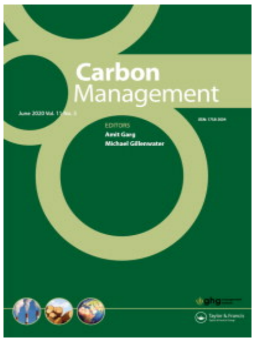

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Temporal and spatial variability of nitrous oxide emissions from agriculture in Argentina

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ABSTRACT

Agricultural activities constitute the main N_2O emission source in Argentina. Although GHG inventories have been developed at the national and provincial level, emissions have not been thus far estimated at a higher spatial resolution. We estimated the time series 2000–2012 of N_2O emissions at national, provincial and district levels. National N_2O emissions in 2012 amounted to 105.1 Gg (95% CI: 73.0–200.7), with manure deposited on pasture accounting for 59.8%, crop residues 24.0%, N-fertilizers use 14.3%, manure management 1.7% and agricultural waste burning 0.2%. Beef cattle excreta followed by soybean crop residues were the major sources of N_2O. The time series of N_2O emission estimated at district level allowed identifying the effect of the frequent displacement of crops and livestock indicative of the variability of the intensity and location of the emission sources. The observed annual variability of emissions and the identification of the main drivers indicate the convenience of using surrogate methods to estimate emissions when activity data cannot be acquired on annual basis. This type of inventory would be of interest for decision makers and stakeholders when discussing environmental policies and measures in light of the responsibility of agricultural activities occurring in the territory of their concern.

KEYWORDS
Emission factors; GHG Inventories; Nitrous oxide; South America; agriculture

Introduction

Among the well-mixed greenhouse gases, N_2O is the third anthropogenic largest contributor to radiative forcing, which is the concept used to evaluate and compare the strength of the various mechanisms that affect the Earth's radiation balance and cause climate change [1]. In their study of the global N_2O budget 1500–2006, Syakila and Kroeze [2] considered natural sources (the oceans), anthropogenic sources (agriculture, biomass burning, energy and industry) and sinks (the uptake at the Earth's surface by soils, aquatic systems and riparian areas). According to their estimates, global annual N_2O emissions, expressed in mass of N, have increased from 11.6 Tg in 1500 to 18.8 Tg in 2006, with anthropogenic emissions accounting for 4.3% in 1500 and 44.1% in 2006. Agriculture is the largest emitter among anthropogenic sources, with annual emissions increasing from 0.4 Tg N_2O-N in 1500 to 5.3 Tg N_2O-N in 2006, accounting for 80% and 63% of total N_2O emissions, respectively. The decrease of the relative share of agriculture in global N_2O anthropogenic emission was largely due to the relative increase in emissions from energy and to a lesser degree in the increase of emissions from biomass burning.

Nitrous oxide (N_2O) emissions from agriculture arise from nitrogen inputs to soils, nitrogen contained in managed manure and the burning of agriculture residues. Main N-inputs to soils from agricultural practices occur through manure deposited on pasture, N-fertilizer application and crop residues. The microbial processes of nitrification (oxidation of ammonium–NH_4^+ to nitrate–NO_3^-) and denitrification (reduction of NO_3^- to the gaseous species nitric oxide–NO, N_2O or N_2) appear to be the dominant N_2O sources in most natural systems [3]. N_2O emissions from soils and manure management occur through both a direct pathway (i.e. directly from the N that is added to soils or present in managed manure), and through two indirect pathways, *i.e.* through volatilization as

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Figure 1. Argentine provinces, where 1: Jujuy, 2: Salta, 3: Formosa, 4: Misiones, 5: Catamarca, 6: Tucumán, 7: Santiago del Estero, 8: Chaco, 9: Corrientes, 10: La Rioja, 11: San Juan, 12: Córdoba, 13: Santa Fe, 14: Entre Ríos, 15: Mendoza, 16: San Luis, 17: La Pampa, 18: Buenos Aires, 19: Ciudad de Buenos Aires, 20: Neuquén, 21: Río Negro, 22: Chubut, 23: Santa Cruz, 24: Tierra del Fuego.

ammonia (NH_3) and nitrogen oxides (NO_x) and subsequent redeposition, and through leaching and runoff [4]. Burning of agriculture residues constitutes a much smaller N_2O source. It also emits NH_3 and NO_x, which contribute to indirect N_2O emissions.

Methods to estimate N_2O emissions fall under bottom-up or top-down approaches. Bottom-up inventories estimate emissions arising from agriculture considering: (*i*) the various pathways associated with the application of N on soils using emission factors based on soil surface gas flux information and/or models that account for the role of other factors (*e.g.* plant N demand. climate and soil variables) influencing direct soil emissions [5]; (*ii*) the N excreted by livestock animals and treated in the different steps of the manure management continuum (generation, storage, treatment and land application) using emission factors based on manure composition, manure production rates, biogeochemical reaction rates, temperature, pH, and moisture content [6, 7]; and (*iii*) the mass of agricultural crop residues burnt on-site taking into account the fractions removed from burning, *i.e.* those that decayed in the field or were destined to other uses [8]. The top-down approach infers anthropogenic N_2O emissions from the relationship between concentration growth of N_2O as a proxy for overall emissions and known atmospheric removal rates [9]. The relevance of bottom-up inventories has been pointed out by Reay *et al.* [10] in their assessment of the role of agriculture on recent and prospective global N_2O emissions. The authors indicated the need to improve the accuracy of national and sub-national estimates.

Achieving the target of the Paris Agreement limiting the increase in the global average temperature will not be possible without significant reductions of GHG emissions in all sectors [11]. Before the adoption of the Paris Agreement, countries reported their intentions on post-2020 climate actions through their Intended Nationally Determined Contributions (INDCs). Although the emission reduction commitments assumed by countries are national in scope, the actions will take place regionally. In this regard, many Argentine provinces have become interested in developing their own GHG emission inventories consistent with the national inventory [12, 13]. The availability of such inventories would enable provinces to undertake region-specific analyses to contribute information to national and local decision makers and stakeholders [14].

