of the spatial distribution of NH_3 emissions is shown in Figure 5, which depicts emissions from (*i*) excreta from beef cattle, (*ii*) use of N-fertilizers and (*iii*) total NH_3 emissions from agriculture. The emissions are shown as emitted mass of NH_3 for 2000 while for 2004, 2008 and 2012; Figure 5 shows the differences between the emitted mass in the corresponding year and that emitted in 2000.

Total emissions from beef cattle manure increased from 62.6 Gg (2000) to 72.6 Gg (2004) reaching 77.0 Gg (2008) and later decreasing to 68.1 Gg (2012). The change in emissions between 2000 and 2012 can be seen as (*i*) a general increase ($\leq$ 50 Mg NH_3 per district) practically covering the entire national territory in 2008 and 2012, (*ii*) patches of further increases (> 50 Mg NH_3 per district), particularly external to the Pampas region and (*iii*) zones of decreases especially for those districts located in the Pampas. The shift in the spatial distribution of emissions reflects the displacement of cattle farming to less fertile soils, particularly in the northern regions of the country, pushed by the taking of prime land by soybean cultivation. The higher level of emissions in 2008 with respect to 2000 and 2012, discussed in section 3.1 and shown in Figure 3, is clearly evident in Figure 5.

As discussed before, both fertilizer consumption and the associated NH_3 emissions exhibited a significant variability in 2000–2012. Emissions in selected years in Figure 5 were: 83.8 Gg (2000), 128.9 Gg (2004), 111.0 Gg (2008) and 134.6 Gg (2012). In general, emissions in the different districts exhibited significant increases (> 50 Mg NH_3 per district) with respect to 2000. Figure 5 depicts the relatively lower emissions level in 2008 in relation with those of 2004 and 2012; this was mainly a consequence of the drought of 2008 and 2009, which led to a reduction in planted areas (Figure 4) and production loses in maize, wheat and sunflower (Barros et al 2014). In spite of the specific decrease in 2008, the general increasing trend in NH_3 emissions can be appreciated in Figure 5. Increase in crop yields in the country since the 1960s can be attributed to several factors: (*i*) climate trends particularly increase in rainfall, (*ii*) increasing application of modern technology and management practices, and (*iii*) enhanced global demand and higher prices of cereals and grains. New lands for agriculture were included to allow the significant increase in annual crops; they did not extend homogenously throughout the country but exhibited an expansion from the Pampas towards the western and northwestern zones (Barros et al., 2014; Viglizzo et al., 2011b). This expansion of cultivated areas is reflected in the changing spatial pattern of NH_3 emissions shown in Figure 5. In 2004, a decrease in NH_3 emissions in the southeastern Pampas can be seen in Figure 5, this is consistent with a retraction of cultivated areas discussed by Viglizzo et al (2011b).

Total NH_3 emissions represented in Figure 5 are the sum of the two sources discussed above plus the emissions from agricultural waste burning and excreta from the other animals considered in this work. The sum of these additional contributions represents ~33% of the total annual NH_3 emissions, amounting to 79.7 Gg (2000), 93.6 Gg (2004), 103.3 Gg (2008) and 110.0 Gg (2012) and did not suffer significant geographical displacements with the only exception of dairy cattle. The burning of agricultural waste took place almost exclusively in a few districts of the NW region (~99%) where also relatively high amounts of N-fertilizers were used for the crops whose residues were burnt. Because of the relatively high levels of N-fertilizer use in these districts it is not possible to clearly identify the contribution of agriculture waste burning from Figure 5 (bottom) since the NH_3 levels from fertilizers are already represented in the center graph with the maximum value of the color scale. Similarly, the contribution from excreta from dairy cattle, swine and poultry (largely located in the Pampas) cannot be distinguished since they belong to the area with the highest and most concentrated emission level. Contributions of emissions from manure from other animals are particularly noticeable in Patagonia and in some districts of the SA and NW regions (Figure 5, year 2000, bottom). These emissions were largely contributed by sheep (particularly in Patagonia) and horses, which are distributed across the country. In addition, specific districts are distinguishable because of their relatively high emission levels: (*i*) the southwestern extreme of the semiarid region, associated with the high number of goats and horses and (*ii*) the northwestern districts of the NW, associated with the emissions from manure from camelid.

