



Past acidification and recovery of surface waters, soils and ecology in the United Kingdom: Prospects for the future under current deposition and land use protocols

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ABSTRACT

The dynamic hydrogeochemical model MAGIC has been applied extensively throughout Europe and North America and has proven to be a valuable tool for predicting the chemical response of catchment soils and surface waters to future changes in atmospheric pollution and land use. Here we present an application of MAGIC to 22 sites in the UK Acid Waters Monitoring Network (UKAWMN) that incorporated uncertainty in model calibration (using 20 years of surface water observations) to evaluate past acidification and prospects for future recovery in surface waters, soils and key ecological indicators. Simulated ANC in 1860 indicated that prior to industrialisation, all UKAWMN surface waters were above the critical acid neutralising capacity (ANC) limit of $20 \mu\text{eq l}^{-1}$, with the exception of a naturally acid site, underlain by granite with large expanses of bare rock, scree and eroded peat in Northern Ireland (Blue Lough); this site had a baseline ANC value $<20 \mu\text{eq l}^{-1}$. The significant increase in acidic deposition from 1860 to its peak during the 1970s resulted in surface water acidification at 14 of the study sites ($\text{ANC} < 20 \mu\text{eq l}^{-1}$). The rate and magnitude of chemical recovery in soil is highly variable among sites owing to differences in factors such as soil mineral weathering and base saturation. Despite the significant reductions in sulphur and to a lesser extent nitrogen deposition since 1970, the simulated soil base saturation at all study sites either continued to decline or remained stable until the late 1980s, with marginal recovery detected at some sites in the past decade. Predictions were based on an emission reduction scenario (Gothenburg protocol) to 2020, and land use scenarios to 2050 based on approved Forestry Commission plans at the five afforested sites. Model predictions indicated that, in general, surface water acid status will continue to improve during the next decade and beyond under the Gothenburg protocol. Surface water recovery was primarily attributed to the significant decline in sulphate concentrations from a present day mean of $58.5 \mu\text{eq l}^{-1}$ – $43.8 \mu\text{eq l}^{-1}$ in 2020. The contribution of nitrate (NO_3^-) leaching to the total acid status of surface water was small and predicted to decrease from $10.9 \mu\text{eq l}^{-1}$ in 2007 to $9.6 \mu\text{eq l}^{-1}$ in 2020. By 2100, NO_3^- concentrations increased slightly to $11.8 \mu\text{eq l}^{-1}$ having a small confounding influence on the rate of chemical recovery at most sites in the network. The future response of soil base saturation to reductions in acid deposition and land use change was mixed and the difference in behaviour between sites is unclear. Whilst recent studies (Malcolm et al., 2014a,b; Monteith et al., 2014) have demonstrated that forest practices have contributed to the acidification of surface waters, in this study, there was no evidence from the model predictions that forested sites will follow a different recovery trajectory to moorland sites. Planned reductions in coniferous forest cover amounting to approximately 13% across the five afforested sites resulted in a slight increase in ANC and pH. Ecological predictions highlighted the sensitivity of three indicator species (*Baetis Rhodani*, *Achnanthes minutissima*

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and salmonids) to changes in the acid status of surface water in the past and into the future. There was clear evidence of biological recovery by 2015 with some sites returning close to their pre-industrial biological status (Allt a'Mharcaidh) while at others (including Loch Grannoch and River Etherow) predicted recovery was incomplete for one or more of the studied indicator species.

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1. Introduction

Deterministic simulation models are important management tools used to mimic or predict the state of hydrological, hydro-geochemical and other complex environmental systems under different (typically future) scenarios. The Model of Acidification of Groundwater in Catchments (MAGIC) has been applied extensively in scientific and management studies throughout Europe and North America and used in policy related assessments in support of sulphur (S) emission reduction protocols and legislation. The model was developed and tested using data from long-term monitoring sites, and from manipulation experiments (Cosby et al., 1985, 2001); MAGIC (version 7.77ext February 2008) includes a conceptual model of nitrogen (N) retention and, as such, is capable of simulating the acid–base response of catchments to both S and N deposition. Moreover, MAGIC simulations of surface water and soil chemical variables have been widely used as indices of the health status of fisheries, forest vitality, and surface water acidification or eutrophication. Deterministic (dynamic) hydrogeochemical models, such as MAGIC, provide an indication of the time to geochemical reversibility and biological recovery in response to emissions reductions. Moreover, dynamic models offer the only opportunity to determine the level of deposition reduction required to achieve a given chemical restoration target, and hence a biological response, within a given time scale. Given the importance of 2015 for the implementation of both the Water Framework Directive (Water Framework Directive, 2000) and UNECE protocols (UNECE, 2012); dynamic models provide a methodology to evaluate the combined environmental consequences of both pieces of legislation.

