

## A global high-resolution emission inventory for ammonia

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**Abstract.** A global emissions inventory for ammonia ( $\text{NH}_3$ ) has been compiled for the main known sources on a  $1^\circ \times 1^\circ$  grid, suitable for input to global atmospheric models. The estimated global emission for 1990 is about 54 Tg  $\text{N yr}^{-1}$ . The major sources identified include excreta from domestic animals (21.6 Tg  $\text{N yr}^{-1}$ ) and wild animals (0.1 Tg  $\text{N yr}^{-1}$ ), use of synthetic N fertilizers (9.0 Tg  $\text{N yr}^{-1}$ ), oceans (8.2 Tg  $\text{N yr}^{-1}$ ), biomass burning (5.9 Tg  $\text{N yr}^{-1}$ ), crops (3.6 Tg  $\text{N yr}^{-1}$ ), human population and pets (2.6 Tg  $\text{N yr}^{-1}$ ), soils under natural vegetation (2.4 Tg  $\text{N yr}^{-1}$ ), industrial processes (0.2 Tg  $\text{N yr}^{-1}$ ), and fossil fuels (0.1 Tg  $\text{N yr}^{-1}$ ). About half of the global emission comes from Asia, and about 70% is related to food production. The regions with highest emission rates are located in Europe, the Indian subcontinent, and China, reflecting the patterns of animal densities and type and intensity of synthetic fertilizer use. The overall uncertainty in the global emission estimate is 25%, while the uncertainty in regional emissions is much greater. As the global human population will show considerable growth in the coming decades, food production and associated  $\text{NH}_3$  emissions are likely to increase as well.

### 1. Introduction

The existence of gas-phase ammonia ( $\text{NH}_3$ ) and its interaction with other trace chemical species in the atmosphere has been recognized since the last century. *Smith* [1852] reported that “All the rain was found to contain sulphuric acid in proportion as it approached the town, . . . I do not mean to say that all the rain is acid—it is often found with so much ammonia in it as to overcome the acidity.” Interest in the role of  $\text{NH}_3$  in the atmosphere at this time was focused largely upon its role as a fertilizer [e.g., *Lawes and Gilbert*, 1851]. The release of  $\text{NH}_3$  into the atmosphere has increased in many regions because of anthropogenic activities [*Dentener and Crutzen*, 1994], and ammonium contributes to deposition of nitrogen in areas downwind of major sources [*Asman*, 1994]. This anthropogenic input of nitrogen to the environment may lead to the eutrophication of terrestrial and aquatic ecosystems. Areas receiving high N inputs from deposition may show forest decline as a consequence of nitrogen saturation of the soil [*Aber et al.*, 1989] or soil acidification. It has become evident that although it is basic,  $\text{NH}_3$  may still contribute toward acidification of ecosystems as the input of 1 mol of ammonium sulfate,  $(\text{NH}_4)_2\text{SO}_4$ , can result in the release of 4 mol of acidity ( $\text{H}^+$ ) by nitrification [*Van Breemen et al.*, 1982]. Dry deposition of  $\text{NH}_3$  can also result in the release of one proton by

nitrification. This, however, is a simplification, and the net acidification resulting from atmospheric inputs of  $\text{NH}_3$  or  $\text{NH}_4^+$  is complex and varies according to the nutrient status of the soil [*Galloway*, 1995]. The overall global magnitude of  $\text{NH}_3$  emissions is, potentially, more acidifying than either present emissions of sulfur dioxide ( $\text{SO}_2$ ) or nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) [*Galloway*, 1995].

Although the importance of  $\text{NH}_3$  in atmospheric chemistry has been recognized for some time [*Junge*, 1963], it is only over the last 10–15 years that much attention has been devoted to it. Ammonia reacts with acidic  $\text{H}_2\text{SO}_4$  in the atmosphere. The reaction of  $\text{O}_3$  with  $\text{SO}_2$  is pH limited, thus the presence of  $\text{NH}_3$ , neutralizing the  $\text{H}_2\text{SO}_4$  in the droplet, may allow further  $\text{SO}_2$  oxidation to take place. Second, the availability of  $\text{H}_2\text{SO}_4$  may result in an increased transport distance of  $\text{NH}_x$ , as  $\text{NH}_3$  gas dry deposits faster by a factor of 5–10 than  $\text{NH}_4^+$  particles.

The final consideration for  $\text{NH}_3$  emissions on the global scale is that of climate change. As mentioned above,  $\text{NH}_3$  interacts with the global tropospheric S cycle via atmospheric chemistry by influencing the abundance and chemical composition of sulfate particles, primarily from dimethyl sulfide (DMS) emissions arising from planktonic algae. Sulfate particles have an important negative effect on radiative forcing by reducing the instantaneous heating at the ground either directly by backscattering visible and UV radiation or indirectly by increasing cloud lifetime from the formation of cloud condensation nuclei and resulting in increased cloud albedo [*Charlson et al.*, 1990, 1991]. This effect may counteract the positive forcing exerted by other radiatively important trace gases such as  $\text{CO}_2$ ,  $\text{N}_2\text{O}$ , and tropospheric  $\text{O}_3$ , although there is much uncertainty regarding the magnitude and extent of this effect [*Intergovernmental Panel on Climate Change (IPCC)*, 1995].

A final interaction of  $\text{NH}_3$  emissions with radiative forcing is its potential influence on nitrous oxide ( $\text{N}_2\text{O}$ ) emissions. Application of  $(\text{NH}_4)_2\text{SO}_4$  was found by *Hutchinson and Brams* [1992] to stimulate production of both NO and  $\text{N}_2\text{O}$ .

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Thus increased  $\text{NH}_x$  deposition may lead to increased radiative forcing from tropospheric  $\text{O}_3$  (from  $\text{NO}$  emissions) and  $\text{N}_2\text{O}$ .

Various estimates of the magnitude of global  $\text{NH}_3$  emissions have been made over the past 20 years [Söderlund and Svensson, 1976; Böttger et al., 1978; Warneck, 1988; Schlesinger and Hartley, 1992; Dentener and Crutzen, 1994], but only one attempt has been made to show the spatial distribution of these emissions, at a  $10^\circ \times 10^\circ$  resolution, for a modeling study of the global cycle of  $\text{NH}_3$  [Dentener and Crutzen, 1994]. In this paper we present a  $1^\circ \times 1^\circ$  global inventory of the main emission sources of  $\text{NH}_3$ : animal wastes, fertilizers, biomass combustion, soils and natural vegetation, oceans, and other miscellaneous minor sources. It is intended that these data will provide input for global atmospheric transport and chemistry models.

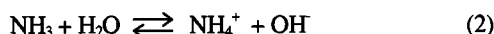
## 2. Emission Source Estimates

### 2.1. Animal Populations

**2.1.1. Domestic animals.** The microbial breakdown of urea and uric acid in animal excreta to  $\text{NH}_4^+$  and its subsequent volatilization to  $\text{NH}_3$  represent the most important source of atmospheric  $\text{NH}_3$ . Ammonium ions are formed as the result of the microbiological hydrolysis of urea by urease:



This process starts when the urea in the voided urine comes in contact with the enzyme urease contained in the excreted faeces. The enzyme urease is also active in the soil, where residual urea and other nitrogenous compounds are broken down. In aqueous solutions there exists an equilibrium between  $\text{NH}_3$  and  $\text{NH}_4^+$ :



The ratio of dissolved  $\text{NH}_3$  to total ammoniacal N ( $\text{NH}_3 + \text{NH}_4^+$ ) in solution increases with increasing pH and temperature. The dissolved  $\text{NH}_3$  may then be lost from the substrate (stored manure and soil) by volatilization to the surrounding air, which has a lower concentration.

The calculation of emission of  $\text{NH}_3$  from domestic animal waste is based upon an average N excretion for different domestic animal categories and subsequent  $\text{NH}_3$  losses during housing, storage of wastes outside the building, grazing, and application of the wastes to grassland or arable land. Some European countries have formulated country-specific emission factors for  $\text{NH}_3$  from animal wastes, for example the Netherlands [Van Der Hoek, 1994], Germany [Isermann, 1990], and the United Kingdom [Pain et al., 1997]. On the basis of the emission factors derived from experimental data from the Netherlands, European [Asman, 1992] and global inventories for  $\text{NH}_3$  emissions from domestic animal populations [Schlesinger and Hartley, 1992] have been made.

In the estimates presented in this study, emission factors were developed for  $\text{NH}_3$  that account for broad differences in regional feeding situations, animal characteristics, and management conditions based on the work of Van Der Hoek and Couling [1996] and Van Der Hoek [1997] for two world regions representing the developed and developing countries. The distinction of these two regions to reflect differences in cattle characteristics is justified by the relatively small difference in N excretion rates between continents [Bouwman et al., 1995] and the large uncertainty on  $\text{NH}_3$

emission rates due to the scarcity of measurement data and lack of information on waste management practices.

The categories nondairy cattle, buffalo, and camels include subclasses such as young and adult animals. The animal characteristics used include the mean live weight and for dairy cattle the mean daily milk production (Table 1). The data for cattle, buffalo, and camels were taken from a study on global methane emissions, based upon animal feed intake [Environmental Protection Agency (EPA), 1994].

Nitrogen excretion is calculated as the difference between N intake by the animal and the N fixation in meat, milk, and eggs. The N intake is assumed to be the minimum protein requirement of the animal for maintenance and production. Protein requirements for maintenance are proportional to the metabolic weight of the animal, whereas growth, lactation, pregnancy, and production of eggs are additional protein requirements. Animals used for labor and grazing animals that travel long distances to collect feed need additional energy and thus have a higher feed and protein intake.

The N excretion rates for the different subclasses of cattle, buffalo, and camels are calculated using standard feeding tables and standard values for N content of animal products [McDonald et al., 1988]. The excretion rates for the subclasses and the share of the subclasses to the animal category are used to calculate the average excretion data for the animal categories on the basis of the animal characteristics (Table 1).

For sheep, pigs, and poultry, European data for N excretion were used [Van Der Hoek and Couling, 1996]. The excretion data for pigs and poultry originate from the Dutch situation [Van Eerd and Olsthoorn, 1994], as it is assumed that the biological efficiency for producing meat and eggs does not differ much on a global scale. The excretion of the young sows and piglets is not presented separately as this is already included in the excretion of the sow.

The emission of  $\text{NH}_3$  arising during housing and storage of the animal waste outside the stable is calculated by multiplying the N excretion by a specific volatilization rate (Table 2). The remaining N in the animal waste can partly volatilize during and after spreading. Part of the N is excreted in the meadow, and a specific volatilization factor is used for this component of the  $\text{NH}_3$  loss (Table 2).

For region I the specific volatilization rates for the different emission sources are comparable to European agriculture [Van Der Hoek and Couling, 1996], and the rates were derived from measurements at farm and field scale [Van Der Molen et al., 1989; Isermann, 1990; Jarvis and Bussink, 1990; De Bode, 1991; Oosthoek et al., 1991; Kroodsma et al., 1993; Groot Koerkamp, 1994]. For emission of  $\text{NH}_3$  from animal wastes present in the stable and that from wastes stored outside the stable, a combined volatilization rate of 20% was used (Table 2). For spreading of wastes from cattle, pigs, and poultry a volatilization rate of 20% was used, assuming that no precautions are taken to prevent emission of  $\text{NH}_3$ . For the wastes from the remaining animal categories a volatilization rate of 10% was used because of the lower content of volatilizable N in the waste. For cattle, pigs, and poultry the combined volatilization rate from stable and storage (20%) and spreading (20%) was 36%, and for the other animal categories the combined rate was 28% (20% and 10%) (Table 2).

The volatilization rate of 15% for N excreted in the meadow for region II is assumed to be twice that of region I as higher temperatures result in greater  $\text{NH}_3$  emissions of the excreted urine (Table 2). For the stable emissions in region II a number of con-

**Table 1.** Characteristics and N Excretion for Different Animal Categories for Two World Regions

Animal Category	Region	Animal Characteristics for Subclass <sup>a</sup>	N Excretion, kg head <sup>-1</sup> yr <sup>-1</sup>	
			Subclass	Category
Dairy cattle	I	500 kg LW, 15 L milk d <sup>-1</sup>	80	80
	II	400 kg LW, 5 L milk d <sup>-1</sup>	60	60
Nondairy cattle	I	50% young cattle, 250 kg LW	30	45
		30% suckling cows, 500 kg LW, 10 L milk d <sup>-1</sup>	70	
		20% beef cattle 400 kg LW	25	
	II	50% young cattle, 200 kg LW	25	40
		30% suckling cows, 400 kg LW, 5 L milk d <sup>-1</sup>	60	
		20% draft cattle, 400 kg LW	40	
Buffalo	I and II	40% female adult, 400 kg LW	60	45
		20% male adult, 500 kg LW	40	
		20% young, 200 kg LW	30	
		20% young, 400 kg LW	40	
Camels	I and II	69% female adult, 500 kg LW	64	55
		3% male adult, 550 kg LW	40	
		14% weaned, 187 kg LW	30	
		14% immature, 356 kg LW	40	
Horses	I and II	assumed equal to buffalo		45
Sheep	I and II	based on 1 ewe and 1-1.5 adherent lambs		10
Goats	I and II	assumed equal to 90% of sheep		9
Pigs	I and II	50% fattening pigs	14	11
		10% sows	36	
		40% piglets and young sows	0	
Poultry	I and II	50% laying hens	0.6	0.5
		50% broilers	0.6	

Region I, developed countries (Europe, the former USSR, North America, Australia and New Zealand, Israel, and Japan); region II, developing countries (Latin America, Oceania excluding Australia and New Zealand, Africa, and Asia excluding the former USSR).

<sup>a</sup> LW, live weight; percent, share of subclass in total of animal category.

siderations were made. First, the ratio of volatilizable (mineral N) to nonvolatilizable N (organic N) in the animal waste is lower in region II than in region I, because of lower feeding levels and lower N content of the feed. Second, in region II a smaller portion of the animals are housed in stables than in region I, and it is assumed that confinement in enclosures results in similar emission of NH<sub>3</sub>. The resulting volatilization rates for region II for emission of NH<sub>3</sub> from stables including waste storage outside the stable and spreading of wastes are assumed to equal those in region I, based on the assumption that the higher temperatures in region II are counteracted by a lower volatilizable fraction of the N in the animal waste (Table 2).

In both regions I and II, grazing buffaloes and camels are found generally in regions with warm climates. As N excretion is equal in regions I and II, we therefore assumed equal NH<sub>3</sub> emission rates for regions I and II for grazing buffaloes and camels (Table 2).

The distributions of the domesticated animals are taken from *Lerner et al.* [1988] and for poultry from *Bouwman et al.* [1995] and were updated with country statistics on animal populations [Food and Agriculture Organization (FAO), 1991] to produce grid-based estimates for 1990. The total annual emission of NH<sub>3</sub> for region I is 7 Tg NH<sub>3</sub>-N and for region II is 14.7 Tg, and the global total emission is 21.6 Tg NH<sub>3</sub>-N (Table 3 and Plate 1a).