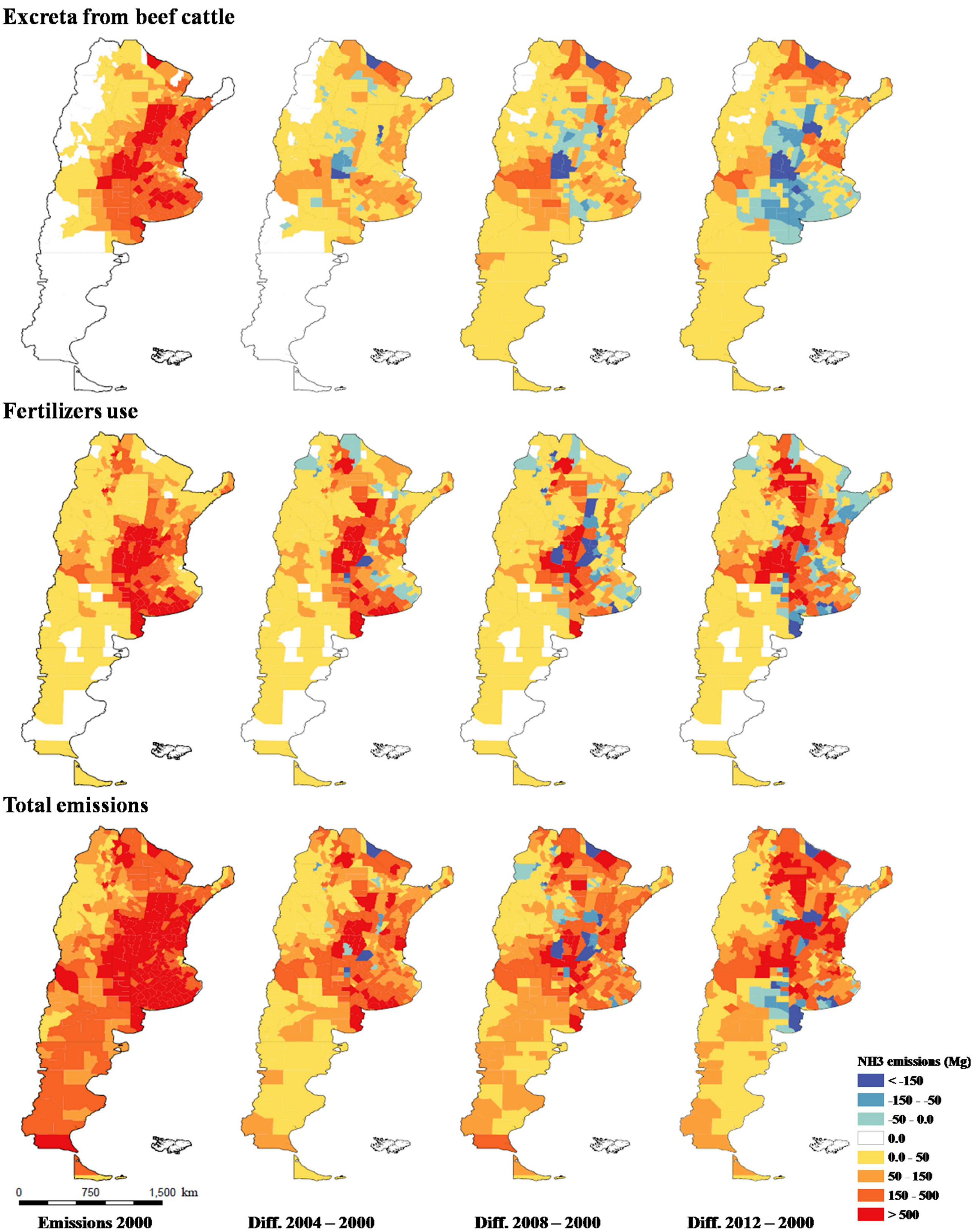

Figure 5: Spatial disaggregation and time variability (2000 to 2012) of NH_3 emissions. Total emissions (top), emissions from fertilizer consumption (center) and emissions from excreta from beef cattle (bottom).

3.3. Verification and quality control checks

3.3.1. *Implicit emission factors*

For verification purposes, and for the emissions that were estimated using a tier 2 approach namely, N-fertilizers use, and manure from livestock, we calculated the implicit emission factors $\overline{EF_i}$ (Eq. (5)) as the ratio of estimated emissions and activity data (N-fertilizer consumption or animal stock).

(5) $\overline{EF_i} = E_i / AD_i$

The $\overline{EF_i}$ thus calculated were set against tier 1 default EFs by EMEP (2013). In general, there is a good agreement between the tier 1 default EFs and the resulting $\overline{EF_i}$ from our study, with values of the $\overline{EF_i}$ within the same order of magnitude than the tier 1 EFs but with lower values, except for the case of fertilizers and cattle in feedlots (Table 3). The differences are discussed below.

