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Exchange of atmospheric ammonia with European grasslands

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A new European scale programme has been established to address the biosphere-atmosphere exchange of ammonia (NH_3) with grasslands. GRAMINAE (GRassland AMmonia INteractions Across Europe) has set up a transect of six field sites from Scotland to Hungary and Greece to measure NH_3 fluxes using micrometeorological methods. At each site measurements are made to interpret fluxes in relation to atmospheric conditions, grassland management, sward characteristics and soil chemistry. These measurements are supported by laboratory and field studies of NH_3 exchange and bioassays of the NH_3 'stomatal compensation point' (χ_s). The aerodynamic gradient method (AGM), applying sensitive continuous NH_3 detectors, provides the core micrometeorological approach. Relaxed eddy accumulation (REA) is also being applied to enable flux measurements at one height; this is relevant to help address flux divergence due to gas-particle inter-conversion or the presence of local sources in a landscape.

Both the field and laboratory measurements demonstrate the bi-directional nature of NH_3 fluxes, with typically daytime emission and small nocturnal deposition. They also confirm the existence of enhanced NH_3 emissions following cutting of intensively managed swards. Further increased emissions follow fertilization), although in both cases most of the emission appears to be due to foliar emissions from the remaining sward. Measurements using REA support these patterns. However, the advantage of measuring at one height is partly offset by a requirement for greater analytical precision than with the AGM.
The results are being used to develop resistance models applying a 'canopy compensation point' approach to reproduce the bi-directional NH_3 fluxes. Key applications of the work include the incorporation of the resistance models in wider models of local and regional NH_3 dispersion and to assess scenarios of global change.

Introduction

Ammonia (NH_3) occurs naturally in the atmosphere, but most of the European emission (>90%) is of anthropogenic origin. The largest source is volatilization from livestock wastes with a significant fraction from fertilized agricultural land. NH_3 is important since:

- with emissions of SO_2 and NO_x, it contributes to long-range pollutant transport and acidic deposition - "acid rain",

- with emissions of NO_x it causes eutrophication of sensitive N-poor ecosystems - changing species composition of threatened habitats.
- atmospheric reaction of NH_3 produces ammonium aerosol (NH_4^+), which contributes to a negative global warming potential.

While NH_4^+ may be transported for >1000 km, deposition of NH_3 is largest in source areas. Hence relict natural habitats in intensive agricultural landscapes are particularly at risk.

Policy context

The 1979 Geneva Convention on Long-Range Transboundary Air Pollution (CLRTAP) was established under the auspices of the UNECE to tackle European air pollution as an international problem. In 1984 the first Sulphur Protocol ("30% club") was signed, and this was followed in 1991 by the NO_x Protocol, and in 1994 with the Second Sulphur (Oslo) Protocol. The agreements have now shifted from simple % cuts, to an effects- based approach minimizing the European exceedance of deposition over ecosystem sensitivity thresholds ('critical loads').

Since 1994, the Convention has been negotiating revision of the NO_x protocol and to deal with the linked issues is taking a multi-pollutant, multi-effect approach, including effects of NO_x, NH_3, SO_2 and VOCs. Hence, international abatement of NH_3 is being negotiated for the first time, with the protocol expected to be finalized in late 1999.

In parallel, the EC has developed its Acidification Strategy, which will lead to the National Emissions Ceilings (NEC) Directive, again including SO_2, NO_x, NH_3 and VOCs.

Objectives

GRAMINAE is a 2nd tranche TERI project running between 1998 and 2001 that has been designed to address these issues. It is widely recognized that there are major uncertainties in the quantification of the atmospheric NH_3 cycle. A key element is the biosphere-atmosphere exchange process. Data on this aspect are essential to parametrize the European transboundary models used in both the UNECE and EC negotiations. With this in mind, GRAMINAE has the following objectives:

- to quantify emissions from different sources (including land area sources such as intensive agricultural land)
- to quantify the rates of NH_3 deposition to receiving ecosystems
- to understand & quantify the biospheric controls on these processes
- to understand the controls on NH_3-NH_4^+ dynamics
- to develop models both to explain the processes and for incorporation in the European policy models used in the negotiations.