**2.1.2 Wild animals.** Data on populations of wild animals have not been collected consistently. There are data available on populations of larger herbivores including moose in North America (900,000 animals), Scandinavia (600,000), and the former USSR (1,000,000) and reindeer in North America

(16,500,000), Scandinavia (800,000), and the former USSR (2,700,000) [Hudson *et al.*, 1989]. Many smaller herbivores and carnivores occur in forest ecosystems, and their NH<sub>3</sub> emissions are included in the estimate for NH<sub>3</sub> from natural ecosystems. Population data for these smaller animals is given by *Roth and Merz* [1997]. Assuming that moose and reindeer excrete 40 and 25 kg N yr<sup>-1</sup>, respectively, and an NH<sub>3</sub> volatilization coefficient equal to that of sheep and goats for grazing animals of 0.04, the total emission from moose and reindeer would be 0.024 Tg yr<sup>-1</sup> for North America, Scandinavia, and the former USSR. For Africa the estimates of populations of wild bovids range from 100 to 500 million animals [Van Soest, 1994]. Assuming the lower end of the range, the NH<sub>3</sub> emission from 100 million animals would be 0.3 Tg N yr<sup>-1</sup>, based on an excretion rate of 40 kg N yr<sup>-1</sup> and a volatilization coefficient of 0.08 (similar to grazing emissions from sheep and goats in the tropics).

In order to obtain a second estimate of the spatial distribution of the possible contribution of wild animals to regional NH<sub>3</sub> emissions, the method proposed by *Warneck* [1988] was modified. *Warneck* [1988] assumed that 3–10% of the net primary production (NPP) is consumed by wild animals and that 0.006 g urea is formed for each gram of biomass consumed. Finally, 10% of urea is converted to gaseous NH<sub>3</sub>.

We used the global modeled NPP map from *Klein Goldewijk et al.* [1994], who estimated a global NPP of 60 × 10<sup>15</sup> g C yr<sup>-1</sup>, combined with spatial distributions of ecosystems from *Olsen et al.* [1983]. We based our estimates on the

**Table 2.** N Excretion and NH<sub>3</sub>-N Volatilization Rates for Different Animal Categories and Two Regions for the Animal Waste Produced in the Meadow and Stable<sup>a</sup> Period

Animal category	Region I		Region II	
	N Excretion, kg N head <sup>-1</sup> yr <sup>-1</sup>	NH <sub>3</sub> -N Loss, %	N Excretion, kg N head <sup>-1</sup> yr <sup>-1</sup>	NH <sub>3</sub> -N Loss, %
Dairy cattle				
Stable	50	36	40	36
Meadow	30	8	20	15
Total	80	25.5	60	29.0
Non-dairy cattle				
Stable	15	36	10	36
Meadow	30	8	30	15
Total	45	17.3	40	20.3
Buffalo				
Stable	15	28	15	28
Meadow	30	15	30	15
Total	45	19.3	45	19.3
Camels				
Stable	18	28	18	28
Meadow	37	15	37	15
Total	55	19.3	55	19.3
Horses				
Stable	20	28	15	28
Meadow	25	8	30	15
Total	45	16.9	45	19.3
Sheep				
Stable	1	28	1	28
Meadow	9	4	9	8
Total	10	6.4	10	10.0
Goats				
Stable	1	28	1	28
Meadow	8	4	8	8
Total	9	6.4	9	10.0
Pigs <sup>b</sup>	11	36.0	11	36.0
Poultry <sup>b</sup>	0.5	36.0	0.5	36.0

See Table 1 for definitions of regions.

<sup>a</sup> Emissions from the stable include those from waste stored outside the stable and from spreading of animal waste.<sup>b</sup> Pigs and poultry are assumed to live only in housing units.**Table 3.** Global Population of Domesticated Animals and Their Individual and Total NH<sub>3</sub>-N Emission for Two World Regions for 1990

Animal Category	Population, Head × 10 <sup>6</sup>		NH <sub>3</sub> -N Emission, kg N head <sup>-1</sup> yr <sup>-1</sup>		Total NH <sub>3</sub> -N Emission, Tg N yr <sup>-1</sup>	
	I	II	I	II	I	II
Dairy cattle	103	125	20.4	17.4	2.1	2.2
Nondairy cattle	287	778	7.8	8.1	2.2	6.3
Buffalo	1	138	8.7	8.7	0.0	1.2
Camels	0	19	10.6	10.6	0.0	0.2
Horses	16	45	7.6	8.7	0.1	0.4
Sheep	528	687	0.64	1.0	0.3	0.7
Goats	27	560	0.58	0.90	0.0	0.5
Pigs	339	516	4.0	4.0	1.4	2.1
Poultry	4427	6335	0.2	0.2	0.8	1.1
Total					7.0	14.7

See Table 1 for definitions of regions.

following assumptions: (1) In ecosystems with tree cover, 20% of the NPP is consumable grass or leaves, and the remainder is not consumed because it is tree biomass or nondigestible. (2) Emissions from wild animals are insignificant in Latin America, the Middle East, the Indian subcontinent, and east Asia. In these regions, grazing is dominated by domesticated animals in extensive livestock production systems (see section 2.2.1). (3) A fraction of 3–10% of the consumable NPP is eaten, as indicated by Warneck [1988]. (4)  $\text{NH}_3$  emissions from wild animals' excreta are found only in tundras, tropical savannas, scrub and shrub lands, and grassland complexes, with an estimated NPP of about 25 Pg C (calculated from Olsen *et al.* [1983] and Klein Goldewijk *et al.* [1994]). (5) In forest ecosystems,  $\text{NH}_3$  arising from herbivore excreta is assumed to be insignificant since it is likely that most of the  $\text{NH}_3$  is taken up by the forest canopy and does not escape to the free atmosphere, as forests have a relatively low ventilation rate, for example of the order of hours for nighttime conditions and <1 hour for daytime conditions in a rain forest [Trumbore *et al.*, 1990].

The results presented here for the second NPP-based estimate are based on the lower end of 3% of the range given by Warneck [1988] for consumption of consumable NPP. The global emission from excreta from wild animals estimated thus is 0.12 Tg N  $\text{yr}^{-1}$ ; this includes emissions from the former USSR (0.02 Tg N  $\text{yr}^{-1}$ ), North America (0.02 Tg N  $\text{yr}^{-1}$ ), Australia (0.01 Tg N  $\text{yr}^{-1}$ ), China (0.02 Tg N  $\text{yr}^{-1}$ ), and Africa (0.04 Tg N  $\text{yr}^{-1}$ ). The NPP-based estimates for North America, Scandinavia, and the former USSR are only slightly higher than the population-based estimates. However, for Africa the population-based estimate is about a factor of 10 higher than the NPP-based estimate of 0.04 Tg N  $\text{yr}^{-1}$ . Considering the strongly decreasing populations of wild animals observed in Africa (H. Prins, personal communication, 1996), the uncertainty regarding animal populations and the fraction of NPP that is consumed, we judge that the NPP-based gridded inventory is acceptable as a first-order estimate. The results show that wild animals may be important sources of  $\text{NH}_3$  only in remote regions in the northern hemisphere.

## 2.2. Synthetic Nitrogen Fertilizers

The application of synthetic N fertilizers is known to result in the release of  $\text{NH}_3$  into the atmosphere. The  $\text{NH}_3$  loss from fertilizers is a function of the type of fertilizer, soil properties (pH, calcium content, water content, buffer capacity, porosity, etc.), meteorological conditions (temperature, wind speed, and precipitation), and management (timing and mode of fertilizer application). Hence emissions are likely to exhibit large spatial and temporal variability. However, as insufficient information is available to describe losses as a function of the above factors, we have adopted the simple emission factors of Asman [1992] for Europe which depend on the fertilizer type only (Table 4). The emission factors used for the most important fertilizers in this paper are very similar to the ones proposed by Schlesinger and Hartley [1992] and used by Matthews [1994]. (Matthews applied both the emission factors from Schlesinger and Hartley [1992] and those of Buijsman *et al.* [1987] to the 1984 fertilizer statistics.)

Here we present factors for an additional number of categories not used by Schlesinger and Hartley [1992]. For the

use of urea in the tropics, anhydrous ammonia and ammonium bicarbonate, new emission factors were developed in this study, as will be discussed in detail below.

The emission factors of Asman [1992] are based primarily on the work of Whitehead and Raistrick [1990], who made laboratory measurements of the volatilization of  $\text{NH}_3$  during application of different fertilizers to different soil types. This method has the advantage that the emission from different fertilizers was measured under the same conditions, and at least the ranking should reflect the difference in volatilization rates. It must be stressed that the uncertainty in the emission factors is large. For example, the emission factor for  $(\text{NH}_4)_2\text{SO}_4$  is 8% of the N content (Table 4). However, when it is applied to soils with a pH of < 5.5, the emission may be less than 2% and as large as 50% when applied to calcareous soils [Whitehead and Raistrick, 1990].

Increased temperatures promote  $\text{NH}_3$  emission in the same way as described in section 2.1. The temperature effect, however, could only be taken into account for urea, for which sufficient experimental data were available to make a rough distinction between tropical and temperate conditions, and ammonium bicarbonate.

**2.2.1. Urea.** In developing countries, approximately 55% of the N fertilizer used is in the form of urea [International Fertilizer Industry Association (IFA), 1994], an important part being applied to flooded rice fields [De Datta, 1987;

Table 4.  $\text{NH}_3$  loss from synthetic N fertilizers

Fertilizer Category <sup>a</sup>	$\text{NH}_3$ Loss, % of N Content
Ammonium sulphate	8
Urea, temperate zones <sup>b</sup>	15
Urea, tropical zones <sup>b</sup>	25
Ammonium nitrate	2
Calcium ammonium nitrate	2
Anhydrous ammonia, direct application <sup>b</sup>	4
Nitrogen solutions	2.5
Other straight nitrogen	
Other straight nitrogen	
China, ammonium bicarbonate, temperate zones <sup>b</sup>	20
Other straight nitrogen	
China, ammonium bicarbonate, tropical zones <sup>b</sup>	30
Total straight nitrogen <sup>c</sup>	4
Monoammonium phosphate	2
Diammonium phosphate	5
Other NP-N <sup>d</sup>	3
NK-N <sup>d</sup>	2
NPK-N <sup>d</sup>	4
Compound N <sup>c</sup>	4

All emission factors are from Asman [1992] unless indicated otherwise.

<sup>a</sup> Fertilizer categories from International Fertilizer Industry Association (IFA) [1994].

<sup>b</sup> Emission factors for urea, anhydrous ammonia, and ammonium bicarbonate were developed for this study.

<sup>c</sup> Only used if no information was available on the fertilizer consumption for the individual categories.

<sup>d</sup> NP-N, compound nitrogen-phosphorous fertilizers; NK-N, compound nitrogen-potassium fertilizer; NPK-N, compound nitrogen-phosphorous-potassium fertilizer.

*Bouwman*, 1991]. Urea is extensively used in the tropics because of its relatively high N content and low transportation costs per unit of N. Two applications of urea to rice crops are made, the first at 15–30 days after transplanting and the second at maximum tillering at 45–55 days. Up to 4 times more  $\text{NH}_3$  may be volatilized after the first application than the second (B.H. Byrnes, personal communication, 1995), because at the second application the crop canopy is high enough to reduce turbulence over the water, restricting the surface exchange process. Moreover,  $\text{NH}_3$  emitted from the floodwater is taken up readily by the aerial parts of the plants. Research on volatilization losses has therefore focused on these initial applications.

Ammonia losses from rice fields after the first application, based on a range of experimental techniques, are presented in Table 5. Earlier experiments were made mostly using enclosure techniques, with or without artificial ventilation, and are not representative of field conditions. More recently, the mass balance micrometeorological technique or the bulk aerodynamic method have been applied in the field. These techniques do not disturb the natural conditions, and the results are therefore more useful than those from enclosure methods.

The usual practice in Asia is to broadcast the fertilizer grains (top-dress after transplantation) into 0.05-m-deep floodwater [*Frenay et al.*, 1990]. Methods or fertilizer types that lead to reduced emissions are not yet commonly used. For this reason an emission factor of 35% was assumed for the first application for flooded rice fields on the basis of measurements under field conditions (Table 5). The uncertainty range in the emission factor for the first application could be 10–55% (Table 5). For the second application an average loss of 15% was adopted, resulting in an overall loss of 25%, which may be conservative.

For application of urea to other tropical crops we also assumed an emission rate of 25%, because of the temperature dependence of volatilization. Moreover, there are other crops than rice where urea is not incorporated adequately into the soil, such as banana, oil palm, and sugar cane (B.H. Byrnes, personal communication, 1995).

**2.2.2. Anhydrous ammonia.** Information on the emission of  $\text{NH}_3$  from anhydrous (liquid)  $\text{NH}_3$  injected into the soil was provided by S.G. Sommer (personal communication, 1995). The  $\text{NH}_3$  emission is negligible if anhydrous ammonia is applied to moist soils under average Danish meteorological conditions. Emission may occur, however, if the injector does not penetrate deep enough or when the soil is either too wet or dry. *Stanley and Smith* [1956] measured losses of  $\text{NH}_3$  from a Putnam silt loam soil in the United States of America as influenced by depth of applications and soil moisture. These experiments indicated that the capacity of soils to retain  $\text{NH}_3$  increases with soil moisture content until an optimum is reached. If the moisture content is further increased, the capacity decreases. This capacity is larger for clay which has a finer texture than for sandy soils with large pores which promote diffusion. At least 50% of the  $\text{NH}_3$  holding capacity of the soil is attributable to organic matter. *Stanley and Smith* [1956] also found that the spacing between the injection points influenced emissions. At the same N application per unit area, close spacing resulted usually in higher  $\text{NH}_3$  retention and lower  $\text{NH}_3$  volatilization than large spacing as the

concentration per unit volume of the soil will be reduced with more injection points per unit area. *Stanley and Smith* [1956] found losses of between 1 and 12%. The emission factor of directly applied liquid  $\text{NH}_3$  in this inventory was arbitrarily set to 4% of the N content.

**2.2.3. Ammonium bicarbonate.** Ammonium bicarbonate ( $\text{NH}_4\text{HCO}_3$ ) is an important fertilizer in China. Its emission factor should be larger than that of urea because of its volatility. Urea is not volatile itself, but urease (equation (1)) will hydrolyze the urea to  $\text{NH}_4\text{HCO}_3$  at values of soil pH encountered by the urease enzyme. Urea hydrolyses completely within 2–3 days and will then become volatile, whereas  $\text{NH}_4\text{HCO}_3$  is already volatile. The emission factor for  $\text{NH}_4\text{HCO}_3$  is therefore high and likely to be higher than that of urea [*Zhu et al.*, 1989; *Roelike*, 1994].

