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AMMONIA EMISSIONS TO AIR IN WESTERN EUROPE

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SUMMARY

Anthropogenic ammonia (NH_3) emissions to the atmosphere in western Europe (EEC + EFTA countries) are estimated to be between 2.8 and 5.2 Mt NH_3-N/year in the year 1990, with 4.0 Mt NH_3-N/year as best estimate. Waste from farm animals (cattle, sheep, goats, pigs, poultry and horses) is the principal source of atmospheric NH_3, as shown in Table 1.

Table 1 Origin and Quantity of Ammonia Emissions to Air in Western Europe in 1990

	Emission (Mt NH_3-N/y)	Range (Mt NH_3-N/y)	Origin (% of total)
Animal husbandry			
stables and manure	1.4		34
storage	1.3		32
manure spreading	0.3		8
grazing			
Subtotal	3.0		74
Fertiliser			
production	0.02		0.5
application	0.5		12.5
Leaf emission from crops	0.2		5
Miscellaneous	0.3		8
Total	4.0	2.8-5.2	100

The estimate of NH_3 emissions from animal husbandry is based on calculated and measured N excretions and losses, adjusted for national conditions when data are available. Fertiliser derived emissions are estimated from consideration of reactions between fertiliser compounds and soil, taking typical national characteristics of soil into account. Crops can both take up and emit NH_3 from the leaves; net emission is probably about 1.5 kg NH_3-N/ha/year, but can be larger when the weather during ripening is adverse. A variety of other minor sources also contribute to atmospheric NH_3: fur animals and other minor groups of farm animals, pets, exhalations from human beings, domestic use of ammonium products, refrigeration, combustion, treatment of waste water and disposal of sludge. Such emissions are included in "miscellaneous emissions".

The emissions can only be crudely estimated, as they vary greatly with circumstances. Thus the estimates for the Netherlands have a range of uncertainty of approximately 30%. The range of

uncertainty for other nations with fewer data useful for making emission estimates is probably even larger. The only source where the magnitude of NH_3 emission is accurately known is that of the fertiliser industry, as most plants measure the emissions and report them to authorities.

The estimate of NH_3 emissions in western Europe from the animal sector is about 15% higher than the lowest of other recent estimates. This is well within the range of uncertainty of such estimates. The estimate of total emissions is about 20 to 25% higher than other recent estimates as items not covered by most other estimates are included (crops and miscellaneous).

Published estimates indicate that NH_3 emissions in western Europe increased by about 50% between 1950 and 1980. The emissions probably peaked around 1990. There are no EEC regulations for NH_3 emissions, but national efforts and regulations are increasingly specifying rapid manure incorporation into soil after spreading; animal feed composition is better adjusted to avoid excessive N intake, and storage conditions for manures are being improved. Furthermore, fertiliser consumption is falling. Such measures should gradually reduce the emissions by about 20 to 30% of the 1990 level.

SECTION 1. INTRODUCTION

1.1 THE ISSUE

It is known that ammonia (NH_3) emissions to the atmosphere mainly originate from agriculture, especially from farm animals and their manures (Buijsman *et al*, 1986, 1987; Isermann, 1990a; Asman, 1992; Klaassen, 1992a,b).

Ammonia emission is an environmental issue because:

- NH_3 concentrations in the air in the vicinity of point sources (large animal husbandry units) can damage vegetation;
- increased aerial deposition of NH_3 and ammonium (NH_4^+) contributes to water and soil acidification and may be part of the complex of factors causing forest damage;
- NH_3 emissions are one of the principal sources for increased nitrogen (N) supply to natural areas; increased N supply to such areas can change the flora, contribute to eutrophication of terrestrial and aquatic ecosystems (e.g. the North Sea), and increase the fluxes of climate relevant gases (mainly N_2O) to the atmosphere.

