

Estimation of global NH₃ volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands

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[1] One of the main causes of the low efficiency in nitrogen (N) use by crops is the volatilization of ammonia (NH₃) from fertilizers. Information taken from 1667 NH₃ volatilization measurements documented in 148 research papers was summarized to assess the influence on NH₃ volatilization of crop type, fertilizer type, and rate and mode of application and temperature, as well as soil organic carbon, texture, pH, CEC, measurement technique, and measurement location. The data set was summarized in three ways: (1) by calculating means for each of the factors mentioned, in which findings from each research paper were weighted equally; (2) by calculating weighted median values corrected for unbalanced features of the collected data; and (3) by developing a summary model using linear regression based on weighted median values for NH₃ volatilization and by calculating global NH₃ volatilization losses from fertilizer application using 0.5° resolution data on land use and soils. The calculated median NH₃ loss from global application of synthetic N fertilizers (78 million tons N per year) and animal manure (33 million tons N per year) amount to 14% (10–19%) and 23% (19–29%), respectively. In developing countries, because of high temperatures and the widespread use of urea, ammonium sulfate, and ammonium bicarbonate, estimated NH₃ volatilization loss from synthetic fertilizers amounts to 18%, and in industrialized countries it amounts to 7%. The estimated NH₃ loss from animal manure is 21% in industrialized and 26% in developing countries. *INDEX TERMS*: 0315 Atmospheric Composition and Structure: Biosphere/atmosphere interactions; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 1615 Global Change: Biogeochemical processes (4805); 3210 Mathematical Geophysics: Modeling; *KEYWORDS*: ammonia, application of animal manure and N fertilizer, NH₃, nitrogen

1. Introduction

[2] Yields of crops and forage species are often constrained by the supply of nitrogen (N) for growth [Laegreid *et al.*, 1999]. In many parts of the world, fertilizer N is routinely applied to food and cash crops and, increasingly, to grasslands. In many countries, fertilizer N application rates of >100 kg ha⁻¹ yr⁻¹ are common [Food and Agriculture Organization (FAO)/International Fertilizer Industry Association (IFA)/International Fertilizer Development Center (IFDC), 1999]. At present, the annual global use of synthetic fertilizers is 78 million tons N [IFA, 1999], and large quantities of animal manure are used to fertilize crops and grasslands [Lee *et al.*, 1997]. The use of N fertilizer and the production of animal wastes are expected to increase in the coming decades, particularly in developing countries [Bouwman, 1998; Bouwman and Van Der Hoek, 1997].

[3] Crop uptake commonly amounts to only 50% of the fertilizer N applied [Peoples *et al.*, 1995]. The main cause of this low N use efficiency is the loss of N from the plant-soil system via gaseous emissions, leaching, runoff, or erosion. The importance of each of these pathways varies from site to site and from year to year. Generally, gaseous N loss is the dominant mechanism in many

agricultural production systems, the major loss processes being volatilization of ammonia (NH₃) and denitrification [Peoples *et al.*, 1995; Smil, 1999].

[4] In this paper we will focus on NH₃ volatilization resulting from applying synthetic N fertilizers and animal manure. Ammonia is an important atmospheric pollutant, with a wide variety of impacts. In the atmosphere, NH₃ neutralizes a great portion of the acids produced by oxides of sulfur and nitrogen. A great part of atmospheric aerosols, acting as cloud condensation nuclei, consist of sulfate neutralized to various extents by NH₃. Essentially, all emitted NH₃ is returned to the surface by deposition, which is known to be one of the causes of soil acidification since the early 1980s [Van Breemen *et al.*, 1982]. The role of NH₃ as a fertilizer was already known more than a century ago [Lawes and Gilbert, 1851]. In the last few years, there has been growing concern about the eutrophication of natural ecosystems and loss of biodiversity due to N deposition [Bouwman and Van Vuuren, 1999].

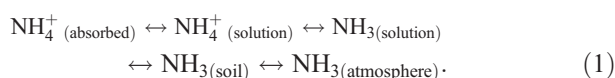
[5] Volatilization of NH₃ from fertilizer use and animal excreta is a major source of atmospheric NH₃ (Table 1). Reviews of NH₃ volatilization from flooded rice fields [e.g., Fillery and Vlek, 1986; Frenay and Denmead, 1992], from fertilizer use in general [e.g., Peoples *et al.*, 1995], and from grazing systems [e.g., Bussink and Oenema, 1998] indicate the presence of many factors regulating the NH₃ loss from soil-plant systems to the atmosphere, depending on the crop, and management of soils, water, and fertilizers.

Table 1. Global Sources of Atmospheric NH₃ in 1990^a

Source	NH ₃ -N emission, Mt yr ⁻¹
Fossil fuel combustion, including aircraft	0.1
Industrial processes	0.2
Animal excreta	21.7
Fertilizer use	9.0
Croplands	3.6
Biomass burning, including biofuel combustion	5.9
Human excreta	2.6
Soils under natural vegetation	2.4
Oceans	8.2
Total	53.6

^aData are based on work by *Bouwman et al.* [1997].

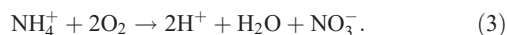
[6] Ammonia is constantly formed in soils from biological degradation of organic compounds and from ammonium (NH₄⁺) yielding synthetic and organic fertilizers. Since NH₃ is a gas, any of it that is present in the soil, water, or fertilizer can volatilize to the atmosphere; its reactions in water are fundamental in regulating the rate of loss. After NH₃ is applied to the soil, the NH₄⁺ can be retained on the exchange sites, nitrified to nitrate (NO₃⁻), or converted to NH₃, depending on soil and environmental conditions:



Ammonia volatilization is driven by the difference in NH₃ partial pressure between the air and soil atmosphere or floodwater. The partial pressure of NH₃ in soil is controlled by the rate of removal of ammonium or NH₃ in solution or by displacing any of the equilibria in equation (1) in some other way. Wind speed, temperature and the reaction (pH) of the soil solution or irrigation water, the soil's pH buffering capacity, and cation exchange capacity (CEC) all affect the partial pressure of NH₃.

[7] The fertilizer N application rate can be expected to influence the ammoniacal N concentration. However, some studies indicate that NH₃ volatilization rates are not related to the N application rate [e.g., *Dhyani and Mishra*, 1992; *Saravanan et al.*, 1987], while other studies indicated lower [e.g., *Thompson et al.*, 1990] or higher [e.g., *Black et al.*, 1985; *Fenn et al.*, 1987] NH₃ volatilization rates at high N application rates than at low rates. Apparently, different factors and processes interact. For example, *He et al.* [1999] found the relationship between N application rate and NH₃ volatilization to depend on the fertilizer type.

[8] Wind speed regulates the exchange of NH₃ between the soil/floodwater and the air. Increasing temperature increases the relative proportion of NH₃ to NH₄⁺ present, decreases the solubility of NH₃ in water, and increases the diffusion of NH₃ away from the air-water or air-soil interface [*Denmead et al.*, 1982; *Fillery et al.*, 1984; *Freney et al.*, 1981]. The pH affects the equilibrium between NH₄⁺ and NH₃, as the relative concentration of NH₃ increases from 0.1 to 50% as pH increases from 6 to ~9 [*Freney et al.*, 1983]. Both the volatilization process itself and the nitrification process can reduce NH₃ volatilization by decreasing NH₄⁺ availability and by producing acidity [*He et al.*, 1999]:



The CEC influences the ammoniacal N concentration through the reaction of positively charged NH₄⁺ with the negatively charged cation exchange sites. Hence soils with low CECs are more prone to high NH₃ volatilization losses than are soils with high CECs.

[9] Plants affect the NH₃ volatilization in various ways. Uptake of NH₄⁺ decreases the quantity of ammoniacal N in soil solution and increases acidity. The plant cover also influences the wind speed, temperature, and moisture conditions at the soil or water surface. For example, temperatures are generally lower and conditions are generally more humid under a grass cover, leading to lower NH₃ volatilization than from bare soil surfaces.

[10] Further factors that regulate NH₃ volatilization include the level of urease activity, producing alkalinity (in the case of urea and urine application), the availability of moisture (rainfall during or just after fertilizer application reduces NH₃ volatilization), soil organic carbon and texture (both determining soil moisture characteristics and CEC), and the presence of plant residues [*Al-Kanani and MacKenzie*, 1992; *Grant et al.*, 1996; *Keller and Mengel*, 1986; *McInnes et al.*, 1986].

[11] Wetland rice systems have a number of specific characteristics important for NH₃ volatilization. In flooded rice fields the pH of floodwater appears to be synchronized with the cycles of photosynthesis and net respiration by aquatic biota (algae, azolla), i.e., the depletion and addition of CO₂ to the floodwater regulating the floodwater pH [*Fillery et al.*, 1984; *Mikkelsen et al.*, 1978]. Water pH values as high as 10 and diurnal variations of 2–3 units can occur in shallow floodwaters populated by aquatic biota under high solar radiation, rising during midday and dropping at night [*Mikkelsen et al.*, 1978].

[12] In floodwater the assimilation of NH₄⁺ by algae and weeds (in competition with rice plants) can decrease the quantity of ammoniacal N. Apart from influencing the NH₄⁺ concentration by uptake, plants exert effects on the NH₃ exchange process proper. Many authors have reported that broadcasting urea or ammonium sulfate to the floodwater 2–3 weeks after transplanting the rice leads to higher NH₃ volatilization loss than does application a few days before panicle initiation of the rice (commonly, ~50–60 days after sowing) or at booting (~65–70 days after sowing) [*Bacon et al.*, 1988; *Fillery et al.*, 1984; *Freney et al.*, 1981; *Humphreys et al.*, 1988; *Patel and Mohanty*, 1989; *Santra et al.*, 1988]. The reason for this is that the rice crop reduces wind speed and thus reduces the NH₃ exchange between the water surface and the air. Furthermore, the crop shades the floodwater, thus reducing algal growth, causing lower pH levels and smaller amplitudes in the daily pH cycle than in fields with no or small rice plants.

[13] Incorporating or broadcasting urea to puddled soil in the absence of standing floodwater also reduces NH₃ volatilization, when compared to broadcasting onto floodwater [*Cai et al.*, 1986; *De Datta et al.*, 1989; *Freney et al.*, 1981; *Humphreys et al.*, 1988; *Obcemea et al.*, 1988; *Zhu et al.*, 1989]. This reduction may be caused by absorption of the NH₄⁺ ions at the cation exchange sites and by immobilization by microorganisms [*De Datta et al.*, 1989]. Consequently, the reduction is caused by reduced algal growth as a result of the lower ammoniacal concentration of the floodwater.

