Science of the Total Environment 645 (2018) 205–219

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

A new framework to estimate spatio-temporal ammonia emissions due to nitrogen fertilization in France

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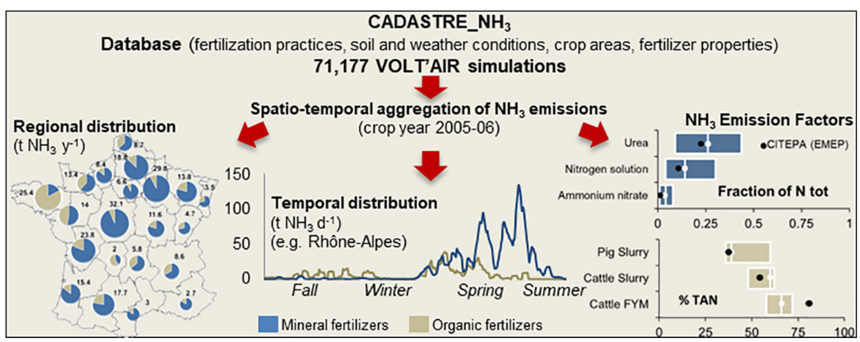
HIGHLIGHTS

- Estimated NH_3 emissions from N fertilization are quantified using a new method.
- 2005/06 NH_3 emissions in France are 29% lower than the official French inventory.
- Spatial variability is due to N fertilizer use and type and pedoclimatic conditions.
- Temporal variability depends on seasonal timing of N fertilizer applications.
- Specific French emission factors are estimated per fertilizer and per region.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 16 November 2017
Received in revised form 16 June 2018
Accepted 16 June 2018
Available online xxxx

Editor: Elena Paoletti

Keywords:
National inventory
Cultural practices
Volt'Air model
CADASTRE_NH_3
Emission factors
France

ABSTRACT

In France, agriculture is responsible for 98% of ammonia (NH_3) emissions with over 50% caused by nitrogen (N) fertilization. The current French national inventory is based on default emission factors (EF) and does not account for the main variables influencing NH_3 emissions. To model the spatio-temporal variability of NH_3 emissions due to mineral and organic N fertilization, we implemented a new method named CADASTRE_NH_3. The novelty lies in the combined use of two types of resources: the process-based Volt'Air model and geo-referenced and temporally explicit databases for soil properties, meteorological conditions and N fertilization. Simulation units are the Small Agricultural Regions. Several sources of information were combined to obtain N fertilization management: census and surveys of the French Ministry of Agriculture, statistics on commercial fertilizer deliveries, and French expertise on physicochemical properties of organic manure. The practical interest of this new framework was illustrated for France during the crop year 2005/06. Aggregation at crop year level showed a reasonable agreement between estimated values derived from CADASTRE_NH_3 and those from the French inventory method, for N and ammoniacal-N (TAN) application rates, total NH_3 emissions and NH_3 EF. Discrepancies were large for organic manure only; national TAN application rates and NH_3 emissions were 62–63% lower with CADASTRE_NH_3. This was due to divergences in the representation of cattle farm yard manure and in the TAN:N ratio of solid manure. Annual emissions for fertilization in France were estimated to be 270 Gg NH_3, 29% lower than the French national inventory estimate. At the regional level, organic manure contributed to 73% of field NH_3 emissions in intensive livestock husbandry areas and to 41% in the other areas. The

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https://doi.org/10.1016/j.scitotenv.2018.06.202

CADASTRE_NH_3 framework can be seen as a Tier 3 approach able to estimate specific regional EF for different mineral fertilizers and organic manure.

1. Introduction

Ammonia (NH_3) is a major atmospheric air pollutant. It contributes to the formation of fine particulate matter in the atmosphere (Erisman and Schaap, 2004), an air pollutant having adverse impacts on human health (Moldanová et al., 2011). NH_3 is also deposited on semi-natural ecosystems. Brought in excess, it contributes to water eutrophication and plant biodiversity decrease (Van Breemen et al., 1984; Sutton et al., 1995; Bobbink et al., 1998; IPCC, 2006); NH_3 nitrification leads to soil acidification, and denitrification that follows leads to indirect emissions of the greenhouse gas nitrous oxide.

Emissions of NH_3 are covered by the recently revised European Union National Emission Ceilings Directive 2016/2284 (EU, 2016) and by the Gothenburg protocol under the United Nations Convention on Long-Range Transboundary Air Pollution (LRTAP) (UNECE, 1999, revised in 2012). Both regulations set NH_3 emission ceilings: the 2012 revision of the Gothenburg protocol imposes emission reduction targets for 2020 in relation to the emissions reported in 2005 by all member states. France is committed to reduce annual NH_3 emissions to 685 Gg by 2020, which represents a 4% reduction from 2005. Generally, Europe Union legislation is associated with programs and/or action plans identifying country-specific measures to reduce NH_3 emissions.