These topics are reviewed by Ellenberg (1985); Istas *et al* (1988); Skeffington and Wilson (1988); Van Breemen and Van Dijk (1988); BML (1989); Malanchuk and Nilsson (1989); Fabry *et al* (1990); Bartels and Gehrmann (1990); Tamm (1991); Isermann (1993a); Sutton *et al* (1993) and Granli and Bøckman (1994).

Reports on the effects of NH_3 on man, animals, fish and plants have been issued by WHO (1986); the Institution of Chemical Engineers (1988); the US-EPA (1989); BML (1989) and the Fertiliser Institute (TFI, 1990). An assessment of NH_3 as a water quality factor has been published by Seager *et al* (1988).

The WHO/IPCS Environmental Health Criteria document on NH_3 (IPCS, 1986) discusses sources releasing NH_3 into the air. The document refers to estimates of such emissions published for the USA and the Netherlands, and states: "It must be emphasized that substantial uncertainties are associated with these estimates, which are given for rough comparison only." Further, the reports from the EMEP (co-operative programme for monitoring and evaluation of the long-range

transmission of air pollutants in Europe) (Iversen *et al* 1991; Sandnes and Styve, 1992) express a need for improved national emission estimates.

It is the purpose of this report to provide a more detailed estimate of these emissions than that given in the IPCS document and hence update this and similar documents in this respect.

1.2 BACKGROUND

The transformations of N between the many forms present in nature form a complex web of processes and reactions, the N cycle.

Man exerts a major influence on the N cycle through:

Agriculture

- soil usage (e.g. tillage, irrigation and drainage) that influence chemical and biological processes in the soil
- increased N input to soils through use of mineral fertilisers, growing of legumes (e.g. clover, beans, peas that can fix atmospheric N) and movement of N from one area to another through import of animal feeds
- animal husbandry, which plays a key role in transforming crops into products useful for humans, but also gives rise to large volumes of manures with associated N emissions.

Burning of fossil fuels in industrial, transport, military and civil activities. This generates N oxides that enter the N cycle as a general increase of nitrate (NO_3^-) input to soils and waters.

The N cycle has been reviewed by Söderlund and Rosswall (1982); Jenkinson (1990a,b) and Isermann (1993a) where further details can be found. A brief description of some of the processes in the N cycle is given in Appendix A.

Man-made changes in the N cycle raise environmental issues (Bøckman *et al*, 1990). ECETOC have earlier addressed one of these topics, nitrate in drinking water (ECETOC, 1988). The present report concerns another aspect of man's impact on the N cycle, namely NH_3 emissions.

NH_3 emissions are here defined as gaseous losses to the atmosphere.

NH_3 emissions as particulate substances to the atmosphere and to water are also mentioned when appropriate. However, NH_3, in the form of NH_4^+, is rather immobile in most soils because of ion exchange by clay particles and soil organic matter retards movement of NH_4^+ in soil, and NH_4^+ dissolved in soil water is prone to conversion to NO_3^- through microbial nitrification. There is thus insignificant leaching of NH_4^+ from soils. NH_4^+ bonded to soil particles may reach waters through soil erosion, and through run-off from intensive animal husbandry. Soil erosion is an important environmental issue, but outside the field covered by this report, as is NH_4^+ discharge to surface waters through sewage.

The topic of this report is NH_3 emissions in western Europe taken as the members of the European Community (EEC) together with European Free Trade Association (EFTA) members: Austria, Switzerland and the Nordic Countries of Finland, Norway and Sweden. Iceland is not included as it is so far out in the Atlantic that its small NH_3 emissions do not notably influence other European nations (Iversen *et al*, 1991). The reason for this restriction is that detailed information about sources is more easily available from the western than from the eastern parts of Europe.

NH_3 emissions in Europe is a complex topic, as agricultural practice and conditions for emissions vary with regions and countries. In order to bring out the main features without losing the details, the factors influencing emissions are discussed in the main text, while the national details are given in Appendix C.

