Recovery and non-recovery of freshwater food webs effects of acidification

Authors:

Clare Gray[1,2] – clare.gray@imperial.ac.uk

Alan Hildrew[2, 3] – a.hildrew@qmul.ac.uk

Xueke Lu[4] - xueke.lu@qmul.ac.uk

Athen Ma[4] - athen.ma@qmul.ac.uk

David McElroy[5] - david.mcelroy@sydney.edu.au

Don Monteith[6] - donm@ceh.ac.uk

Eoin O'Gorman[1] - e.ogorman@imperial.ac.uk

Ewan Shilland[7] - e.shilland@ucl.ac.uk

Guy Woodward[1]* - guy.woodward@imperial.ac.uk

*Corresponding author

1. Department of Life Sciences, Imperial College London, Silwood Park Campus, Buckhurst Road, Ascot, Berkshire SL5 7PY, UK
2. School of Biological and Chemical Sciences, Queen Mary University of London, London E1 4NS, UK
3. Freshwater Biological Association, The Ferry Landing, Ambleside, Cumbria, LA22 OLP
4. School of Electronic Engineering and Computer Science, Queen Mary University of London, London E1 4NS, UK

5. Centre for Research on Ecological Impacts of Coastal Cities, University of Sydney, 2006, Australia
6. Centre for Ecology & Hydrology, Lancaster Environment Centre, Lancaster LA1 4AP, UK
7. Environmental Change Research Centre, University College London, Gower Street, London WC1E 6BT, UK

SUMMARY

Many previous attempts to understand how ecological networks respond to and recover from environmental stressors have been hindered by poorly resolved and unreplicated food web data. Few studies have assessed how the topological structure of large, replicated collections of food webs recovers from perturbations. We analysed food web data taken from 23 UK freshwaters, sampled repeatedly over 24 years, yielding a collection of 442 stream and lake food webs. Our main goal was to determine the effect of acidity on food web structure and to analyse the way food web structure recovered from the effects of acidity over time.

Long-term monotonic reversals of acidification were evident at many of the sites, but the ecological responses were generally far less evident than chemical changes, or absent. Across the acidity gradient, food web linkage density and network efficiency declined with increasing acidity, while node redundancy (i.e. trophic similarity among species within a web) decreased. Within individual sites, connectance, linkage density, trophic height, resource vulnerability and network efficiency tended to increase over time as sites recovered from acidification, while consumer generality and node redundancy tended to decrease. There was evidence for a lag in biological recovery, as those sites showing a recovery in both their biology and their chemistry were a nested subset of those which only showed a chemistry trend.

These findings support the notion that food web structure is fundamentally altered by acidity, and that inertia within the food web may be hindering biological recovery. This suggestion of lagged recovery highlights the importance of long-term monitoring when assessing the impacts of anthropogenic stressors on the natural world. This temporal dimension, and recognition that species interactions can shape community dynamics, is missing from most national biomonitoring schemes, which often rely on space-for-time proxies.

I. INTRODUCTION

Natural ecosystems are increasingly exposed to anthropogenic stressors, such as habitat modification, pollution and global climate change (Steffen *et al.* 2011; Smith & Zeder 2013; Sutherland *et al.* 2016). A deeper understanding of how they respond to and recover from such perturbations is important if we are to manage our natural resources effectively in the coming decades (Pimm *et al.* 1995; Woodward *et al.* 2010a).

Biological recovery from the effects of stressors does not necessarily follow from the removal of that stressor, as there may be time lags or ecological hysteresis, even to the extent that alternative equilibria are possible for otherwise identical environmental conditions (O'Neill 1998; Scheffer & Carpenter 2003; Feld *et al.* 2011; Battarbee *et al.* 2014; Murphy *et al.* 2014). These may arise via species interactions, which can alter the rate and/or trajectory of recovery (Scheffer & Carpenter 2003) and confound attempts to scale up predictions made from individuals or species populations to the whole-community or ecosystem, because of the increasing scope for "ecological surprises" to be manifested via complex indirect pathways in the food web (Ings *et al.* 2009; Thompson *et al.* 2012). For instance, artificially high nutrient concentrations can trigger regime shifts in shallow lakes, which may persist even long after nutrient loads have been reduced (Scheffer & Carpenter 2003).