[14] A range of different measurement techniques has been employed in NH₃ volatilization studies, of which the two major ones are enclosure techniques and micrometeorological approaches. Within the group of enclosure techniques, there are a wide variety of concepts and designs. The main types of enclosures used for estimating NH₃ volatilization rates are closed systems with forced flow-through (forced draft) or closed systems without airflow. In forced draft techniques the effect of air exchange on NH₃ volatilization is often maximized [*Sommer and Ersboll*, 1996; *Vlek and Craswell*, 1979; *Vlek and Stumpe*, 1978], although the airflow-through rates that are used vary among the different studies, which makes comparisons between measured volatilization rates difficult. To minimize the disturbance of natural conditions, *Vallis et al.* [1982] and *Lockyer* [1984] developed "wind tunnel" enclosures,

in which the airflow through the tunnel can be adjusted to the wind speed outside the tunnel.

[15] Micrometeorological techniques that use analyses of NH_3 in air and meteorological measurements such as wind speed, wet bulb and dry bulb air temperatures, net radiation, and heat fluxes do not disturb the environment. These techniques can be used to determine field-scale fluxes. Different micrometeorological methods are discussed by *Denmead et al.* [1977] and *Denmead* [1983].

[16] In most enclosure and micrometeorological techniques, atmospheric NH_3 concentrations are determined by chemical trapping, followed by elution of the NH_3 with distilled water and measurement of its concentration. Various types of NH_3 traps and analytical techniques have been used.

[17] Turning to scales larger than the measurement site or field, we can make various estimates of country emissions on the basis of local measurements [e.g., *Jarvis et al.*, 1991, 1989]. At a yet larger scale, inventories of NH_3 emissions from fertilizers have been made for Europe, based on so-called “expert judgments” [*European Centre for Ecotoxicology and Toxicology of Chemicals (ECETOC)*, 1994] or laboratory experiments from one single research paper [*Asman*, 1992], and have been made for the world, based on *Asman* [1992] and additional expert judgments [*Bouwman et al.*, 1997].

[18] It is generally very difficult to predict gaseous emissions from soils, as a consequence of the complexity observed in a soil’s regulating factors. Models are widely used tools in bottom-up approaches for scaling soil gaseous emissions [*Schimmel and Panikov*, 1999]. Models are used for extrapolation of measurements to wider temporal and spatial coverage [*Bachelet and Neue*, 1993; *Bouwman et al.*, 1999]. Therefore a key issue in any upscaling of soil gaseous emissions is the need to decrease the functional complexity of the main regulating factors and processes, as observed at the site level, in relation to the adopted scale of mapping. Large-scale patterns of soil gaseous emissions, when aggregated to prolonged (e.g., seasonal or annual) timescales, may have a strong element of predictability. This is because at such spatial and temporal scales, integrated gas fluxes may be strongly related to “average” biophysical conditions [*Schimmel and Panikov*, 1999]. Therefore, if estimation of seasonal or annual emissions is the objective, the use of empirical relationships between gas fluxes and environmental and management conditions represents a suitable approach for bridging the gap between site and landscape scales.

[19] The first objective of this study was to summarize the available literature on NH_3 volatilization from application of synthetic N fertilizers and animal manure to crops and grasslands, so as to assess the factors regulating NH_3 volatilization. The second objective was to describe the relationships between regulating factors and NH_3 volatilization rates in an empirical “summary” model and to calculate global NH_3 volatilization losses. By using more measurement data and an approach based on regulating factors, the work presented in this paper is hoped to be an improvement of previous estimates of NH_3 volatilization [*Bouwman et al.*, 1997] based on emission factors.

[20] The methods used for analyzing and summarizing the literature data and the approaches used for upscaling to the global scale will be presented in section 2. Section 3 presents the results, while conclusions are discussed in section 4.

2. Data and Methods

2.1. Handling Measurement Data From the Literature

[21] We used data for ~1900 NH_3 volatilization measurements reported primarily in the peer-reviewed literature. This data set is described in detail by *FAO/IFA* [2001]. Those measurements that

included the use of chemicals such as algicides, urease, and nitrification inhibitors were excluded from our study because their use is still very limited on the global scale [*Trenkel*, 1997]. The resulting data set comprises 1667 measurements from 148 different studies. These include both laboratory and field experiments based on a range of different measurement techniques to measure NH_3 volatilization rates for different crops and uncropped systems, different soil types, climates, fertilizer types and rates, and methods and timing of fertilizer application.

[22] The information collected in the data set for measurements of NH_3 volatilization rates in upland and flooded systems is described in detail by *FAO/IFA* [2001]. The data set includes literature reference, soil type, texture/other soil properties, soil organic carbon content, soil drainage, pH, CaCO_3 content, CEC, temperature and precipitation during measurements, flooding (if applicable), residues left in the field, crop and fertilizer type, fertilizer application method and form, N application rate, NH_4^+ rate (for organic fertilizers), NH_3 loss (total over measurement period), length of measurement period, measurement technique, frequency of measurements, NH_3 volatilization rate (percent of N application and percent of N application accounting for control), and additional relevant information on measurement (e.g., volume of air flowing through forced draft chambers, specific characteristics of fertilizer used, and specific weather events important for explaining measured volatilization rates). Additional information collected for wetland rice systems included floodwater pH and presence of azolla.

[23] Some factors, like rainfall after fertilizer application and wind speed, are directly related to weather conditions, while others (e.g., algal growth and floodwater pH in wetland rice systems) are indirect. Such factors cannot be used for making predictions because geographical information is not available to scale up possible relationships found and because they should represent average conditions, excluding the effects of weather events.

[24] Further factors not used are related to the measurement technique, like frequency of measurements (which is inherently related to the measurement technique used) and the length of the measurement period, because most measurements were intended to determine the total fertilizer N lost by NH_3 volatilization). For some other factors the data were scant. These factors (soil type, soil drainage, calcium carbonate content, and crop residue management) were also excluded from the data summary. The factors that were selected for the data summary include the measurement technique and location (field or laboratory), crop type, fertilizer type, application rate, mode and timing of application, climate, soil pH, CEC, organic carbon content, and texture.

[25] Crop types were grouped into broad classes, i.e., grass, upland crops, and flooded systems (mainly wetland rice). In upland systems, fertilizer application at seeding was assumed; hence the soil surface is bare. This meant that all field and laboratory experiments with nonflooded soils were considered as systems with bare soils.

[26] Differences in soil conditions are described using functional groupings based on soil texture, soil organic carbon content, CEC, and soil reaction (pH). Although soil analytical methods vary between laboratories [*Pleijster*, 1989], these differences could not be considered explicitly. Four broad classes for soil pH were used; these were $\text{pH} \leq 5.5$, $5.5 < \text{pH} \leq 7.3$, $7.3 < \text{pH} \leq 8.5$, and $\text{pH} > 8.5$, commensurate with the classes considered on the global soil pH database [*Batjes*, 1997], and they are available for the upscaling exercise. Similarly, CEC values were placed into four main classes: $\text{CEC} \leq 16$, $16 < \text{CEC} \leq 24$, $24 < \text{CEC} \leq 32$, and $\text{CEC} > 32 \text{ cmol kg}^{-1}$. Soils were grouped for organic carbon content (SOC) as follows: $\text{SOC} \leq 1$, $1 < \text{SOC} \leq 2.5$, $2.5 < \text{SOC} \leq 5$, and $\text{SOC} > 5\%$. Soil texture was classified as coarse (including sand, loamy sand, sandy loam, loam, silt loam, and silt), medium (sandy clay loam, clay

loam, and silty clay loam), and fine (sandy clay, silty clay, and clay). Temperatures during the measurements were put into two classes, i.e., $<20^{\circ}\text{C}$ and $\geq 20^{\circ}\text{C}$. Lack of information on such items as soil pH, carbon content, texture, CEC, N application rate, and temperature was indicated in the data set by flagging as NR.

[27] Working from the broad classification of data for the different factors, the data set was summarized using Genstat 5 [Payne *et al.*, 1993] in three ways:

1. Weighted mean NH_3 volatilization rates were calculated for each class of selected factors. Because values from one source are probably not independent, each data source was given equal weight to calculate these means. The weighting has no systematic influence on the result in the case of independent values. It should be noted that by weighting the NH_3 volatilization rates, only 148 degrees of freedom remain instead of the 1667 of the full data set. The weight representation given for each factor class depends on the number of studies reporting NH_3 volatilization rates for this factor class and on the number of NH_3 volatilization rates reported in each study. Since results of 148 different studies are included in the data set, the maximum value of the weight representation is 148. When, for example, a factor class occurs with nine others in only one study, the weight representation is 0.1.

2. Balanced weighted medians were calculated to correct for unbalanced features in the data. First, the NH_3 volatilization rates were log-transformed to reduce the influence of outliers. Subsequently, balanced weighted mean values of the log-transformed NH_3 volatilization rates were calculated for each class of the selected factors using the residual maximum likelihood (REML) directive of Genstat. The means calculated this way can be considered to be mean effects of factor levels adjusted for any lack of balance in the other factors, i.e., the means that can be expected if the data had been orthogonal [Payne *et al.*, 1993].

The residual distribution of the log-transformed values is closer to a normal distribution than that of the original values. Back-transformation yields values that can be considered as balanced weighted median values of the NH_3 volatilization rate. Median values were considered to be more representative of the published data and therefore were more appropriate in view of the objective of this study than mean values.

We did not look to see if specific combinations of factor classes give different NH_3 volatilization rates (interaction effects). Reasons for this were that (1) analyzing the data set for all such combinations is complicated given the number of factors and classes and (2) we did not have a priori knowledge of such combinations.

3. A linear regression model was derived for log-transformed weighted values of NH_3 volatilization rates. We will refer to this as the summary model, developed with the aid of the stepwise regression technique whereby only factors having a variance ratio >4 were included [Payne *et al.*, 1993]. Thus only those factors having a clear influence on NH_3 volatilization rates were selected. The model was based on 1600 measurements. Seven measurements considered too extreme, even after log transformation, were excluded because they had large residuals or had both large residuals and leverages (that is, their influence on the model was considered to be very large).

2.2. Information and Assumptions for Scaling Up

[28] The summary model was used in a geographic information system to calculate NH_3 volatilization losses from fertilizer and animal manure application on the global scale. Various sources of statistical data and geographical information were combined in this study, including data on land use and application of synthetic fertilizers and animal manures to croplands and grasslands. These data were used in combination with assumptions regarding the

location and extent of different types of grasslands and fertilizer management. The approach used is discussed briefly here. Details are given by *FAO/IFA* [2001].

[29] The land cover/use distribution was taken from *Zuidema et al.* [1994], who constructed their global database on the basis of maps of natural vegetation, soil properties, and climate in combination with statistical information from *FAO* [1992]. The data on spatial distribution used from *Zuidema et al.* [1994] are the areas of grassland, rice, and other crops. These areas were updated with 1995 data from the Food and Agriculture Organization (FAOSTAT Database Collections, available at <http://apps.fao.org/page/collections>) (hereinafter referred to as FAOSTAT database). As the resolution is 0.5° , there must be errors in the gridded data for grid cells with only a partial land coverage. For example, the total land area within a 0.5° grid cell may be $<50\%$ of the grid cell area and thus is not represented. Such problems may occur on islands and coastal areas.