Argentina is a South American country with sovereignty over a geographic surface of 27,80,400 km^2 distributed in 24 provinces and subdivided into a total of 527 districts, including 15 districts belonging to Buenos Aires City, where it can be considered that agricultural activities do not occur (see Figure 1). The country belongs to the top six agricultural exporters of the world [15], and according to the Third National Communication of Argentina to the United Nations Convention on Climate Change (UNFCCC), in 2012 N_2O emissions contributed with 17% of GHG, of which >95% arose from the agriculture sector [16].

In this work, we estimated the time series 2000–2012 of N_2O emissions from agriculture in Argentina at national, provincial and district levels aimed at studying the temporal and spatial variability in a context of changes in agricultural practices occurred during this period, which exhibited the highest expansion rates and agricultural

Table 1. **Annual N_2O emissions from the Argentine agriculture sector. Values in brackets represent the 95% confidence interval of the estimated emissions.**

N_2O emissions (Gg)	Manure management	Manure on pasture	Synthetic N-fertilizers	Crop residues	Agricultural waste burning	Total inventory
2000	0.72	65.57	9.43	17.98	0.19	93.88
	(0.48-1.81)	(36.80-153.33)	(4.02-24.71)	(6.80-48.88)	(0.09-0.48)	(62.55-188.18)
2001	0.81	66.36	10.18	18.88	0.19	96.42
	(0.54-2.03)	(37.53-154.31)	(4.34-26.69)	(7.15-51.34)	(0.09-0.48)	(64.75-191.62)
2002	0.91	68.84	9.56	19.12	0.16	98.59
	(0.60-2.26)	(38.57-161.18)	(4.08-25.08)	(7.24-51.99)	(0.08-0.44)	(65.61-197.84)
2003	1.01	70.96	12.38	19.65	0.19	104.20
	(0.66-2.51)	(39.66-166.45)	(5.28-32.47)	(7.44-53.43)	(0.09-0.50)	(69.85-207.46)
2004	1.11	71.73	14.61	20.23	0.19	107.88
	(0.73-2.76)	(40.15-168.09)	(6.23-38.31)	(7.66-55.01)	(0.09-0.49)	(72.86-213.04)
2005	1.21	72.42	13.69	23.28	0.17	110.78
	(0.80-3.01)	(40.49-169.83)	(5.84-35.90)	(8.81-63.30)	(0.08-0.48)	(74.85-218.42)
2006	1.32	73.96	16.60	22.21	0.19	114.28
	(0.86-3.27)	(41.21-173.87)	(7.08-43.53)	(8.41-60.38)	(0.09-0.52)	(77.48-224.58)
2007	1.42	74.08	18.78	25.84	0.19	120.31
	(0.93-3.52)	(41.30-174.08)	(8.01-49.24)	(9.78-70.27)	(0.09-0.52)	(82.24-233.91)
2008	1.64	70.53	12.98	26.80	0.19	112.14
	(1.07-4.03)	(39.69-164.64)	(5.54-34.03)	(10.14-72.86)	(0.09-0.52)	(76.30-219.04)
2009	2.01	66.42	11.80	20.10	0.20	100.53
	(1.31-4.91)	(37.85-153.57)	(5.04-30.93)	(7.61-54.65)	(0.09-0.53)	(68.62-196.25)
2010	1.63	61.33	16.32	25.90	0.17	105.36
	(1.06-4.08)	(35.38-140.52)	(6.97-42.76)	(9.80-70.43)	(0.08-0.46)	(73.41-200.00)
2011	1.71	60.68	17.89	28.89	0.20	109.36
	(1.11-4.27)	(35.00-139.02)	(7.63-46.88)	(10.94-78.53)	(0.10-0.52)	(76.39-206.57)
2012	1.74	62.84	15.06	25.28	0.22	105.14
	(1.13-4.38)	(36.12-144.35)	(6.43-39.46)	(9.57-68.73)	(0.11-0.55)	(72.96-200.72)

production since the 1940s [17, and references therein]. The effects of these changes on ammonia emissions were deeply analyzed by Castesana *et al.* [18] while the consequences on N_2O emissions are the focused of this work.

In this study, we estimated N_2O emissions from manure, N-fertilizer application, crop residues and open burning of agricultural residues. We considered (*i*) manure from 10 different livestock species, (*ii*) all N-fertilizers used in the country according to their application on 13 crop types and (*iii*) N-inputs to soils from 18 crops and (*iv*) the burning of two crop residues. A bottom-up approach such as that by the Intergovernmental Panel on Climate Change [8] was used to estimate emissions. For the particular case of direct N_2O emissions arising from N-application to managed soils of mineral fertilizers, two approaches were used: the tier 1 IPCC default method, which assumes a linear relationship of 1% between N-input and N_2O emissions [8], and the model by Shcherbak *et al.* [19] who investigated the relationship between the N_2O emissions and the levels of N-inputs to soils on the basis of 78 studies containing emission data arising from at least three N-input levels obtained in 84 locations worldwide, including one data set in Argentina.

Methodology

The 2000–2012 series of N_2O emissions from agriculture in Argentina were estimated based on the 2006 IPCC guidelines. We used a tier 1 approach, except for emissions from beef cattle excreta that were estimated using a tier 2 approach due to the importance of this source in the country and the availability of information.

Excepting emissions from fires, direct (E_{direct}) and indirect ($E_{indirect}$) emissions were estimated using Equations 1 and 2, respectively. Total N_2O emissions from each source are given by $E_i = E_{direct} + E_{indirect}$.

$$E_{direct} = AD_i \cdot EF_{D_i} \tag{1}$$

$$\begin{aligned} E_{indirect} &= E_{ATD} + E_L \\ &= (AD_i \cdot Frac_{GAS} \cdot EF_{ATD_i}) \\ &\quad + (AD_i \cdot Frac_{LEACH} \cdot EF_{L_i}) \end{aligned} \tag{2}$$

where:

- AD: activity data expressed as the amount of nitrogen contributed by each source i (animal excreta, synthetic fertilizers and crops residues),
- E_{ATD}: emissions from atmospheric deposition of N volatilized,
- E_L: emissions from leaching and runoff,
- $Frac_{GAS}$ and $Frac_{LEACH}$: fraction of N contribution that volatilizes as NH_3 and NO_x, and fraction lost through leaching and runoff, respectively,
- EF_{D_i}, EF_{ATD_i} and EF_{L_i}: emission factors of N_2O emitted directly and indirectly from atmospheric deposition and N leaching and runoff, respectively.

