QUANTIFYING UNCERTAINTIES IN SULPHUR AND OXIDISED N DEPOSITION TO WALES (UK) MODELLING USING HARM

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INTRODUCTION

In this paper we quantify the uncertainty associated with deposition estimates for 25 sites in Wales, in the west of the UK, made using HARM (Hull Acid Rain Model), within the Generalised Likelihood Uncertainty Estimation (GLUE) framework of Beven and Binley (1992). GLUE allows uncertainties to be assessed using observed data to condition model predictions. The approach does not attempt to obtain a single parameter set for a given model structure, but recognises that there may be a number of parameter sets which may be identified as 'behavioural' in describing the system, based on the fit to the observed data. The goodness of fit is measured by likelihood measures or objective functions. All behavioural parameter sets are used in the calculation of prediction bounds, yielding a range of model responses. Monte Carlo sampling of pre-specified parameter ranges is used to derive combinations of multiple parameters. The model is then run with each randomly chosen parameter set and its performance evaluated as being behavioural (or not) based on the likelihood measures. Model output was initially constrained by comparison with gas concentrations, and wet and dry sulphur and oxidised and reduced nitrogen depositions at 25 sites across Wales for 1995. The robustness of the parameter sets found to perform well for 1995 was then tested by re-running them for wet deposition measured at 44 sites in 1984.

MODELLING DEPOSITION

The performance of long range transport models is commonly evaluated by comparing their outputs with results from national monitoring networks (gas and precipitation concentrations and best estimates of deposition). National monitoring networks are limited to relatively few locations, with the UK's Acid Rain Monitoring Network currently comprising only 32 sites. In 1984, however, as part of a study by the then Welsh Water Authority (WWA), precipitation concentrations were measured at 44 sites across Wales to assess acid inputs. In 1995, there was a second, smaller survey (Welsh Acid Waters Survey) which measured precipitation concentrations at 25 sites and gas concentrations at slightly more sites, to form the basis for estimating wet and dry deposition of S and oxidised N species (Stevens *et al*., 1997). The results of these surveys represent the best data sets available for model validation.

HARM is a receptor orientated Lagrangian trajectory model which has been used extensively in recent years to assist the UK Department of the Environment (now DEFRA) and the Environment Agency (EA) in policy formulation. The model version used here provides estimates of annual deposition of S, oxidised (and reduced) N to the UK on a 10 km x 10 km grid. The model follows the composition of air parcels crossing both the EMEP (50 km x 50 km) and the UK (10 km x 10 km) emission grids. Inputs to each receptor cell are simulated along 72 trajectories, which are weighted using a wind-rose. HARM employs a simplified representation of meteorological conditions, including constant windspeed and constant drizzle. It has a coupled chemical scheme and includes a parameterisation of orographic enhancement which is believed to make a significant contribution to wet deposition in upland Britain. For scenario modelling HARM employs a long term (30 year average) rainfall field for the UK, but in this exercise we have assigned measured rainfall values to the 25 grid cells in Wales

containing the monitoring sites, whilst retaining average values elsewhere. An initial assessment of uncertainties in HARM modelled S deposition indicated that across the UK, the model was most sensitive to changes in emissions, but that there was considerable spatial variability in both the magnitude and major source of uncertainty.

SAMPLING OF EMISSIONS AND PARAMETERS

In our analysis, an estimate of uncertainty in emissions (SO_2, NO_x and NH_3) was combined with varying the values assigned to 9 model parameters. The emissions scaling factors and parameter ranges used are summarised in Table 1.

Table 1 a. Emissions scaling factors using best estimates.

Parameter	Minimum	Maximum
SO_2 emissions	0.7	1.3
NO_x emissions	0.7	1.3
NH_3 emissions	0.6	1.6

Table 1b. Parameter ranges.

