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Evaluation of ammonia fluxes into the free atmosphere over Western Germany

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ABSTRACT

The upward flux densities of gaseous ammonia and particulate ammonium through a layer 400 m above ground were determined by aircraft measurements over a rural area in Western Germany. The flux densities were calculated according to the gradient method. The fluxes amounted to 118 ± 49 $\mu g/m^2h$ NH_3–N and 93 ± 47 $\mu g/m^2h$ NH_4^+–N in summer and to 7 ± 14 $\mu g/m^2h$ NH_3–N and 23 ± 34 $\mu g/m^2h$ NH_4^+–N in winter.

When integrated over a year, the sum of these ammonia fluxes can be maintained by NH_3 volatilization from domestic animal excrements. The NH_3 liberation from mineral fertilizer and natural soils contributes only a small amount to the overall production rate. Most of the NH_3 and NH_4^+ diffusing upwards into the middle troposphere will eventually be incorporated in rain. For the conversion of gaseous NH_3 to particulate NH_4^+ in the first 400 m of the atmosphere a lower limit for a pseudo first-order reaction rate constant of $1\text{–}2 \times 10^{-5}\ s^{-1}$ was deduced from flux density considerations.

1. Introduction

Ammonia in the atmosphere is found as gaseous NH_3 and as NH_4^+ ions in suspended matter. It is part of the atmospheric cycle of nitrogen compounds and interacts with other cycles of atmospheric trace substances. Apart from this geochemical aspect, atmospheric ammonia can be of interest for plant and soil scientists studying nutrient budgets in the biosphere.

Up to now four different production mechanisms for atmospheric ammonia have been postulated: decay of animal excrements (e.g. Hall, 1915; Doak, 1952; Healy et al., 1970; Buchner and Sturm, 1973), transformation of organic material in natural soils (e.g. Erikson, 1952; Allison, 1955; Robinson and Robbins, 1970; Hooker et al., 1973; Dawson, 1977; Georgii and Lenhard, 1978), volatilization from nitrogen fertilizer (e.g. Jewitt, 1942; Nommik, 1966; Förster and Lippold, 1975; Mikkelsen et al., 1978), and combustion of coal (e.g. Erikson, 1952; Georgii, 1963; Söderlund and Svenson, 1976). A detailed discussion of these processes is given by Söderlund and Svenson (1976) and Böttger et al. (1978).

The total source strength for atmospheric ammonia has been estimated by evaluating the chemical composition of precipitation (Robinson and Robbins, 1970; Böttger et al., 1977; Dawson, 1977; Georgii and Lenhard, 1978). These wet-deposition rates represent minimum total source strengths because other possible removal mechanisms of atmospheric ammonia like absorption by plants (Hutchinson et al., 1972; Denmead et al., 1976) and oxidation by OH radicals (McConnell, 1973) are not included. Production and removal rates by different mechanisms were compared on a global scale by Böttger et al. (1978).

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A difficulty in evaluating atmospheric source strengths of ammonia for a larger area is the transfer of laboratory results to natural outdoor phenomena and the generalization of results gained under special restricted conditions. To overcome this problem an atmospheric investigation was started to characterize more representative conditions for budget considerations. The aim of this investigation was to determine the direct upward flux into the free atmosphere of gaseous NH_3 and particulate NH_4^+ for a larger area from aircraft measurements. Fluxes were calculated according to the gradient method. A gradient approach had already been used to determine the amount of ammonia diffusing into the first few metres of the atmosphere from pastures (Denmead et al., 1976) and plant fields (Lemon, 1975). Their results characterize the limited source areas investigated under special conditions. Using aircraft measurements, as we have done, all the different NH_3 emissions are integrated over a larger area and at the same time over varying conditions. This way it is possible to directly determine the overall inflow rate into the atmospheric reservoir over a region of about 10^4 km^2 including numerous individual release processes. The results of our measurements can be compared with theoretical estimations of ammonia emissions from the ground by natural and antropogenic processes. In addition to source strength data derived from budget considerations these values will help to improve our knowledge of the atmospheric ammonia cycle.

2. Aircraft measurements

Flights were performed about 100 km northeast of Frankfurt in a rural area with a homogeneous pattern of villages and towns. Areas covered with pastures, cropland and forests are found in roughly equal proportions.