Emission factors and parameters were selected from the IPCC methodology considering the types of sources and technologies in the country.

Activity data were adopted from (*i*) Vázquez Amabile *et al.* [20], (*ii*) additional data to complete gaps in the time series 2000–2012 [18], and (*iii*) improved description of poultry and swine farming [18].

We used the disaggregation by categories provided by the Intergovernmental Panel on Climate Change [8] to compare our results with those reported in the Third National Communication [16]. Further details of the applied methodology are presented below. Comprehensive information of the activity data, emissions factors and parameters used is provided in Table A.1 in the Supplemental Material.

Livestock

One of the main control factors of N_2O emissions from animal husbandry is the N present in excreta, arising from N-rich protein in feed that has not been completely converted into animal products. This amount of N ($AD_{Livestock}$) can be related to practices where manure is (*i*) managed, reported under manure management (MM) in this work (IPCC subcategory 3A2), or (*ii*) deposited on pasture, reported as manure deposited on pasture (MP) under agricultural soils in this work (IPCC subcategories direct–3C4 and indirect–3C5). In Argentina, those types of livestock for which manure is managed (or at least a proportion thereof) are: beef cattle in feedlots, poultry in solid based systems (with and without bedding), and dairy cattle and swine in anaerobic lagoons. Unless better information is available, the proportion of manure that is managed can be attributed to the fraction of the year the animals spend in buildings or yards, while the rest of the time (as well as the rest of types of livestock) animals can be considered to be farmed under grazing systems, the most widely used system in the country.

Livestock was organized in 10 species subdivided into subclasses with similar characteristics in regard to feeding, excretion and weight, defined by Castesana *et al.* [18]: beef cattle (59 subclasses), dairy cattle (2), poultry (2), swine (2) and other livestock (6, including sheep, buffalos, goats, camelids, horses and asses/mules). Emissions from all subclasses of livestock were estimated from Equations 1 and 2, where $AD_{Livestock}$ is the amount of N contributed by this source, calculated as the number of head of each subclass (H_{sub}) of livestock by the annual average amount of N excreted (Nex_{sub}). Emission estimates from all beef cattle subclasses, estimated using a tier 2 approach, require detailed information on the characterization of livestock and manure management practices to determine the country-specific Nex_{sub} rates. Country-specific N_{ex} values for beef cattle were taken from the information compiled by Vázquez Amabile *et al.* [20], while those for the rest of animals were adopted from Castesana *et al.* [18], based on excreted N-rates reported by IPCC [8] and local conditions. Values for H_{sub} of each subclass and each year of the 2000–2012 period, disaggregated at district level, are those reported on Castesana *et al.* [18], as well as the information about the period of the year that the animals spent in buildings, on yards and/or during grazing.

Fertilizers

We considered only the application of mineral fertilizers since according to local experts the use of manure as a fertilizer for crop production is not common practice in the country and therefore its possible use was disregarded. Fertilizer consumption (AD_F, expressed as kg N in synthetic fertilizers) differentiated by product was adopted from the information provided by the Chamber of the Argentine Industry of Fertilizers and Agrochemicals [21]. This information indicates that urea (the fertilizer with the highest N-content) was the main N-fertilizer used in the country, accounting for ~60% of the total N-consumption, followed by the use of urea ammonium nitrate solution (UAN) accounting for ~20% for 2005–2012.

Direct and indirect (E_{ATD_F} and E_{L_F}) N_2O emissions from crop fertilization with synthetic nitrogenous products, were estimated from the Equations 1 and 2. To estimate direct emissions, we adopted the suggested IPCC default value of 1% for the relationship between N_2O emissions and N-additions to soils of mineral fertilizers, except for flooded rice fields (EF_{F_rice} =0.3%). As an alternative to the IPCC default value, we evaluated N_2O direct emissions from N-fertilizers applied to soils for all crops except rice using the non-linear model proposed by Shcherbak *et al.* [19], as follows:

$$E_{F_D model_j} = \left(6.58 + 0.0181 \cdot Input_j\right) \cdot Input_j \qquad (3)$$

where $Input_j$ is the N-input to soils from fertilizers (kg ha^{-1}) for each crop *j*, and $E_{F\_D\_model}$ are direct N_2O emissions associated with N-fertilization of that crop (g N_2O-N ha^{-1}). To determine the $Input_j$ values, the following steps were considered:

1. Assignation of the N-consumption by each crop *i* (N_i) based on the methodology described in Castesana et al. [18]. Note that wheat and corn together represent ~60% of N-consumption; sunflower, malting barley, sorghum, and pastures account for ~22%; they constitute the six major crops in the country [22]. Remaining crops, hereinafter denominated *minor crops*, covered in this section consist of fruit trees, citruses, grapes, potatoes, sugarcane and tobacco, summing a total of 12 crops which accounted for ~96% of N-consumption.
2. N_i was spatially disaggregated based on the data on annual production of the six major crops in each district (2000–2012) [23]. This decision was based on expert judgment, indicating that the fertilizer dose applied is not homogeneous throughout the territory but depends on soil conditions and humidity, and production is a better indicator of the dose applied than the planted area. For the rest of the crops there is no information on annual production by district, spatial disaggregation of N_i was therefore based on planted area as described in Castesana et al. [18].
3. $Input_i$ was calculated as $N_i/area_i$. The planted area by each crop ($area_i$) was obtained from (*i*) data on annual planted area in each district for each major crop in the whole period [23], and (*ii*) estimates on annual planted area in each district for the rest of the crops, taken from Castesana et al. [18].

Our analysis and results are based on the emission estimates obtained using the IPCC methodology while the results obtained using the alternative model are used for comparative purposes.

Crop residues

Direct and indirect N_2O emissions from crop residues were estimated from the Equations 1 and 2, where indirect emissions only occur from leaching and runoff pathway ($E_{ATD} = 0$).