A. Food web recovery research

Due to the difficulties in constructing highly-resolved food webs, however, very few studies have examined how trophic network structure responds to, and recovers from, perturbations, and fewer still have a replicated design. Field experiments have revealed how replicated freshwater food webs respond to drought, through the loss of rare and rare-for-size consumers, as well as the larger taxa high in the food web (e.g. eight stream food webs; Ledger *et al.* 2012; Woodward *et al.* 2012). Other studies have economised on effort and increased their sample sizes, and hence ability to detect responses statistically, by making assumptions about the diet of consumers. Thus, O'Gorman and Emmerson (2010) used 144 marine food webs in a mesocosm study across a range of experimental treatments to investigate how their structure responded to the removal of keystone species. Very few examples exist where the recovery of replicated, natural food webs following a perturbation has been studied, although McLaughlin, Emmerson and O'Gorman (2013) constructed a collection of 96 terrestrial food webs which tracked the recovery of sixteen riparian food webs after a flood over the course of a year. Often, a space-for-time substitution approach is used: for instance, Layer et al (2010b) studied the structure of 20 freshwater food webs sampled once from 20 sites distributed across a wide pH gradient, and this was subsequently set in the context of long-term change in the single model system of Broadstone Stream over four decades of rising pH (Layer *et al.* 2011). However, no studies of which we are aware have analysed the long-term recovery from perturbation of replicated food webs distributed across wide ecological gradients.

B. Freshwater acidification

Freshwater acidification is usually caused by atmospheric pollution (though there are naturally rather acidic systems), in which strong mineral acids emitted from industrial sources are deposited on the landscape (e.g. Driscoll *et al.* 2001). Where soils and geology have an insufficient supply of base cations to buffer acidity, runoff to streams and lakes become strongly acidic. At a pH of 5.5, alkalinity falls to zero and inorganic aluminium concentration rises to become toxic to many forms of life, including almost all fish (Sutcliffe & Hildrew 1989). Such anthropogenic acidification has profound ecological impacts, including the loss of many acid-sensitive species from all trophic levels (e.g. Dillon *et al.* 1984; Schindler 1988). Evidence for the causes and consequences of acidification and its effect on species assemblages is long-standing and overwhelming (e.g. Likens & Bormann 1974; Schindler 1988; Hildrew & Ormerod 1995) and comes primarily from north-western Europe and much of north and eastern USA and Canada. This ranges from palaeolimnological reconstructions of lake pH (e.g. Battarbee *et al.* 1988), widespread surveys and descriptions (e.g. Townsend, Hildrew & Francis 1983; Henriksen *et al.* 1990), experimental acidification of whole systems (e.g. Hall *et al.* 1980; Webster *et al.* 1992; Findlay *et al.* 1999), and biogeochemical modelling (e.g. Jenkins *et al.* 1990). In the UK, intensive research on the long-term and large-scale ecological consequences of acidification on running waters was concentrated in three main study systems: Llyn Brianne in south-west Wales (Durance & Ormerod 2007), the Ashdown Forest of south-eastern England (Hildrew 2009), and via the nationally covering UK Acid Waters Monitoring Network (UKAWMN), which interdigitated to a limited extent by sharing three sites with the other two (Layer et la 2010).

In the face of such evidence, reductions of polluting emissions were agreed upon in both Europe and North America, which have resulted in dramatically reduced depositions since the 1970s (Stoddard *et al.* 1999; RoTAP 2012). In 1988, the UK Government set up UKAWMN (incorporated within the latterly expanded Upland Waters Monitoring Network – or UWMN, in 2014) to assess the effectiveness of these measures, which came at considerable economic and social costs. The

Network was designed explicitly to detect any recovery in the quality of surface waters at its 23 stream and lake sites, as well as any shifts in their biology and ecology that might be expected to accompany chemical recovery. The network's sites are distributed across acid sensitive (base poor) regions of the UK (Figure 1), mainly in the uplands areas of the north and west where precipitation and wet deposition of acidity tend to be high, although some are in small, acid sensitive areas in the south and east. A few sites are located in the extreme north and west of Scotland and Northern Ireland which were thought sufficiently remote from industrial pollutant sources to have been significantly affected by acid deposition (Patrick *et al.* 1991).