1.2.1 Units and Nomenclature

NH_3 emissions are reported in the literature both as NH_3 and as NH_3-N. The form NH_3-N is used throughout the report. Literature data expressed as NH_3 have been converted to N using the conversion factor:

$$0.822 \times NH_3 = NH_3\text{-}N \qquad \text{(Eq. 1)}$$

In aqueous solution, NH_3 and NH_4^+ are related through the reaction:

$$NH_4^+ + H_2O \ll NH_3 + H_3O^+ \qquad \text{(Eq. 2)}$$

Below pH 8 (that is in almost all soils of agronomic interest in Europe) NH_4^+ is by far the dominant form. In this report the forms NH_3 and NH_4^+ are used according to which form is dominant in the process under discussion.

NH_3 concentrations in air are reported as μg NH_3-N/m^3 or mg NH_3-N/m^3 as appropriate. Conversion of ppm and ppb to mg/m^3 and μg/m^3 depends somewhat on temperature and pressure. The following calculation factors are used:

$$1 \text{ ppm} = 0.7 \text{ mg } NH_3/m^3 = 0.58 \text{ mg } NH_3\text{-}N/m^3 \quad \text{(Eq. 3)}$$

$$1 \text{ ppb} = 0.7 \text{ μg } NH_3/m^3 = 0.58 \text{ μg } NH_3\text{-}N/m^3 \quad \text{(Eq. 4)}$$

The concept of NH_3 emissions is used to describe net losses of NH_3 to the atmosphere from buildings, fields and crops. Circulation within enclosed systems (e.g. emitted from the soil but taken up by the crop canopy) is not regarded here as NH_3 emission.

1.2.2 Ammonia Concentrations in the Air

It has been known since 1804 that rainwater and thus air contains NH_3 (Sutton *et al*, 1993). NH_3 is present in air both as NH_3 gas and as its reaction product with acidic air components e.g. SO_2, giving NH_4^+ salts as aerosol and dissolved in droplets. These reactions are briefly discussed later in Section 4.2. Techniques for measuring NH_3 in air are reviewed by Kessel (1990). The subject of NH_3 and NH_4^+ in air has been reviewed by Warneck (1988); BML (1989) and Grünhage *et al* (1990).