The fertilizer  $\text{NH}_4\text{HCO}_3$  is included in the category “other straight nitrogen” (ZSN) of IFA [1994]. More than 90% of the global use of ZSN occurred in China in 1990, and practically all ZSN (8.9 Tg N  $\text{yr}^{-1}$ ) in China is  $\text{NH}_4\text{HCO}_3$  (K. Isherwood, personal communication, 1995). In analogy to urea, different climatic zones are distinguished. An emission factor of 30% (the average of the first and second applications) is estimated for ZSN applications in Chinese tropical zones, and 20% is estimated in temperate zones in China.

**2.2.4. Global  $\text{NH}_3$  emission from synthetic fertilizers.** Fertilizer use was distributed on the basis of a  $1^\circ \times 1^\circ$  country database [*Lerner et al.*, 1988] and the 1990 N fertilizer mix for 90 countries from IFA (1994), representing > 98% of the global synthetic N fertilizer consumption. For 60 other countries the total N fertilizer consumption for 1990 from FAO [1991] was used, assuming that the mix of fertilizer types in these countries is identical to the regional data from IFA (1994). For 34 minor countries, N fertilizer data are not available from IFA [1994] nor FAO [1991]. For each fertilizer type the average N application was calculated as N fertilizer use / (area arable land plus permanent crops) [FAO, 1991]. The  $\text{NH}_3$  emission, based on the emission rates for the various fertilizer types (Table 4), was simply assigned to arable grid boxes given by *Olson et al.* [1983], corrected with the country data from FAO [1991] for arable land and permanent crops by using country-specific correction factors.

The global emission of  $\text{NH}_3$  from fertilizers is 9 Tg  $\text{yr}^{-1}$ . The distribution of emission rates is presented in Plate 1b. This global estimate is higher than that of *Schlesinger and Hartley* [1992] and *Matthews* [1994] mainly because of more detailed data on fertilizers by type and higher emission factors for the tropics. Globally, urea is the dominant source, followed by other straight nitrogen, which is mainly  $\text{NH}_4\text{HCO}_3$  used in China (Table 6). The emission in tropical regions is about 2.5 times that of temperate regions (Table 6) mainly because higher temperatures result in larger losses and because of the widespread use of urea and  $\text{NH}_4\text{HCO}_3$  in tropical regions.

## 2.3. Biomass Burning

There is evidence that during biomass burning significant amounts of  $\text{NH}_3$  are injected into the atmosphere [*Warneck*, 1988]. Several sources of burning are considered: burning during forest clearing, savanna burning, agricultural waste burning, and combustion of biofuels for energy purposes. Many estimates of the global magnitude of biomass burning have

**Table 5.** NH<sub>3</sub> Volatilization Losses From Urea Applied to Flooded Rice Fields for the First Split Fertilizer Application From Field and Laboratory Experiments

Soil Type	Site, Details	N Rate kg N ha <sup>-1</sup>	Applica- tion Method <sup>a</sup>	N Loss %	Measure- ment Method <sup>b</sup>	Reference <sup>c</sup>
Mahas clay	Philippines, soil only, laboratory	50	B	8.1	Cv	1
Mahas clay	Philippines, soil only, laboratory	200	B	11.0	Cv	1
Mahas clay	Philippines, soil only, laboratory	50	BI	3.5	Cv	1
Mahas clay	Philippines, soil only, laboratory	200	BI	7.2	Cv	1
Luisiana clay	Philippines, soil only, laboratory	50	B	3.1	Cv	1
Luisiana clay	Philippines, soil only, laboratory	200	B	10.1	Cv	1
Luisiana clay	Philippines, soil only, laboratory	50	BI	5.3	Cv	1
Luisiana clay	Philippines, soil only, laboratory	200	BI	4.5	Cv	1
Acid sulphate soil	Philippines, soil only, laboratory	50	B	1.2	Cv	1
Acid sulphate soil	Philippines, soil only, laboratory	200	B	0.3	Cv	1
Acid sulphate soil	Philippines, soil only, laboratory	50	BI	0.3	Cv	1
Acid sulphate soil	Philippines, soil only, laboratory	200	BI	0.1	Cv	1
Silo silt loam	Philippines, soil only, laboratory	50	B	19.0	Cv	1
Silo silt loam	Philippines, soil only, laboratory	200	B	15.7	Cv	1
Silo silt loam	Philippines, soil only, laboratory	50	BI	5.9	Cv	1
Silo silt loam	Philippines, soil only, laboratory	200	BI	5.3	Cv	1
Mahas clay	Los Baños, Philippines	100	B	8.2	C-	2
Mahas clay	Los Baños, Philippines	100	BI	3.6	C-	2
Mahas clay	Los Baños, Philippines, pot experiment, labora-	90	B	18	Cv	3
Mahas clay	Los Baños, pot experiment, field, wet	90	B	6	Ohnv	3
Decatur silt loam	Muscle Shoals, Alabama, USA, laboratory	95	BI	50	Cv	4
Decatur silt loam	Muscle Shoals, Alabama, USA, laboratory	47	BI	25	Cv	4
Grey clay soil	Griffith, New South Wales, Australia	80	B	11	MB	5
Maligaya silty clay loam	Muñoz, Philippines	80	BI	15	MB	6
Mahas clay	Los Baños, Philippines	60	BI	13	MB	6
Maligaya silty clay loam	Muñoz, Philippines	80	B	47	MB	6
nd	Los Baños, Philippines	60	B	27	MB	6
Maligaya silty clay loam	Muñoz, Philippines	40	PI	11	MB	6
Acidic lacustrine clay	Lianhu farm, Dan Yang (Jiangsu), China	90	BI	8.8	MB	7
Maligaya silty clay loam	Muñoz, Philippines	58	B	36	MB	8
Angeles loamy sand	San Marcelino, Philippines	53	B	26	MB + BA	9
Calumpang clay	Calauan, Philippines	53	B	27	MB + BA	9
Calumpang clay	Calauan, Philippines	53	BI	18	MB + BA	9
Calumpang clay	Calauan, Philippines	53	BIdry	7-10	MB + BA	9
Angeles loamy sand	San Marcelino, Philippines	80	B	36	MB + BA	9
Angeles loamy sand	San Marcelino, Philippines	80	BI	26-30	MB + BA	9
Angeles loamy sand	San Marcelino, Philippines	n.d.	BIdry	10	MB + BA	9
Silty clay loam	Kalyani, India (Haplustalfs)	90	B	6-13	Ohnv	10
Guthrie silty loam	Muscle Shoals, Alabama, USA, laboratory	100	BIdry	34-36	Cv	11
Alluvial sandy clay-loam	Cuttack, India (Haplaquept)	100	BIpud	5.8	Ohnv	12
Calcareous sandy loam	Pandian Commune (Henan), China	90	BI	30.1	MB + BA	13
nd	Ludhiana, India, laboratory	200	BIpud	9.6	Cv	14
Maligaya silty clay	Mabitac, Philippines	53	B	48	BA	15
Calumpang clay	Calauan, Philippines	53	B	27	BA	15
Alaminos silty loam	Aguilar, Philippines	53	B	14	BA	15
Angeles loamy sand	San Marcelino, Philippines	53	B	26	BA	15
Maligaya silty clay	Mabitac, Philippines	80	B	56	BA	15
Calumpang clay	Calauan, Philippines	80	B	23	BA	15
Alaminos silty loam	Aguilar, Philippines	80	B	10	BA	15
Angeles loamy sand	San Marcelino, Philippines	80	B	36	BA	15
Maligaya silty clay	Mabitac, Philippines	53	BI	42	BA	15
Calumpang clay	Calauan, Philippines	53	BI	12	BA	15
Alaminos silty loam	San Marcelino, Philippines	53	BI	10	BA	15
Angeles loamy sand	San Marcelino, Philippines	53	BI	30	BA	15
Maligaya silty clay	Mabitac, Philippines	80	BI	43	BA	15
Calumpang clay	Calauan, Philippines	80	BI	12	BA	15
Alaminos silty loam	Aguilar, Philippines	80	BI	10	BA	15
Angeles loamy sand	San Marcelino, Philippines	80	BI	26	BA	15
Maligaya silty clay	Mabitac, Philippines	53	BIdry	11	BA	15

Table 5 (continued)

Calumpang clay	Calauan, Philippines	53	BIdry	11	BA	15
Alaminos silty loam	Aguilar, Philippines	53	BIdry	4	BA	15
Angeles loamy sand	San Marcelino, Philippines	53	BIdry	14	BA	15
Maligaya silty clay	Mabitac, Philippines	80	BIdry	7	BA	15
Calumpang clay	Calauan, Philippines	80	BIdry	16	BA	15
Alaminos silty loam	Aguilar, Philippines	80	BIdry	5	BA	15
Angeles loamy sand	San Marcelino, Philippines	80	BIdry	10	BA	15
Calumpang clay	Calauan, Philippines	80	B	54	MB	16
Calumpang clay	Calauan, Philippines	53	B	46	MB	16
Laterite soil w. sandy	Pattambi, India	90	B	8.7	C-	17
Sandy loam	Hisar, India	240 <sup>d</sup>	BI	4.1-5.2	Cv	18

Abbreviation nd indicates no data.

<sup>a</sup> B, surface broadcast applied to flood water, which is the usual practice (in most cases 14-21 days after transplanting); PI, surface broadcast applied 5-7 days before panicle initiation; BI, broadcast and incorporated in the soil (by this method the emission is somewhat reduced, depending on the time period between application and incorporation); pud, puddled soil; dry, dry soil.

<sup>b</sup> BA, bulk aerodynamic technique; Cv, closed system with ventilation; C-, closed system without ventilation; MB, mass balance micrometeorological technique; Ohnv, semi-open system with natural ventilation.

<sup>c</sup> References are as follows: 1, *MacRae and Ancajas* [1970]; 2, *Ventura and Yoshida* [1977]; 3, *Mikkelsen et al.* [1978]; 4, *Vlek and Craswell* [1979]; 5, *Simpson et al.* [1984]; 6, *Fillery et al.* [1984, 1986]; 7, *Cai et al.* [1986]; 8, *Fillery and De Datta* [1986]; 9, *Obcemea et al.* [1988]; 10, *Santra et al.* [1988]; 11, *Katyal and Carter* [1989]; 12, *Panda et al.* [1989]; 13, *Zhu et al.* [1989]; 14, *Khind et al.* [1990]; 15, *Frenay et al.* [1990]; 16, *De Datta et al.* [1991]; 17, *Kumar and Menon* [1992]; 18, *Sharma et al.* [1993].

<sup>d</sup> Fertilizer application rate expressed as mg N kg<sup>-1</sup> soil.

been made [e.g., *Crutzen et al.*, 1979; *Seiler and Crutzen*, 1980; *Hao et al.*, 1990; *Andreae*, 1991; *Hao and Liu*, 1994] based mostly upon estimates from the 1970s–1980s. The spatial disaggregation of biomass burning emissions commonly used [e.g., *Langner and Rodhe*, 1991; *Dentener and Crutzen*, 1994; *Bouwman et al.*, 1995] is based on the tropical inventory of *Hao et al.* [1990]. However, the extent of natural and prescribed fire extends well into temperate zones [*Levine*, 1991]. Although revised biomass inventories are being developed, which include tropical and temperate burning [*Graedel et al.*, 1995], none were available at the time of compiling this ammonia inventory. In this paper we use the inventory of savanna burning of *Hao and Liu* [1994] with additional data for Australia, and for worldwide deforestation we use the inventory of *FAO* [1995].

**2.3.1. Savanna burning.** The annual amount and distribution of savanna burning of 2375 Tg dry matter (dm) was taken from *Hao and Liu* [1994]. For savanna burning in Australia the inventory of *Walker* [1981] of 300 Tg dm yr<sup>-1</sup> was used and distributed on the basis of the relationship between structural vegetation types and vegetation types for Australia listed by *Galbally et al.* [1992] using the 1° × 1° vegetation/land use database of *Matthews* [1983].

**2.3.2. Deforestation.** The rates of deforestation for the period 1981–1990 (globally 16.3 Mha yr<sup>-1</sup>) and biomass per unit area used in this inventory are from *FAO* [1995]. This study includes estimates for large countries such as China and Australia, with a global loss of biomass of 2100 Tg dm yr<sup>-1</sup>. Country data for deforestation and loss of biomass due to burning were distributed on the basis of forest types defined by *Olsen et al.* [1983]. The distribution by state for Brazil was taken from *Fearnside* [1993]. The advantage of this approach, which is similar to that of *Hao and Liu* [1994] for the tropics, is the inclusion of data for some important temperate countries such as China and Australia.

**2.3.3. Agricultural waste burning.** In many regions, part of the agricultural residues is used as a fuel, and part is simply burnt in the field. The volumes involved in both categories are highly uncertain. In this study the amount of available agricultural residues was estimated on the basis of crop production data from *FAO* [1991]. The total availability of residues from cereals and pulses was calculated as 1.5 times the grain production; waste from root and tuber crops was calculated as 60% of the produce [*FAO*, 1981]; waste from sugar cane and sugar beet was calculated as 10% of the biomass production plus 25% as bagasse [*Barnard*, 1991]; and waste from other crops, including coconuts, fruits, vegetables, sisal, jute, rubber, etc., was calculated as 10% of the biomass production. Burning of residues from oil extraction from oilseeds, an important feedstuff, is assumed to be negligible.

For the developing countries we assumed that 40% of the resulting residual biomass is burnt each year according to *Hao and Liu* [1994]. This is a factor of 2 lower than the estimated fraction of 0.8 proposed by *Andreae* [1991]. For developed countries the fraction of wastes actually burnt was assumed to be 0.2 on the basis of the estimate of *Lee and Atkins* [1994] for Europe, instead of the 50% used by *Andreae* [1991]. For Oceania the fraction of residues burnt was assumed to be 0.4 to achieve consistency with *Galbally et al.* [1992]. The estimate for China (393 Tg dm yr<sup>-1</sup>) was taken from *Zhuang et al.* [1995].

The energetic use of agricultural wastes is taken from *Hall et al.* [1994], and the field burning of residues was calculated as the total residue burning minus the energy use. The assumption that 40% of residues is burnt in North Africa yields an estimate that is lower than the independent data from *Hall et al.* [1994] on the energetic use of agricultural wastes. Therefore, for North Africa it was assumed that the total burning of agricultural wastes equals the energetic use taken from *Hall et al.* [1994] and that there is no uncontrolled field bur-



**Table 6.** Global Consumption of Synthetic N Fertilizers and Ammonia Emissions for Temperate and Tropical Zones

Fertilizer Type	Global Consumption <sup>a</sup> , Tg N yr <sup>-1</sup>	Temperate Zones, Gg N yr <sup>-1</sup>	Tropics, Gg N yr <sup>-1</sup>	Total, Gg N yr <sup>-1</sup>
Ammonium sulphate	2.6	34	169	203
Urea	29.2	1632	4137	5769
Ammonium nitrate	8.2	25	141	166
Calcium ammonium nitrate	4.1	9	72	82
Anhydrous ammonia	5.2	18	190	208
Nitrogen solutions	4.2	11	93	104
Other straight N	9.5	802	1189	1991
Ammonium phosphates	3.7	35	113	147
Other NP-N	3.2	18	77	95
NK-N	0.1	0	1	1
NPK-N	6.6	40	219	259
No N type	0.4	2	7	9
Total	77.0	2626	6409	9035

<sup>a</sup> Based on country estimates from IFA [1994] and FAO [1991].

ning. This implies that 70% of the available crop residues are burnt to generate energy.