Concentrations were determined by sampling NH_3 in 0.1 N H_2SO_4 solutions in specially designed teflon scrubbers (Süssenguth, 1976). Particulate ammonium was separated from the air stream with PVC membrane filters (Sartorius No. 12806). Laboratory tests were performed to evaluate the possible absorption or desorption of gaseous NH_3 by the aerosol-laden filters (Süssenguth, 1976): Ten filters were loaded with atmospheric aerosol in a parallel sampling run of 1 m^3 outdoor air near Frankfurt. Six of the filters were analysed, yielding an average of 4.8 ± 0.1 μg NH_4^+ per filter. In an additional run 500 l of aerosol-free air containing 2.6 μg/m^3 s.t.p. NH_3 were sucked through each of the remaining four filters at 100% R.H. The average NH_4^+ content of these filters was found to be 4.5 ± 0.4 μg. The resulting very small difference of 0.3 μg (about 6%) between these two filter sets can be attributed to the statistical variance of the filter loads. This conclusion can be substantiated by comparing the variance of all 10 filter loads with the reproducibility of our NH_4^+ sampling and analysis method. This reproducibility was separately determined by parallel filter measurements to be 6.9%. The variance of all 10 filters (treated with additional gaseous ammonia as well as untreated filters) was 7%, indicating no deviation from the variance typically found with filters sampled in parallel runs. Therefore negligible interference of the aerosol filters with the determination of gaseous ammonia was found.

The detection limit for both NH_3 and NH_4^+ was 0.5 μg/m^3 s.t.p. for a 2-hour sampling period at a flow rate of 600 l/h. The collection efficiency for gaseous NH_3 absorption was determined to be 85% for airborne sampling conditions. Our concentration values were corrected accordingly. The chemical analysis, leading to vertical concentration distributions, has already been published (Georgii and Lenhard, 1978).

In Table 1 the average concentration values for gaseous ammonia and particulate ammonium in

Table 1. *Concentration of gaseous and particulate ammonia at two altitudes in a rural area of Western German*

	Summer (6 flights)		Winter (5 flights)	
	NH_3 (μg m^{-3} s.t.p.)	NH_4^+ (μg m^{-3} s.t.p.)	NH_3 (μg m^{-3} s.t.p.)	NH_4^+ (μg m^{-3} s.t.p.)
100 m a.g.	4.8 ± 0.8	4.9 ± 0.8	1.9 ± 0.6	2.6 ± 0.9
700 m a.g.	2.5 ± 0.4	2.9 ± 0.4	1.5 ± 0.4	2.0 ± 0.5

summer and winter are given for the two altitudes of 100 m and 700 m above ground.

3. Determination of vertical flux densities

Under steady-state conditions to the vertical flux of a trace substance into the first few hundred metres of the atmosphere is proportional to the gradient of the mixing ratio if it can be assumed that sources and sinks are homogeneously distributed at the earth's surface and higher than 1 km in the troposphere, respectively. We have calculated the NH_3 and NH_4^+ gradients at an altitude of 400 m above ground from concentration values at 100 m and 700 m for the two ammonia components. In this altitude range concentration gradients can be resolved. The measurements were grouped together for winter and summer seasons so that individual results for the eddy diffusion coefficient did not vary much. Thus, it can be assumed that the flux calculated from the product of the average gradient of the mixing ratio and the average eddy diffusion coefficient instead of the average product approximates the true flux.

The vertical coefficients of turbulent diffusion for the meteorological conditions encountered during aircraft measurements were derived from radiosonde data of the German Weather Service stations at Essen and Stuttgart. Although these soundings were not performed in the immediate vicinity of our measurement flights, we assume that, on the average, they give representative descriptions of turbulent diffusion parameters. The value of the turbulent diffusion coefficient was calculated from temperature and wind velocity profiles according to Wu (1965). The quantities needed for these calculations are potential temperature profiles and wind shears. They were taken from radiosonde data in vertical steps of 10 m. To give the reader an impression of the broad meteorological conditions, mean values and the variance of the individual results for these quantities are given in Table 2. The method of calculating the vertical eddy diffusivity is based on several assumptions so that individual results will probably be uncertain by a factor of about two. However, the variance of the seasonal average given in Table 3 is only about 17%. Also in Table 3 the thus derived fluxes are given. The variance of the fluxes in winter do not exclude downward flows. This is the result of a greater variance of the concentration gradients observed during winter.