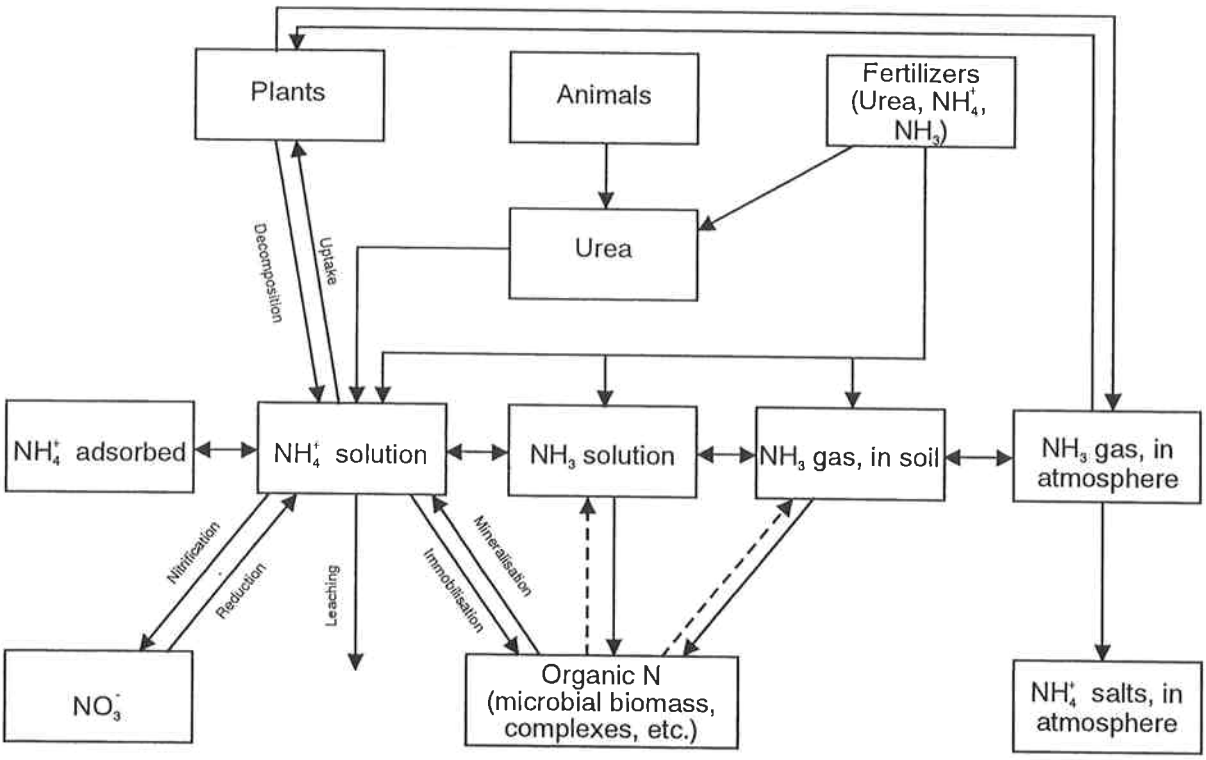
Concentrations over oceans and at high remote mountains are mostly below 1 μg NH_3-N/m^3. Concentrations over land are higher. Indoor concentrations in 10 homes in the UK averaged 36 μg NH_3-N/m^3 (Atkins and Lee, 1993). In urban areas concentrations of about 16 μg NH_3-N/m^3 are said to be typical (IPCS, 1986). Rural areas generally have lower concentrations within the range from 1-14 μg NH_3-N/m^3, with about 1-6 μg NH_3-N/m^3 as typical. Allen *et al* (1988) found that in rural areas in the UK, atmospheric concentrations were mostly around 2 μg NH_3-N/m^3. That urban areas have somewhat higher NH_3 concentrations than rural areas is supported by data listed by Atkins and Lee (1993). There are conflicting reports, as others find no difference between rural and urban air in this respect (Kruse *et al*, 1989). This topic deserves further investigation as urban areas may be an underrated source of NH_3. The concentrations tend to be higher in summertime than during winter. In areas of intensive animal husbandry NH_3 concentrations can be much higher, e.g. 50 μg NH_3-N/m^3 (Grünhage *et al*, 1990). Concentrations are commonly about 1 mg NH_3 - N/m^3 at a height

of 1-1.5 m above a field shortly after spreading of slurry (Bussink, 1993). Sensitive human beings can probably detect NH_3 at a concentration of about 2.2 mg NH_3-N/m^3, but most require a concentration about 27 mg NH_3-N/m^3 (46.8 ppm) to identify NH_3 by its odour (IPCS, 1986). The Occupational Exposure Limit (OEL) varies between countries, but are generally 15 mg NH_3-N/m^3 (18 mg NH_3/m^3 or 25 ppm, in Germany 50 ppm). Thus if the odour of NH_3 is noticeable, the concentration is too high. However, manure also emits other odorous materials (e.g. H_2S and organic substances) and perception of odour can not be used to measure NH_3 concentrations.

1.2.3 The Mechanism of Ammonia Emissions

This topic is reviewed in detail by Freney *et al* (1983) and Jayaweera and Mikkelsen (1990, 1991). A useful discussion is provided by Ross (1989). NH_3 is a gas at normal atmospheric temperatures and pressures, but with high solubility in water. NH_3 reacts with water and this reaction determines the concentration of NH_3 in the solution and hence the potential for NH_3 volatilisation. These relationships can be represented by Figure 1.

Figure 1 Some Biological and Chemical Reactions Affecting Ammonia Volatilisation (after Freney *et al*, 1983)



The NH_4^+ in soil originates as manures, fertilisers, and mineralisation (decomposition) of organic matter, e.g. plant residues. NH_3 can also be formed, e.g. in the rumen of animals and in the soil, by