

Atmospheric fluxes current estimates and uncertainties

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Abstract

Within the LOIS programme, estimates of atmospheric fluxes are of primary interest to RACS(A) and RACS(C). The major objectives of RACS(A) include exploration of land-sea fluxes of nutrients and pollutants, net fluxes of climatically significant trace gases from estuarine and coastal systems and the characterisation of chemical transformations. In this presentation, the emphasis is on pollutants related to potential acidification and eutrophication; the estimation of land-sea fluxes and their source attribution and the calculation of atmospheric inputs to catchments.

The nature of emissions sources, their spatial distribution and trends through time are highly variable. There are also important differences in the precision of emissions estimates; sulphur dioxide (SO₂) emissions are believed to be the most reliable ($\pm 5-10\%$), while ammonia (NH₃) emissions may only be accurate to $\pm 50\%$. Some of the pollutants of interest to LOIS are poorly quantified and are believed to show big inter-annual and seasonal variability (e.g. DMS). National estimates of deposition are based on data from monitoring networks and models for dry deposition, cloud water deposition and orographic enhancement. Uncertainty estimates for national deposition fields for 1992-94 range from $\pm 40\%$ in central and eastern England to $\pm 80\%$ in north-west Scotland. Measurements of gas and precipitation concentrations by national network sites within the LOIS study area are quite restricted, with only one site (High Muffles, in North Yorkshire) collecting daily data. Other sites in the area have recorded some of the UK's highest precipitation concentrations of non-sea salt SO₄, nitrate (NO₃), ammonium (NH₄) and HCl. The LOIS study area is one of high sulphur (S), quite high oxidised N (NO_y) and variable ammonia (NH_x) deposition.

Atmospheric pollution transport models can provide flux estimates at higher spatial resolution than available measurement data and be used for source attribution work. In this study we have applied the HARM model (Metcalfe *et al*, 1995) to explore some of these issues. HARM is a simple, coupled-chemistry Lagrangian model which yields annual estimates of the concentrations and depositions of S, NO_y, NH_x and HCl. Two examples of applications of HARM to the LOIS area are given: source attribution of S deposition and modelled HCl deposition. HARM indicates that S deposition in the LOIS area is dominated by land-based sources, primarily coal fired power stations (see Figure). The contribution from DMS is very small, probably 1-2% of the total, although this is likely to show a marked seasonal variation following the emissions. Based on measured chemical loads of rivers in the LOIS area, it has been suggested that there are probably substantial inputs of dry deposited HCl 'missing' from the national scale atmospheric deposition fields (Smith *et al*, in press) (gas concentrations of HCl are not measured by the national monitoring networks); HARM can be used to explore this. The model suggests very high inputs of dry deposited HCl within the study area, with coal fired power stations again being the main source.

HARM is currently under development to improve both its spatial and temporal resolution. A month-by-month version shows very strong variation in deposition fluxes of some pollutants e.g. NO_y . Further work is also in progress on the chemistry in the model, with particular emphasis on DMS and daytime/night-time chemistry, which draws on measurements made at Weybourne.

References

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