4. Discussion

The results given in Table 3 show that the flux of NH_4^+ equals or even exceeds that of NH_3 in both seasons. If we assume all ammonia to be released into the atmosphere as a gas, it can be concluded that the transformation of gaseous NH_3 to particulate NH_4^+ proceeds mainly within the lowest layers of the atmosphere. Different size distributions of

Table 2. *Zonal (u') and meridional (v') component of the wind shear and the potential temperature gradient (θ') at 400 m above ground. The K_z values represent average eddy diffusivities for altitudes of 300, 400 and 500 m*

	u' (s^{-1})	v' (s^{-1})	θ' ($K \cdot cm^{-1}$)	K_z ($m^2 s^{-1}$)
Summer	$3.6 \pm 1.9 \times 10^{-3}$	$2.0 \pm 1.7 \times 10^{-3}$	$4.6 \pm 9.3 \times 10^{-5}$	10.2 ± 3.9
Winter	$2.7 \pm 1.3 \times 10^{-3}$	$3.8 \pm 3.6 \times 10^{-3}$	$4.6 \pm 7.8 \times 10^{-5}$	3.9 ± 1.6

Table 3. *Results of computations for turbulent diffusivities K_z and ammonia–nitrogen flux densities ϕ at an altitude of 400 m above ground over Germany*

	$\phi_{NH_4^+-N}$ ($\mu g\ m^{-2}\ h^{-1}$)	ϕ_{NH_3-N} ($\mu g\ m^{-2}\ h^{-1}$)	$\phi_{NH_3-N+NH_4^+-N}$ ($\mu g\ m^{-2}\ h^{-1}$)	K_z ($m^2 s^{-1}$)
Summer 8.6–5.8	93 ± 47	118 ± 49	211 ± 68	10.2 ± 1.6
Winter 27.1–11.3	23 ± 34	7 ± 14	30 ± 37	3.9 ± 0.7
$\frac{\text{Summer + Winter}}{2}$	58 ± 29	62 ± 26	120 ± 39	

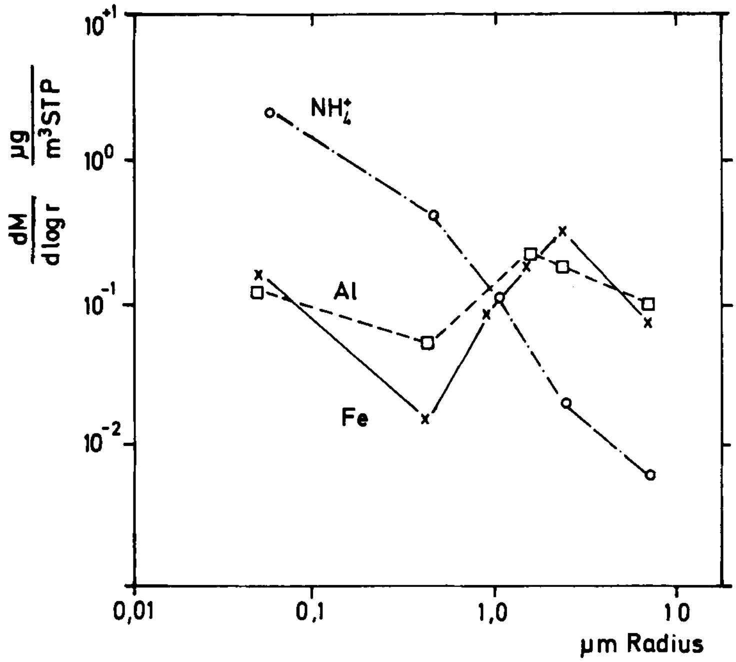

Fig. 1. Mass-aerosol size relationship in a rural area in the F.R. Germany for ammonium, iron and aluminium (after Koch, 1979).