Deterministic models (as with all models) are prone to several sources of uncertainty (Funtowicz and Ravetz, 1990; Saloranta et al., 2003). In general, the greatest source is related to the quality, quantity and representivity of model input data and observations used in calibration, i.e., parametric uncertainty (Blöschl and Grayson, 2000; Uhlenbrook and Sieber, 2005). The extent to which the model describes the 'real world' system, i.e., structural uncertainty is another type. Models are by necessity simplified descriptions of natural systems, and the challenge is to include the important process descriptions (for the problem under investigation) without over parameterisation (Perrin et al., 2001). Given the importance of models in the development of environmental policies, it is necessary to present model forecasts with associated uncertainties. However, most model applications typically ignore uncertainty analyses.

Calibration of input parameters through fitting the corresponding model predictions to observational data is an important step in developing dynamic, process-oriented models (Larssen et al., 2006). Several hydrological modelling studies have assessed the relationship between prediction uncertainty and length of calibration time series data (Anttil et al., 2004; Brath et al., 2004; Xia et al., 2004). Similarly it has been demonstrated that hydro-geochemical model calibrations conditioned upon an increasing number of observational data have significantly reduced uncertainties in predictions (Larssen et al., 2004). A fundamental approach to quantifying uncertainty is to assign probability distributions to the input parameters and thereby obtain probability distributions rather than single values of model outputs. Several approaches have been developed for uncertainty assessment combining Bayesian

techniques and hydrological catchment models (Bates et al., 2003; Kuczera and Parent, 1998; Makowski et al., 2002; Thiemann et al., 2001). Such approaches are less developed for biogeochemical or hydrogeochemical models, as well as other types of integrated water resource models, but are becoming increasingly available (Larssen et al., 2006).

The objective of this study was to evaluate past acidification and prospects for future recovery in surface water chemistry, soils and key ecological indicators in response to emission controls for the UKAWMN. Model calibration was based on long-term surface waters observations (>20 years) using Bayesian techniques. Uncertainty in model prediction was assessed by incorporating uncertainty in the model input. Although structural uncertainty cannot yet be quantified until our models' weaknesses are better characterised, we can still use the standard approach of cautiously interpreting results from poorly calibrated sites. In addition to acidic deposition, forestry is acknowledged to be a contributing factor to the acidification of surface waters owing to enhanced interception of acidic deposition by forest canopies (Forestry Commission, 2003; Harriman et al., 1987) and the loss of neutralising capacity in forest soils as a result of base cation uptake and removal through harvesting. In the current study, an attempt was also made to assess the response of surface waters to future planned forest management under reduced emissions.

2. Materials and methods

2.1. Sites

The UKAWMN was established in 1988 as part of the UK Department of the Environment monitoring programme to assess trends in surface water acidity associated with reduced acidic oxide emissions (Patrick et al., 1995). The network was designed to include all acid sensitive regions (predominantly upland) in the UK, across the range of acidic deposition. It included streams and lakes, afforested and non afforested catchments across a range of altitude, and site geological types. Conifer plantations were present at only five sites and represented the principal land use impact in the network (Shilland et al., in this issue).

2.2. The MAGIC model

MAGIC is a process-orientated model, developed to provide long-term reconstructions and predictions of soil and stream water chemistry in response to scenarios of acidic deposition and land use (Cosby et al., 1985). The model consists of: (i) soil–soil solution equilibrium equations in which the chemical composition of the soil solution is assumed to be governed by simultaneous reactions involving cation exchange, dissolution and speciation of inorganic and organic carbon; and (ii) mass balance equations in which fluxes of major ions to and from the soil and surface water are assumed to be governed by atmospheric inputs, mineral weathering, net uptake by biomass and loss in runoff. Studies on UK upland lakes (e.g., Cooper and Jenkins, 2003) have demonstrated a direct link between S input and output fluxes, confirming that in-catchment processes have a minor impact. Therefore, SO_4^{2-} was treated as 'pseudo-conservative', with surface water output equal to deposition inputs in the current study. Nitrogen dynamics within the model embrace the N saturation concept (Stoddard, 1994); a soil

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