The resulting burning of residues for sub-Saharan Africa including South Africa (14 and 61 Tg dm yr<sup>-1</sup> field burning and energetic use, respectively), North Africa (0 and 35 Tg dm yr<sup>-1</sup>), Middle East in Asia (12 and 33 Tg dm yr<sup>-1</sup>), Latin America (143 and 0 Tg dm yr<sup>-1</sup>), South Asia (68 and 161 Tg dm yr<sup>-1</sup>), East Asia (78 and 11 Tg dm yr<sup>-1</sup>), China (38 and 355 Tg dm yr<sup>-1</sup>), Oceania (14 and 5 Tg dm yr<sup>-1</sup>), Europe (97 and 0 Tg dm yr<sup>-1</sup>), North America (151 and 0 Tg dm yr<sup>-1</sup>) and the former USSR (74 and 0 Tg dm yr<sup>-1</sup>) yield a global total of about 710 Tg dm yr<sup>-1</sup> for field burning and 660 Tg dm yr<sup>-1</sup> for energetic use of agricultural wastes. The global volume of waste burnt of about 1370 Tg dm yr<sup>-1</sup> (including energetic use) is 70% of the estimate presented by *Andreae* [1991]. The regional estimates are consistent with *Barnard and Kristofersen* [1985], with most waste burning occurring in China, India, and Bangladesh, mainly for energy purposes. *Zhuang et al.* [1995], however, report that in China the use of residues for energy is decreasing currently, and the occurrence of field burning is increasing. The distribution of field burning of agricultural residues was based on the spatial distribution of arable land as defined by *Olsen et al.* [1983], while the distribution of energetic use of wastes was based on population densities modified by J. Logan (personal communication, 1993) from *Lerner et al.* [1988].

**2.3.4. Other biofuel burning.** Other biofuel burning includes the use of fuelwood, charcoal, vegetal waste, bagasse, and dung in the residential and industry sectors. The biofuel consumption data were based upon *Hall et al.* [1994], with subdivisions over different fuel types from *Olivier et al.* [1996]. The global amount for these categories is 2513 Tg dm yr<sup>-1</sup>. The distribution used for biofuel use is the database of population densities modified by J. Logan (personal communication, 1993) from *Lerner et al.* [1988].

**2.3.5. Ammonia emission from burning.** Emissions of NH<sub>3</sub> from biomass burning arise principally as a result of the nitrogen content of the fuel. In determining the global emission of NH<sub>3</sub>, emission factors from the literature were re-

viewed (Table 7). However, few such factors are available in comparison to other trace gases such as CO<sub>2</sub>, CO, and NO<sub>x</sub>. A series of observations of wildfires, mostly temperate forest and shrub, have been reported by the University of Washington's research group [*Hegg et al.*, 1988, 1990; *Radke et al.*, 1991; *Laurson et al.*, 1992; *Nance et al.*, 1993]. The emission factors from individual fires were calculated from their ratio to C-containing gases measured in the plumes. This methodology has some uncertainty as *LeBel et al.* [1988] found a poor correlation between CO<sub>2</sub> and NH<sub>3</sub> and considered that an emission factor normalized to CO<sub>2</sub> was inappropriate, whereas *Laurson et al.* [1992] report good correlations between the ratio of NH<sub>3</sub> to the ratio of CO to CO<sub>2</sub> emission factors for most of their observations.

A further uncertainty is the emission of NH<sub>3</sub> by flame type. *Hegg et al.* [1988] reported that one of their fires had an emission ratio of NH<sub>3</sub>/CO that differed from another fire (with an identical fuel) by a factor of 19. The higher emission fire had wetter fuel and was essentially a smoldering fire, rather than a flaming fire. Thus the more moderate heating of the soil would produce NH<sub>4</sub><sup>+</sup> from pyrolysis of organic matter, whereas the higher-temperature flaming fire would oxidize the N through to NO<sub>x</sub> [*Hegg et al.*, 1988]. In a series of laboratory experiments, *Lobert et al.* [1991] found that from the 15 experiments, 85% of the ammonia was released in the smoldering stage, and only 15% was released in the flaming stage, qualitatively in agreement with *Hegg et al.* [1988]. In contrast to these findings the results of *LeBel et al.* [1991] are less equivocal, finding emissions higher in the smoldering stage by factors ranging between approximately 30 and 66% in three of four fires and almost an order of magnitude smaller in another. Furthermore, *Lee and Atkins* [1994] found lower emissions of NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> from an agricultural waste burn with a water content of 40% than from one with 14% water content. The flame temperature in the wetter burn was lower by 300°C. It would appear that there is evidence for higher emission during the smoldering stages of fire but that this evidence is not unequivocal nor yet sufficiently quantitative to extrapolate emissions by flame type.

**Table 7.** Measurements of  $\text{NH}_3$  emission from fires

Fuel Type	N/C, %	Scale	Sampling	$\Delta\text{CO}/\Delta\text{CO}_2$ , %	Emission Ratio, (mmol $\text{NH}_3$ / mol $\text{CO}_2$ )	Emission Factor		Remarks	Reference <sup>a</sup>
						mg N/g C	N, %		
Chaparral, pine, fir	-	30-20,000 ha	aircraft	5.5	1.4	1.6±0.8	—		1
Grass wetland	-	1,250 ha	helicopter	—	0.04-1.24	—	—		2
Boreal forest	-	220 ha	helicopter	—	23 (22-31)	—	—	Lowest value in range for flaming;	3
Wetland	-	500-700 ha	helicopter	—	8 (6-11)	—	—	highest value for smoldering	3
(Sub)-Tropical savanna	0.88	plots	ground based	1.2-5.5	—	—	—		4
(Sub)-Tropical savanna	0.88	15-20 km <sup>2</sup>	aircraft	9.0±2.6	2.9±1.9	2.0±1.1	23±13		4
(Sub)-Tropical savanna	1.1	>500m fronts	aircraft	5.8±2.0	8.6±5.3	0.9±0.6	8.6±5.5		5
Tropical savanna	-	—	—	6.2±1.0	0.7±0.4	—	—		6
Tropical savanna	0.6	4x 1 ha plots, + 10 km <sup>2</sup> fire	ground based	6.1	0.057	0.04	10	flaming only	7
Rain forest	-	large scale	airborne	8.0	—	—	—	flaming only	8
Sagebush	-	plots	aircraft	8.4±4	0.92	—	—	particulate $\text{NH}_4^+$ only	9
Forest slash	-	plots	ground based	14.1-18.1	8.3-8.4	—	—		10
Tropical grasses plus agricultural waste	-	plots	ground based	18.3-19.6	1.4-3.3	—	—		10
			laboratory	—	—	0.8	4±3	75% flaming, 25% smoldering	11

Dash indicates no data available.

<sup>a</sup> References are as follows: 1, Hegg *et al.* [1988, 1990]; 2, LeBel *et al.* [1988]; 3, LeBel *et al.* [1991]; 4, Hurst *et al.* [1994a]; 5, Hurst *et al.* [1994b]; 6, Andreae *et al.* [1997]; 7, Delmas *et al.* [1995]; 8, Lacaux *et al.* [1995]; 9, Andreae *et al.* [1988]; 10, Griffith *et al.* [1991]; 11, Lobert *et al.* [1990].

It is not yet possible to use different emission factors for the various types of biomass burning on the basis of the literature data in Table 7. Therefore the emission ratio of 1.3 mmol NH<sub>3</sub> mol<sup>-1</sup> CO<sub>2</sub> (1.52 × 10<sup>-3</sup> g N/g C) as proposed by *Andreae* [1991] was used, recognizing that this may overestimate emissions from burning for energy generation which generally occur under more controlled conditions. The global total of 5.9 Tg N yr<sup>-1</sup> (Table 8 and Plate 1c) is similar to the estimate by *Schlesinger and Hartley* [1992] and *Dentener and Crutzen* [1994].

## 2.4. Natural Ecosystems

**2.4.1. Soils under natural vegetation.** Many organisms in soils involved in the decomposition of organic matter excrete NH<sub>3</sub> directly or N compounds that readily hydrolyze to NH<sub>x</sub> (NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup>). In equilibrium conditions, which are assumed for natural ecosystems, all aboveground and belowground NPP is turned over in the soil each year. Therefore NH<sub>3</sub> fluxes are related closely to biological activity in the soil (see literature reviews by *Schlesinger and Hartley* [1992] and *Langford et al.* [1992]). Here a simple method is utilized to calculate decomposition in soils and the subsequent emission of NH<sub>3</sub>, accounting for the portion absorbed in the vegetation canopy. The results of this method are compared with available measurements from the literature.

Annual decomposition in soils covered by natural vegetation was based upon the global distribution of NPP from *Klein Goldewijk et al.* [1994], assuming that NPP includes aboveground and belowground production. The C/N ratios for different vegetation types proposed by *Melillo et al.* [1993] were used to derive net N mineralization. The rates of net N mineralization (Table 9) calculated in this way were found to be consistent with those given by *Nadelhoffer et al.* [1991].

All NH<sub>x</sub> generated below several centimeters from the surface may remain in the system, while that produced at or near the surface is vulnerable to loss [*Woodmansee*, 1978]. Hence it is necessary to know how much N is cycled in the topsoil, for which a depth of 10 cm was assumed. The ratio of litter to belowground inputs of organic matter ranges from 30:70 to 70:30 [*Vogt et al.*, 1986; *Trumbore et al.*, 1995]. All litter and 10% of the rhizodeposition is assumed to be decomposed in the soil's surface layer of 10 cm. Hence decomposition in the topsoil ranges from 30% (±7%) to 70% (±3%) of NPP. Therefore approximately 50% (±20%) of the N mineralization occurs in topsoils and is present as NH<sub>x</sub> which is vulnerable to

loss by volatilization at some point in time during the year. In temperate ecosystems most of the N mineralization occurs in summer when the N released is readily taken up by plants. During autumn and spring, N mineralization might exceed plant uptake, so that there may be an increase in soil ammonium. In tropical seasonal climates most N is mineralized at the beginning of the rainy season. In tropical wet climates (rain forest) the N mineralization is a more constant process during the year.

An emission coefficient of 1% was used for the NH<sub>3</sub> escaping from the NH<sub>x</sub> present in the soil moisture and gas phase in the topsoil. This coefficient is lower than those used for ammonium yielding fertilizers and anhydrous ammonia, as the NH<sub>x</sub> concentration in undisturbed soils during the relatively slow decomposition process is expected to be lower than that in recently fertilized soils. The calculated NH<sub>3</sub> emission from soils to the air below the canopy is 5.1 Tg NH<sub>3</sub>-N yr<sup>-1</sup> of the 1010 Tg yr<sup>-1</sup> net N mineralization in all ecosystems considered (Table 9).

The NH<sub>3</sub> emitted from the soil surface may be absorbed by plant leaves. *Denmead et al.* [1976] showed that virtually all NH<sub>3</sub> emitted by the soil was reabsorbed or deposited in the canopy. Dense canopies may absorb NH<sub>3</sub> effectively, as gaseous transport is much faster through stomata than through the soil matrix, and the leaf area of a dense canopy is often much greater than the soil area below. Furthermore, deposition to the canopy can be enhanced in the presence of dew or rainwater on leaves [*Denmead et al.*, 1976]. In order to account for losses of NH<sub>3</sub> to the canopy, we utilize canopy absorption coefficients [*Lee et al.*, 1997a]. These coefficients are not well based physically but are a first-order approximation for the effect. The coefficients are 0.8 for tropical rain forest, 0.5 for other forests and woodlands, and 0.2 for all other vegetation types including tundras, grasslands, shrub lands, and arable land. Moist tropical rain forests have a high leaf area index, and for a large proportion of the time the canopy is wet or moist. Thus the fraction of the NH<sub>3</sub> that is deposited in the canopy may exceed that in dry or open forests. Another reason to assume lower canopy absorption for temperate deciduous and tropical seasonal forests is that an important part of the NH<sub>3</sub> volatilizes from the soil in fall and spring when decomposition is fast, periods during which the leaf area is smaller. The absorption coefficient for grass may be low compared to the observations of *Denmead et al.* [1976]. However, the uncertainties in NPP estimates, C/N ratios, the ratio of aboveground to belowground production, the effect of soil pH, soil wetness, and soil gas conductivity are so large that we have not attempted to arrive at refined estimates for canopy reduction coefficients. In addition, soils can be sinks for NH<sub>3</sub> under certain conditions [*Malo and Purvis*, 1964; *Hannwald*, 1969], although the methods used in these studies may have led to overestimation of the fluxes [*Denmead*, 1990; *Dawson*, 1977]. Data are lacking to quantify soil uptake of NH<sub>3</sub> in natural ecosystems. The result is that about 50% of the soil global emission is captured by canopies, and the amount of NH<sub>3</sub> escaping from the canopies to the atmosphere is 2.4 Tg NH<sub>3</sub>-N yr<sup>-1</sup> (Table 9).