The resulting $\overline{EF_i}$ for N-fertilizer application is about twice the EMEP default tier 1 EF. This difference can be explained considering that the EMEP tier 1 EF is based on the mean fertilizer consumption informed by the International Fertilizer Industry Association (IFA, 2016) for Western, Central, Eastern and Central Asia for 2010, which reported a urea consumption between 6% and 17% (measured according to N contribution) while in Argentina the consumption 2000–2012 of urea (the fertilizer with the largest EFs) was ~60% of the N–fertilizers.

The resulting $\overline{EF_i}$ from our study for beef cattle manure in pasture and feedlot encompassed the EMEP default EF. This is consistent with the fact that the default tier 1 EF by EMEP for emissions from beef cattle manure for dry systems is based on a housing period of 180 days while in our study, and according to the country's situation, we have assumed that pasture animals spend all their time excreting on land (grazing period 365) and that feedlot livestock spend virtually all their time in feedlots until they reach market weight. It is worth noticing that the value of $\overline{EF_i}$ for beef cattle manure management in our study, which lies between those of beef cattle in pasture and beef cattle in feedlots, reflects the fact that this source is composed of both types of disposal.

Table 3: Comparison between implicit emission factors resulting from emissions estimated using a tier 2 approach divided by the corresponding activity data and tier 1 default emission factors from EMEP.

Implicit emission factor ($\overline{EF}_i$)		Tier 1 EMEP default EF		Units
Source	This study	Source	Reported value	
N-fertilizer application	0.17–0.19	N-fertilizer application	0.081	Kg NH_3 kg^{-1} N-fertilizer applied
Beef cattle feedlot	10.6–11.1			
Beef cattle manure managed (feedlot and pasture)	3.0–5.9	Beef cattle manure solid	9.2	Kg NH_3AAP^{-1}
Beef cattle in pasture	1.2–1.3			

3.3.2. Comparison with the EDGAR database

Our estimates of NH_3 emissions were compared with those reported in the EDGARv4.2 global database, aggregated at the national level for each of the activities considered. The comparison of emission estimates 2000–2008 is reported in Table 4 in terms of the difference between the estimates in EDGAR minus our own relative to our estimates.

The estimates by EDGAR of emissions from manure management were higher than our estimates in 2000 and 2001 and they become lower since 2002, varying from +16% (in 2000) to -35% (in 2008, last year available in that database). This difference may be ascribable to a certain disregard by part of EDGAR of (*i*) the intensification in feedlot systems and (*ii*) the relevant increase in the poultry population, discussed in section 3.1. Since the emissions estimates in EDGARv4.2 are based on activity data from FAO (Food and Agriculture Organization of United Nations) (FAO, 2017), we analyzed FAO's information relevant to these activities. We corroborated that the FAO information system (*i*) provides official data for the average annual stock of beef cattle but it does not reflect the proportion of the animals that were farming on grazing or in feedlots and (*ii*) does not provide official data for poultry population but estimates that do not reflect the great increase of this activity.

Emissions from manure in pasture in EDGAR are 96% to 123% higher than those estimated in our results. These differences may arise from the use by part of EDGAR of (*i*) AD from FAO, which provides official information only for cattle and sheep and estimates for the rest of the livestock, and (*ii*) default values of housing periods that are not suitable for the country's practices.

Fertilizer consumption data used in the EDGAR database for Argentina are also based on FAO information. We noted that the data from FAO are overall consistent with national statistics and therefore exhibited the same trend than that showed in Figure 2. Therefore, the trend in emissions 2000–2007 from N-fertilizer use reported in EDGAR were consistent with our results, however the emission levels were higher in EDGAR. We noted that the implicit emission factors resulting from EDGAR estimates were in the range 0.24–0.38 kg NH_3 kg^{-1} N-fertilizer applied, which represent values ~2 times higher than our estimates and 3–5 higher than the default tier 1 EMEP EF. These differences may arise from the possible accounting of manure as a fertilizer by part of EDGAR.

Emissions from biomass burning are reported in EDGAR according to the following disaggregation: agricultural waste burning, savannah burning, forest fires and grassland fires. In Table 4 we have compared our estimates of AWB against the values reported in EDGAR for the same denomination. The differences between estimates are significantly large; our estimates were within the range 4.3–5.5 Gg NH_3 year $^{-1}$ while those of EDGAR were in the range 21.2–31.3 Gg NH_3 year $^{-1}$. According to EDGAR's methodology, estimates are based on fraction of crop residues burned in the field taken from the work of Yevich and Logan (2003). Although this paper has very useful information and an adequate description of Argentina's situation, we found out big differences in the level and in the spatial distribution of crops that are burned. The cited reference provides an overall value of 23% for the fraction of crop residues burned in the field in Latin America countries (excluding Brazil) while only sugarcane and flax waste is burned in Argentina, which together represents less than 4% of the total crop residues.