The main controlling factor in this emission process is the amount of N in crop residues (above and belowground) returned to soils annually (AD_{CR}, in kg N y^{-1}), including N-fixing crops and N from N-fixing and non-N-fixing forages mineralized. Values for AD_{CR} were adopted from [20] and are based on: (*i*) local information, as the total annual area harvested and yield of each crop, or the annual burnt area of those crops burned in fields in Argentina (sugarcane and flax); and (*ii*) IPCC default parameters, as the ratio of above and below-ground residues dry matter to harvested yield, or the N-content of above and below-ground residues, for each crop. The main annual fixing and non-fixing crops, and annual and perennial legume forage species in the country include soybean, wheat, sunflower, corn, barley, sugarcane, sorghum, cotton, birdseed, rice, oats, safflower, rye, rapeseed, flax, peanut, millet and dry bean [20].

Open burning of agricultural residues

In accordance with the Third National Communication, only sugarcane and flax residues are burnt on-field in Argentina as an agricultural practice. Direct emissions agricultural waste burning (AWB) were estimated using Equation 4, based on the generic approach to estimating GHG emissions from fires of 2006 IPCC.

$$E_{D_{AWB}} = \sum\nolimits_c (A_b \cdot M_b \cdot C_f \cdot EF_{D_{AWB}})_c \quad (4)$$

where A_b is the area burnt, M_b is the mass of fuel available for combustion, C_f is the burning factor, and EF_{D_AWB} is the emission factor, all of them referred to each crop *c* burnt in fields. Values of A_b of sugarcane by district are those from Castesana at al. (2018), and values of A_b of flax at national level were taken from Vázquez Amabile *et al.* [20]. Values of EF_{D_AWB} and the product $M_b.C_f$, which represents the amount of fuel actually burnt, are those reported in the IPCC 2006 guidelines for the tier 1 level of detail.

Indirect emissions from biomass burning take place as a result of the deposition onto soils and waters of reactive nitrogen compounds formed from the NO_x and NH_3 emitted during the combustion, in an analogous way to those resulting from their deposition from livestock and N-fertilizers. Because of atmospheric transport, the formation of N_2O may occur in a different location and time than those where combustion took place. N_2O indirect emissions from AWB (E_{ATD_AWB}) were estimated using Equation 5.

$$E_{ATD_{AWB}} = (NO_x{-}N + NH_3{-}N) \cdot EF_{ATD_{AWB}} \cdot 44/28 \quad (5)$$

where NO_x–N is the N-content of NO_x emissions from AWB (assuming that NO_x is reported in NO_2 equivalents), NH_3–N represent the N-content of NH_3 emissions from that source, and EF_{ATD_AWB} is the IPCC default emission factor for N_2O emissions from atmospheric deposition of N on soils and water surfaces. Emissions of NH_3 and NO_x from AWB are those estimated in the context of the 2000–2012 Argentine inventory of ammonia emissions [18], applying the European Monitoring and Evaluation Program (EMEP) approach [24].

Spatial distribution of emissions

Spatial distribution of emissions from livestock and AWB was taken from Castesana *et al.* [18]. In this work the authors estimated emissions as follows: (*i*) beef cattle at district level, (*ii*) livestock other than beef cattle at national level and subsequently disaggregated at district level, and (*iii*) AWB at national level and subsequently disaggregated at district level according to the location of sugar plants with active production and the location of areas planted with flax.

Emissions from crop residues were estimated from the information available at district level on the production of the main crops of the country for the whole period [23]. Emissions from fertilizers were estimated at national level from N-fertilizer consumption, and spatially disaggregated using the annual production of major crops and planted area for the rest of the crops.

Province and district level analysis

Spatial distribution of emissions was analyzed under two different scopes: (*i*) assessing the contribution of each category and province to both the level and trend of the estimated totals, and (*ii*) determining the highest emission districts in terms of emission amount per unit area (emissions fluxes).

The first assessment was grounded in the IPCC methodology for identification of key categories [8], by defining source categories and referring to the N_2O emission totals estimated in our study (Equation 6). We defined 11 single sources as follows: (*i*) five groups of animals (beef cattle, dairy cattle, swine, poultry and other livestock) adding emissions from manure management and manure deposited on pasture, and (*ii*) six groups of crops (soybean, wheat, corn, sunflower, sugarcane and other crops), adding emissions from crop residues, fertilizers and AWB. To determine the contribution of each category and province to the level, emissions from each single source and each province of the country were considered separately, assessing their influence on the national inventory level for each year of the 2000–2012 period.

$$L_{i,j,t} = E_{i,j,t} / \sum_i \sum_j E_{i,j,t} \tag{6}$$

$L_{i,j,t}$ and $E_{i,j,t}$ are the contribution to the level and the emissions, respectively, for the single source *i* and the province *j* in the year *t*. Contributions were summed together in descending order of magnitude, and that hereinafter called "key-levels" were identified using a 75% threshold. To identify those single sources that are not large enough to have received particular attention by the level assessment, but whose trend throughout the studied period is significantly different from the trend of the overall inventory, we performed a trend assessment taking the beginning of the period (2000) as the base year, as follows:

$$T_{i,j,t} = \frac{E_{i,j,0}}{\sum_i \sum_j E_{i,j,0}} \cdot \left| \frac{E_{i,j,t} - E_{i,j,0}}{E_{i,j,0}} - \frac{\sum_i \sum_j E_{i,j,t} - \sum_i \sum_j E_{i,j,0}}{\sum_i \sum_j E_{i,j,0}} \right| \tag{7}$$

where $T_{i,j,t}$ is the contribution to trend for the single source *i* and the province *j* in the year *t*, and the suffix *0* refers to the base year 2000.

Highest emission flux districts were identified for four aggregated sources (livestock, fertilizers, crop residues and AWB) and for total N_2O emissions in 2012 following Van den Heuvel *et al.* [25]. The SSPS software (SPSS 12.0.1, 2003) was used to detect outliers and extreme cases from boxplots of the distribution of N_2O fluxes at different spatial scales. Districts with emission fluxes that were more than 1.5 box-lengths from the edge of their box were classified as outliers, and those that were more than 3 box-lengths away from the edge of their box, were classified as extreme outliers.