Substantial (though not complete) chemical recovery from acidification has now occurred at most sites that had been acidified at the outset (Monteith *et al.* 2014). This has included large reductions in inorganic aluminium concentrations in the most acidified waters and more widespread but gradual increases in water pH, while Acid Neutralising Capacity (ANC) has increased in proportion to the rate of reduction in acid anions. Ubiquitous increases in dissolved organic carbon concentration also appear to be part of the biogeochemical response, leading to a partial replacement of mineral acidity by organic acidity that has tempered the pH response (Evans *et al.* 2008). Evidence for biological recovery, in terms of the establishment of acid-sensitive assemblages of species, has been much less obvious (Battarbee *et al.* 2014), but most evident in the diatoms of the epilithon and from the colonisation of recovering sites by some species of macrophytes. In the last UWMN data interpretation report (Kernan *et al.* 2010), invertebrate assemblages showed signs of partial recovery at around half of the chemically recovering sites. Only two of those sites, both of which were particularly severely acidified at the onset of monitoring, showed any evidence of recovery of salmonid populations, another indicator of decreasing acidity (Murphy *et al.* 2014; Malcolm *et al*. 2014). Similar, "sluggish" biological recovery has also been reported in other acidified systems elsewhere globally (e.g. Nedbalová *et al.* 2006; Keller *et al.* 2007).

Several hypotheses have been put forward to explain these delays in the anticipated simple reversal of acidification, including dispersal limitations, pollutant legacies and attendant recurring acid episodes (e.g. Kowalik *et al.* 2007), interactions with other stressors (e.g. climate change; Johnson & Angeler 2010), and indirect food-web effects (Yan *et al.* 2003; Ledger & Hildrew, 2005; Monteith *et al.* 2005). These mechanisms are not necessarily mutually exclusive explanations and there are differing levels of support for each. For instance, the role of dispersal is still uncertain for differing biological components, with evidence for and against its role:

Gray & Arnott (2011) suggested it may constrain the recovery of lake zooplankton in Canadian lakes, contrary to previous findings (Keller *et al.* 2002; Yan *et al.* 2004). Evidence for more mobile taxa, such as benthic insects with flying adults, however, suggests that dispersal is usually sufficient to allow rapid recolonisation and thus unlikely to explain delayed biological recovery (Masters *et al.* 2007; Hildrew 2009). The type of waterbody also affects both its rate of chemical recovery and its recolonisation potential: lakes are larger and thus better able to absorb spikes of run-off that can create acid episodes than streams (Evans, Monteith & Harriman 2001) and yet they show similarly limited biological recovery and so acidic episodes alone cannot explain the lag in biological recovery. In addition, these hypotheses may operate additively or even synergistically.

To date, no evidence has been found for differing rates of recovery between streams and lakes. Given the evidence that persistent acidic episodes in stream systems can limit biological recovery (Lepori & Ormerod 2005; Kowalik *et al.* 2007) one might expect stream systems to recover more slowly than lakes, although Monteith et al., 2014 found that the magnitude of acid pulses at UWMN sites had declined at a similar rate to mean acidity. Conversely, the dynamic nature of streams, with a natural regime of frequent flow disturbances, and downstream connectivity to pools of less acid-sensitive species in the lower reaches might render them naturally more resilient (e.g. Hildrew & Giller 1994). Thus, we had no clear *a priori* hypothesis or expectations of the relative rates of recovery in the communities of lakes and streams.

C. The recovery of acidified food webs

The possibility that species interactions within the food web might inhibit ecological recovery requires further testing (Webster *et al.* 1992; Frost, Montz & Kratz 1998). Circumstantial evidence from streams suggests that generalist, non-predatory invertebrates (e.g., stoneflies of the families Leuctridae and Nemouridae), that are acid-tolerant and often dominate the benthos of acid streams, may fill the feeding niche of specialist, acid-sensitive grazers and inhibit their return (Ledger & Hildrew 2005; Layer, Hildrew & Woodward 2013). Further, dynamic modelling found that the reticulate acidified food webs are more robust, suggesting that they might be more inherently stable and thus less prone to (re)invasion (Layer *et al.* 2010b). Finally, the common reliance on space-for-time proxies may miss the transient dynamics and the possible existence of alternate stable states of a system responding to stress. For instance, if the strong effects of pH on food web properties reported in the space-for-time survey of 20 sites by Layer *et al.* (2010b) are not evident in systems undergoing actual changes in pH over time, then the expected mapping of the biota onto the environmental template might not be evident, and its absence would indeed indicate ecological inertia. This has wider implications for biomonitoring science in general, which is underpinned by space-for-time approaches and rarely has access to truly long-term (i.e. multidecadal) high quality biological time series (Friberg *et al.* 2011). Mismatches between temporal and space-for-time data may provide evidence for time lags or hystereses resulting from the system's own internal dynamics, although the potential importance of biogeographic constraints should also not be discounted. On the other hand, if the two data types match perfectly then indirect food web effects – which would otherwise reshape the simple biota-environment relationship - can effectively be discounted.

