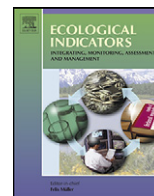




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Ecological Indicators

journal homepage: www.elsevier.com/locate/ecolind



Recovery of macroinvertebrate species richness in acidified upland waters assessed with a field toxicity model

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ARTICLE INFO

Article history:

Received 3 December 2010

Received in revised form 3 October 2011

Accepted 2 November 2011

Keywords:

Acidification
Bioavailability
Chemical speciation
Lakes
Macroinvertebrates
Modelling
Streamwaters
Toxicity

ABSTRACT

The WHAM- F_{TOX} model uses chemical speciation to describe the bioavailability and toxicity of proton and metal mixtures (including Al) to aquatic organisms. Here, we apply the previously parameterised model to 45 UK and Norwegian upland surface waters recovering from acidification, to compare its predictions of the maximum species richness of the macroinvertebrate Orders Ephemeroptera, Plecoptera and Trichoptera (SR-EPT) with time-series observations. This work uses data from two national scale survey programmes, the Acid Waters Monitoring Network in the UK and a lakes survey in Norway. We also investigate data from a long-studied catchment, Llyn Brianne in Wales. For the national surveys, model results relate well with actual trends, with Regional Kendall analysis indicating biological recovery rates for both actual and predicted species richness that are generally consistent (1.2–2.0 species per decade). However, actual recovery rates in AWMN lakes were less than in the rivers (0.6 vs. 2.0 species per decade), whilst predicted rates were similar (1.7 vs. 2.0). Several sites give a very good fit between model predictions and observations; at these sites chemistry is apparently the principal factor controlling limits of species richness. At other sites where there is poorer agreement between model predictions and observations, chemistry can still explain some of the reduction in species richness. However, for these sites, additional (un-modelled) factors further suppress species richness. The model gives a good indication of the extent of these un-modelled factors and the degree to which chemistry may suppress species richness at a given site.

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1. Introduction

Acidification of sensitive freshwaters by acid precipitation has affected large parts of Europe and North America (Rodhe, 1989). After acid deposition peaked in the 1970s and 1980s many countries introduced monitoring programmes for water chemistry and biological indicators with the aim of examining responses to reductions of acidifying emissions of S and N. Measured biological indicators include macrophytes, diatoms, macroinvertebrates and fish. A number of these monitoring programmes have been run-

ning for two decades or more and assessments have been made on the observed long-term data (Raddum et al., 2001; Monteith et al., 2005; Ormerod and Durance, 2009).

Whilst chemical and biological recovery is clearly underway in many acidified regions, biological improvement is often considered to be “modest” or lagging behind the observed reduction in acidity, and various hypotheses have been advanced to explain this apparent hysteresis (Yan et al., 2003; Ledger and Hildrew, 2005). However, as little is normally known of the pre-acidification communities of recovering waters these are often largely subjective judgements, based on comparisons with sensitive ecosystems in areas of little acid deposition, or with neighbouring circumneutral streams. Both are of restricted power, because of biogeographic variation and fundamental biogeochemical differences respectively.

More generally, species-acidity modelling has been largely based on analysis of individual components (pH, Al, DOC, acid neutralising capacity; Andrén and Jarlman, 2008; McFarland et al.,

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2010) and has rarely attempted to represent the complexity of competing and sometimes possibly synergistic toxic effects of hydrogen, aluminium ions and heavy metals on freshwater organisms. The UNECE manual on modelling critical loads and levels exclusively uses acid neutralising capacity-limits to consider avoidance of potential harm to biotic species in surface waters (UNECE, 2004). Other work describes the concept of consensus based 'Threshold Effect Concentrations' above which harmful effects (from metals and organic pollutants) are likely to be observed (e.g. MacDonald et al., 2000). Such techniques have also been used to derive sediment quality guidelines from field-based species sensitivity distributions (MacDonald et al., 2000; Leung et al., 2005). Comprehensive speciation modelling considering multiple chemical conditions is a feature of the Biotic Ligand Model (BLM). However, this gives a single concentration at which a toxic response occurs in a specific organism, rather than a graded species richness response to a range of chemical conditions. BLMs generally only deal with toxicity of a single metal and have not been formulated for proton toxicity.

Consequently, the degree that freshwater biota have kept pace with chemical improvements in acidified waters has not been quantitatively assessed. The development of a new ecotoxicological modelling approach (Stockdale et al., 2010) provides an opportunity to do this for the first time.