Uncertainty estimation

We used the IPCC tier 1 approach for the uncertainty analysis of the estimated emissions, based on the concept of error propagation of the uncertainties in the activity data, emission factors and other estimation parameters. Uncertainty values associated with emission factors and parameters were adopted from IPCC 2006 guidelines, while those associated with activity data were mostly based on expert judgment, in consistency with the considerations done by Vázquez Amabile *et al.* [20] and Castesana *et al.* [18].

Table 2. Annual N_2O emissions from the main livestock in Argentina disaggregated in manure deposited on pasture (MP) and manure management (MM) emissions from each livestock.

Gg N_2O (MP–MM*)	Beef cattle	Dairy cattle	Swine	Poultry	Other livestock	Livestock farming
2000	50.4 – 0.3	9.3 – 0.1	0.4 – 0.1	NO – 0.2	5.5 – NO	66.3
2001	50.5 – 0.4	9.2 – 0.1	0.4 – 0.1	NO – 0.3	6.2 – NO	67.2
2002	53.1 – 0.4	9.2 – 0.1	0.4 – 0.1	NO – 0.3	6.2 – NO	69.7
2003	55.0 – 0.5	9.1 – 0.1	0.4 – 0.1	NO – 0.3	6.5 – NO	72.0
2004	55.5 – 0.6	9.1 – 0.1	0.4 – 0.1	NO – 0.4	6.7 – NO	72.8
2005	56.1 – 0.7	9.0 – 0.1	0.4 – 0.1	NO – 0.4	6.8 – NO	73.6
2006	57.6 – 0.7	9.0 – 0.1	0.5 – 0.1	NO – 0.4	6.9 – NO	75.3
2007	57.7 – 0.8	8.9 – 0.1	0.5 – 0.1	NO – 0.4	7.0 – NO	75.5
2008	54.2 – 1.0	8.8 – 0.1	0.5 – 0.1	NO – 0.5	7.0 – NO	72.2
2009	50.0 – 1.3	9.0 – 0.1	0.5 – 0.1	NO – 0.5	6.9 – NO	68.4
2010	45.4 – 0.9	8.7 – 0.1	0.5 – 0.1	NO – 0.5	6.8 – NO	63.0
2011	44.9 – 1.0	8.5 – 0.1	0.6 – 0.1	NO – 0.6	6.7 – NO	62.4
2012	46.7 – 0.9	8.8 – 0.1	0.6 – 0.1	NO – 0.6	6.7 – NO	64.6

*MM comprises feedlot (beef cattle), anaerobic lagoons (dairy cattle and swine) and solid based systems (poultry).

Results and discussion

National emissions

The time series 2000–2012 of national N_2O emissions (both, direct and indirect) from the Argentinean agriculture sector are summarized in Table 1 according to five main source types manure management, manure from grazing animals, N-fertilizers, crop residues and AWB; the corresponding uncertainty ranges are also indicated. Overall, emissions increased from 94 Gg in 2000 to 105 Gg in 2012 and reached a maximum of 120 Gg in 2007. Annual N-inputs to soils accounted for more than 98% of the emissions with manure deposited on pasture resulting by far the largest emitter (55–70%). Contributions by the other sources were: manure management (1–2%) and AWB (~0.2%). Our estimates of national total N_2O emissions for 2010 and 2012 from agricultural activities are in good agreement with those reported by Argentina in its Third National Communication [16].

Emissions from both manure related activities are reported in Table 2 disaggregated by animal groups. Livestock farming constituted the main emission source, accounting for 70% of the total in the early years and 59% in the late years. Emissions slightly decreased from 66.3 Gg in 2000 to 64.6 Gg in 2012, reaching a maximum of 75.5 Gg in 2007. This trend was mostly driven by the number of grazing beef cattle, which increased from 48 million head in 2000 to 57 million in 2007 and decreased to ~44 million in 2010–2012, in a context of changes in agricultural technologies and practices and market conditions. Beef cattle accounted for ~75% of livestock emissions, with more than 98% arising from manure deposited on pasture. Emissions from dairy cattle and grazing beef cattle experienced a reduction of 3%, unlike the emissions from the other animal groups. Highest increases resulted from feedlot practices in beef cattle (203%, 0.3 to 0.9 Gg N_2O) and poultry farming (157%, 0.2 to 0.6 Gg N_2O). Although these manure management activities are relatively minor N_2O emission sources, they are emergent sources of co-emitted species such as ammonia [18]. Emissions from other livestock were fully attributable to manure deposited on pasture and they were contributed in the order 45% (sheep) > 26% (horses) > 20% (goats).

Emissions from crop residues deposited on soils and the application of N-fertilizers corresponding to five main crops are reported in Table 3, remaining crops has been grouped as "Others". For soybean, it is clearly evident that the sole contribution to N_2O emissions arose from crop residues since this is a N-fixing crop and no N-fertilizers are applied to it. For the entire period 2000–2012 the averaged percent contribution per crop to N_2O emissions from both N sources were in the order: soybean (28%) > wheat (25%) > corn (20%) > others (10%) > sugarcane (9%) > sunflower (4%). Severe drought events that occurred in 2008, 2009 and 2012, and lingering floods in 2012, affected planted areas and harvests, leading to comparatively lower emission levels from crop production related activities [26, 27].

Crop residues deposited above and belowground, constituted the second largest emission source from agriculture (Table 1). These emissions increased at an average rate of ~0.8 Gg N_2O year^{-1} from 18 Gg (2000) to 25 Gg N_2O (2012), while their share increased from ~19% of the total in the early years to 24% in 2012.

The use of N-fertilizers is the third main source, accounting for 10–16% of the totals. Higher levels of N-fertilizer use associated with the displacement of cultivation areas to less fertile soils were the main driver explaining the overall trend in emissions, which increased at an average rate of 0.5 Gg

Table 3. Annual N_2O emissions from the main crops in Argentina disaggregated in crop residue (CR) and fertilizers (F) emissions from each crop.