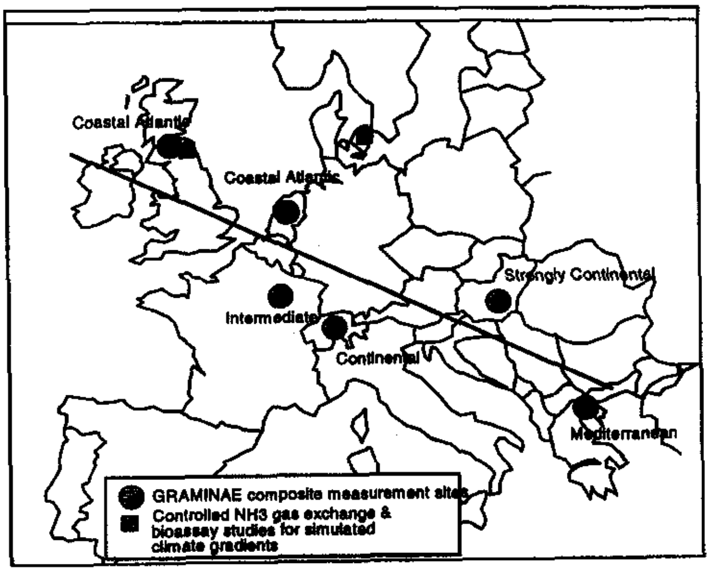

Figure 1: GRAMINAE experimental transect across Europe to address the effect of increasing continentality. It is hypothesized that biospheric controls result in longer atmospheric residence times of NH_3 in continental than in oceanic climates. This has implications for modelling the transboundary transport of NH_3 for example in estimating polluter-receptor 'blame matrices' for different European countries.

Approaches

The objectives of GRAMINAE are being achieved through:
1. Applying state-of-the-art technology to measure NH_3 fluxes with European grasslands using micro-meteorological methods.
2. Exploiting the TERI transect approach to quantify interactions with climate, particularly continentality (Figure 1).
3. Considering both I) intensive grasslands as these impact on the atmosphere (through emissions), II) semi-natural grasslands as these are impacted by the atmosphere (deposition).
4. Making complementary laboratory & interpretative measurements to understand the exchange processes.
5. Developing quantitative process based models of both I) biosphere-atmosphere exchange and II) grassland ecosystem functioning.
6. Providing the models to European policy modellers to underpin the negotations for revision of the UNECE N Protocol, and EC NEC.
7. Engaging in the UNECE and EC negotiations on the potential for and difficulties in NH_3 abatement across Europe.

Novel scientific elements in the project include: Europe-wide analysis of NH_3 exchange with grasslands; linking of micromet approach with laboratory cuvette analysis and plant bioassays; new flux sampling methods (Relaxed Eddy Accumulation, REA) for NH_3; Assessment of gas-aerosol flux interactions; Assessment of biospheric controls on atmospheric NH_3 concentrations; Development of dynamic ecosystem-NH_3flux models able to consider management practice.

Modelling ammonia fluxes

Ammonia exchanges in relation to a compensation point (χ_s) which is the concentration at equilibrium with $[H^+]$ & $[NH_4^+]$ in the apoplastic solution. Models being developed under GRAMINAE, describe the different component fluxes in relation to Ohm's Law: '*Voltage = Current x Resistance*' transposes to '$(\chi_1-\chi_2)$ = *Flux x Resistance*' (Figure 3).

The resistances are: R_a, R_b, atmospheric transfer; R_w, uptake to leaf cuticles; R_s, stomatal transfer; R_{ac}, R_{bl}, in-canopy transfer (for the 3-layer model), while the subscripts p and f denote upper and lower canopy parts. χ is air concentration, with χ_l and χ_c the litter and canopy compensation point, respectively.

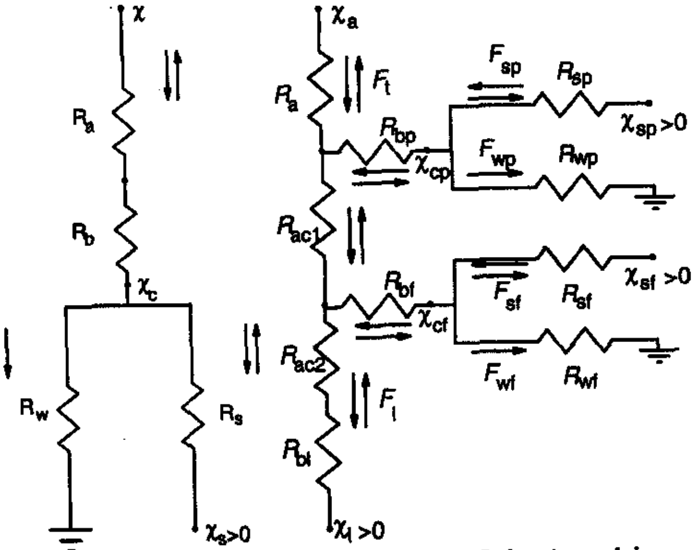

Figure 2: Example NH_3 models under development in GRAMINAE showing a 1-layer and 3-layer model. See text for explanation of concentration (χ), flux (F) and resistance (R) subscripts.