[30] The statistical data available on grassland management is scant. However, in order to extrapolate NH_3 volatilization from fertilizer and manure application, it is necessary to know the location of more-or-less intensively used grasslands. Some simple assumptions were made. First, three types of grassland were defined, i.e., extensive, intensive, and fertilized. Intensively used grasslands receive inputs from animal manure and are defined as grassland located within arable areas, generally in grid cells where arable land makes up at least one third of the area. For many countries, exceptions had to be made to avoid unrealistic application rates of animal manure [see *FAO/IFA*, 2001]. For some countries, we allocated the fertilized grasslands within the areas of intensive grasslands, receiving additional nutrient inputs from synthetic fertilizers, as reported by *FAO/IFA/IFDC* [1999]. Extensive grasslands consisting of the remaining grasslands given by *Zuidema et al.* [1994] were not considered since they are grazed and deliberate application of fertilizers or manure was assumed to be negligible.

[31] For wetland rice fields, the N fertilizer use per hectare was, with a few exceptions, taken directly from *FAO/IFA/IFDC* [1999]. The N fertilizer use for upland crops was calculated as the difference between total N fertilizer use from *IFA* [1999] and the sum of the N use in wetland rice and grasslands, taken from *FAO/IFA/IFDC* [1999]. Corrections were necessary for some countries [see *FAO/IFA*, 2001].

[32] The mix of fertilizers applied was assumed to be the same for grasslands and croplands. Where the fertilizer types applied in grasslands differ from those applied in croplands, errors may occur. However, fertilizer use in grasslands occurs in only a small number of countries, mainly in Europe. Some of the fertilizer categories used by *IFA* [1999] are single N fertilizers (AS, ammonium sulfate; U, urea; AN, ammonium nitrate; CAN, calcium ammonium nitrate; and AA, anhydrous ammonia). Other categories, however, consist of compound fertilizers (NK, NP, and NPK) or of two or more different fertilizer types (AP, ammonium phosphate, which is composed of monoammonium and diammonium phosphate). Information that is not given by *IFA* [1999] was used for the composition of some fertilizer categories. The category "other straight N" occurs mainly (90%) in China, where it is primarily (95%) ammonium bicarbonate (ABC); the remainder of other straight N is ammonium chloride; the global composition (80% DAP, diammonium phosphate, and 20% MAP, monoammonium phosphate) was used for ammonium sulfate, and the compound NK consists of mainly KNO_3 (K. Isherwood, personal communication, 2000). The composition of the fertilizer categories NP-N, NPK-N, and N solutions is uncertain. For the compounds NP and NPK, we assumed a mix of all NP and NPK compound fertilizers present in the data set of measurements. For N solutions, we used the data present in the data set for all N fertilizers applied in liquid form.

[33] For estimating N excretion by animals, we used animal population data from FAO (FAOSTAT database) and estimates of annual N excretion per animal from *Mosier et al.* [1998] for dairy cattle, nondairy cattle, pigs, and poultry. Not all animal manure is available as fertilizer. Animal excreta from cattle, pigs, and poultry that is available for spreading is all excreta, except excretions in grazing areas, use of manure as a fuel, and NH_3 volatilization from stored manure (based on work by *Mosier et al.* [1998]). Sheep and goats are essentially grazing animals, and their manure was considered to be unavailable for spreading. Globally, half the animal excreta used as fertilizer is assumed to be applied in croplands, and the other half is assumed to be applied in grasslands (based on work by *Lee et al.* [1997]). We applied this global distribution over crops and grass to all countries, except for those countries where the area of grasslands from *FAO/IFA/IFDC* [1999] exceeded the area given by *Zuidema et al.* [1994]. In such cases the percentage of the manure applied to grasslands had to be adjusted [see *FAO/IFA*, 2001], leading to adjustments in the manure available for crops. Manure N application rates for wetland rice and upland crops were assumed to be equal. The distribution of manure over crops and grass is uncertain, and errors caused by the above assumptions may occur in the estimated emissions from crops and grasslands, while total emissions and emissions by fertilizer type are not affected.

[34] The most poorly known aspect of the upscaling is fertilizer management. Although much is known about the local and country scales, it is difficult to generalize such information in a global inventory. Much of the fertilizer applied to rice in Southeast Asia is either broadcast directly onto flooded soil 14–21 days after transplanting and incorporated by harrowing or is broadcast directly onto flooded soil after transplanting [*Fillery et al.*, 1986; *Mikkelsen et al.*, 1978; *Obcemea et al.*, 1988]. Broadcasting is also common on grasslands.

[35] Therefore the general assumption is that fertilizers are applied as a basal application by broadcasting. A few exceptions include anhydrous ammonia, which is commonly injected, and fertilizer solutions. For anhydrous ammonia (AA), estimates are based on incorporation, and for N solutions, estimates are based on the application mode, “s”, for N solutions. Animal manure used in rice cultivation is assumed to be incorporated.

[36] It is difficult to estimate the temperature at the time of fertilizer application (generally the beginning of the growing season). Therefore the temperature at fertilizer application was simply assumed to be $\geq 20^\circ\text{C}$ between 40°N and 40°S . At higher latitudes (north and south) the temperature was assumed to be $< 20^\circ\text{C}$. Information on soil properties was taken from *Batjes* [1997], who prepared data sets of, for example, soil pH and CEC with $0.5^\circ \times 0.5^\circ$ resolution (see section 2.1).

3. Results and Discussion

[37] The mean and balanced median values for laboratory studies exceed those for field studies by 47 and 64%, respectively (Table 2). Higher NH_3 volatilization rates from laboratory studies may be caused by various factors, including the measurement technique (laboratory studies generally use forced draft enclosures aimed at determining the maximum NH_3 volatilization). However, the environmental conditions in the laboratory may also favor NH_3 volatilization. Finally, in most cases (except in greenhouse studies) the soils in the laboratory enclosure studies were uncropped; this may also favor NH_3 volatilization. It is, however, difficult to explain why the relative difference in the balanced medians between laboratory and field studies is greater than the difference in the mean values.

[38] The measurement technique used to determine NH_3 volatilization also plays an important role. For example, the N15-based

measurements and indirect open measurement yielded high values for means and yielded balanced medians for the NH_3 volatilization rate in comparison to the other techniques (means for both techniques are higher than 0.4, and balanced medians are higher than 0.1). The mean value of 0.203 for the forced draft technique (in cubic feet per day) is higher than that for micrometeorological techniques (in meters) (0.164), while the balanced medians show the reverse order (0.047 and 0.066 for cubic feet per day and meters, respectively). The estimates for both techniques are represented by a large number of observations in the database (weight representation of 56 for cubic feet per day and 40 for meters), providing a much firmer basis than the data available for the other techniques.

[39] The mean values for different types of crops show clear differences (mean for grass is 20% lower than that for upland crops and is 10% lower than that for flooded systems). The mean values confirm the expectation that NH_3 volatilization rates are generally lower in grasslands than in croplands, but the balanced medians show almost no difference.

[40] The effect of the type of fertilizer applied on the NH_3 volatilization is, as expected, very important. Differences between fertilizer types occur both in the mean and balanced median, with the highest values for ammonium nitrate applied to grassland (mean = 0.280, median = 0.204), manure (mean = 0.212, median = 0.160), and urea (mean = 0.210, median = 0.140), and with the lowest values for calcium nitrate (mean = 0.005, median = 0.01) and anhydrous ammonia (mean = 0.001, median = 0.029).

[41] Broadcasting and application of fertilizer in liquid form result in similar balanced median values (0.103 and 0.086, respectively), while incorporation leads to an important reduction of 50% compared to broadcasting. In rice systems the application of fertilizer before inundation (b/f; i/f) and application at panicle initiation (b/w/pi) have 50% lower balanced median values than broadcasting application to the flooded water (b; b/w). Hence our findings for fertilizer application mode agree with the literature summarized in section 1. Although differences between the mean values for different N fertilizer application rates seem to be consistent with those between the balanced medians, there is no clear relation between N application and NH_3 volatilization rate. The results for soil properties agree to various extents with the expectations based on studies in the literature. The balanced medians for soil pH > 8.5 exceed those for pH 5.5–7.3 by 61% and exceed those for pH < 5.5 by 80%. The balanced medians for soil pH 7.3–8.5 exceed those for pH 5.5–7.3 by 39% and exceed those for pH < 5.5 by 55%. The mean values for the different CEC classes show a consistent pattern, with lower NH_3 volatilization in soils with high CEC than in soils with low CEC. The balanced medians are also ~40% lower for CEC $> 32 \text{ cmol kg}^{-1}$ than for soils with CEC $< 32 \text{ cmol kg}^{-1}$. However, in the balanced medians the relationship between NH_3 volatilization rates and CEC for soils with CEC $< 32 \text{ cmol kg}^{-1}$ has disappeared.

[42] The effect of soil organic carbon content is not clear. Both the mean and balanced median values are greater for the lowest soil organic carbon class (mean = 0.208, median = 0.070) than for the second lowest (mean = 0.164, median = 0.059). The number of observations is small in classes 3 and 4, and estimates are less reliable. The effect of soil texture is not clear, with high means and balanced medians for the NH_3 volatilization rate for medium textured topsoils (mean = 0.207, median = 0.116) and with lower values for both fine (mean 0.188, median 0.096) and coarse textured soils (mean = 0.164, median = 0.085).