Parameter	Minimum	Nominal	Maximum	Units
Boundary layer depth	640	800	960	m
Wind speed	5.2	10.4	15.6	m s^{-1}
Dry deposition velocity	0.7	1	1.3	Factor
Washout removal coefficient	0.5	1	1.5	Factor
SO_2 conversion rate	1.4×10^{-6}	2.8×10^{-6}	4.2×10^{-6}	s^{-1}
All reaction rates	0.5	1	1.5	Factor
OH radical concentration	0.4×10^{-6}	0.8×10^{-6}	1.2×10^{-6}	molecules cm^{-3}
Background O_3 concentration	15	30	45	ppb
Scavenging rate (HNO_3 to NA)	1.5×10^{-5}	3.0×10^{-5}	4.5×10^{-5}	s^{-1}

Estimates of uncertainties in emissions were based on published data (RGAR, 1997; NEGTAP 2001). In the sampling strategy, we tried to allow for correlation between emissions from same source processes (e.g. NO_x and SO_2 from power generation) and to identify that fraction of the emission that would be uncorrelated between the different pollutants. The full analysis was based on two different strategies for emissions uncertainties: one using a range wider than the published estimates and a second using just the RGAR and NEGTAP 'best estimates'. In this paper we refer only to the results of the latter, restricted ranges. The prior likelihood weighting assigned to the different emissions scenarios was propagated through to the final predictive uncertainty estimation. Each emissions scenario was then run using the full range of HARM parameter sets. The parameters chosen (see Table 1) were consistent with previous, more limited, assessments of uncertainties in HARM (e.g. Metcalfe *et al.*, 1995; Page *et al*., submitted). A total of some 200,000 model runs were carried out to estimate deposition at each of the 25 sites, with about 100,000 relating to the best estimates used here.

MODEL RESULTS

Model output from the different parameter sets was assessed (constrained) by comparing measured and modelled gas concentrations and depositions. As well as absolute deposition values, ratios of wet to dry deposition, based on the measurement data, were also used to reject simulations. Parameter sets were initially constrained for each site individually and then by comparison with all sites. The 5th and 95th percentiles of the likelihood-weighted cumulative distribution were selected as measures of the uncertainty prediction bounds. An initial combination of likelihood measures across all 25 sites and using the broad emissions ranges,

resulted in all model runs being rejected as non-behavioural. It was found that four sites (Llyn Brianne, Ogwen, Waunfawr and Ynysfro Reservoir, see Figure 1) were problematic and these were excluded from the subsequent analyses. Using the restricted emissions uncertainty range (the published best estimates), 2101 parameter sets were found to yield behavioural results. In general terms, wet S deposition is well simulated by the model, while wet oxidised N is systematically overestimated. Model performance for wet reduced N deposition was generally acceptable, but was given less weighting in the assessment due to the uncertainties relating to many aspects of this pollutant. The spatial pattern of wet S uncertainty prediction bounds for the 25 sites is illustrated in Figure 1.

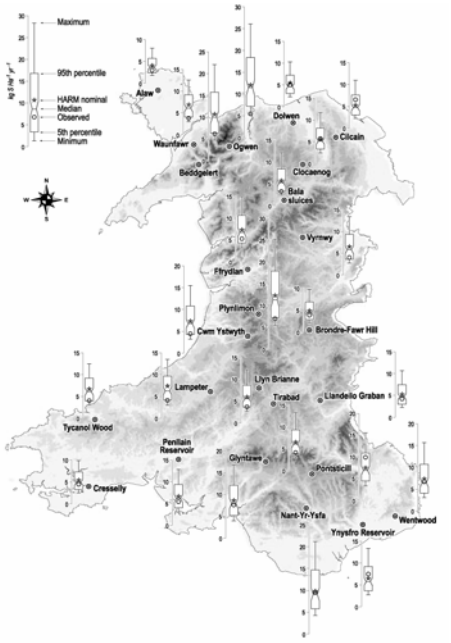

Figure 1. Box and whisker plots showing 1995 HARM estimates of wet deposited S combined across all sites using restricted emissions ranges.