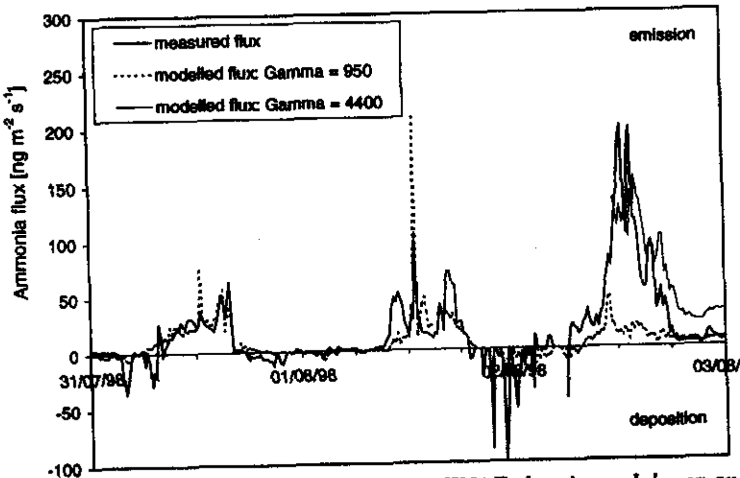

Figure 3: Example NH_3 models under development in GRAMINAE showing a 1-layer and 3-layer model. The one layer model is applied to grassland in the right hand graph.

The measurements shown in Figure 3 are for the Scottish GRAMINAE site. The period is after the second cut of the field, but before fertilization, with the grass being lifted on 2 August. The model (see Figure 2 – one layer model) is parametrized with $R_w = 30 \exp^{[100-\%RH]/7}$ (s m^{-1}) with the ratio of apoplastic $[NH_4^+]/[H^+] = \Gamma = 950$. The emissions following lifting of the grass would be consistent with $\Gamma > 4000$.

These results demonstrate both the close coupling of NH_3 fluxes to N turnover in the grassland, and the performance of the 1-layer model in rapidly changing conditions. A key challenge is to generalize these results for scaling up in time and for different European grasslands. In this respect GRAMINAE are coupling the NH_3 resistance modelling approaches with dynamic grassland ecosystem models in order to develop a functional prediction of NH_3 fluxes in relation to C and N cycling.

Current policy debate - the 'Ammonia Gap'

As Europe moves towards NH_3 emissions abatement for the first time, it is important to ensure that the investment will result in environmental benefits. To do this, it is necessary to show the link between changing emissions and monitored deposition of NH_3 and NH_4^+.
Over the last century the link is clear, with deposition estimates more than doubling at remote locations (e.g. ice cores, long-term precipitation chemistry).
However, NH_3 emissions should already have been reduced in two cases:

- *Eastern Europe* (e.g. Hungary), due to the collapse of the agricultural sector (reduced animal numbers and fertilizer consumption).
- *The Netherlands*, due to the implementation of an NH_3 abatement policy.

In both cases, analyses by GRAMINAE partners in the Netherlands, Hungary & UK have shown that the expected reductions are difficult to observe.

This has important implications for the policy negotiations, as it indicates uncertainty in our understanding of the controls on NH_3 levels. In recent discussions for the UNECE Protocol (5/1999) it was agreed that countries should proceed with caution using a selection of well established abatement techniques, but that further measurements are essential to investigate these issues.

One of the messages of GRAMINAE, is that a feedback exists (via the NH_3 'compensation point') where ecosystems can control NH_3 concentrations as well as being impacted by NH_3 deposition. In a further linkage it needs to be recognized that NH_3 abatement will be much less effective if N inputs to the system are also not reduced. Together with interactions with SO_2 chemistry, inter-annual variation and possible reduced efficiency of some abatement measures, this may go some way to explaining the ammonia gap.

Acknowledgements

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For further details see the following example publications:

Fowler D., Coyle M., Flechard C., Hargreaves K.J., Nemitz E., Storeton-West R., Sutton M.A., Erisman J.W. (2001). Advances in micrometeorological methods for the measurement and interpretation of gas and particle nitrogen fluxes. *Plant and Soil* **228** (1):117-129.

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