[43] After we studied the effect of each factor separately, the next step was to calculate the combined effect of different factors determining NH_3 volatilization rates by means of linear

Table 2. Means and Balanced Weighted Medians of NH₃ Volatilization Rates and Weight Representation for Different Factors Analyzed^a

	Mean	Balanced Weight Median	Weight Representation
<i>Measurement Location</i>			
Field	0.159	0.054	101
Laboratory	0.235	0.089	47
<i>Measurement Technique^b</i>			
c	0.114	0.049	13
cf _d	0.203	0.047	56
cs _o	0.140	0.044	16
ioc	0.445	0.113	1
m	0.164	0.066	40
N15	0.400	0.116	2
Nbal	0.289	0.082	5
ocb	0.260	0.089	1
wt	0.175	0.057	15
<i>Crop type</i>			
Upland	0.197	0.068	77
Grass	0.159	0.070	33
Flood	0.174	0.070	38
<i>Fertilizer Type^c</i>			
AS	0.187	0.112	12
U	0.210	0.140	74
AN	0.081	0.048	4
CAN	0.022	0.022	2
AA	0.001	0.029	1
Nsol	0.044	0.034	0
CN	0.005	0.010	0
ABC	0.152	0.130	3
UAN	0.124	0.105	5
MAP	0.094	0.025	0
DAP	0.138	0.089	3
U + DAP	0.194	0.122	1
U + MAP	0.057	0.036	1
UP	0.089	0.054	1
UUP	0.170	0.111	1
Manure	0.212	0.160	18
Grazing	0.058	0.038	3
Urine	0.147	0.142	6
AN + grazing	0.280	0.204	1
Uc	0.134	0.093	4
U + KC1	0.177	0.099	2
U + Ca	0.264	0.154	2
UCN	0.062	0.045	1
U + FYM	0.143	0.095	2
<i>Application Mode^d</i>			
b; b/w	0.203	0.103	94
i	0.138	0.051	20
s	0.179	0.086	22
b/f; i/f	0.119	0.053	6
b/w/pi	0.059	0.050	2
NR	0.116	0.092	6
<i>N Application Rate (N_{appl}), kg N ha⁻¹</i>			
N _{appl} ≤ 50	0.134	0.060	13
50 < N _{appl} ≤ 100	0.193	0.074	58
100 < N _{appl} ≤ 150	0.170	0.071	24
150 < N _{appl} ≤ 200	0.158	0.060	17
200 < N _{appl} ≤ 250	0.190	0.070	11
N _{appl} > 250	0.174	0.079	18
NR	0.278	0.072	8
<i>Temperature, °C</i>			
< 20	0.178	0.069	37
≥ 20	0.212	0.071	26
NR	0.176	0.068	85

Table 2. (continued)

	Mean	Balanced Weight Median	Weight Representation
<i>Soil pH</i>			
pH ≤ 5.5	0.153	0.051	22
5.5 < pH ≤ 7.3	0.174	0.057	66
7.3 < pH ≤ 8.5	0.215	0.079	32
pH > 8.5	0.221	0.092	2
NR	0.188	0.075	25
<i>Soil CEC, cmol kg⁻¹</i>			
CEC ≤ 16	0.190	0.079	30
16 < CEC ≤ 24	0.187	0.075	24
24 < CEC ≤ 32	0.183	0.083	12
CEC > 32	0.175	0.050	12
NR	0.180	0.065	70
<i>Soil Organic Carbon, Content %</i>			
SOC ≤ 1%	0.208	0.070	25
1 < SOC ≤ 2.5%	0.164	0.059	42
2.5 < SOC ≤ 5%	0.183	0.074	13
SOC > 5%	0.163	0.085	6
NR	0.187	0.062	61
<i>Soil Texture</i>			
Coarse	0.164	0.085	30
Medium	0.207	0.116	63
Fine	0.188	0.096	29
Organic	0.026	0.025	1
NR	0.145	0.068	25

^aMean and balanced weight median are expressed as a fraction of the fertilizer N applied. NR means not reported.

^bAbbreviations are c, closed chamber technique; cfd, chamber technique with forced draft; cso, semi-open chamber technique; ioc, indirect open measurement in air with comparison with standard fluxes; m, micrometeorological methods; N15, ¹⁵N isotope recovery; Nbal, N balance method (N balance of soil-crop system); Ndif, N difference method (input at $t = 0$ minus Nsoil at $t = t$ for laboratory experiments with bare soil); ocb, open and closed containers where the difference in N content in soil plus water is assumed to be equal to the NH₃ volatilization; wt, wind tunnel.

^cAbbreviations are AS, ammonium sulfate; U, urea; AN, ammonium nitrate; CAN, calcium ammonium nitrate; AA, anhydrous ammonia; Nsol, nitrogen solutions; CN, calcium nitrate; ABC, ammonium bicarbonate; UAN, urea ammonium nitrate; MAP, mono-ammonium phosphate; DAP, diammonium phosphate; UP, urea phosphate; UUP, urea urea phosphate; Manure, animal manure; Grazing, animal excretion during grazing in pasture; urine, application of urine solution; Ue, coated urea; KCl, potassium chloride; Ca, calcium; UCN, urea-calcium nitrate; FYM, farm yard manure.

^dAbbreviations are b, broadcast; b/w, broadcast to floodwater; i, incorporated; s, solution; b/f, broadcast and then flooded; i/f, incorporated and then flooded; b/w/pi, broadcast to floodwater at panicle initiation.

regression. Weather-related factors were excluded a priori from the regression (see section 2.1). From the factors selected for the data summary (see Table 2), we used only those in the regression that had a clear influence on NH₃ volatilization, except for the measurement technique and location. Despite the clear differences between both the means and balanced medians calculated for the different measurement techniques and locations (with a strong relation to the technique used, as discussed above), we decided to ignore these factors. Measurement techniques cannot be used for predictions, since the a priori knowledge for judging their accuracy is lacking, while the influence of the measurement location cannot be explained on the basis of the data set used. The regression model therefore yields an average value for NH₃ volatilization rates applying to all measurement techniques and locations included in the data set. Since our data set is dominated by forced draft and micrometeorological techniques (Table 2), the estimated volatilization rates will be somewhat higher than those based on only micrometeorological techniques and will be somewhat lower than those based on forced draft techniques.

[44] The factors N application rate, soil organic carbon content, and soil texture were not used in the regression because of the absence or lack of consistent relationships with NH₃ volatilization

rates in the data set used. Moreover, soil carbon and texture are the main determinants of soil CEC. Therefore the influence of soil C and texture on NH₃ volatilization is assumed to be included in the factor CEC.

[45] The factors that we selected for the regression on the basis of the data summary discussed above include the type of crop, fertilizer type and application mode, temperature, soil pH, and CEC. The fitted parameters for the different factor classes of the resulting summary model are presented in Table 3. The values for several individual fertilizer types were recalculated for the fertilizer categories used by IFA [1999]. The variance accounted for by the model is 28%. This implies that individual NH₃ volatilization values from research papers differ, on the average, ~15% less from the means calculated by the model than from their common mean. The model is therefore not suitable for predicting NH₃ volatilization rates from measurements in individual research papers for specific sites. The mean NH₃ volatilization rates for factor class combinations is, however, of more interest for upscaling to “landscape” conditions than are individual measurements.

[46] It should be noted that when used to predict NH₃ volatilization rates, the regression should, in fact, be performed on the basis of the data used in the upscaling. However, for laboratory

Table 3. Factor Class Values for Type of Crop, Fertilizer Type, Application Mode, pH, CEC and Climate Determined by Linear Regression^a

Factor	Value
<i>Factors related to Management</i>	
Crop type	
Upland crops	-0.045
Grass	-0.158
Flooded crops	0
Fertilizer type ^b	
Ammonium sulfate	0.429
Urea	0.666
Ammonium nitrate	-0.35
Calcium ammonium nitrate	-1.064
Anhydrous ammonia	-1.151
Other straight N	-0.507
N solutions	-0.748
Ammonium phosphates	0.065
Other compound NP	0.014
Compound NK	-1.585
Compound NPK	0.014
Ammonium bicarbonate	0.387
Animal manure	0.995
Application mode ^c	
b	-1.305
i	-1.895
s	-1.292
b/f; i/f	-1.844
b/w/pi	-2.465
<i>Factors Related to Environmental Conditions</i>	
Soil pH	
pH ≤ 5.5	-1.072
5.5 < pH ≤ 7.3	-0.933
7.3 < pH ≤ 8.5	-0.608
pH > 8.5	0
Soil CEC	
CEC ≤ 16	0.088
16 < CEC ≤ 24	0.012
24 < CEC ≤ 32	0.163
CEC > 32	0
Climate	
Temperate climate	-0.402
Tropical climate	0

^aWeighted log-transformed values are given. NH₃ volatilization rate is calculated as $\exp(\text{factor value for crop type} + \text{fertilizer type} + \text{application mode} + \text{soil pH} + \text{soil CEC} + \text{climate})$. For example, for grass fertilized with U by broadcasting fertilizer on soil with $5.5 < \text{pH} \leq 7.3$ and CEC of $16 < \text{CEC} \leq 24$ in a temperate climate, volatilization rate is calculated as $\exp(-0.158 + 0.666 - 1.305 - 0.933 + 0.012 - 0.402) = \exp(-2.120) = 0.120$. Hence volatilization as a fraction of urea N application is 0.120.

^bFactor value for ammonium phosphates is calculated on basis of global composition of 80% DAP and 20% MAP; compound NK (mainly KNO₃) uses value for CN. Factor values for other compound fertilizers are based on various compound NP and NPK fertilizers in data set of measurements; for N solutions, we used data collected for all N fertilizers applied in solution.

^cSee Table 2 for explanation.

measurements, this is not possible. Furthermore, the resolution of $0.5^\circ \times 0.5^\circ$ in the maps used allows only a generalized representation of environmental and management conditions on the landscape scale and is therefore not suitable to be used in combination with local field measurements.

[47] The summary model was used to calculate global NH₃ volatilization rates, with results summarized in Tables 4 and 5 and Figures 1 and 2. The global annual NH₃ loss from synthetic fertilizers is 11 million tons N, which is 14% of the applied N. The results indicate that NH₃ volatilization loss in developing countries exceeds that in industrialized countries by a factor of 4.4. The global NH₃ loss from the 11.8 million tons of synthetic fertilizer N used annually in wetland rice cultivation amounts to 2.3 million tons, or 20% of the N application. Most of this loss occurs in developing countries (97%). In upland crops a global 14% of the 61.7 million tons of synthetic fertilizer N is lost as NH₃, with higher loss rates in developing (18%) than in industrialized countries (8%). In grasslands the annual global use of synthetic fertilizer N is only 4.3 million tons, with estimated loss rates of 13% for developing and 6% for industrialized countries. Close to 100% of the synthetic N fertilizer use in grasslands is in industrialized countries.

[48] Higher NH₃ losses from N fertilizers in developing countries are due to high temperatures and due to the widespread use of urea (60% of N fertilizer use) and ammonium bicarbonate (19%), which are both prone to high NH₃ volatilization losses. In industrialized countries the use of urea makes up only 16% of fertilizer use; ammonium bicarbonate is not used at all.

[49] The global annual NH₃ loss from the use of 12.4 million tons N in animal manure in grasslands amounts to 2.7 million tons N, i.e., 22% of the N application (20% in industrialized and 25% in developing countries). About 60% of the global NH₃ volatilization loss stems from industrialized countries. In upland crops a global 26% of the 17.4 million tons N from the annual application of animal manure is lost as NH₃, with higher loss rates in developing (29%) than in industrialized countries (22%). The volume of animal manure applied annually to upland crops is 8.6 million tons N in industrialized and 8.8 million tons N in developing countries. In wetland rice systems the estimated annual use of N from animal manure is 3.3 million tons, mainly in developing countries. As incorporation of animal manure is assumed to prevail in rice cultivation, the NH₃ loss rates are lower than for upland crops (17% in developing and 16% in industrialized countries).