Gg N_2O (CR – F)	Soybean	Wheat	Corn	Sunflower	Sugar cane	Others
2000	5.6 – NO	4.2 – 4.0	2.8 – 2.6	1.5 – 0.3	2.2*– 0.2	1.7* – 2.3
2001	7.2 – NO	4.4 – 4.6	2.6 – 2.7	0.8 – 0.3	2.2*– 0.2	1.8* – 2.4
2002	8.0 – NO	4.3 – 4.0	2.5 – 2.7	1.0 – 0.3	2.1*– 0.3	1.4*– 2.3
2003	9.1 – NO	3.5 – 5.2	2.5 – 3.6	1.0 – 0.4	2.5*– 0.3	1.3* – 3.0
2004	9.0 – NO	4.0 – 6.2	2.5 – 4.1	0.8 – 0.4	2.7*– 0.4	1.4* – 3.5
2005	10.1 – NO	4.4 – 5.4	3.4 – 4.2	0.9 – 0.5	3.0*– 0.3	1.8* – 3.3
2006	10.7 – NO	3.5 – 6.7	2.4 – 5.0	1.0 – 0.6	3.3*– 0.2	1.5* – 4.0
2007	12.2 – NO	4.0 – 7.7	3.6 – 5.4	0.9 – 0.7	3.5*– 0.2	1.9* – 4.8
2008	12.0 – NO	4.4 – 4.7	3.7 – 4.1	1.2 – 0.4	3.7*– 0.1	2.0* – 3.6
2009	9.5 – NO	2.4 – 3.6	2.2 – 4.2	0.7 – 0.4	3.7*– 0.1	1.8* – 3.5
2010	13.6 – NO	2.5 – 5.5	3.7 – 5.0	0.6 – 0.5	3.5*– 0.3	2.2* – 5.0
2011	13.1 – NO	4.2 – 5.0	4.0 – 6.1	0.9 – 0.6	3.8*– 0.3	3.2* – 5.9
2012	11.3 – NO	3.8 – 3.6	3.6 – 5.2	0.8 – 0.5	2.7*– 0.3	3.2* – 5.4

*The amount includes emissions from burning of crop residues, which represents 5–8% for sugar cane and less than 0.5% for flax among "other crops".

N_2O year^{-1}. A larger rate of increase was exhibited from crop residues (0.8 Gg N_2O year^{-1}) It is plausible to attribute this increase to the contribution of increasing soy production.

Spatial disaggregation

From the development of the national N_2O emission inventory for agricultural sector, emissions were analyzed by province and by district. It was found that, for the entire period, three provinces (Buenos Aires, Córdoba and Santa Fe, hereinafter *main provinces*) systematically accounted for more than 60% of the total emissions. However, the increase in the national emission level between 2000 and 2012 (12%) was higher than those of these three provinces, while emissions in Buenos Aires and Santa Fe increased by 7% and 5% respectively, emissions in Córdoba decreased by 6% and increases in other 20 provinces were above the national total.

In the three main provinces, beef cattle farming has historically prevailed over crop production. However, since the 90 s drivers as market conditions that favored the production of crops over livestock, technological asymmetries between different crops, and changes in rainfall pattern, have promoted important changes in agricultural practices in the country. As consequence, (*i*) croplands expanded and (*ii*) soybean production increased, competing for lands with traditional crops of the country (such as wheat and corn) and modifying the dynamics of livestock farming mainly through relocation of beef cattle in lower-performance areas. The role of these drivers on N_2O emissions discussed below is equivalent to that on ammonia emissions, which were deeply analyzed by Castesana *et al.* [18].

Emissions at district level from (*i*) animal excreta, (*ii*) crop residues, (*iii*) use of N-fertilizers, and (*iv*) total N_2O emissions from agriculture, are depicted in Figure 2. For 2000, emissions are expressed as emitted mass, while for 2004, 2008 and 2012 are shown in terms of the difference between that emitted in each year and 2000. Decreasing levels of manure-related emissions in the central region and increasing levels towards the northern region can be linked to the displacement of livestock farming pushed by the taking of prime land by, mainly, soybean production. In particular, the decreases between 2000 and 2012 in the contribution of N_2O emissions arising from beef cattle in each main province were: Buenos Aires (61%49%), Santa Fe (45%40%) and Córdoba (41%27%). Contrariwise, emissions from crop residues increased in these three provinces as follows: Buenos Aires (3%11%), Santa Fe: (13%15%) and Córdoba (12%19%). This trend can be associated with the magnitude and rate of soybean expansion in these three provinces that concentrated ~80% of soybean production. Reflecting the levels of N-fertilizers applied, associated emissions exhibited increases with respect to 2000 in most districts of the country. Although emissions from crop residues and N-fertilizers use arise from crops, the time series of spatially distributed emission patterns were different. This difference lies mainly in the different role of soybean production in these emissions, while it has a dominant role on emissions from crop residues, its contribution to emissions from N-fertilizers is nil because as N-fixing crop does not require this type of fertilizers. Note also that emissions from N-fertilizers applied to the so called *minor crops* mostly grown in the western and southern regions were estimated on the basis of amount of fertilizer used while emissions from the corresponding crop residues were not estimated because of lack of adequate activity data. Nevertheless, it is expected that these emissions would be a minor contributor of