ammonium on the one side and aluminium and iron on the other (Fig. 1) suggest that particulate ammonium in the atmosphere is not a soil-derived component as it is aluminium and iron in the size range larger than 0.8 μm radius but rather a product of reactions of gaseous ammonia within the atmosphere. A pseudo first-order rate constant for the transformation reaction can be roughly estimated by the following: The combined flux of ammonia and ammonium in 400 m $\phi_{NH_4^+ + NH_3}$ from Table 4 is taken as the total initial source strength of NH_3 near the ground. ϕ_{NH_3} is the flux leaving the first 400 m layer of the atmosphere. $\phi_{NH_4^+}$ represents a measure of the transformation rate of NH_3 to NH_4^+ within these 400 m if no additional sources and sinks are assumed. The values from Table 3 yield rate constants of 1.2×10^{-5} s^{-1} in winter and 2.1×10^{-5} s^{-1} in summer. These are average rate constants for a layer of 400 m and give only a lower limit, since the transformation could already be completed within, for instance, the first 100 m. The nearly parallel decrease of the concentrations of NH_3 and NH_4^+ in the lower layer (Georgii and Lenhard, 1978) indicates that ammonium ions in the atmosphere are formed mainly below 100 m altitude, also suggesting a faster transformation rate than averaged over the total altitude range of 400 m.

Table 4. *NH_3–N volatilization rate for assumed N percentages lost to the atmosphere from organic fertilizer in the Federal Republic of Germany (F.R.G.) (Buchner and Sturm, 1973) and from cattle excrements in two German states neighbouring one another in the measuring area. Unit μg NH_3–N m^{-2} h^{-1}*

% N loss	10%	30%	45%
F.R. Germany (organic fertilizer)	60	180	270
Hessen (cattle excrements)	38	115	173
Bayern (cattle excrements)	60	180	270

Table 3 shows a marked difference in the flux densities of total ammonia (NH_3 + NH_4^+) between summer and winter, indicating different release conditions in the course of the year. Higher NH_3 production rates in summer are also reflected by the annual concentration trend of the sum of particulate and gaseous ammonia in the city of Frankfurt and on top of a mountain (Kl. Feldberg), 700 m above Frankfurt and at 20 km distance (Fig. 2).

The upward fluxes represent an average NH_3 leakage from the vegetation layer of the atmosphere into the lower troposphere. Absorption rates of gaseous ammonia by plants, filtration of particulate ammonium by vegetation, dry deposition to the ground and gas phase conversion have to be added to the upward flux through the 400 m layer to get total initial NH_3 production rates at the ground. As mentioned above, flights were performed in a more rural area. It should therefore be reasonable to consider decaying animal excrements, volatilization of ammonia from mineral fertilizers and the soil as predominent ammonia source mechanisms in this region.

In Table 4, release rates of NH_3 by decaying excrements are estimated for different assumed percentages of nitrogen lost as NH_3–N to the atmosphere. About 110,000 tons per year (16% of excrement nitrogen) are lost as NH_3 before application to the farmland in the Federal Republic of Germany (Buchner and Sturm, 1973). Additional volatilization of NH_3 after application will increase the ammonia loss. For comparison we estimated ammonia release rates from cattle excrements for the two German states Bavaria and Hessen, neighbouring one another in our flight area. Cows produce more than 80% of excrement nitrogen from domestic animals in the flight area. Their number were taken from data of the statistical council of Germany (Statistisches Jahrbuch, 1977) and the average amount of excrement nitrogen produced by each animal (235 g per day) from Mangold (1929) and Nehring (1963). An extensive survey of laboratory and field experiments on the volatilization of organic fertilizer nitrogen by Böttger et al. (1978) revealed that the percentage of nitrogen lost to the atmosphere as ammonia most frequently covered a range from 10 to 45% depending on farmyard practice and soil and weather conditions. The different columns of Table 4 show ammonia release rates as a function of the percentage of total nitrogen loss assumed by different authors (Healy, 1970, 10%; Böttger et al., 1978, 30%; Hall, 1915, 25–45%; Buchner and Sturm, 1973, 45%).