[50] The global mean NH₃ volatilization rates for the fertilizer types distinguished by IFA [1999] and based on the summary model are compared with other inventories from the literature in Table 6. It should be noted that the volatilization rates resulting from the global upscaling differ from the balanced median values. This is because the summary model used includes the influence of the selected environmental and management factors, while in the balanced median these influences are eliminated.

[51] The global loss rate calculated in this study for synthetic fertilizers is 25% higher than the estimate of Bouwman *et al.* [1997]. This is caused by differences in the global estimates for some of the individual fertilizer types. Our results, summarized in Table 6, are in general agreement with the estimates used by ECETOC [1994] and Bouwman *et al.* [1997], except for ammonium sulfate, ammonium bicarbonate, and the compound fertilizers including MAP and DAP. The estimate for AS has a weight representation of 12. This is based on 176 measurements from a great number of different research papers and from different sites with different conditions; the results can be considered to be representative for a wide range of environmental and management conditions. The data on ammonium bicarbonate represent a smaller but still considerable number of measurements.

[52] For the compound fertilizers (except NK, which is primarily KNO₃), lack of data on the exact composition causes uncertainty. However, the volatilization rates found for all P-containing N fertilizers in the data set are higher than the estimates presented by ECETOC [1994] and Bouwman *et al.* [1997] (Tables 2 and 6).

Table 4. Area, Use of Synthetic N Fertilizers, and NH₃ Volatilization Loss for Fertilized Grasslands, Upland Crops, and Wetland Rice for Different World Regions for 1995

Region ^a	Fertilized Grasslands			Upland Crops			Wetland Rice		
	Area, Mha	N use, ^b kt	NH ₃ -N loss, kt	Area, Mha	N use, ^b kt	NH ₃ -N loss, kt	Area, Mha	N use, ^b kt	NH ₃ -N loss, kt
1 Canada	0	0	0	46	1576	140	0	0	0
2 U.S.A.	0	0	0	189	10982	788	1	168	15
3 Central America	1	25	3	40	1392	215	0	32	5
4 South America	1	12	1	109	2049	348	3	234	43
5 North Africa	0	0	0	21	1126	214	1	78	16
6 Western Africa	0	0	0	73	130	19	1	26	4
7 Eastern Africa	0	0	0	40	109	17	1	1	0
8 Southern Africa	3	31	3	42	477	51	0	3	0
9 OECD Europe	37	3074	156	90	6384	448	0	32	3
10 Eastern Europe	3	210	13	48	1834	123	0	1	0
11 Former U.S.S.R.	33	760	59	229	1856	157	1	14	1
12 Middle East	4	17	3	57	2305	422	1	71	18
13 South Asia	0	0	0	162	8295	1828	44	4646	1031
14 East Asia	0	0	0	69	19855	3318	26	4490	829
15 Southeast Asia	0	0	0	52	2405	421	35	1811	335
16 Oceania	20	175	23	49	639	108	0	12	2
17 Japan	0	27	4	2	265	36	2	171	24
Total	103	4331	265	1319	61678	8654	117	11788	2328

^aRegions designated for global change research, as defined by *Kreileman et al.* [1998] and as indicated in Figures 1 and 2.

^bSomewhat lower here than total presented by *IFA* [1999], because of scaling errors.

This may be caused by the influence of phosphate, which may change the environment to favor increased NH₃ loss by precipitating Ca [*Fenn et al.*, 1990]. Both the formula (pH) and form of the phosphate added can influence its reaction with Ca and therefore can influence NH₃ volatilization.

[53] Uncertainties in the results of the upscaling stem from uncertainties in the summary model as such and from uncertainties caused by scaling errors. From the standard errors we estimated the range of NH₃ volatilization rates for the various fertilizers. The standard errors depend on the combination of

factor classes selected, but appeared to be very similar for individual fertilizer types across crop types, climate, and soil conditions. Therefore we have calculated the range of volatilization rates for a number of combinations of factor classes for each crop type and fertilizer type. This was done on the basis of twice the standard errors, to include 95% of the observations, and applied the average deviation from the model results to the estimated global NH₃ volatilization loss for each fertilizer category (Table 6). The resulting range in the estimates for the global NH₃ volatilization loss from all fertilizers is 10–19% for

Table 5. Area, Use of Animal Manure N, and NH₃ Volatilization Loss for Intensively Used Grasslands, Upland Crops, and Wetland Rice for Different World Regions for 1995^a

Region ^b	Intensive Grasslands			Upland Crops			Wetland Rice		
	Area, Mha	N use, kt	NH ₃ -N loss, kt	Area, Mha	N use, kt	NH ₃ -N loss, kt	Area, Mha	N use, kt	NH ₃ -N loss, kt
1 Canada	20	207	41	46	207	45	0	0	0
2 U.S.A.	84	1583	366	189	1573	394	1	10	2
3 Central America	22	351	92	40	349	105	0	3	0
4 South America	59	1051	271	109	1020	291	3	32	5
5 North Africa	10	34	11	21	33	12	1	3	1
6 Western Africa	48	137	35	73	137	39	1	3	1
7 Eastern Africa	26	148	40	40	143	43	1	6	1
8 Southern Africa	24	78	20	42	78	23	0	2	0
9 OECD Europe	50	3085	561	90	3402	684	0	7	1
10 Eastern Europe	18	737	130	48	757	149	0	0	0
11 former U.S.S.R.	177	2389	511	229	2378	555	1	15	2
12 Middle East	13	167	53	57	177	64	1	2	1
13 South Asia	10	425	114	162	2850	917	44	965	175
14 East Asia	29	1404	324	69	3500	953	26	1650	276
15 Southeast Asia	15	477	114	52	544	147	35	396	62
16 Oceania	20	52	14	49	63	19	0	0	0
17 Japan	0	59	16	2	186	51	2	175	28
Total	625	12386	2712	1319	17396	4492	117	3270	555

^aAnimal manure N estimated as all excretion from cattle, pigs, and poultry, except part excreted during grazing, use of manure as fuel, and storage losses of NH₃.

^bSee Table 4.

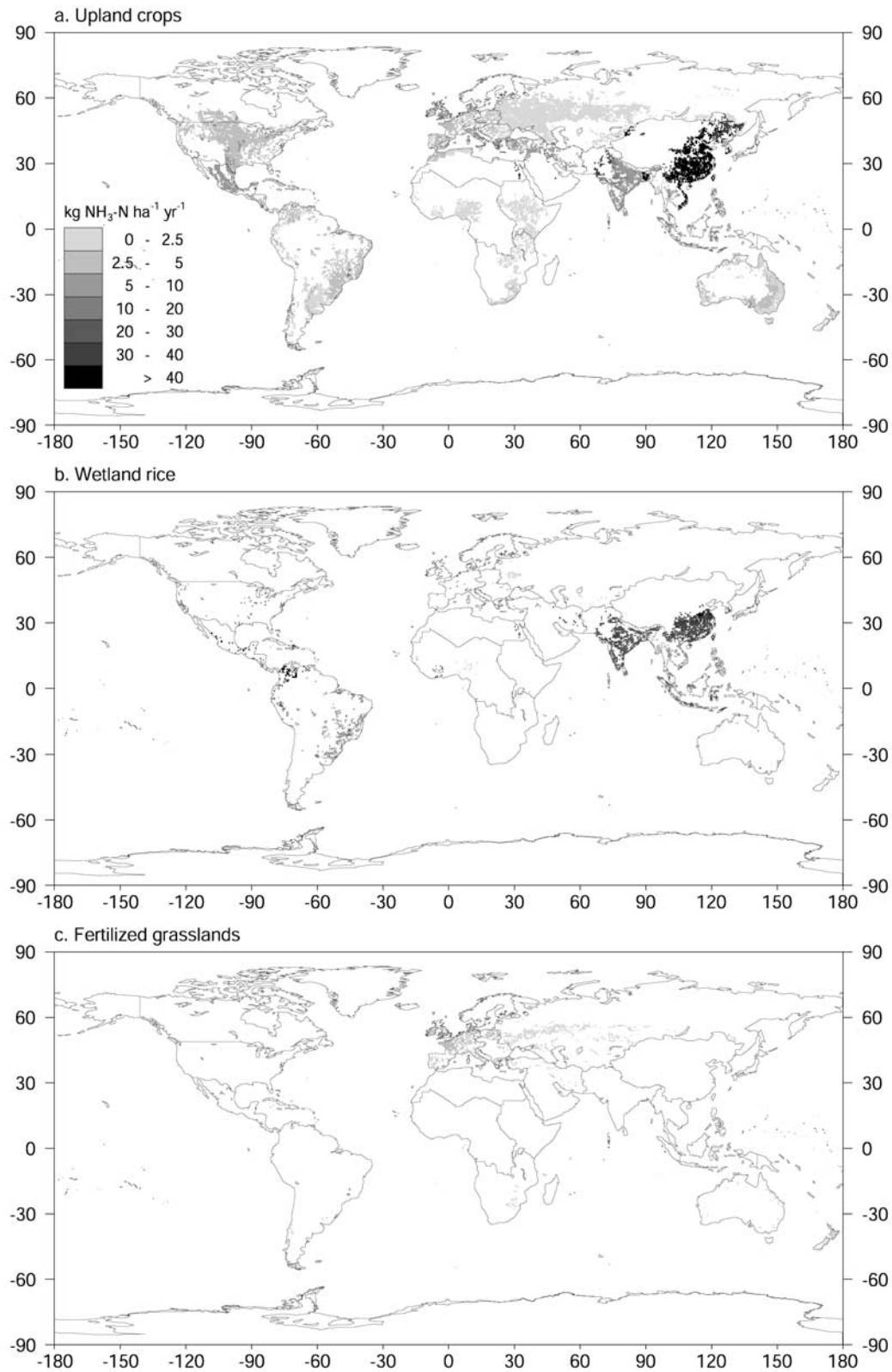


Figure 1. Estimated annual NH_3 volatilization loss for 1995 from synthetic fertilizers used in (a) upland crops, (b) wetland rice fields, and (c) grasslands. Note that emission is an annual estimate. High values may be caused by high cropping intensities, such as in China. Low values may be the result of low cropping intensities. Emission rates per hectare of harvested land may thus differ from those presented here. See color version of this figure at back of this issue.