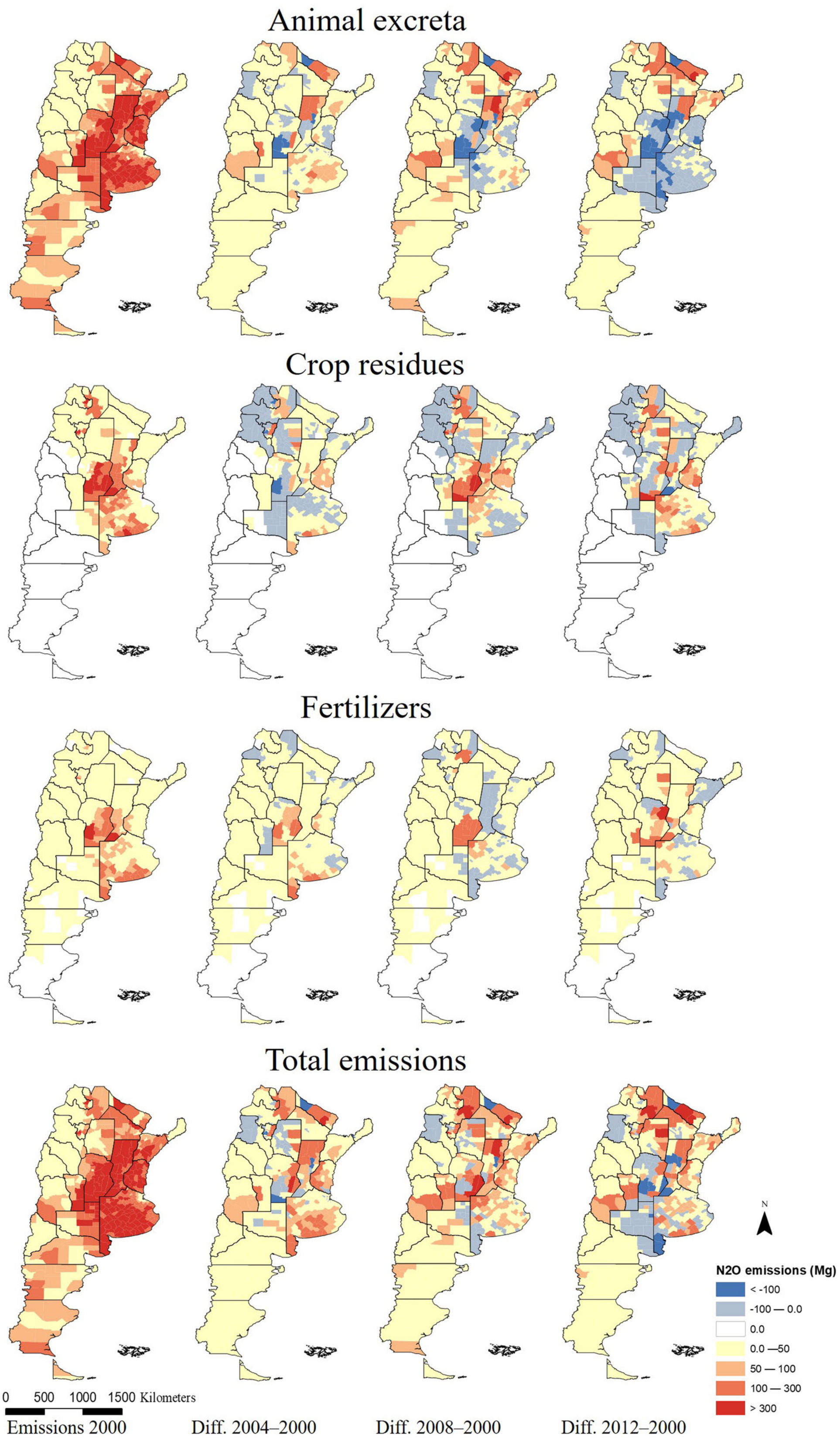

Figure 2. Spatial disaggregation and time variability (2000 to 2012) of N_2O emissions from (i) animal excreta (top), (ii) crop residues (including agricultural waste burning, upper center), (iii) N-fertilizer consumption (lower center), and (iv) total emissions from agriculture in Argentina (bottom). For each item, the first column shows the 2000 N_2O emissions (Mg) while the adjacent columns show from left to right the differences in emissions: 2004–2000, 2008–2000, 2012–2000.

N returned to soils in relation with the main crops.

In addition to the role of main emission drivers, the effects of the previously mentioned droughts and floods over N_2O emissions from crop production activities are also reflected in Figure 2. These include the relatively lower emission levels from fertilizers use in 2008 and lower emission levels from crop residues in 2012, mainly in Córdoba province and the north of Santa Fe associated with

Table 4. Contribution of single sources by province to the 75% of the emission levels in 2012, together with their contribution to the trend.

75% of 2012 N_2O emissions	Key-levels	Contribution to the trend*
Buenos Aires		
Beef cattle	17%	(15%)
Soybean	4%	8%
Wheat	4%	(5%)
Corn	3%	2%
Dairy cattle	2%	1%
Malting barley	2%	6%
Córdoba		
Beef cattle	4%	(10%)
Soybean	3%	2%
Dairy cattle	3%	(3%)
Corn	2%	2%
Santa Fe		
Beef cattle	6%	(3%)
Dairy cattle	3%	(1%)
Soybean	2%	<1%
Corn	1%	2%
Wheat	1%	(1%)
Entre Ríos		
Beef cattle	4%	(3%)
Corrientes		
Beef cattle	4%	1%
La Pampa		
Beef cattle	3%	(4%)
Chaco		
Beef cattle	2%	1%
Tucumán		
Sugar cane	2%	1%
Formosa		
Beef cattle	1%	1%
San Luis		
Beef cattle	1%	<1%
Santiago del Estero		
Beef cattle	1%	<1%

*Values between brackets correspond to decreasing trends.

severe drought. This last finding is in good agreement with local studies on economic losses from soybean production, due to extreme weather events [27].

Burning of sugarcane wastes, represented in Figure 2 together with emissions from crop residues, took place almost exclusively in few districts of the northwest region and ~65% of emissions occurred in Tucumán province.

Table 4 reports the contribution of single sources by province to the 75% of the emission levels in 2012, and the contribution of these key-levels to the trend. Content of this table reinforces the previous analysis, but also discloses the following information for 2012:

- Beef cattle farmed in Buenos Aires was the biggest single contributor to the level (17%).
- Beef cattle showed decreasing trends in provinces belonging to the central region of the country.
- Except in Tucumán, where sugarcane production is the main agricultural activity, beef cattle was the dominant single source in all those provinces that contribute to the 75% of agricultural emissions.
- Although soybean does not consume N-fertilizers, it emerged as the dominant crop in main provinces, with an important contribution to the trend in Buenos Aires (8%), due to emissions from crop residues.
- Although relatively small, the contribution by malting barley was identified as an emerging source in Buenos Aires because of its significantly increasing trend throughout the studied.

In 2012, only five of the 527 districts in the country were detected as *highest emission flux districts*, which all belong to the Tucuman province. Two of those districts were classified as outliers with emission fluxes of 318 and 331 kg N_2O km^{-2}, and three of them as extreme outliers with fluxes ranging from 370 to 547 kg N_2O km^{-2}, while the average for all districts of the entire country and for the whole period rounded ~50–60 kg N_2O km^{-2}. In that year, N_2O emissions coming up of activities related to the production of sugarcane (adding those from N-fertilizers, crop residues and AWB) accounted for 85–99% of the total emissions in each district. Maps of highest emission flux districts, both those corresponding to total emissions and those of each aggregated source can be found in the Supplemental Material (Figure A.1).