Comparing the annual mean flux density found in aircraft measurements (Table 3) to these proposed release rates, we find that the upward flux corresponds roughly to a 30% release of nitrogen as ammonia from decaying excrements. This value

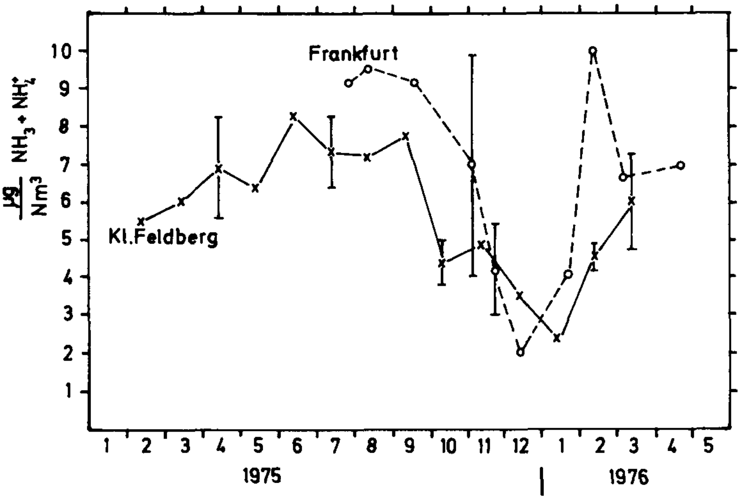

Fig. 2. The seasonal concentration trend for the sum of gaseous and particulate ammonia in the city of Frankfurt (crosses, *ca.* 45 measurements per month, each lasting 2 h) and on Kl. Feldberg (circles, 700 m above Frankfurt in 20 km distance, *ca.* 18 measurements during 2 weeks). The vertical bars indicate the standard deviation of the data.

falls well within the range of possible N losses from domestic animal excrements. An uptake of ammonia in the vegetation layer by plants would imply a higher release percentage or other additional production mechanisms.

Ammonia can also volatilize from mineral nitrogen fertilizers. The loss depends mainly on environmental conditions. From the results of different investigations it can be said that it increases with increasing soil pH values and temperature (Jewitt, 1942; Allison, 1955; Gerritzen and de Hoop, 1957; Kresge and Satchell, 1959; Fenn and Kissel, 1974) and decreases with growing moisture content, depth of fertilizer intrusion and fertilizer granular size (Musa, 1968; Nommik, 1973; Förster and Lippold, 1975).

For Germany a representative value of 1–2% has been estimated by Buchner (1977, personal communication). Values up to 20% were measured by Förster and Lippold (1975) for urea application. Urea makes up a few per cent (2%) of total fertilizer nitrogen used in the Federal Republic of Germany, but releases, however, four times as much nitrogen than mineral fertilizer (Buchner, 1977, personal communication). Therefore a range of 1–5% can be taken as a representative NH_3 loss from mineral nitrogen fertilizer to the atmosphere in this area. In Table 5, annual mean production rates are given for this loss range.

The amounts of applied mineral nitrogen fertilizer were taken from Statistisches Jahrbuch (1977). These volatilization fluxes are annually averaged values. Ammonia releases resulting from mineral fertilizer application are restricted, however, to the time of fertilizer application in the growing season (March–June). During this period the contribution to the total input can be four times larger and thus contribute at most an equal amount to the total source strength as volatilization from excrements.

Table 5. *NH_3 volatilization rates from mineral nitrogen fertilizer for assumed loss percentages*

NH_3 loss as percentage of mineral nitrogen fertilizer	F.R. Germany ($\mu g\ m^{-2}\ h^{-1}$)	Hessen ($\mu g\ m^{-2}\ h^{-1}$)	Bavaria ($\mu g\ m^{-2}\ h^{-1}$)
1	8	5	6
5	40	25	30

Finally we want to consider the volatilization of ammonia from soils by natural microbial processes. Source strengths of 1–10 $\mu g/m^2$ h for western European conditions have recently been estimated from field experiments, in which concentration changes within a closed chamber covering several representative soils were interpreted as an overall effect of NH_3 desorption and absorption at the soil surface (Georgii and Lenhard, 1978).

The decay of animal excrements makes, therefore, the largest contribution to the total NH_3 source strength of this region and NH_3 volatilization from mineral fertilizer and natural soils adds less than 25 and 10%. The larger values for the fluxes observed during the summer season can be explained by the fact that cattle are kept inside during winter, so that the conditions for ammonia escape into the atmosphere are less favourable. Higher temperatures during summer should accelerate the conversion of urea into ammonia.