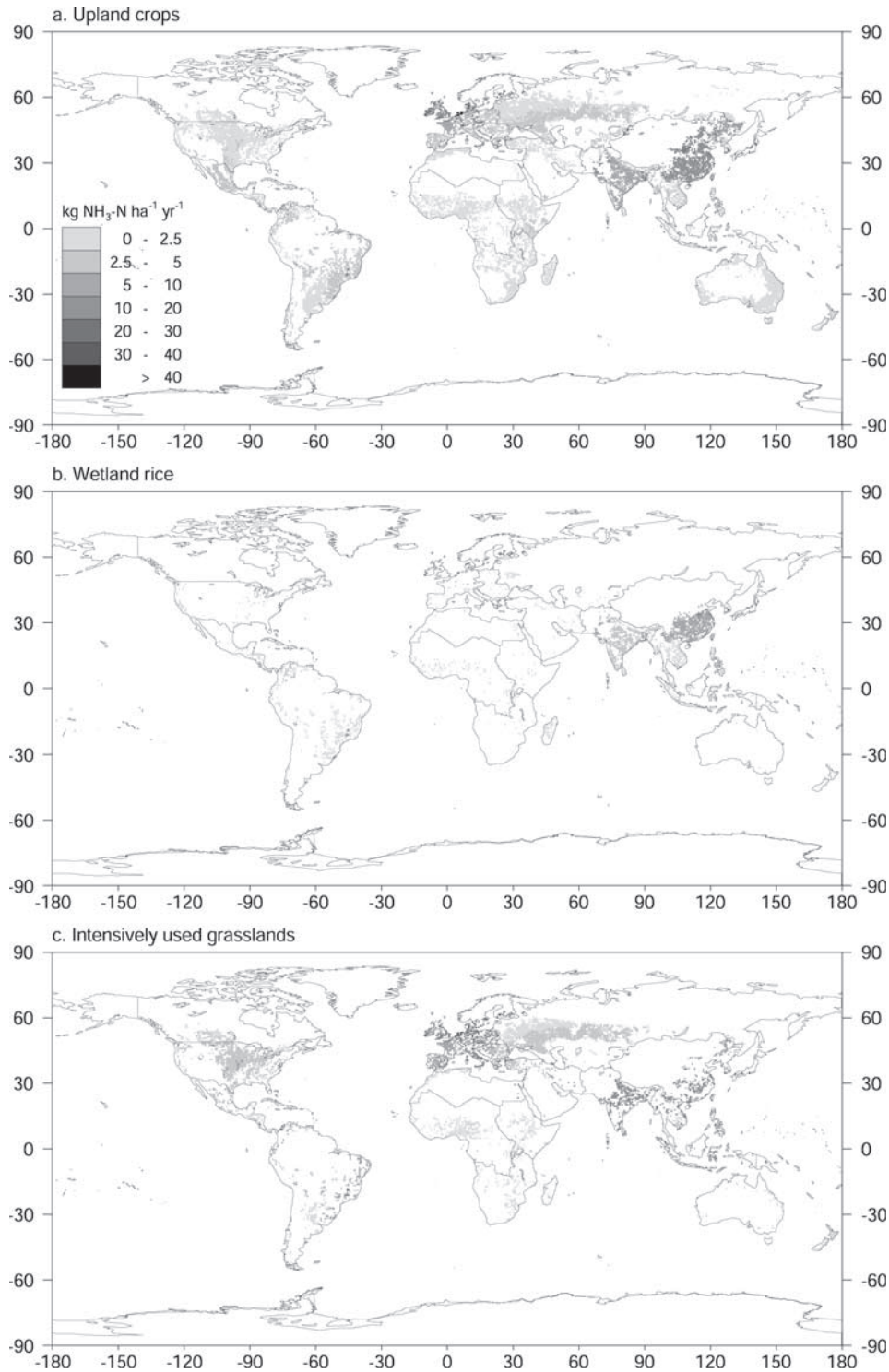


Figure 2. Estimated annual NH_3 volatilization loss for 1995 from animal manure used in (a) upland crops, (b) wetland rice fields and (c) grasslands. See color version of this figure at back of this issue.

all synthetic fertilizers and is 19–29% for animal manure. The range for individual fertilizers may be much wider, depending on the number of representations in the data set (Table 6). It should be noted that the calculated ranges neither account for the omissions in the summary model nor for scaling errors.

[54] It is very difficult to quantify uncertainties associated with scaling. Uncertainties in fertilizer use stem primarily from the grouping of different N fertilizer types into one category. The data on fertilizer use by crops have a varying degree of reliability; data are more complete and probably more certain in industrialized

Table 6. Global Consumption and Estimated Mean and Range Of NH₃ Volatilization Loss Rates for Synthetic Fertilizer Categories From IFA [1999] and for Animal Manure for 1995, Compared With Estimates of NH₃ Loss Rates Taken From Literature

Fertilizer Category	Use, Mt	NH ₃ Volatilization Loss				Compared With <i>Bouwman et al.</i> [1997], %	Compared With <i>ECETOC</i> [1994], %
		Total, Mt	This Study ^a				
			Mean, %	Range %			
Ammonium sulfate	2.4	0.4	16	12–20	8	5–15	
Urea	34.4	7.3	21	18–26	15–25	10–20	
Ammonium nitrate	7.5	0.5	6	5–9	2	1–3	
Calcium ammonium nitrate	3.6	0.1	3	2–4	2	1–3	
Ammonia, direct application	4.6	0.1	2	1–3	4	4	
Nitrogen solutions	4.0	0.2	5	2–11	2.5	–	
Other straight nitrogen ^b	10.1	1.5	15	10–22	20–30	–	
Ammonium phosphates	4.1	0.5	11	5–25	2–5	5	
Other compound NP-N	1.7	0.2	11	6–19	–	–	
Compound NK-N	0.0	0.0	2	1–5	–	–	
Compound NPK-N	6.1	0.5	9	5–16	2–4	1–5	
Total synthetic fertilizers	78.5	11.2	14	10–19	–	–	
Animal manure	33.1	7.8	23	19–29	20	20	

^aMode of application: broadcast, except for AA (incorporated), N solutions (application mode “s” for solution), and animal manure in flooded systems (incorporated).

^bIncludes use of ABC in China, accounting for 90% of global “other straight N.”

countries than in developing countries. Unfortunately, there are no statistics on fertilizer management. In the upscaling, assumptions had to be made on the mode of application of fertilizers and manure. Although it is known that application rates vary between crops and farmers within countries *FAO/IFA/IFDC* [1999], we do not know the spatial distribution of fertilizer application rates. Moreover, it is not known if certain fertilizer types are preferentially used for specific crops or grasslands. In any case, the fertilizer application rate was found not to influence the NH₃ volatilization rate, and this allowed us to scale up NH₃ losses on the basis of average country fertilizer and manure application rates. Hence we recognize that there are errors in the spatial distribution of the mix of fertilizers and their application rates and of their combination with soil conditions. However, for the 0.5° resolution used, this error was considered to be acceptable.

[55] The distribution of arable lands is known fairly well, but the distribution and the management of grasslands are highly uncertain. Although information is available on the use of synthetic fertilizers in grasslands, the application of animal manures in grasslands is based on a global estimate. For different world regions and individual countries, the application of animal manures is highly uncertain.

[56] The uncertainty in the data on animal populations is probably <10% [*Bouwman et al.*, 1997]. Most of the uncertainty in the NH₃ volatilization rate for animal manure stems from the assumptions on N excretion and waste management. For developing countries, in particular, there are no reliable data on waste management practices and on the use of animal manure in grasslands and arable lands. Therefore the largest uncertainty is probably found in tropical countries and the smallest is probably found in western Europe [*Bouwman et al.*, 1997].

[57] The emission estimates based on regulating factors presented in this paper are an update of the global estimates of NH₃ volatilization from fertilizers and animal manure based on emission factors presented by *Bouwman et al.* [1997]. The set of measurement data and references used and the emission files can be obtained from <http://www.rivm.nl/iweb>, under databases.

4. Conclusions and Recommendations

[58] The major conclusions drawn on the basis of the data summary relate to the global NH₃ volatilization estimates and to uncertainties and recommended methods to reduce uncertainties.

[59] The global NH₃ loss from synthetic fertilizers is 11 million tons N yr⁻¹, which is 14% of the applied fertilizer N. The NH₃ loss from synthetic N fertilizers used in wetland rice cultivation is 20% of the 11.8 million tons N used annually. In upland systems the NH₃ loss is 14% of the 61.7 million tons of synthetic fertilizer N used per year. In grasslands the annual global use of synthetic fertilizer N is only 4.3 million tons, mostly in industrialized countries, with estimated NH₃ loss rates of 6%. High NH₃ losses from synthetic N fertilizers in developing countries are due to high temperatures and due to the widespread use of urea (60% of N fertilizer use) and ammonium bicarbonate (19%). Both of these are prone to high NH₃ volatilization losses. In industrialized countries the use of urea makes up only 16% of N fertilizer use, and ammonium bicarbonate is not used at all.

[60] The global annual NH₃ loss from the use of 12.4 million tons N in animal manure in grasslands amounts to 23% of the N application. In upland crops a global 26% of the 17.4 million tons N from annual application of animal manure is lost as NH₃. In wetland rice systems the estimated annual use of N from animal manure is 3.3 million tons and the global NH₃ loss is 17%. In wetland rice we assumed that animal manure is incorporated, giving lower volatilization rates than for broadcasting in upland soils under grass or crops.

[61] The global results are in good agreement with recent global and European estimates of NH₃ losses for some fertilizer categories, except for ammonium sulfate, ammonium bicarbonate, and the different compound fertilizers. The data collected indicate that ammonium bicarbonate (primarily used in China) is less prone and ammonium sulfate and compound fertilizers are more prone to NH₃ volatilization than was thought earlier.

[62] There are many uncertainties in this study, including uncertainties related to the summary model and in the global upscaling. The major uncertainty in the summary model is caused by the omission of the measurement technique employed and of many factors known to be crucial controls of NH₃ volatilization, for example, wind speed, rainfall events in the period of fertilizer application, and floodwater pH in wetland rice systems.

[63] Major uncertainties occurring during upscaling are caused by the scarcity of data on the spatial and temporal distribution of application of synthetic fertilizers and animal manure by crop and on the prevailing management conditions. These uncertainties can be reduced in several ways. First, field measurements are required,

particularly in tropical agroecosystems; these are still underrepresented in the available data from the literature. Second, the upscaling can be improved significantly by using country data on fertilizer and crop residue management from an inventory of countrywide and regional data. Third, further model development may be done on spatial scales of fields. To make it applicable to that scale, the regression model would need to be modified, for example, by including measurement technique and weather-related factors. Validation of the model against micrometeorological measurements, also using atmospheric modeling combined with atmospheric measurements, would improve the empirical relationships found in this study. Simplification of such a model by upscaling and downscaling, and validation between field and landscape scales, would result in a revised summary model appropriate for global upscaling.