Application of alternative model to estimate emissions from N-fertilizers

N-input per area were estimated for 12 crops to apply the model by Shcherbak *et al.* [19]. In Argentina, and during the entire period, the areal consumption of fertilizers for all analyzed crops, except potato and tobacco, fall below the cutting point of 188.95 kg ha^{-1} (Figure 3), indicating that emissions per unit mass of fertilizer applied are lower than the IPCC default emission factor of 1%. The likely improvement in accuracy when using the model by Shcherbak instead of the IPCC default value would represent the following reductions in estimates throughout the period:

- 23% in emissions from N-fertilizers;
- 2–3% in emissions from agriculture sector at national level;
- ~4% (Córdoba) > 3% (Buenos Aires) > 2% (Santa Fe) in overall emissions in each *main province*;
- ~3% in overall emissions in each *highest emission district*; and

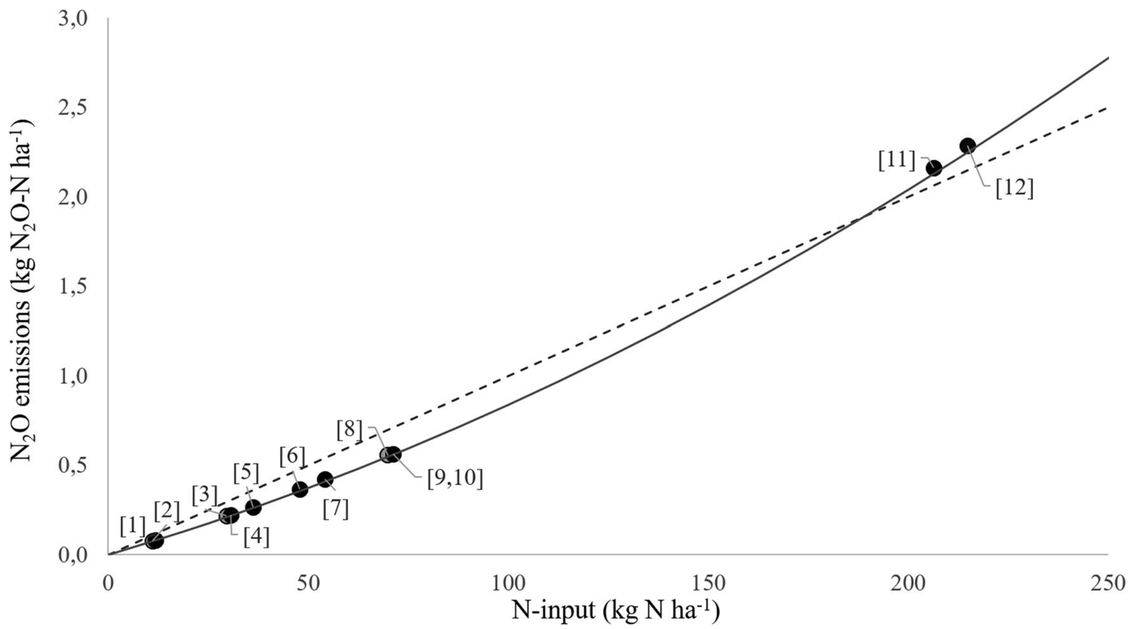

Figure 3. N_2O areal emissions (kg N_2O-N ha-1) as a function of N-input (kg ha^{-1}) obtained by using (i) the IPCC default value for direct emissions from N-fertilizers, and (ii) the Shcherbak *et al.* model for 12 crops in Argentina throughout 2000–2012, where 1: sunflower, 2: pastures, 3: sorghum, 4: fruit trees, 5: barley, 6: wheat, 7: corn, 8: sugarcane, 9: grapes, 10: citrus, 11: tobacco and 12: potatoes.

- 0.1% of the 2012 national GHG emissions reported in the Third National Communication [16].

These outcomes showed that the differences between N_2O emission estimates using the IPCC emission factor and the model by Shcherbak *et al.* were all within the uncertainty range of these estimates. The balance between the effort made to estimate N-input per area and the relatively small difference in the results obtained using this information in a more refined model may be of interest for inventory compilers in prioritizing efforts to improve the accuracy of their national GHG inventories [28].

Conclusions

We developed the 2000–2012 N_2O emission inventory from agriculture activities in Argentina, with a spatial resolution at district level. Total emissions from agriculture in 2012 were 105.14 Gg, being manure related activities (64.6 Gg) and nitrogen inputs to soils from crop residues (25.3 Gg) the largest contributors, while nitrogen fertilizer application accounted for 14% and agricultural waste burning for less than 1%. Uncertainty of the total emissions was around one order of magnitude, being manure deposited on pasture the greatest contributor, the availability of locally determined emission factors would be key to narrow the overall uncertainty range.

The estimation of spatially disaggregated emissions adds value to the estimation of the national totals, which are consistent with those reported in the Third National Communication of Argentina to the UNFCCC. Our results served to identify that only three out of 24 provinces accounted for >60% of the total emissions. The disaggregation by districts allowed identifying the role of main drivers (expanding soybean planted area displacing livestock and other crops, occurrence of droughts and floods), which conducted to an annual relocation of emission sources. As a consequence, we showed how the emissions have varied spatially across the studied period. One caveat of this finding concerns filling out data gaps in ensuring a consistent time series, indicating that interpolation would be most likely not advisable when activity data are not available on an annual basis. A surrogate method based on underlying variables such as spatial determinants (soil quality and climate), agricultural practices, land-use change and economic conditions capable of explaining the temporal and spatial variability of the source activities may constitute a better option to simulate the trend in emissions [17].

The availability of time series of spatially distributed N_2O emissions from agriculture is of interest for policy purposes and decision making in light of changes in agricultural practices, market trends in the agriculture and farming industry and the occurrence of extreme weather events. This type of GHG emissions inventories is helpful in supporting mitigation strategies by providing accurate quantitative information for reconciling potentially contradictory interests at the district, province and national level regarding food supply, economic growth and environmental commitments.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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