Overall, the prediction bounds bracketed the observed values. HARM wet S deposition estimates are close to, or slightly less than, observed values (e.g. Ynysfro Reservoir, Dolwen) close to the major source areas of south and north east Wales. Elsewhere, model depositions are more than observed, with this becoming most pronounced in the highest deposition areas (e.g. Ogwen). Wet oxidised N and wet reduced N show similar patterns with overestimation at the wettest sites and better estimates close to source areas. The overestimation of wet oxidised N deposition was systematic across most sites (Figure 2) with measured values for many sites lying below the 5th percentile of the HARM prediction bounds and some even below the minimum behavioural value estimated by HARM. Values for reduced N were closer to the observed, generally bracketed by the prediction bounds. Although high deposition values can be caused by either proximity to source or/and high rainfall, the patterns that emerge suggest that the standard parameterisation of orographic enhancement used in HARM, may not be appropriate for individual regions of the UK, such as Wales.

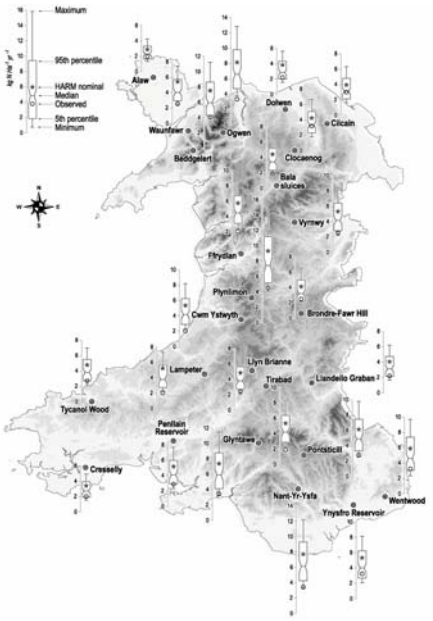

Figure 2. Box and whisker plots showing 1995 HARM estimates of wet deposited oxidised N combined across all sites using restricted emissions ranges.

HARM's predictive capabilities were tested by applying the same 2101 parameter sets identified as behavioural for 1995, to 1983 emissions to compare with the larger number of measured values from the 1984 WWA survey (1984 emissions were not available). For wet S, the spatial pattern of under prediction was similar between the two years, but there was more variability in over prediction, with more sites close to source areas falling into this category in 1984. Patterns for oxidised N were similar between 1984 and 1995, although overestimation was less pronounced for 1984. Based on sites common to both surveys, there is more variability in oxidised N wet deposition between years than there is in wet S deposition. Overall, the model did less well in relation to reduced N deposition in 1984 than in 1995, although the pattern of over and underestimation was similar.

CONCLUSIONS

Modelled and measured S and N depositions across Wales for 1995 have been compared using uncertainty prediction bounds derived from combined emissions and parameter uncertainty estimates. Overall, model performance was better when using the median values of the prediction distributions constrained by the observed data than when the standard parameter set was used. This is unsurprising as the model's standard parameters have been set based on values in the published literature, not tuned to a particular region. HARM is generally able to make estimates of wet S and reduced N deposition that bracket the observed values, but for Wales, there was systematic overestimation of wet oxidised N deposition. This suggests that there will be systematic bias in the standard model outputs for Wales for this pollutant and that we might need to regionalise the values of some model parameters (e.g orographic enhancement) to achieve better results.

HARM was not able to capture the full spatial variability of deposition recorded by the 25 sites in the 1995 Welsh Acid Waters Survey using any single combination of parameter sets. In order to retain some predictive capability, four sites were excluded from the analysis. This result is not unexpected as we have employed a very stringent, site specific approach to testing HARM. Again the issue of the how a model represents the spatial complexity of orographic enhancement needs to be considered.

Employing the 5th and 95th percentile values of wet S and wet oxidised N deposition gives large ranges of values compared with the standard HARM model run. Such variations have considerable implications for the calculation of critical loads exceedances and hence any assessment of the likely value of emissions reduction policies. It is also clear from this analysis, that for HARM to be used for modelling future emissions scenarios (beyond 2010) when climate change might become more apparent, then a more detailed assessment of model sensitivity to rainfall distribution and wind speed is needed.

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