In this work we compared, at an annual time step, temporal trends of a biological metric (species richness of Ephemeroptera, Plecoptera and Trichoptera; SR-EPT) with theoretical maximum values determined by a mixture toxicity model, which incorporates chemical speciation modelling. The model relates streamwater concentrations of cationic metallic species and protons to a field ecological index of biodiversity (SR-EPT) in order to determine the extent to which the macroinvertebrate communities of chemically recovering waters may be limited by contemporary chemical conditions. Datasets were drawn from the UK Acid Waters Monitoring Network, studies of the experimental catchment Llyn Brienne and monitoring of Norwegian lake outlet streams. The EPT index is employed as a measure of biological integrity in North America and is sensitive to a wide range of stressors (Plafkin et al., 1989). It was appropriate for the present work in view of data availability, established sensitivity, widespread distribution, and ecological importance of these Orders in stream ecosystems and sufficient variability to indicate graded responses to acidification. Taxa richness measures have been shown to be superior to alternatives in terms of sensitivity, variability and statistical power (Carlisle and Clements, 1999).

Aluminium becomes more soluble and, therefore, potentially more toxic to aquatic organisms at acidic pH (Gensemer and Playle, 1999). Toxic effects will be modified by interactions with dissolved organic matter (DOM) and competition for DOM binding sites from other cations including protons. A recent study by Moe et al. (2010) demonstrated statistical links between a range of macroinvertebrate metrics and pH, but also the importance of humic substances (DOM) in ameliorating effects on macroinvertebrates, as has previously been demonstrated for salmonids (McCartney et al., 2003; Hesthagen et al., 2008). This suggests that DOM may protect against hydrogen ion and metal toxicity and should be considered in the assessment of chemical relationships with macroinvertebrate metrics.

WHAM- F_{TOX} (Stockdale et al., 2010), is a toxicity model that, through chemical speciation, relates the observed field variable SR-EPT to the combined toxic stress of a hydrochemical cocktail of protons, aluminium (Al), heavy metals, dissolved organic carbon (DOC) and base cations. Thus it attempts to predict the hydrochemical SR-EPT "carrying capacity" of a freshwater, i.e. the maximum number of EPT species that can be supported in that specific chemical environment. The degree to which observations of SR-EPT

fall below those predicted will depend upon the intensity of un-modelled factors. Data falling below the model prediction are the result of depleted species richness caused by factors not included in the model. Thus, general biological assessment requires additional knowledge over chemistry and our model takes this into account. We model the 90th quantile so any reference to "maximum values" refers to the value below which 90% of data will fall. It is important to emphasise that the model is not designed to predict actual EPT, but how it may be affected by (inorganic) chemistry.

This work sought to compare the parameterised model predictions with observations for acidified surface waters, in order to help interpret the biological signals. This is an alternative to conventional correlation type analysis (e.g. plotting EPT species numbers against pH) and offers a more mechanistic approach. It should be recognised that we are not attempting to fit actual data but to show how well the model predicts the effects of chemistry, whilst also recognising that other un-modelled factors may suppress species richness. Predictions will be less than the theoretical maximum in many cases due to modelled chemical factors and in ~90% of cases predictions should be greater than observed data because of un-modelled factors. We firstly assessed recovery trends, comparing model predictions and observations of species richness using Regional Kendall testing to assess any potential recovery in these sites from acid deposition. Secondly, time-series for individual sites were examined to explore the extent to which chemistry can explain suppression of species richness.

2. Methods

2.1. The WHAM- F_{TOX} model

WHAM- F_{TOX} was developed in order to relate complex chemical mixtures, including protons, dissolved organic carbon (DOC), major ions and cationic trace metals, to toxicity in the field. Accumulation of metals by macroinvertebrates, akin to the binding of cations to conventional ligands, is the measure of proton and metal exposure (discussed further in Section 2.1.2). A full description of the model development and parameterisation is given in Stockdale et al. (2010); a concise description is included here.

2.1.1. Chemical speciation

WHAM (Tipping, 1994) is a chemical speciation model that calculates the equilibrium distributions of dissolved components based on input parameters, such as temperature, pH, pCO_2 , and the concentrations of DOC, major ions and trace metals. The model incorporates Humic Ion-Binding Model VI (Tipping, 1998). Model VI uses a structured formulation of discrete, chemically plausible, binding sites for protons, allowing the creation of regular arrays of bidentate and tridentate binding sites for metals. Metal aquo ions (Al^{3+} , Fe^{3+} , etc.) and their first hydrolysis products ($AlOH^{2+}$, $FeOH^{2+}$, etc.) compete with each other, and with protons, for binding. The same intrinsic equilibrium constant is assumed to apply to the aquo ion and its first hydrolysis product (Tipping et al., 2008). The humic ion-binding model is combined with an inorganic speciation model, the chemical species list and constants for which are specified in Tipping (1994). In calculating aqueous chemical speciation, the concentrations of Na, Mg, K, Ca, Cl, NO_3 and SO_4 were assumed to represent truly dissolved components, as were concentrations of filterable trace metals (Ni, Cu, Zn, Cd and Pb). Where reactive Al was measured this was used for the calculations. Where filtered Al was measured the activity of Al was calculated from the measured total filtered concentration and also from the generalised equation derived by Tipping (2005), with the lower of the two values being adopted. This avoided over-estimation of Al activity in filtrates containing colloidal forms of the element. The generalised

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