5. Comparison of NH_3 fluxes and wet deposition of ammonia

The total upward NH_3 and NH_4^+ flux measured in this area should be matched, on the average, by a downward flux of the same amount of nitrogen resulting from ammoniacal nitrogen. Rates of wet deposition of NH_4^+ ions in this region have been reported to range from 32 to 70 μg N/m^2 h in winter and summer (Georgii and Lenhard, 1978) and bulk deposition should be larger than 35 μg N/m^2 h according to Böttger et al. (1978). It seems, therefore, that the measured NH_3 and NH_4^+ flux of 120 ± 39 NH_4^+/m^2 into the troposphere in this area in somewhat larger than the amount of NH_4^+ deposited in rain on the ground. This difference could be due to the meteorological situations encountered during the flight measurements. The flights were performed under clear conditions without rainfall for at least 1 day prior to the measurements. The soils were therefore drier than average and more NH_3 volatilizes under these conditions as shown by Förster and Lippold (1975) in an investigation on the relation between ammonia release and rain frequency.

Furthermore, precipitation in this area most frequently falls in maritime air masses coming from the North Atlantic and having lower ammonia concentrations. The wet ammonium deposition rate should therefore be smaller than the overall NH_3 production rate.

Another mechanism which might reduce the amount of NH_3 and NH_4^+ nitrogen reaching the ground from the middle troposphere as ammonium nitrogen is the reaction of NH_3 with OH radicals. This would destroy roughly 0.1 $\mu g/m^2h$ NH_3—N in an air column above 400 m in the region of our flight measurements. This value is based on measured NH_3 concentration profiles (Georgii and Lenhard, 1978), calculated OH concentrations (Röth and Ehhalt, 1978) and a reaction rate constant of 1.5×10^{-13} cm^3 molecule^{-1} s^{-1} (Stuhl, 1973; Hack et al., 1974; Smith and Zellner, 1975; Perry et al., 1976). This NH_3 destruction rate represents less than 1% of the total upward flux through the 400 m layer and cannot explain the difference between upward flux of particulate and gaseous ammonia and downward flux in rainwater. It seems therefore that—averaged over the year—less ammonia and ammonium is advected horizontally into the flight area than flows out of it. During the time of flight measurements, however, horizontal homogeneity can be assumed for the first kilometre in the atmosphere. A comparison of upward and downward vertical fluxes shows that roughly half of the upward total ammonia flux is not rained out in the same area. A similar regional outflow of total ammonia was presented by Söderlund (1976) for NW Europe. On a larger scale, however, nearly all of the particulate and gaseous ammonia diffusing into the mid troposphere will be incorporated into rain-water. This is the result of an evaluation of global precipitation chemistry and atmospheric source strength data for ammonia (Böttger et al., 1978).

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ОЦЕНКА ПОТОКОВ АММИАКА В СВОБОДНУЮ АТМОСФЕРУ НАД ЗАПАДНОЙ ГЕРМАНИЕЙ

Самолетными измерениями над сельской местностью в Западной Германии определялись плотности вертикальных потоков аммиака в газовой и твердой фазе через слой высотой 400 м над поверхностью. Плотности потоков вычислялись по градиентному методу. Потоки достигали 118 ± 49 мкг/м^2 час для NH_3—N и 93 ± 47 мкг/м^2 час NH_4^+—N летом и 7 ± 14 мкг/м^2 час NH_3—N и 23 ± 34 мкг/м^2 час NH_4^+—N зимой.

Будучи проинтегрированной за год, сумма этих потоков аммиака может поддерживаться улетучиванием аммиака из экскрементов домашних животных. Высвобождение аммиака из минеральных удобрений и естественных почв дает лишь малый вклад в суммарную скорость производства аммиака. Большая часть NH_3 и NH_4^+, диффундирующих вверх в среднюю тропосферу, со временем попадает в дождевые капли. Из рассмотрений плотностей потоков для превращения газового NH_3 в твердый NH_4^+ в первых 400 м атмосферы установлен нижний предел на константу скорости реакции псевдопервого порядка в $(1 \div 2) \cdot 10^{-5}$ с$^{-1}$.