The method presented above is much simpler than that of *Dawson* [1977], who used NPP, decomposition, and nitrification to estimate soil exchangeable NH<sub>4</sub><sup>+</sup>, modeled soil pH,

**Table 8.** Global Estimates of Biomass Burning and NH<sub>3</sub> Emission

Activity	Material Burnt		NH <sub>3</sub> Emission, Tg N yr <sup>-1</sup>
	Pg C yr <sup>-1</sup>	Pg dm yr <sup>-1</sup>	
Deforestation	1.0	2.1	1.4
Savanna burning	1.2	2.7	1.8
Agricultural residues			
Field burning	0.3	0.7	0.5
Energetic use	0.3	0.7	0.5
Other biofuels	1.1	2.5	1.7
Total	3.9	8.6	5.9

**Table 9.** Global Areas, Estimated N Mineralization, and NH<sub>3</sub> Volatilization With and Without Canopy Absorption for Soils Under Natural Vegetation

Type	Area, Mha	N <sub>min</sub> <sup>a</sup> , Tg yr <sup>-1</sup>	NH <sub>3</sub> -N volatilization <sup>b</sup>			
			Soil, Tg yr <sup>-1</sup>	Canopy, Tg yr <sup>-1</sup>	Soil, g m <sup>-2</sup> yr <sup>-1</sup>	Canopy, g m <sup>-2</sup> yr <sup>-1</sup>
Closed tropical forests	1,671	392	2.0	0.4	0.12	0.03
Open tropical forests	349	62	0.3	0.2	0.09	0.04
Tropical savannas	672	130	0.7	0.3	0.10	0.05
Temperate forests	2,232	119	0.6	0.3	0.03	0.01
Shrub lands	604	59	0.3	0.2	0.05	0.04
Tundras <sup>c</sup>	1,225	16	0.1	0.1	0.01	0.01
Deserts	1,872	28	0.1	0.1	0.01	0.01
Grasslands <sup>d</sup>	2,376	204	1.0	0.8	0.04	0.03
Arable lands <sup>e</sup>	1,429					
Intensively used grasslands <sup>e,f</sup>	752					
Total	13,185	1010	5.1	2.4	0.04	0.02

<sup>a</sup> N<sub>min</sub>, N mineralization.<sup>b</sup> Soil denotes the flux from the soil, Canopy denotes the flux from the canopy (equals soil flux - canopy absorption).<sup>c</sup> Including Siberian parks and marshes/swamps.<sup>d</sup> The delineation of grasslands used is from *Bouwman et al.* [1995] on the basis of *Olsen et al.* [1983]. The "natural" grassland complex of 2376 Mha includes Mediterranean grazing areas, warm grass/shrub, and cool grass/shrub complexes.<sup>e</sup> Emissions from decomposing plant material in croplands and intensively used grasslands are included in the estimates for emissions resulting from grazing animals and decomposing crop residues.<sup>f</sup> The 752-Mha difference between 2181 Mha of total agricultural land of *Olsen et al.* [1983] and 1429 Mha of arable land (including fallow land) was assumed to be part of the intensively used grassland complex. The total area of grasslands including the "natural" grasslands is thus 3128 Mha, which is somewhat less than the global area of grasslands given by *FAO* [1991] for 1990. Correction was not considered justifiable because of the uncertainties in estimates of extent and use of grasslands.

and NH<sub>3</sub> concentration in the soil atmosphere to calculate a global NH<sub>3</sub> flux to the atmosphere of 39 Tg NH<sub>3</sub>-N yr<sup>-1</sup>. The estimate presented here of 2.4 Tg N yr<sup>-1</sup> is more than a factor 10 lower. Obvious reasons for this difference are (1) *Dawson* [1977] assumed that most ammonium is concentrated in the top 10 cm of the soil, while in our calculations 50% of all N in NPP is assumed to be cycled within the top 10 cm, and (2) *Dawson* [1977] assumed no canopy reduction and even stated that in natural ecosystems re-absorption of NH<sub>3</sub> is less important than in cultivated land.

Contrary to *Dawson* [1977], who used modeled soil pH in his calculations for NH<sub>3</sub> volatilization, we decided not to use pH because of the lack of correlation between climate and soil pH [*Langford et al.*, 1992] and because of the paucity of data on soil pH on the global scale, as discussed by *Bouwman et al.* [1993]. In addition, there is evidence that NH<sub>3</sub> volatilization rates may be high even in acid soils [*Blasco and Cornfield*, 1966], and soil pH varies in space and time [e.g., *Van Cleve and Powers*, 1995].

The data on soil NH<sub>3</sub> emissions from soil under natural vegetation from the literature (Table 10) are presented on an annual basis to allow for comparison with the computed emissions. The emissions range considerably, between 0 and 3.5 g N m<sup>-2</sup> yr<sup>-1</sup>. Most of the expert judgements of soil

emission rates from *Schlesinger and Hartley* [1992] are consistent with our computed emission rates, except those for tropical savannas and temperate forests, for which *Schlesinger and Hartley* [1992] proposed much higher rates; their estimates may have been biased by information obtained at a single site. The data used by *Schlesinger and Hartley* [1992] to derive emission rates for tropical savanna are from *San José et al.* [1991] for the wet season from a few days of observations only. As NH<sub>3</sub> emissions from the soil are closely related to biological activity, annual values accounting for low emission during the dry season may be much lower than the wet season values. The data from Korea may overestimate the true emission rate because of the continuous trapping measurement technique used (see Table 10). The remaining flux data for temperate forests for summer and winter conditions (Table 10) could be used to infer an annual flux close to our computed flux of 0.03 g N m<sup>-2</sup> yr<sup>-1</sup>. On the basis of these considerations it is concluded that the computed soil emissions (Table 9) are not inconsistent with the measured emissions (Table 10).

The flux between the canopy and atmosphere is the net result of the soil emission, absorption within the canopy, and upward and downward fluxes between canopy and atmosphere. The absorption component cannot be isolated easily.

**Table 10.** Measurements of NH<sub>3</sub> Emission from Undisturbed Soils Under Natural Vegetation

Vegetation Type	Location	Flux, g N m <sup>-2</sup> yr <sup>-1</sup>	Season, Length of Period of Measurements, days	Reference <sup>a</sup>
<i>Tropical open forest</i>				
Tropical savanna	Venezuela	3.5	July/August, [wet season], 3	1
<i>Temperate forests</i>				
Coniferous forest	Oslo, Norway	0.1	summer, 72	2
Coniferous forest	Canada	0.002	autumn/winter, 41	3
<i>Shrub lands</i>				
Shrub steppe	Washington, United States of America	<0.05	annual <sup>b</sup>	4
<i>Tundras</i>				
Tundra -alpine tundra	United States of America - Sweden	0	annual <sup>b</sup>	5
<i>Deserts and Semideserts</i>				
Desert grassland	southern. New Mexico, United States of America	<0.05	annual <sup>b</sup>	4
Temperate desert	New Mexico, United States of America	0.005-0.03 <sup>c</sup>	summer 1988/ 1989, 7	6
Cold winter semi-desert	Utah, United States of America	0.1 <sup>d</sup>	annual estimate, based on 10 week measurements	7 8
<i>Grasslands</i>				
Mixed prairie	southern Dakota, United States of America	0.1	annual <sup>b</sup>	4
Shortgrass steppe	NE Colorado, United States of America	<0.05	annual <sup>b</sup>	4, 9
Shortgrass steppe	Colorado, United States of America	<0.01 <sup>e</sup>	17 months	10
Shortgrass steppe	northern Texas, United States of America	0.05	annual <sup>b</sup>	4
Tallgrass prairie	Oklahoma, United States of America	0.2	annual <sup>b</sup>	4

Schlesinger and Hartley [1992] presented a number of flux measurements from the literature. Many of these measurements were made in disturbed or agricultural systems or grazed pastures [Erisman *et al.*, 1988; Georgii and Lenhard, 1978; Goethel quoted in Warneck, 1988; Harrison *et al.*, 1989; Hooker *et al.*, 1973; Kissel *et al.*, 1977; Lenhard and Gravenhorst, 1980; Lightner *et al.*, 1990; Ruess and McNaughton, 1988; Ruess *et al.*, 1989; Ventura and Yoshida, 1977], and some data represent water-air fluxes [Murphy and Brownlee, 1981]. Finally, the measurements by Kim [1973] were based on an enclosure method with continuous trapping that could create a gradient from soil to air and thus overestimate the emission of NH<sub>3</sub> [Bowden, 1986].

<sup>a</sup> References are as follows: 1, San José *et al.* [1991]; 2, Overrein [1968]; 3, Marshall and Debell [1980]; 4, Woodmansee [1978]; 5, Van Cleve and Alexander [1981]; 6, Schlesinger and Peterjohn [1991]; 7, West and Skujins [1977]; 8, Skujins and Eberhardt [1973]; 9 Woodmansee *et al.* [1978]; 10, Schimel *et al.* [1986].

<sup>b</sup> Compilation based on International Biological Program data.

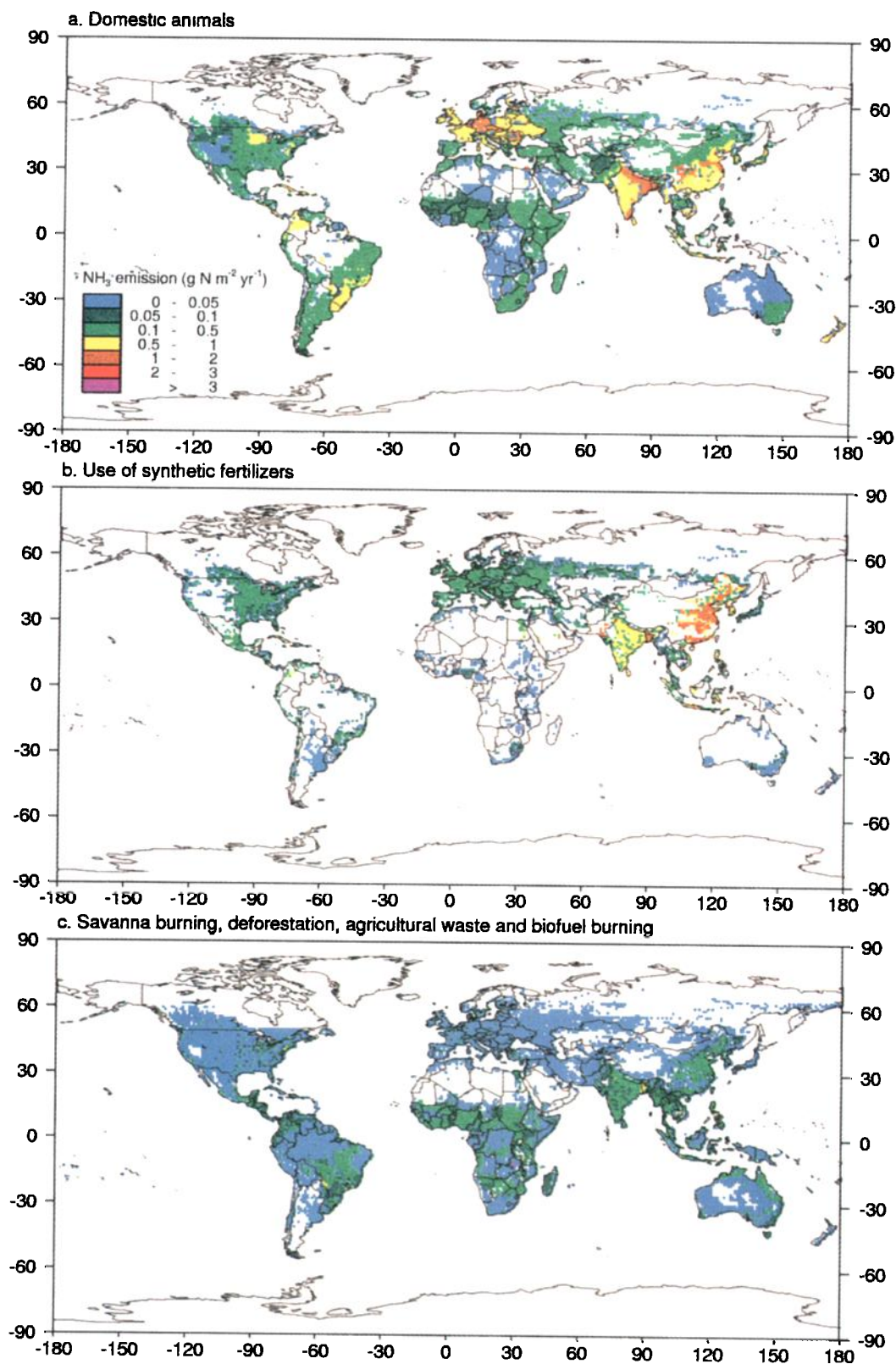
<sup>c</sup> Pretreatment flux was 0.004 (one observation).

<sup>d</sup> N fixation due to occurrence of legumes.

<sup>e</sup> Includes effects of grazing.

The only attempt to account for canopy absorption was made by Denmead *et al.* [1976], who estimated that most of the NH<sub>3</sub> emitted from the soil was absorbed by ryegrass/clover canopy. Other studies discussed by Langford *et al.* [1992] indicate negative (deposition) or zero fluxes from grasslands and a temperate forest. Our emission estimate of 0.03 g N m<sup>-2</sup> yr<sup>-1</sup> for natural grasslands is consistent with the estimate of <0.06 g N m<sup>-2</sup> yr<sup>-1</sup> presented by Langford *et al.* [1992] and

Heckathorn and Delucia [1995], but our estimate of 0.01 g N m<sup>-2</sup> yr<sup>-1</sup> for temperate forests is lower than the 0.04 g N m<sup>-2</sup> yr<sup>-1</sup> for U.S. temperate forests estimated by Langford *et al.* [1992]. It should be noted that in our computations there is variation within each vegetation complex due to differences in NPP. The global value for temperate forest may be low because of the low NPP in large unproductive forested areas, and in our calculations the fertilizing effect of additions from N depo-



**Plate 1.** Distribution of NH<sub>3</sub> emission in g N m<sup>-2</sup> yr<sup>-1</sup> from three sources. (a) Excreta from domesticated animals. The global emission from this source is 21.6 Tg NH<sub>3</sub>-N yr<sup>-1</sup>, (b) Synthetic fertilizer use. The global emission from this source is 9.0 Tg NH<sub>3</sub>-N yr<sup>-1</sup>, (c) Biomass burning (savanna burning, deforestation, agricultural waste burning, and biofuel combustion) The global emission from this source is 5.9 Tg NH<sub>3</sub>-N yr<sup>-1</sup>