In Europe, agriculture is responsible for about 90% of total NH_3 emissions (EEA, 2014). As the leading agricultural producer in the Europe Union, France is one of the largest emitters of NH_3 in Europe. The French organism in charge of national inventories, the *Centre Interprofessionnel Technique d'Étude de la Pollution Atmosphérique* (CITEPA) estimated that 718 Gg of NH_3 were lost in 2013: 98% were attributed to agriculture (CITEPA, 2015). Mineral fertilizer applications account for 32% of the total French agricultural NH_3 emissions, while livestock production accounts for 68%, manure spreading contributing 40% of total NH_3 emissions from livestock production (see CITEPA internal calculation files used for the national inventories (CITEPA, 2015)). Clearly, emissions due to field application of either manure or N fertilizers represent a sizable proportion of total NH_3 emissions in France: 59%.

Reliable estimates of NH_3 emissions are essential to identify efficient mitigation strategies at both the national and regional level. The European Monitoring and Evaluation Programme (EMEP) guidebook (EMEP/EEA, 2013a, 2013b) proposes NH_3 emission inventory methodologies for countries that have to report NH_3 emissions to the United Nations Economic Commission for Europe (UNECE) and/or European Commission. Estimated NH_3 emissions are obtained by multiplying activity data by their respective emission factors (EF). Activity data refer to the characteristics of emission sources, such as the total amount of fertilizer applied and the total amount of livestock manure excreted. Three levels of emission factors are proposed by the EMEP guidebook: (i) Tier 1, which is based on one single default EF for all mineral N fertilizers and on one EF per livestock category and manure type (solid or slurry) for organic products; (ii) Tier 2, which is based on an intermediate level of default EF; and (iii) Tier 3, which is based on modeling. Tier 2 and Tier 3 emission factors require detailed and comprehensive activity data. In general, emission factors are not available specifically for each country: they are obtained by compiling all the available experimental measurements of volatilization performed in various field conditions thanks to an international literature review.

The French inventory methodology implemented by the CITEPA is based on the 2009 Tier 2 method (EMEP/EEA, 2009): CITEPA uses various types of activity data, mainly drawn from French agricultural statistics (CITEPA, 2014), but details on cultural practices and on fertilizer and manure properties are neglected. The use of one constant emission factor per fertilizer and per manure does not allow accounting for the effects of soil and meteorological conditions on NH_3 emissions in the field (EMEP/EEA, 2013a, 2013b; Sommer et al., 2003; Garcia et al., 2012; Le Cadre, 2004; Génermont and Cellier, 1997; Smith et al., 2009a; Huijsmans et al., 2014). Given these limitations, the French inventory methodology cannot provide the spatial distribution in France or the dynamics over the year of NH_3 emissions. Representing the spatial and temporal variability of NH_3 emissions are however essential to obtain an accurate evaluation of (i) the impact of NH_3 emissions on the formation of fine particulate matter using current chemical transport models such as CHIMERE (Hamaoui-Laguel et al., 2014) or (ii) the effectiveness of mitigation techniques at fine spatial and time scales.

Our objective was to explicitly and realistically represent the spatial and temporal distributions of NH_3 emissions in France over one crop year. We developed the CADASTRE_NH_3 framework based on the process-based Volt'Air model (Garcia et al., 2011; Garcia et al., 2012; Génermont and Cellier, 1997; Le Cadre, 2004) and on a description of the activity data in France at the Small Agricultural Region (SAR) level. The Volt'Air model was chosen because (i) it is process-based and incorporates current knowledge on NH_3 volatilization after application of the main types of organic manure and mineral N fertilizers in the field, (ii) it uses major factors known to influence NH_3 volatilization in the field (soil properties, weather conditions, cultural practices and properties of mineral fertilizers and organic products) as input data, and (iii) it has been successfully assessed in different studies for a relatively large range of agro-pedo-climatic conditions and organic products and mineral fertilizers (Le Cadre, 2004; Le Cadre et al., 2008; Smith et al., 2009b; Garcia et al., 2012; Huijsmans et al., 2014; Langevin et al., 2015). Furthermore, Volt'Air has already been applied at the national scale in England and Wales, U.K. (Theobald et al., 2005), France (Hamaoui-Laguel et al., 2014) and at the European scale (Theobald et al., 2014). In this paper, we describe the CADASTRE_NH_3 framework, and we implement it in France to derive emission factors at both national and regional levels and per fertilizer and manure type. The EF derived with the new framework are compared with the default EF currently used by the French national inventory.

2. Materials and methods

2.1. General framework

Fig. 1 presents the general flowchart of CADASTRE_NH_3 based on the process-based Volt'Air model.

As the input data required to run Volt'Air were not available at the field scale in France, we defined an intermediate geographical unit in which all information was allocated: the Small Agricultural Regions (SAR). These geographical units are homogenous agricultural regions delineated in the early 50 s according to criteria mainly related to soil and climatic conditions. The number of SAR units in continental France is 713, with a size ranging from 1096 to 440,650 ha. Data were processed using a Geographical Information System in order to be integrated into each SAR unit. All input data required by Volt'Air (detailed in 2.2.4) were geographically overlaid and intersected with the Geographical Information System to generate input combinations in each SAR. Each input combination was considered as one situation, and used as the input data for a virtual field for a simulation using Volt'Air. As Volt'Air is sensitive to field size, the size of the virtual field was calculated as the mean of the surface areas of the French agricultural fields declared in the Land Parcel Identification System in 2006: it was 9 ha i.e., 300 m × 300 m. Input data as well as output emissions were stored