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References

- Al-Kanani, T., and A. F. MacKenzie, Effect of tillage practices and hay straw on ammonia volatilization from nitrogen fertilizer solutions, *Can. J. Soil Sci.*, **72**, 145–157, 1992.
- Asman, W. A. H., Ammonia emission in Europe: Updated emission and emission variations, National Institute for Public Health and the Environment, Bilthoven, 1992.
- Bachelet, D., and H. U. Neue, Methane emissions from wetland rice areas of Asia, *Chemosphere*, **26**, 219–237, 1993.
- Bacon, P. E., E. H. Hoult, L. G. Lewin, and J. W. McGarity, Ammonia volatilization from drill sown rice bays, *Fert. Res.*, **16**, 257–272, 1988.
- Batjes, N. H., A world dataset of derived soil properties by FAO-UNESCO soil unit for global modelling, *Soil Use Manage.*, **13**, 9–16, 1997.
- Black, A. S., R. R. Sherlock, N. P. Smith, K. C. Cameron, and K. M. Goh, Effects of form of nitrogen, season, and urea application rate on ammonia volatilisation from pastures, *N. Z. J. Agric. Res.*, **28**, 469–474, 1985.
- Bouwman, A. F., Nitrogen oxides and tropical agriculture, *Nature*, **392**, 866–867, 1998.
- Bouwman, A. F., and K. W. Van Der Hoek, Scenarios of animal waste production and fertilizer use and associated ammonia emission for the developing countries, *Atmos. Environ.*, **31**, 4095–4102, 1997.
- Bouwman, A. F., and D. Van Vuuren, Global assessment of acidification and eutrophication of natural ecosystems, *Rep. UNEP/DEIA and EW/TR.99-6; RIVM/4002001012*, U. N. Environ. Programme, and Natl. Inst. for Public Health and the Environ., Nairobi, Kenya; Bilthoven, the Netherlands, 1999.
- Bouwman, A. F., D. S. Lee, W. A. H. Asman, F. J. Dentener, K. W. Van Der Hoek, and J. G. J. Olivier, A global high-resolution emission inventory for ammonia, *Global Biogeochem. Cycles*, **11**, 561–587, 1997.
- Bouwman, A. F., R. G. Derwent, and F. J. Dentener, Towards reliable global bottom-up estimates of temporal and spatial patterns of emissions of trace gases and aerosols from land-use related and natural sources, in *Scaling of Trace Gas Fluxes in Ecosystems*, edited by A. F. Bouwman, pp. 1–26, Elsevier Sci., New York, 1999.
- Bussink, D. W., and O. Oenema, Ammonia volatilization from dairy farming systems in temperate areas: A review, *Nutrient Cycling Agroecosystems*, **51**, 19–33, 1998.
- Cai, G. X., Z. L. Zhu, A. C. F. Trevitt, J. R. Freney, and J. R. Simpson, Nitrogen loss from ammonium bicarbonate and urea fertilizers applied to flooded rice, *Fert. Res.*, **10**, 203–215, 1986.
- De Datta, S. K., A. C. T. Trevitt, J. R. Freney, W. N. Obcemea, J. G. Real, and J. R. Simpson, Measuring nitrogen losses from lowland rice using bulk aerodynamic and nitrogen-15 balance methods, *Soil Sci. Soc. Am. J.*, **53**, 1275–1281, 1989.
- Denmead, O. T., Micrometeorological methods for measuring gaseous losses of nitrogen in the field, in *Gaseous Loss of Nitrogen From Plant-Soils Systems. Developments in Plant and Soil Science*, vol. 9, edited by J. R. Freney and J. R. Simpson, pp. 133–157, Martinus Nijhoff, The Hague, 1983.
- Denmead, O. T., J. R. Simpson, and J. R. Freney, A direct field measurement of ammonia emission after injection of anhydrous ammonia, *Soil Sci. Soc. Am. J.*, **41**, 1001–1004, 1977.
- Denmead, O. T., J. R. Freney, and J. R. Simpson, Dynamics of ammonia volatilization during furrow irrigation of maize, *Soil Sci. Soc. Am. J.*, **46**, 149–155, 1982.
- Dhyani, B. P., and B. Mishra, Effect of nitrogen-application schedule on ammonia volatilization from field of rice (*Oryza sativa*), *Indian J. Agric. Sci.*, **62**, 73–73, 1992.
- European Centre for Ecotoxicology and Toxicology of Chemicals, *Ammonia Emissions to Air in Western Europe*, 196 pp., Brussels, Belgium, 1994.
- Fenn, L. B., H. L. Malstrom, and E. Wu, Ammonia losses from surface-applied urea as related to urea application rates, plant residue and calcium chloride addition, *Fert. Res.*, **12**, 219–227, 1987.
- Fenn, L. B., G. Tatum, and G. Horst, Ammonia losses from surface-placed mixtures of urea-calcium-potassium salts in the presence of phosphorus, *Fert. Res.*, **21**, 125–131, 1990.
- Fillery, I. R. P., and P. L. G. Vlek, Reappraisal of the significance of ammonia volatilization as an N loss mechanism in flooded rice fields, *Fert. Res.*, **9**, 79–98, 1986.
- Fillery, I. R. P., J. R. Simpson, and S. K. de Datta, Influence of field environment and fertilizer management on ammonia loss, *Soil Sci. Soc. Am. J.*, **48**, 914–920, 1984.
- Fillery, I. R. P., J. R. Simpson, and S. K. de Datta, Contribution of ammonia volatilization to total nitrogen loss after applications of urea to wetland rice fields, *Fert. Res.*, **8**, 193–202, 1986.
- Food and Agriculture Organization, *User Manual, AGROSTAT-PC Comput. Info. Ser.*, Rome, 1992.
- Food and Agriculture Organization/International Fertilizer Industry Association/International Fertilizer Development Center, *Fertilizer Use by Crop*, 4th ed., Rome, 1999.
- Food and Agriculture Organization/International Fertilizer Industry Association, Global estimates of gaseous emissions of NH₃, NO, and N₂O from agricultural land, report, 106 pp., Rome, 2001.
- Freney, J. R., and O. T. Denmead, Factors controlling ammonia and nitrous oxide emissions from flooded rice fields, *Ecol. Bull.*, **42**, 188–194, 1992.
- Freney, J. R., O. T. Denmead, I. Watanabe, and E. T. Craswell, Ammonia and nitrous oxide losses following applications of ammonia sulfate to flooded rice, *Aust. J. Agric. Res.*, **32**, 37–45, 1981.
- Grant, C. A., S. Jia, K. R. Brown, and L. D. Bailey, Volatile losses of NH₃ from surface applied urea and ammonium nitrate with and without the urease inhibitors NBPT or ammonium thiosulphate, *Can. J. Soil Sci.*, **76**, 417–419, 1996.
- He, Z. L., A. K. Alva, D. V. Calvert, and D. J. Banks, Ammonia volatilization from different fertilizer sources and effects of temperature and soil pH, *Soil Sci.*, **164**, 750–758, 1999.
- Humphreys, E., J. R. Freney, W. A. Muirhead, O. T. Denmead, J. R. Simpson, R. Leuning, A. C. F. Trevitt, W. N. Obcemea, R. Wetselaar, and C. Gui-Xin, Loss of ammonia after application of urea at different times to dry-seeded, irrigated rice, *Fert. Res.*, **16**, 47–57, 1988.
- International Fertilizer Industry Association, Nitrogen-Phosphate-Potash, IFADATA statistics from 1973/74–1973 to 1997/98–1997 including separately world fertilizer consumption statistics, Paris, 1999.
- Jarvis, S. C., D. J. Hatch, and D. H. Roberts, The effects of grassland management on nitrogen losses from grazed swards through ammonia volatilization: The relation to excretal N returns from cattle, *J. Agric. Sci.*, **112**, 205–216, 1989.
- Jarvis, S. C., D. J. Hatch, R. J. Orr, and S. E. Reynolds, Micrometeorological studies of ammonia emission from sheep grazed swards, *J. Agric. Sci.*, Cambridge, **117**, 101–109, 1991.
- Keller, G. D., and D. B. Mengel, Ammonia volatilization from nitrogen fertilizers surface applied to no-till corn, *Soil Sci. Soc. Am. J.*, **50**, 1060–1063, 1986.
- Kreileman, E., J. Van Woerden, and J. Bakkes, RIVM Environmental Research, *CIM Rep. M025/98*, Natl. Inst. for Public Health and the Environ., Bilthoven, 1998.
- Laegreid, M., O. C. Bockman, and O. Kaarstad, *Agriculture and Fertilizers*, 294 pp., CAB Int., New York, 1999.

- Lawes, J. B., and J. H. Gilbert, On agricultural chemistry, *J. R. Agric. Soc. England*, 12, 1–40, 1851.
- Lee, D. S., A. F. Bouwman, W. A. H. Asman, F. J. Dentener, K. W. Van Der Hoek, and J. G. J. Olivier, Emissions of nitric oxide and ammonia from grasslands on a global scale, in *Gaseous Nitrogen Emissions From Grasslands*, edited by S. C. Jarvis and B. F. Pain, pp. 353–371, CAB Int., New York, 1997.
- Lockyer, D. R., A system for the measurement in the field of losses of ammonia through volatilization, *J. Sci. Food Agric.*, 35, 837–848, 1984.
- McInnes, K. J., R. B. Ferguson, D. E. Kissel, and E. T. Kanemasu, Ammonia loss from applications of urea-ammonium nitrate solution to straw residue, *Soil Sci. Soc. Am. J.*, 50, 969–974, 1986.
- Mikkelsen, D. S., S. K. de Datta, and W. N. Obcemea, Ammonia volatilization losses from flooded soils, *Soil Sci. Soc. Am. J.*, 42, 725–730, 1978.
- Mosier, A. R., C. Kroeze, C. Nevison, O. Oenema, S. Seitzinger, and O. Van Cleemput, Closing the global atmospheric N₂O budget: Nitrous oxide emissions through the agricultural nitrogen cycle, *Nutrient Cycling in Agroecosystems*, 52, 225–248, 1998.
- Obcemea, W. N., J. G. Real, and S. K. de Datta, Effect of soil texture and nitrogen management on ammonia volatilization and total nitrogen loss, *Philippian J. Crop Sci.*, 13, 145–153, 1988.
- Patel, S. K., and S. K. Mohanty, Relative ammonia loss from urea-based fertilizers applied to rice under different hydrological conditions, *Fert. Res.*, 19, 113–119, 1989.
- Payne, R. W., et al., *Genstat 5 Release 3. Reference Manual*, Clarendon, Oxford, England, 1993.
- Peoples, M. B., J. R. Freney, and A. R. Mosier, Minimizing gaseous losses of nitrogen, in *Nitrogen Fertilization and the Environment*, edited by P. E. Bacon, pp. 565–602, Marcel Dekker, New York, 1995.
- Pleijsters, K., Variability in soil data, in *Land Qualities in Space and Time*, edited by J. Bouma and A. K. Bregt, pp. 89–98, Pudoc, Wageningen, 1989.
- Santra, G. H., D. K. Das, and L. N. Mandal, Loss of nitrogen through ammonia volatilization from flooded rice soils, *J. Indian Soc. Soil Sci.*, 36, 652–659, 1988.
- Saravanan, A., V. Velu, and K. M. Ramanathan, Ammonia volatilization loss in rice soils of Cauvery Delta, *Int. Rice Res. Notes*, 12, 59–60, 1987.
- Schimel, D. S., and N. S. Panikov, Simulation models of terrestrial trace gas fluxes at soil microsites to global scales, in *Scaling of Trace Gas Fluxes in Ecosystems*, edited by A. F. Bouwman, pp. 185–202, Elsevier Sci., New York, 1999.
- Smil, V., Nitrogen in crop production: An account of global flows, *Global Biogeochem. Cycles*, 13, 647–662, 1999.
- Sommer, S. G., and A. K. Ersboll, Effect of air flow rate, lime amendments, and chemical soil properties on the volatilization of ammonia from fertilizers applied to sandy soils, *Biol. Fert. Soils*, 21, 53–60, 1996.
- Thompson, R. B., B. F. Pain, and Y. J. Rees, Ammonia volatilization from cattle slurry following surface application to grassland, II, Influence of application rate, wind speed and applying slurry in narrow bands, *Plant Soil*, 125, 119–128, 1990.
- Trenkel, M. E., Improving fertilizer use efficiency: Controlled-release and stabilized fertilizers in agriculture, Int. Fert. Ind. Assoc., Paris, 1997.
- Vallis, I., L. A. Harper, V. R. Catchpool, and K. L. Weier, Volatilization of ammonia from urine patches in a subtropical pasture, *Aust. J. Agric. Res.*, 33, 97–107, 1982.
- Van Breemen, N., P. A. Burrough, E. J. Velthorst, H. F. v. Dobben, T. d. Wit, T. B. Ridder, and H. F. R. Reijnders, Soil acidification from atmospheric ammonium sulphate in forest canopy throughfall, *Nature*, 299, 548–550, 1982.
- Vlek, P. L. G., and E. T. Craswell, Effect of nitrogen source and management on ammonia volatilization losses from flooded rice-soil systems, *Soil Sci. Soc. Am. J.*, 43, 352–358, 1979.
- Vlek, P. L. G., and J. M. Stumpe, Effects of solution chemistry and environmental conditions on ammonia volatilization losses from aqueous systems, *Soil Sci. Soc. Am. J.*, 42, 416–421, 1978.
- Zhu, Z. L., G. X. Cai, J. R. Simpson, S. L. Zhang, D. L. Chen, A. V. Jackson, and J. R. Freney, Processes of nitrogen loss from fertilizers applied to flooded rice fields on a calcareous soil in north-central China, *Fert. Res.*, 18, 101–115, 1989.
- Zuidema, G., G. J. Van Den Born, J. Alcamo, and G. J. J. Kreileman, Simulating changes in global land cover as affected by economic and climatic factors, *Water Air Soil Pollut.*, 76, 163–198, 1994.