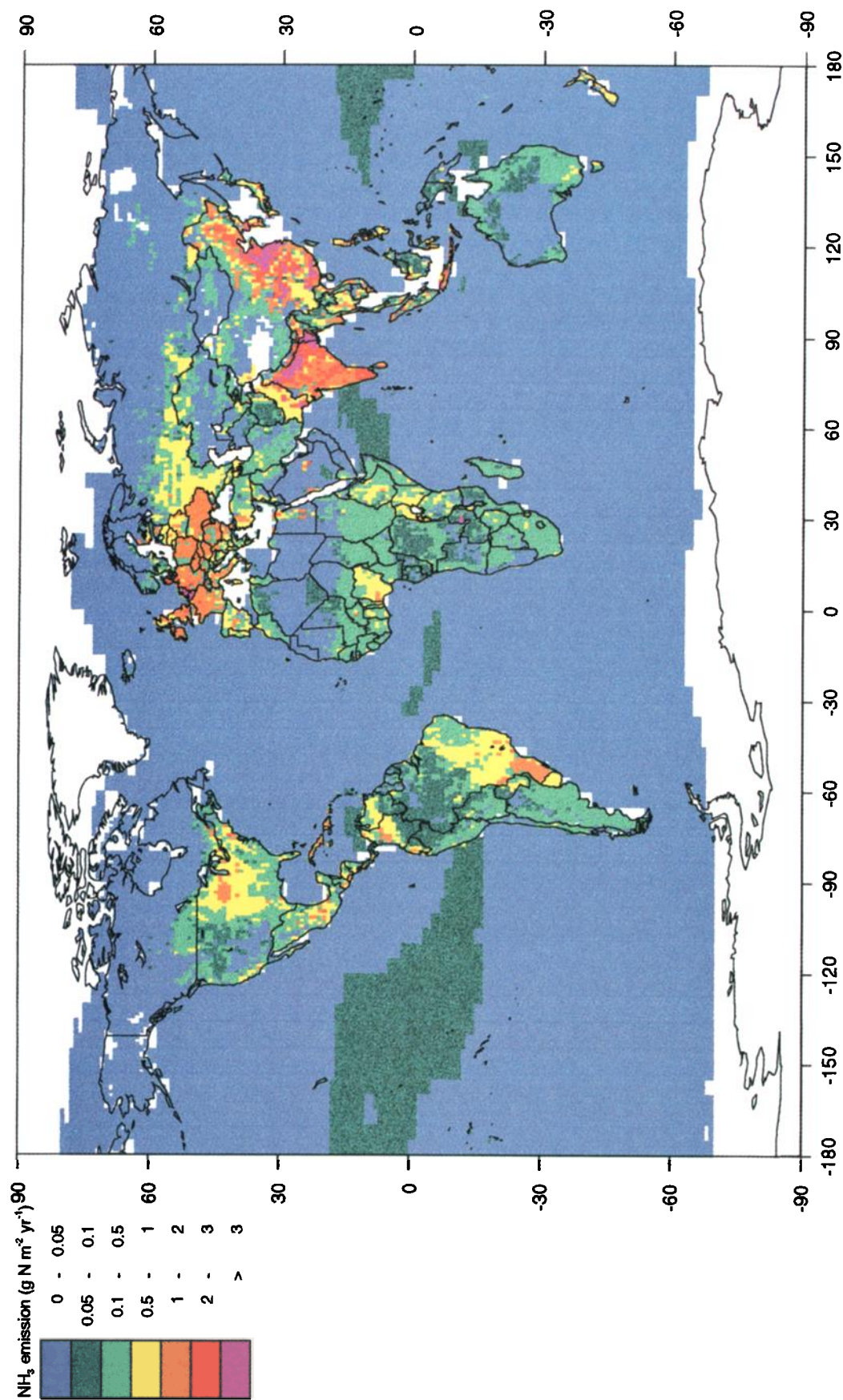


Plate 2. Distribution of  $\text{NH}_3$  emission in  $\text{g N m}^{-2} \text{yr}^{-1}$  from all terrestrial sources and the oceans. The global total emission is 54 Tg  $\text{NH}_3\text{-N yr}^{-1}$ .

sition to the soils is not accounted for. Despite the lack of measurement data we have attempted to constrain this source on the basis of analysis of the amount of  $\text{NH}_x$  in soils that is vulnerable to loss, soil emissions, and canopy absorption. This has led to a much lower global total than that of *Schlesinger and Hartley* [1992], which also included canopy emissions and emissions from wild animals.

**2.4.2. Natural vegetation.** In addition to emissions from the soils under natural vegetation the canopy may also act as a source of atmospheric  $\text{NH}_3$ . Recently, *Langford and Fehsenfeld* [1992] showed that under circumstances of high atmospheric  $\text{NH}_3$  concentrations a pine forest acted as a sink for  $\text{NH}_3$  and at low atmospheric  $\text{NH}_3$  concentrations acted as a source. The atmospheric concentration above which plants assimilate and below which they emit  $\text{NH}_3$  is defined as the "compensation point". The compensation point is strongly dependent on temperature, mesotrophic ammonium, concentration, and pH. *Langford et al.* [1992] showed that because of the existence of a compensation point, the fluxes between the canopy and the atmosphere are nearly always smaller than those dictated by the soil equilibrium discussed in section 2.4.1.

One further aspect of soil emissions is worth noting here. *Lau and Charlson* [1977] noticed that in the relatively sparsely populated middle and western part of the United States sulfates were almost completely neutralized by  $\text{NH}_3$ , whereas acidic sulfates prevailed in the industrialized eastern United States. Hence, in regions with relatively low ratios of acidic to alkaline emissions another constraint to the  $\text{NH}_3$  emission from soils and (natural) vegetation may be given by the availability of free acidity. In fact, higher "emissions" would lead to atmospheric  $\text{NH}_3$  concentration above equilibrium and hence immediate "deposition" resulting in no net exchange. This may lead to higher N emissions in the case of increasing acidic concentrations, but apparently the neutralizing potential of  $\text{NH}_3$  emissions from soils/vegetation is limited.

*Dentener and Crutzen* [1994] calculated that  $5.2 \text{ Tg N yr}^{-1}$  could be emitted from natural vegetation worldwide, assuming a temperature-dependent compensation point. However, they did not consider soil emissions. Hence the emission from the canopy could be the remainder of soil emissions minus the uptake by the canopy. The uptake of the soil emissions within the canopy is then determined by the compensation point. The effective emission from the soil-vegetation complex depends on the net soil equilibrium concentration, the turbulent transport through the canopy, the mesotrophic compensation point, and the exchange between the plant leaves and the atmosphere (stomatal resistance). As the compensation point of natural vegetation is highly uncertain and the "soil" emissions are somewhat better constrained, we take only the soil emissions into consideration in this inventory. We recognize that by using the "canopy" absorption factor, the exchange with the vegetation is implicitly accounted for.

## 2.5. Oceans

The potential for oceans to be a source of atmospheric  $\text{NH}_3$  has been recognized for some time [*Kawamura*, 1968], although some debate ensued as to whether the source was adjoining land masses or the sea itself [*Tsunogai*, 1971; *Georgii and Gravenhorst*, 1977]. Much of the earlier work on sea-

surface/atmosphere exchange focused upon oceans as being a sink of  $\text{NH}_3$  [e.g., *Hicks and Liss*, 1976], and it was not until the work of *Quinn et al.* [1987, 1988, 1990, 1992, 1996] that the potential for oceans to be a significant global source of  $\text{NH}_3$  was discussed. *Schlesinger and Hartley* [1992] derived a global flux of  $13 \text{ Tg N yr}^{-1}$  by extrapolating a Pacific emission flux of  $7 \mu\text{mol m}^{-2} \text{ d}^{-1}$ , reported by *Quinn et al.* [1990], to the global oceanic area. *Dentener and Crutzen* [1994] subsequently made a more refined estimate by scaling the emission of oceanic  $\text{NH}_3$  to DMS emissions according to the relationship proposed by *Liss and Galloway* [1993], resulting in a global emission of  $7 \text{ Tg N yr}^{-1}$ . However, it has been found that  $\text{NH}_3$  may be either deposited to or emitted from seawater [*Lee et al.*, 1997b; *Asman et al.*, 1994]. The direction of the flux depends upon a number of parameters, including atmospheric concentration of  $\text{NH}_3$ , seawater  $\text{NH}_x$ , salinity, and temperature. This bidirectional exchange is similar in concept to the so-called compensation point exhibited by some plant surfaces [*Farquhar et al.*, 1980], whereby if atmospheric concentrations fall below the compensation point, the surface may emit. *Asman* [1994] has presented a model which predicts the atmospheric concentration of  $\text{NH}_3$  ( $c_s^*$ ) over sea surfaces according to the following equation:

$$c_s^* = \frac{[\text{NH}_x]}{RTH_{\text{NH}_3} \left( \frac{1}{\gamma_{\text{NH}_3}} + \frac{10^{-\text{pH}}}{\gamma_{\text{NH}_4} K_{\text{NH}_4}} \right)} \quad (3)$$

where:

- $c_s^*$   $\text{NH}_3$  concentration in air which would be in equilibrium with  $[\text{NH}_x]$  ( $\text{mol m}^{-3}$ );
- $[\text{NH}_x]$   $\text{NH}_x$  concentration in the surface ( $\text{mol m}^{-3}$ );
- $H_{\text{NH}_3}$  Henry's law coefficient ( $\text{mol L}^{-1} \text{ atm}^{-1}$ );
- $\gamma_{\text{NH}_3}$  activity coefficient  $\text{NH}_3$  in water;
- $\gamma_{\text{NH}_4}$  activity coefficient  $\text{NH}_4^+$ ;
- $R$  gas constant ( $8.0275 \times 10^{-2} \text{ atm} \cdot \text{L mol}^{-1} \cdot \text{K}$ );
- $T$  temperature of seawater ( $^{\circ}\text{K}$ );
- $\text{pH}$  pH in seawater;
- $K_{\text{NH}_4}$  dissociation constant for  $\text{NH}_4^+$  ( $\text{mol L}^{-1}$ ).

This model has been used to show that some polluted coastal waters in the North Sea may emit  $\text{NH}_3$  [*Asman et al.*, 1994] but that for the North Sea as a whole, surface air concentrations of  $\text{NH}_3$  are high enough and seawater  $\text{NH}_x$  is low enough that it is likely that the net flux is downward [*Lee et al.*, 1997b].

In other more remote oceanic regions, measurements show that the sea can be a source of  $\text{NH}_3$ , although the dynamics are not well understood. In order to estimate the  $\text{NH}_3$  flux into the atmosphere the Hamburg Model of the Ocean Carbon Cycle (HAMOCC3), modified to include plankton cycling [*Six and Maier-Reimer*, 1996], has been used to generate sea-surface concentrations of  $\text{NH}_4^+$  and equilibrium gas phase concentrations  $c_s^*$  (equation (3)) (K. D. Six and F. J. Dentener, Exchange of dimethylsulphide and ammonia between the ocean and the atmosphere calculated using a global oceanic and atmospheric model, manuscript in preparation, 1997). In this ocean chemical-transport model, seawater  $\text{NH}_x$  is diagnostically calculated from phosphate variations due to plankton activity applying the Redfield ratio of N:P of 16. Ammonium



is released as a result of zooplankton metabolism during grazing on phytoplankton or the decay of organic material. This  $\text{NH}_4^+$  is then available to phytoplankton and bacteria as a nutrient. A small fraction of  $\text{NH}_4^+$  leaves the ocean as gaseous  $\text{NH}_3$  from exchange with the atmosphere.

The calculated oceanic  $\text{NH}_4^+$  concentrations are highly uncertain because of the nearly unknown magnitude of bacterial activity in the ocean. However, a preliminary comparison of modeled open ocean  $\text{NH}_4^+$  with measurements did not indicate large discrepancies, except in coastal regions [Quinn *et al.*, 1996]. The choice of parameters, their associated uncertainties, and a comparison with measurements is given elsewhere (Six and Dentener, manuscript in preparation, 1997). Major uncertainties involve the bacterial uptake rate of  $\text{NH}_4^+$ , the calculated plankton concentrations, the rate of sea-atmosphere exchange of  $\text{NH}_3$ , and the rate of removal by chemical reaction and upward transport of  $\text{NH}_3$  in the atmosphere.

The chemical removal rate of the emitted  $\text{NH}_3$  in the atmosphere is, in turn, a strong function of DMS emissions and consequent conversion to  $\text{SO}_2$  and sulfuric acid. The estimated global emission of  $8.2 \text{ Tg N yr}^{-1}$  is therefore highly uncertain but compares favorably with oceanic and atmospheric measurements [Quinn *et al.*, 1996; Dentener and Crutzen, 1994].

It should be noted that the above mentioned global emission of  $8.2 \text{ Tg N yr}^{-1}$  is not yet corrected for the existence of nonzero atmospheric  $\text{NH}_3$  concentrations. Preliminary results indicate that the net sea-atmosphere flux may be reduced by a factor of 2, when these atmospheric concentrations are taken into account (Six and Dentener, manuscript in preparation, 1997).

## 2.6. Fossil Fuel Combustion

Information on  $\text{NH}_3$  emissions from fuel combustion and industrial processes (i.e., noncombustion sources) is either scarce or quite uncertain, in particular, emission factors for fuel combustion. As agricultural sources such as animal waste and fertilizer application are the dominating sources of ammonia, these minor sources are often neglected. Here the discussion has been limited to those sources which contribute significantly to this 'miscellaneous' category.

For fossil fuel combustion, emission factors for  $\text{NH}_3$  are very scarce. Only three recent reports are available: Dianwu and Anpu [1994], Klaassen [1992], and McInnes [1996]. The emission factors given by Dianwu and Anpu [1994] appear to be excessively large, as can be determined when applied to national conditions for countries which have reported national ammonia inventories, such as the Netherlands. Klaassen [1992] and McInnes [1996] cite only emission factors for road transport and some data for stationary sources, mainly for coal combustion. In particular, the emission factor for petrol-engined cars equipped with three-way catalysts is greater than that for cars with no emission controls. Here emission factors of 0.07, 4.26, and  $0.31 \text{ g N GJ}^{-1}$  (equivalent to 1, 60, and  $3 \text{ mg N km}^{-1}$ , respectively) have been assumed for uncontrolled petrol-engined cars, three-way catalyst cars, and for diesel vehicles, respectively. In 1990, catalyst-equipped car fleets were confined mainly to the United States of America, Canada, Japan, and to some extent also Australia, Germany, and the Netherlands.

Available data indicate that the emission factor for combustion of coal and natural gas is of the order of  $1 \text{ g N GJ}^{-1}$  or less, yielding a negligible global emission. We have not included stationary oil combustion, because no emission factors were available. For coke production, emission factors of  $3 \text{ g N GJ}^{-1}$  for uncontrolled production in countries not a part of the Organization for Economic Cooperative Development (OECD) and  $0.3 \text{ g N GJ}^{-1}$  for OECD countries (assuming controlled production) were used (based on Buijsman *et al.* [1987]; EPA [1985]; Eggleston [1992]; Allemand [1992]; Gaudioso and Trozzi [1992]; European Centre for Ecotoxicology (ECETOC) [1994]). Essentially, all fossil energy data were taken from International Energy Agency / Organization for Economic Cooperation and Development data (Energy statistics of OECD countries and non-OECD countries 1971-1992, data diskettes dated 17-03-1994 and 28-07-1994 for OECD and non-OECD countries, respectively, conversion factor diskettes dated 06-09-1994 and 28-07-1994, respectively, Paris, 1994), supplemented with data for road transport from Olivier *et al.* [1996], resulting in a total global emission of  $0.1 \text{ Tg N yr}^{-1}$  with an uncertainty range of 0.0–0.3  $\text{Tg N yr}^{-1}$ . Road transport is responsible for 85% of this total. The emissions were distributed according to the human population densities modified by J. Logan (personal communication, 1993) from Lerner *et al.* [1988].