In Table 4, we have also compared our estimates of emissions from OBB against the sum of emissions from savannah burning, forest fires and grassland fires as reported in EDGAR. As it is rather difficult to compare the emission estimates in this disaggregated manner as they may not include the same sources, it is better to estimate the national emissions from biomass burning, independently of sectoral allocation (whether agriculture or land use). Our estimates were in the range 59.8–104.6 Gg NH_3 year $^{-1}$ (2000–2003) and in the range 8.7–20.4 Gg NH_3 year $^{-1}$ (2004–2008) while those of EDGAR were in the range 43.1–73.3 Gg NH_3 year

$^{-1}$ (2000–2008). The noticeable differences between the early years (2000–2003) and late years (2004–2008) of our results are mainly due to the discontinuity in the data collection methodology, already discussed (see section 3.1).

Table 4: Comparison between ammonia emissions estimates reported by the EDGAR global database and this work (relative error %: (EDGAR/our work -1) x 100).

Differences EDGAR NH_3 (%)	2000	2001	2002	2003	2004	2005	2006	2007	2008
Manure management	16%	10%	-9%	-16%	-18%	-23%	-27%	-31%	-35%
Manure in pasture	123%	110%	102%	108%	104%	101%	97%	96%	102%
Fertilizers	72%	80%	89%	59%	55%	75%	56%	43%	113%
Agricultural waste burning	395%	415%	420%	401%	429%	450%	393%	477%	472%
OBB (this work) versus savannah, grassland and forest fires (in EDGAR)	-43%	-57%	-40%	-24%	329%	85%	144%	260%	141%
Total without OBB	91%	87%	83%	72%	66%	73%	61%	55%	83%
Total with OBB	60%	45%	60%	53%	74%	74%	64%	57%	84%

4. Conclusions

We developed a new inventory of ammonia emissions from agriculture activities in Argentina. Annual emissions 2000–2012 from animal excreta, fertilizer application and agricultural waste burning were estimated with a spatial resolution at district level. Total ammonia emissions from agriculture in 2012 were 0.31±0.08 Tg, being manure related activities (0.17±0.06 Tg) and nitrogen fertilizer application (0.13±0.05 Tg) the largest contributors while agricultural waste burning accounted for less than 2%. Uncertainty of the total emissions was in the range 26–31%, being manure in pasture the greatest contributor.

Despite Argentina is one of the top ten agro-exporters of the world, prior to our inventory the only available ammonia emission estimates for the country were those reported in global databases. However, during the last decades there have been important changes in the agricultural practices of the country that were not reflected in the global inventories. This study was carried out within the framework of a national project on atmospheric aerosols, which identified the need of having more accurate estimates of spatially distributed ammonia emissions for air quality modeling purposes.

Activity data used contain high resolution in terms of (*i*) manure from beef cattle, dairy cattle, poultry, swine, sheep, buffalos, goats, camelids, horses and asses/mules, with beef cattle subdivided into 59 sub-classes, and dairy cattle, poultry and swine subdivided into 2 sub-classes each of them, (*ii*) fertilizer type (four main products plus one group accounting for those less used) subdivided according to their application on three groups of crops and (*iii*) waste burning of two crops (sugar cane and flax). Ammonia emissions from these activities were assigned to the districts in the Argentinean territory from which the emissions originate. The spatiotemporal resolution of the key activity data allowed identifying the sensitivity of the estimated emissions to three main drivers: (*i*) expansion of croplands associated with increased rainfall in regions outside the Pampas, (*ii*) dominance of soybean cultivation, competing for lands with N-fertilized crops such as wheat, corn and sunflower and (*iii*) changes in the dynamics of livestock farming including the relocation of cattle in lower-performance areas and the increasing implementation of feedlot systems.

This inventory constitutes an important component of mitigation and air quality policies however it is only a step towards highly resolved ammonia emission estimates in Argentina and there is room for improvement especially if it is to be used as input data by for chemical transport modeling. Temporal resolution can be refined considering seasonal and monthly patterns in agricultural practices and climate conditions, particularly ambient air temperature. Spatial disaggregation can be refined beyond district level by using land use information on fertilizer application and manure disposal practices on specific areas within each district.

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Highlights:

- New ammonia emission inventory from agricultural activities in Argentina, 2000-2012
- We used high resolution activity data regarding types of livestock and fertilizers
- Disaggregated emissions reflect the effect of changing technologies and practices
- Main drivers include changes in: rainfall, cropland areas and livestock farming