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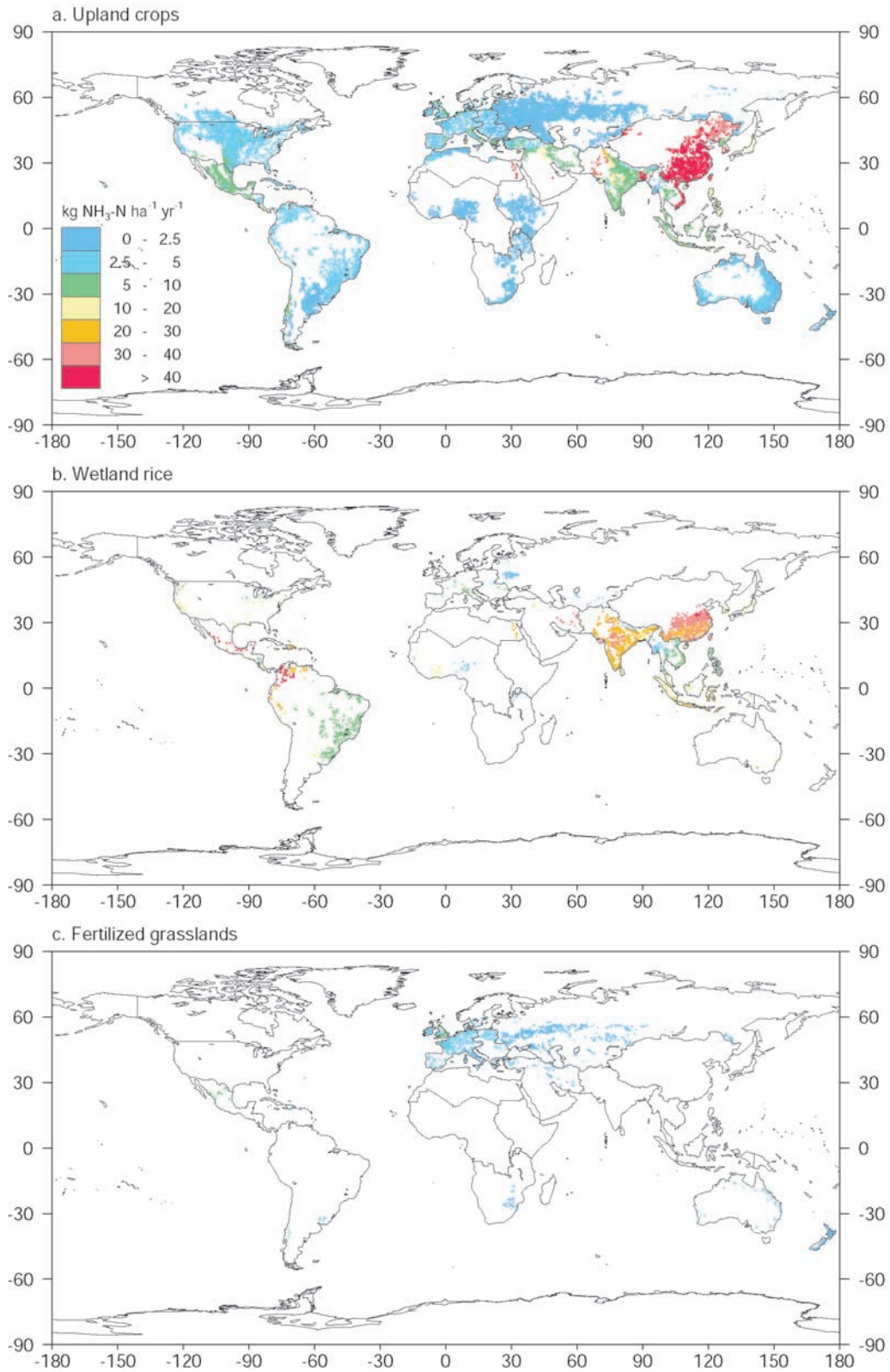


Figure 1. Estimated annual NH_3 volatilization loss for 1995 from synthetic fertilizers used in (a) upland crops, (b) wetland rice fields, and (c) grasslands. Note that emission is an annual estimate. High values may be caused by high cropping intensities, such as in China. Low values may be the result of low cropping intensities. Emission rates per hectare of harvested land may thus differ from those presented here.

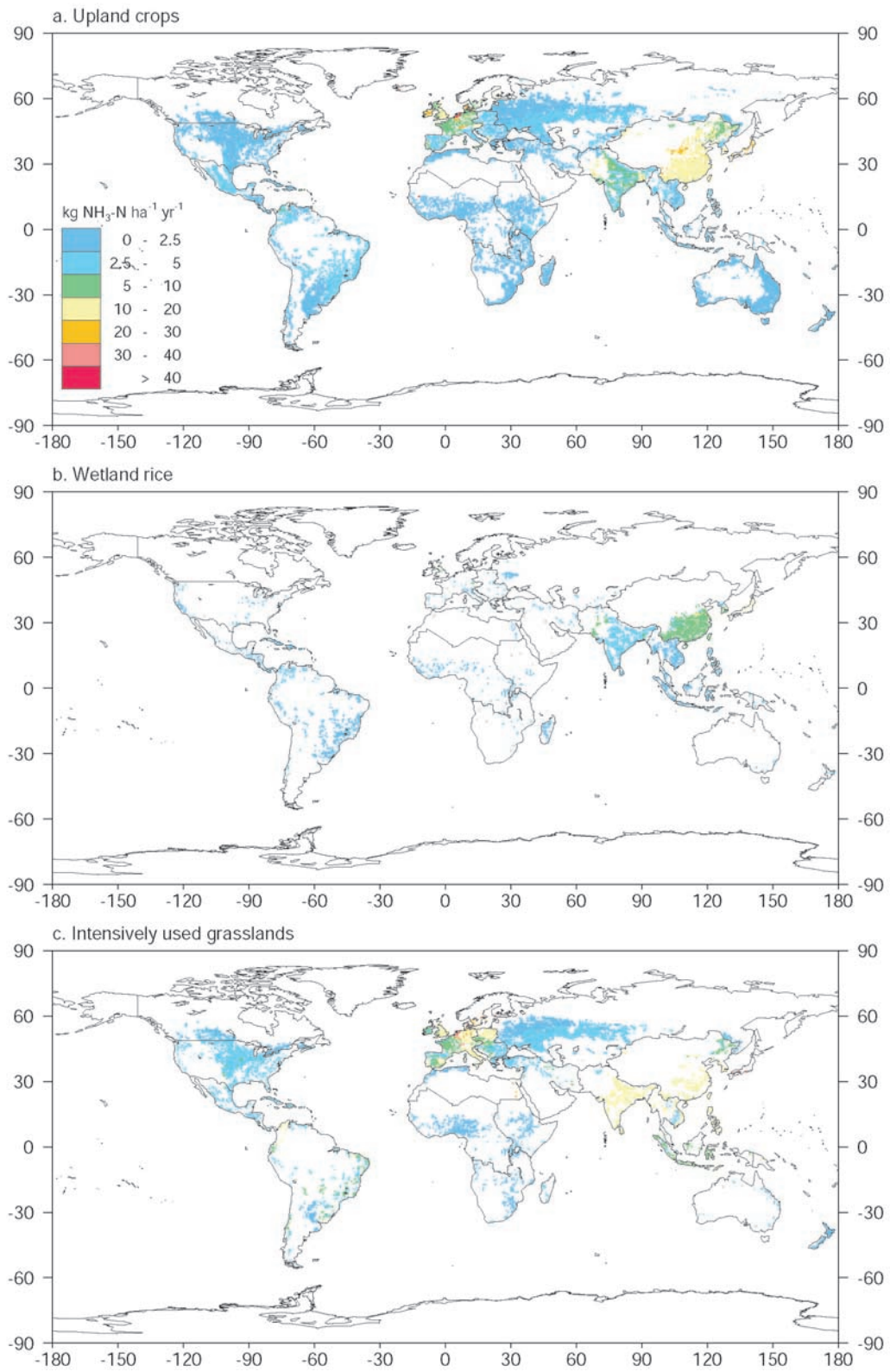


Figure 2. Estimated annual NH₃ volatilization loss for 1995 from animal manure used in (a) upland crops, (b) wetland rice fields and (c) grasslands.