## 2.7. Industrial Processes

More data are available on  $\text{NH}_3$  emissions from industrial processes, particularly the production of  $\text{NH}_3$  and of nitrogen fertilizers, nitric acid, and urea [e.g., Buijsman *et al.*, 1987; EPA, 1985; ECETOC, 1994; Eggleston, 1992; Allemand, 1992; Gaudioso and Trozzi, 1992]. Some reports also give emission factors for the production of caprolactam, acrylonitrile, calcium-ammonium nitrate, and zinc. For the global inventory, estimates were confined to the larger sources within this category for which production data were readily available. The sources included are the production of N fertilizer,  $\text{NH}_3$ , and nitric acid, for which globally uniform emission factors of 1647, 800, and  $41 \text{ kg N per Gg N}$ , respectively, were assumed in the product. Using production data from the United Nations (Industrial Commodity Production Statistics, International Environmental data Service, Geneva, data on diskette, 1993 and Industrial Commodity Production Statistics Database 1970-1992, New York, data on diskette, 1995) the total global emissions from industrial processes is  $0.2 \text{ Tg N yr}^{-1}$  with an uncertainty range of 0.1–0.3  $\text{Tg N yr}^{-1}$ , of which 65% stems from N fertilizer production and 35% stems from ammonia production. The spatial distribution for industrial sources of  $\text{NH}_3$  is the human population database modified by J. Logan (personal communication, 1993) from Lerner *et al.* [1988].

Emissions from the production of explosives may be compared with emissions from other nitrogen processing plants, notably N fertilizer production. As the world production of explosives is much less than that of N fertilizers, the emissions can be estimated to be much less than the  $0.2 \text{ Tg N}$  for identified industrial processes. Emissions from use of explosives may be related the emission of  $\text{N}_2\text{O}$  from this source, which according to Granli and Bøckmann [1994] is insignificant (less than  $0.01 \text{ Tg N yr}^{-1}$ ).

## 2.8. Humans

It is well documented that  $\text{NH}_3$  is released from humans, from breath, sweat, and excretion as a normal metabolic process [Lee and Dollard, 1994]. However, the source strength of humans is not well determined. Möller and Schieferdecker [1989] gave a factor of  $1.3 \text{ kg N person}^{-1} \text{ yr}^{-1}$ , which compared with indoor measurements of  $\text{NH}_3$ , assumed to be the result of human sweat and breath seemed too high [Atkins and Lee, 1993]. The assumption of indoor  $\text{NH}_3$  being the result of human sweat and breath seems valid, as concentrations of indoor  $\text{NH}_3$  are consistently up to an order of magnitude greater than those outdoors and show a significant correlation with occupancy (D. S. Lee, unpublished data, 1992). However, these data would imply a source strength of approximately  $0.01$  to  $0.2 \text{ kg N person}^{-1} \text{ yr}^{-1}$ . The other source of human-produced  $\text{NH}_3$  in many countries is the sewerage system. Sewage treatment plants represent a local source of atmospheric  $\text{NH}_3$  [Lee et al., 1992] which is relatively small and cannot justify the use of a total human emission factor as high as proposed by Möller and Schieferdecker [1989]. Another source of  $\text{NH}_3$  emissions closely associated with humans is that of domestic cats and dogs which are found in a ratio of approximately 1:10 pets:humans in the United Kingdom and the Netherlands [Lee and Dollard, 1994; Erisman, 1989]. The only emission factor proposed for these animals is that of Cass et al. [1982], who based their nitrogen excretion rates on rates given by Altman and Dittmer [1968]. These emission rates of  $0.66$  and  $1.98 \text{ kg N animal}^{-1} \text{ yr}^{-1}$  for cats and dogs, respectively, seem improbably high despite their very high nitrogen diet. Further difficulties exist in assigning a human emission factor. Western diets are relatively high in N which gives rise larger excretion rates of urea. Furthermore, sanitary arrangements and customs differ greatly across the world. However, it would be incorrect to ignore this source completely, and an emission factor of  $0.5 \text{ kg N person}^{-1} \text{ yr}^{-1}$  has been assigned without any regional differences and encompasses a consideration of  $\text{NH}_3$  emissions from domestic pets. The resultant global total is  $2.6 \text{ Tg N yr}^{-1}$ . The spatial distribution is assumed to be according to the human population densities modified by J. Logan (personal communication, 1993) from Lerner et al. [1988].

## 2.9. Crops

The emission of  $\text{NH}_3$  from agricultural crops depends on chemical, biological, and meteorological factors [Schjørring, 1991; Holtan-Hartwig and Bøckman, 1994]. The relatively few measurements that are available show that the emission varies with the plant species. In addition, the emission from plants varies during growth and development during the growing season. The emission is likely to increase with temperature, and it is stimulated by conditions of stress caused by plant diseases, pests, or adverse weather conditions, such as drought [Heckathorn and DeLucia, 1995]. At present, insufficient data are available to differentiate between the emission in temperate and tropical zones. For that reason the same emission factor of  $2.5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  is used for all agricultural crops. The emission factor is rather uncertain and could vary between  $0$  and  $5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ . The global total is  $3.6 \text{ Tg N yr}^{-1}$ . The  $\text{NH}_3$  emissions were simply assigned to the arable grid boxes given by Olsen et al. [1983] with corrections on the

basis of country-specific data on areas of annual and permanent crops taken from FAO [1991].

## 3. Discussion

The global estimate of  $\text{NH}_3$  emission from all sources is  $54 \text{ Tg N yr}^{-1}$  (Table 11). The sources identified include excreta from domestic animals, wild animals, synthetic nitrogen fertilizers, oceans, biomass burning, emissions from soils under natural vegetation, human population, crops and decomposition of crop residues, fossil fuel combustion, and industry. All sources are presented on a  $1^\circ \times 1^\circ$  grid suitable for input to global atmospheric transport/chemistry models (Plate 2).

The most important source identified is excretion from domesticated animals (dairy cattle, nondairy cattle, buffalo, camels, horses, sheep, goats, pigs and poultry), contributing  $21.6 \text{ Tg N yr}^{-1}$  (40% of the total; Table 11). This confirms earlier estimates for this source presented, for example by Schlesinger and Hartley [1992]. The second and third most important sources are nitrogen fertilizer use and oceans, contributing 17% and 15%, respectively (Table 11). The  $\text{NH}_3$  loss from fertilizers occurs primarily in developing countries and represents more than 10% of the N in the world's fertilizer usage. Biomass burning (11%), crops and decomposing crop residues (7%), human population (5%), and soils under natural vegetation (4%) form another important group (Table 11). Minor sources include industrial processes, stationary and mobile fossil fuel combustion, and wild animals.

The global  $\text{NH}_3$  emissions are dominated by human activities related to food production. Domestic animals, fertilizer use, deforestation, savanna burning, and field crop residue burning together constitute a source of  $38 \text{ Tg N yr}^{-1}$  or 70% of the global source.

### 3.1. Geographical Distributions

The areas with highest emission rates per square meter are located in Europe, the Indian subcontinent, and eastern China (Plate 2). The emission densities closely follow the patterns of the animal populations, with dense populations of cattle in India, Pakistan, and Bangladesh and dense populations of pigs and chickens in China. As highest emissions from synthetic fertilizers occur in the Indian subcontinent and eastern China, because of the dominant use of urea and ammonium bicarbonate, the pattern resulting from animal populations is amplified further in these regions. The third most important terrestrial source, biomass burning, occurs mainly in the tropical countries. Emissions from crops and decomposing crop residues occur in arable lands, and emissions from human populations are highest in urban areas (Plate 2). All other sources are from large areas of natural ecosystems, natural grasslands, and some local point sources from industrial plants.

Most of the  $\text{NH}_3$  from oceans (both the total emission and the emission rate per square meter as shown in Plate 2) stems from equatorial regions of the Pacific Ocean, Atlantic Ocean, and Indian Ocean and low emissions toward the polar regions. This contrasts with the patterns observed for  $\text{N}_2\text{O}$ , which show highest emission densities only in upwelling zones such as the Antarctic Ocean [Bouwman et al., 1995],  $\text{NH}_3$  emissions be-

**Table 11.** Overall Emission Terms and Uncertainty Ranges

Source	Emission, Tg N yr <sup>-1</sup>	Uncertainty Range, Tg N yr <sup>-1</sup>
Excreta of domesticated animals		
dairy cattle	4.3	
nondairy cattle	8.6	
buffalo	1.2	
camels	0.2	
horses	0.5	
sheep	1.0	
goats	0.5	
pigs	3.4	
poultry	1.9	
subtotal	21.6	10-30
Excreta of wild animals	0.1	0-1
Synthetic fertilizer use	9.0	4.5-13.5
Biomass burning		
deforestation	1.4	
savanna burning	1.8	
field burning of crop residues	0.5	
biofuels	2.2	
subtotal	5.9	3.0-7.7
Soils under natural vegetation	2.4	0-10
Oceans	8.2	3-16
Fossil fuel combustion	0.1	0.0-0.2
Industrial processes	0.2	0.1-0.3
Human excreta and pets	2.6	1.3-3.9
Crops and decomposition of crops	3.6	1.4-5.0
Total	54	40-70

ing correlated strongly with sea-surface temperatures and biological activity in the surface mixed layer of the ocean.

About half of the global terrestrial NH<sub>3</sub> source is located in the Asian continent (including the former USSR; Table 12). Latin America and Africa each contribute 16 and 14%, respectively, of the global source, while Europe, North America, and Oceania add 9, 8, and 2%, respectively, of the global source. As most NH<sub>3</sub> comes from food production, it is clear that these numbers are closely related to human population densities. The latitudinal distribution of terrestrial and oceanic emissions (Table 12) indicates that 75% of the global emission of NH<sub>3</sub> comes from the northern hemisphere, with 37% from temperate and 38% from tropical zones. The estimated emission for the southern hemisphere is about 25%, with about 18% from the tropical zone and 7% from the southern temperate zone. This roughly reflects the distribution of human population, which is most dense in the northern hemisphere (90%) [Lerner *et al.*, 1988].

### 3.2. Uncertainties

The emission fields represent estimates of the “potential” flux of ammonia to the atmosphere. The potential flux does not account for the difference between the ambient NH<sub>3</sub> concentrations and the compensation concentrations regulating the exchange of NH<sub>3</sub> between the surface and the atmosphere. The rationale for presenting potential emissions is that all atmospheric models using emission fields have their own specific representation of atmospheric transport and chemistry. Hence producing a set of net emission fields is of less value to any atmospheric model than potential emissions and, indeed,

will vary between models. However, we recognize that the estimates and data used for the different sources vary in nature. Some estimates are derived from measurements (animals, nitrogen fertilizer use, biomass burning, industrial processes, and fossil fuel combustion), others are derived from modeling (oceans), and a third category is derived from constraints on the emission (human population, crops, natural soils, and wild animals). Even in the case of measurement-based estimates the measurement technique used may be important. In micrometeorological flux determinations both the upward and downward fluxes are determined, and the net flux is presented. When measurements are made in enclosures, the net flux is determined, but the technique suffers from various kinds of disturbances. The most reliable gross flux estimates are perhaps from animal housings, for example, where ventilation air is sampled. However, many housing systems are naturally ventilated which makes flux determination very difficult.

It is difficult to estimate the uncertainty in the global NH<sub>3</sub> emission. We have attempted to produce uncertainty ranges (Table 11) based on expert, and therefore subjective, judgments. These uncertainties are not the grid-by-grid uncertainties, which may be far larger than the uncertainty of the global estimate presented in Table 11. The uncertainty for the different sources depends on the uncertainty in the basic data and in the emission factors used. The basic data used for the different sources vary in quality.

Animal populations are quite well known, although there is some uncertainty caused by the season in which censuses are made. The overall uncertainty caused by the animal population is probably <10%. The uncertainty in the emission factor for

**Table 12.** Total NH<sub>3</sub> Emission for Continents and Latitudinal Zones

	Emission, Tg NH <sub>3</sub> -N yr <sup>-1</sup>	Percent of Total
Terrestrial emissions		
Total	45	100
Europe	4.1	9
Africa	6.3	14
North America	3.6	8
Latin America	7.2	16
Asia including former USSR	23.4	51
Oceania	1.0	2
Terrestrial and oceanic emissions		
Total	54	100
30°N-90°N	19.8	37
0°-30°N	20.4	38
0°-30°S	9.6	18
30°S-90°S	3.7	7

animals stems from the assumptions on N in the feed and waste management. For developing countries, in particular, there are no data on waste management practices. Therefore the uncertainty is probably great in tropical countries and smallest in western Europe. The overall uncertainty for this source is estimated to be  $\pm 50\%$ .

The nitrogen fertilizers statistics used in this study are reliable in comparison to other statistics of economic activity. Minor uncertainties in fertilizer use stem mainly from the grouping of different nitrogen fertilizer types into one category. However, the timing and mode of fertilizer application (single or split application; injection, superficial or dissolved in irrigation water, etc.) and the fertilizer dose determine the gross emission. Unfortunately, there are no statistics on fertilizer management. Furthermore, the published measurements do not cover all management conditions. The overall uncertainty in the gross NH<sub>3</sub> loss from nitrogen fertilizers amounts to  $\pm 50\%$ . The greatest uncertainty stems from lack of measurements of NH<sub>3</sub> emissions in tropical cropping systems.

Estimates of oceanic emissions are based on modeled NH<sub>4</sub><sup>+</sup> concentrations in the surface water and assumptions on the piston velocity. The overall uncertainty may amount to a factor 2–3, although the relative source distribution may be somewhat more reliable than the absolute emissions.

For biomass burning the basic data are the areas and volumes of burning, which are rather uncertain [e.g., Seiler and Crutzen, 1980; Hao and Liu, 1994]. The uncertainty in the volumes for deforestation, savanna burning, and field crop residue burning amounts to perhaps  $\pm 50\%$ . Biofuel use includes an uncertainty of the order of  $\pm 25\%$ . However, the uncertainty in the estimate for the emission factor for biofuel burning is greater than that for the other biomass burning categories. The overall uncertainty in the estimate for biomass burning is  $-50\%$  to  $+30\%$ .

The estimated uncertainty in the other sources is  $\pm 50\%$ , except for the wild animals for which a rather high upper limit is maintained as a result of the uncertainty in African animal populations (Table 11).

It is difficult to produce an independent estimate of the upper limit for the global source. An upper limit to the upward flux could be given by the deposition. Unfortunately, the estimates for deposition are highly uncertain. Wet deposition is relatively easy to measure, but measurements are of variable quality. Measurements of dry deposition are very sparse. A test made with the Model of the Global Universal Tracer Transport in the Atmosphere (MOGUNTIA) model showed that the emission fields in Europe and North America can be validated against deposition measurements [Dentener and Crutzen, 1994]. However, the estimates for other parts of the world (Asia, Africa, South America, and Oceania) are highly speculative because of the sparsity of deposition measurements.

We estimate that the uncertainty in the global total emission is  $\pm 25\%$ . It must be stressed, however, that the uncertainty in regional and grid-by-grid estimates may be much higher than that of the global emission.

### 3.3. The Use of Ammonia Emission Fields in Global Chemistry Transport Models

The emissions are presented on a  $1^\circ \times 1^\circ$  grid. At the equator the grid boxes are largest ( $\sim 12,320 \text{ km}^2$ ), and at  $45^\circ \text{ N}$  or  $\text{S}$  the area is 70% of that at the equator, while at  $70^\circ \text{ N}$  or  $\text{S}$  the area is reduced to 34%. Hence most countries are represented by a small number of grid boxes. Each grid box includes a great variation of environmental and management conditions. The paucity of data makes it necessary to generalize. For each grid box the conditions have been aggregated to obtain one single emission rate, usually by treating the landscape as a composite of average farms with average management and average weather conditions or by treating populations as a group of identical average members. Such generalizations may lead to errors due to averaging procedures. For example, the result from a population of average farms may differ geographically and temporally from the sum of all individual farms [Bouwman and Asman, 1997]. Aggregation may also cause unknown errors in the resulting temporal distribution.

In addition to the problem of spatial scales, there is a difficulty to produce temporal distributions of emissions. Except for the oceanic emissions, which are available as monthly estimates, the estimates presented are annual emissions. Atmospheric transport models require temporal distributions of the emissions, for example, on a monthly, decadal, or daily scale, depending on the type of model used. On the basis of the available data it is very difficult to produce time distributions. Therefore the distribution in time must be derived indirectly from expert knowledge or inferred from knowledge about the emissions of other gases. Seasonal distributions for emissions from animal excreta could be derived from data on waste management, for biomass burning derived from climatic data as proposed by Hao *et al.* [1990], for fertilizers and emissions derived from decomposition of crop residues from cropping calendars indicating the period of planting and the number of crops per year [FAO, 1978] (for rice [Matthews *et al.*, 1994]).

In dealing with net fluxes the bidirectionality of fluxes is ignored. In most models, either small- or large-scale emission and deposition are treated as simultaneous processes. Field measurements have shown that this is not the case: the so-called “compensation point” (which is strictly for stomata) or

equilibrium concentration controls whether deposition or emission occurs. In the model of *Dentener and Crutzen* [1994] this approach was used. However, the field measurements of the compensation point show that it varies with many environmental parameters, and any value, or values, used in a model have rather a large uncertainty. In their model, part of the emission is dry deposited within the same grid box as emission: the so-called "alpha factor". This factor is usually incorporated into atmospheric transport models because of low resolution in the vertical domain and may differ between models with different resolutions and transport descriptions.

*Dentener and Crutzen* [1994] have compared their global emissions to a host of observations using the coarse grid MOGUNTIA model. Their global emissions were somewhat smaller than estimated in this study. They generally found an agreement within a factor of 2 of model results with observations. However, because of the coarse grid resolution and the treatment of the averaged meteorology in the model, the differences may be derived from inaccuracies in model transport (and chemistry) as well as the emission fluxes applied. To our knowledge this model is still the only global model which has been applied to model the global  $\text{NH}_x$  cycle. Two of the authors (F. J. Dentener and D. S. Lee) are presently involved in incorporating  $\text{NH}_3$  emissions and chemistry in finer resolution and meteorologically more realistic models which we hope will provide a better constraint on the  $\text{NH}_3$  emissions.

### 3.4. Global N Mobilization

The global total emission of  $\text{NH}_3$  of  $54 \text{ Tg N yr}^{-1}$  exceeds  $\text{NO}_x$  emissions estimated recently to be  $44 \text{ Tg N yr}^{-1}$  [Lee *et al.*, 1997a]. More than two thirds of the present-day atmospheric  $\text{NH}_3$  input into the atmosphere stems from human activities related to food production, causing major changes in the composition and processes in the atmosphere. The population in developing countries is expected to grow from the current 4 billion to 6.8 billion in 2025 [United Nations, 1994]. This will entail a necessary increase in food production. Parallel to this, consumption patterns will shift along with economic development toward more consumption of animal products per capita. Increasing animal production will cause growing animal feed production and fertilizer use, and this will cause an important intensification of the cycling of reduced nitrogen [Bouwman, 1997]. The likely magnitude of effects of increasing  $\text{NH}_3$  inputs and its potential enhancement of aerosol formation is not yet known.

For some of the sources, mitigation could be implemented. For example, selection of other fertilizer types would reduce the emissions in Asia, the major global source region for  $\text{NH}_3$  from fertilizers in 1990. Reductions of emissions from animal waste could be achieved by reducing the amount of nitrogen involved in livestock production and by limiting volatilization losses (e.g., by increasing the efficiency of N use, reduction of  $\text{NH}_3$  volatilization during spreading of animal manure, etc.).

## 4. Conclusions

The major conclusions drawn on the basis of the compilation of the global  $\text{NH}_3$  emission inventory described in this paper relate to the source strengths and their uncertainties and recommended methods to reduce uncertainties and validation.

### 4.1. Global Emissions

The global emission of  $\text{NH}_3$  estimated in this study is approximately  $54 \text{ Tg NH}_3\text{-N yr}^{-1}$  for the base year 1990. The sources that contribute to this source include excreta from domestic animals (40%,  $10\text{--}30 \text{ Tg yr}^{-1}$ ), synthetic fertilizers (17%,  $4.5\text{--}13.5 \text{ Tg yr}^{-1}$ ), biomass burning (11%,  $3.0\text{--}7.7 \text{ Tg yr}^{-1}$ ), natural ecosystems (4%,  $0\text{--}10 \text{ Tg yr}^{-1}$ ), oceans (15%,  $3\text{--}16 \text{ Tg yr}^{-1}$ ), human excreta and pets (5%,  $1.3\text{--}3.9 \text{ Tg yr}^{-1}$ ), and crops and decomposition of crop residues (7%,  $1.4\text{--}5.0 \text{ Tg yr}^{-1}$ ). Minor sources include stationary and mobile fossil fuel combustion, a number of industrial processes, and wild animals. About 75% of the global  $\text{NH}_3$  emission comes from the northern hemisphere, and 25% comes from the southern hemisphere. About 56% of the global  $\text{NH}_3$  emission comes from the tropics, and 37% comes from the developed countries in the northern hemisphere. Food production is responsible for 70% of the global  $\text{NH}_3$  emission.

The global emission from all sources calculated in this study is somewhat higher than that of *Dentener and Crutzen* [1994] but is much lower than that of *Schlesinger and Hartley* [1992]. The major difference between this study and that of *Schlesinger and Hartley* [1992] is caused by our lower emissions estimates from animal waste and undisturbed ecosystems (Table 13). Our estimate for emissions from animal waste is similar to that of *Dentener and Crutzen* [1994]. The major differences between our inventory and that of *Dentener and Crutzen* arise from higher estimates for  $\text{NH}_3$  from fertilizers (2.6 Tg higher) and biomass burning (a factor of 2 higher) (Table 13).

**Table 13.** Global Estimates of Emissions of Atmospheric Ammonia ( $\text{NH}_3$ ) for the Various Sources in Million Tons  $\text{NH}_3\text{-N yr}^{-1}$

	1 <sup>a</sup>	2	3
Cattle including buffaloes	19.9	14.2	14.0
Pigs	2.0	2.8	3.4
Horses/mules/asses	1.8	1.2	0.5
Sheep/goats	4.1	2.5	1.5
Poultry	2.4	1.3	1.9
Wild animals <sup>b</sup>	—	2.5	0.1
Total animals	32.3	24.5	21.7
Synthetic fertilizers	8.5	6.4	9.0
Undisturbed ecosystems	10	5.1	2.4
Croplands	—	—	3.6
Biomass burning	—	—	—
including biofuels	5	2.0	5.7
Human excrement	4	—	2.6
Sea surface	13	7.0	8.2
Fossil fuel combustion	2.2 <sup>c</sup>	—	0.1
Industry	—	—	0.2
Total emission	75	45.0	53.6

Dash indicates data not available.

References for columns are as follows: 1, *Schlesinger and Hartley* [1992]; 2, *Dentener and Crutzen* [1994]; 3, this study.

<sup>a</sup> Values for a number of sources are reported with precision of  $10^{12} \text{ g N}$ .

<sup>b</sup> *Schlesinger and Hartley* [1992] included the estimate for wild animals in undisturbed ecosystems.

<sup>c</sup> Mainly coal combustion.

It is difficult to produce estimates of the uncertainties for the various individual sources as they are all from a different nature. For some sources the uncertainties are due to the sparsity or unreliability of statistical data, while uncertainties in other estimates are caused by lack of measurements. The data used are globally available data, except for data on deforestation in Brazil and agricultural residue burning in China. In one case (emissions from natural ecosystems) the estimate is purely based on hypotheses on soil N cycling, exchange with atmosphere below the canopy, and canopy reabsorption. The estimated uncertainty in the global total source is  $\pm 25\%$ .

## 4.2. Methods to Reduce Uncertainties

With the currently available data it is difficult to improve the global estimates. In general, the uncertainty in the estimates could be reduced by using national statistics, data, regional studies, and measurements. Unfortunately, for most of the sources national data are scarce. On the basis of the inventory presented in this paper, we propose a number of methods that could lead to a reduction of the large uncertainties in the regional and global estimates.

**4.2.1. Animal waste.** For ammonia from animal waste we need to know animal populations, N intake, meat and milk production, excretion, and waste management and associated emissions in relation to climatic conditions. Most measurement data are from the Netherlands, Denmark, the United Kingdom and Germany. Outside Europe and North America both measurement and statistical data pose difficulties. For many countries only the total number of animals within a category is available, and the data within a category on age and weight class are scarce. Data on some animal categories, such as buffaloes, horses, donkeys, camels, and data on housing and the type and form of manure are simply unavailable. Estimates for regions within countries may be available, but sometimes they do not correspond to the official statistics, or they are outdated. Geographic data on the application rate of manure, soil conditions, and the variability in timing of application and weather conditions during application are not available. Information on the N intake, distributed over consumption of roughages and concentrates, and the management of the waste (the excretion during the grazing period, storage of animal waste, type of storage, and practices during the spreading of wastes) is very sparse and scattered, in particular in developing countries. National inventories in countries outside Europe and the United States of America coupled with some experiment on ammonia losses in different types of stables and measurements after spreading of animal waste under tropical conditions would form a major improvement.

**4.2.2. Fertilizers.** For fertilizer use, good statistics by country on total use by category are available, but the application rates, methods, and timing by crop are not reliable. Field measurements in various tropical agroecosystems are required to reduce the uncertainties in the emission factors in particular for systems with split fertilizer applications.

**4.2.3. Biomass burning.** Data on biomass burning are not based on statistics, and the data found in literature are contradictory [see, e.g., *FAO*, 1995; *Hao and Liu*, 1994; *Andreae*, 1991]. Areal extent, volumes of burning coupled with observations of the type of burning, and associated emissions are currently being studied in various projects. An inventory of

biomass burning is planned to be produced within the Global Emission Inventories Activity (GEIA).

**4.2.4. Natural ecosystems.** The estimate of the emissions from natural ecosystems includes various uncertainties. Improvements can be achieved by investigations on the relation between N mineralization rates and soil  $\text{NH}_4^+$ , the exchange process between the soil surface and the air, and canopy absorption rates for different vegetation types under different tropical and temperate climates.

**4.2.5. Oceans.** The estimate for oceanic emissions is a model calculation based on scarce concentration measurements in the world oceans. Validation of model predictions is very difficult, since the fluxes cannot be measured directly and measurements of ammonia in seawater and atmospheric ammonia are sparse. Yet the model results presented in this paper suggest the possibility of a major source. The only way to reduce the uncertainty in the oceanic ammonia flux is to increase the number of concurrent seawater and atmospheric ammonia observations in a variety of representative ocean masses, including open ocean, coastal waters and continental shelves, and productive versus oligotrophic regions. The possibility of scaling of  $\text{NH}_4^+$  to a parameter such as ocean productivity should be given research attention.

## 4.3. Validation

An indirect check on the emissions is the comparison of atmospheric observations with model results. Measurements are needed over at least a full year to include possible seasonal cycles. Three types of measurements are needed: (1) Measurement of  $\text{NH}_3(\text{g})$  concentrations is needed. Owing to the relatively short lifetime of ammonia, only those measurements that are representative for a relatively large scale (100 x 100 km) are really useful. Other measurements should at least include some statement on the representativity and an estimate of the average value at a larger scales. (2) Measurement of  $\text{NH}_4^+$  aerosols is needed. As there are only 1 or 2 steps between atmospheric  $\text{NH}_3$  and formation of  $\text{NH}_4^+$  aerosols, measurement of  $\text{NH}_4^+$  aerosols is more useful for validation of emission estimates than  $\text{NH}_3(\text{g})$ . Such measurements are also representative for larger areas than  $\text{NH}_3$  concentration measurements, because the lifetime of these aerosols is longer than that of  $\text{NH}_3(\text{g})$ . (3) Measurement of  $\text{NH}_4^+$  wet deposition is needed. It is difficult to interpret wet deposition data for  $\text{NH}_4^+$  as the modeled deposition also depends on an accurate description of wet scavenging (both in cloud and below cloud). The stations in the current Global Atmospheric Watch (GAW) (and also the Co-operative programme for monitoring and evaluation of the long range transmission of air pollutants in Europe, EMEP, and American) networks provide a first useful step for validation, but the quality is not always assured.

Wet and dry deposition are simulated in the various atmospheric transport models in different ways. Therefore the emission fields in this inventory represent the potential or gross flux of  $\text{NH}_3$  to the atmosphere. The potential flux does not account for the difference between the ambient  $\text{NH}_3$  concentrations and the compensation concentrations regulating the exchange of  $\text{NH}_3$  between the surface and the atmosphere. Hence local deposition is not accounted for.

It is hoped that the emission fields reported in this study provide input for modeling the role of  $\text{NH}_3$  in global cycles of

S species and its role in radiative forcing of climate by aerosols. The results also provide impetus for reducing the uncertainties associated with the emission estimates. We hope that other modeling groups will use this work and encourage them to give feedback on the emission inventory and potential problems. The emission files by source category presented in this paper can be obtained from the National Institute of Public Health by anonymous ftp (node name: info.rivm.nl, sub-directory pub/lae/EDGARV20/DATA/details/NH3) and from the National Center for Atmospheric Research (<http://blue-skies.sprl.umich.edu/geia/index.html>) under the auspices of the Global Emissions Inventory Activity (GEIA), a project of the International Global Atmospheric Chemistry Program [Graedel et al., 1993, 1995].

Regarding the oceanic fluxes, the modeled monthly fluxes of  $\text{NH}_3$  are available; from the piston velocities (for ammonia about equal to aerodynamic resistance) specific to the model used, gas-phase concentrations in equilibrium with oceanic concentrations of  $\text{NH}_4^+$  can be calculated and used to compute fluxes. (In case of zero atmospheric  $\text{NH}_3$  concentrations this yields a flux equal to the ones presented in this paper, while for atmospheric  $\text{NH}_3$  concentrations  $> 0$ , lower fluxes will be obtained.) In addition to the  $\text{NH}_3$  fluxes the gas phase equilibrium  $\text{NH}_3$  concentrations will be available. This is more straightforward than providing the oceanic  $\text{NH}_4^+$  concentrations, because no additional data on seawater temperature, pH, and activity coefficients are needed.

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