Repeated assessment of the topology of a large collection of food webs, as they recover from the effects of acidification, is needed to complement previous work, which dealt with single, 'model' systems (e.g. Layer *et al.* 2010a, 2011), or relied on space-for-time substitutions. At present examples of the former case are lacking, but in the latter, Layer *et al.'s* (2010b) suggested the smaller but more interconnected acidified webs had more stable configurations of trophic linkages across a pH gradient of <5 to >8. Linkage density and connectance are both common measures of web complexity, and an abundance of "redundant" interactions can help stabilise the network's structure in the face of perturbations, by preventing the secondary extinctions that arise when consumers are left without resources (Dunne, Williams & Martinez 2002; Thébault & Fontaine 2010). Indeed, linkage density increased with stream pH across the spatial gradient of the 20 sites, which included 10 of the UWMN streams used in the current study.

Mean food chain length (the number of steps between a basal resource and a particular consumer, see Text Box) gives a measure of the trophic height of the web as a whole (Williams & Martinez 2004), which tends to shorten as environmental stress increases and productivity declines (Woodward *et al.* 2005). Food chains are generally assumed to be shorter than six links, and omnivory, which is commonplace in aquatic systems, tends to truncate them further (Pimm 1980; Hildrew, Townsend & Hasham 1985; Lawton 1989; Yodzis 1989; Williams & Martinez 2000). Although long-term data are still scarce, there is some evidence that suggests that increasing pH leads to higher productivity and an overall lengthening of food chains (Gerson, Driscoll & Roy; Grahn, Hultberg & Landner 1974; Woodward *et al.* 2005; Hildrew 2009). For instance, progressively larger and more acid-tolerant predators have (re)invaded Broadstone Stream as pH has risen since the 1970s, culminating in the recent return of the apex predator, the brown trout (*Salmo trutta*) (Layer *et al.* 2011).

In addition to simple food chain metrics, the range of both resources and consumers each species is connected to in the web has important implications for the overall network's dynamical and structural stability, and its ability to respond to or resist environmental change. *Generality* (see Text Box) is a substructural measure of the dietary breadth of a consumer, derived statistically from its number of resources. If a consumer is a specialist (i.e. narrow diet), then it might be more vulnerable to extinction as the loss of only a few species will leave it with insufficient resources. *Vulnerability* (see Text Box) is the converse measure of *Generality*; it is derived from the number of consumers feeding on a particular resource species, and indicates how important that resource is in terms of the consumers it supports. Freshwater predators are commonly gape-limited generalists, so the size and diversity of prey increases with consumer size (Woodward & Hildrew 2002; Woodward *et al.* 2010b). Similarly, the herbivorous consumers in acidified streams are also generalists, feeding on a wide range of detritus and algae (e.g. Ledger & Hildrew 2005; Layer, Hildrew & Woodward 2013). As acidity decreased and acid sensitive, but more specialist, species re-invade, the average generality of consumers (i.e., normalised to the size of the food web) should decrease. The average vulnerability of resources (again normalised to the size of the food web) should increase with decreasing acidity as the consumer guild becomes more speciose (Layer *et al.* 2010b). However, Layer *et al.* (2010b), found that normalised consumer generality and resource vulnerability did not change systematically, and there was no relationship between either the variation (standard deviation) in consumer generality or resource vulnerability and pH, although the sample size was rather small.

Under acidified conditions, generalist primary consumers can partially occupy the niche left by the loss of specialist herbivorous species, potentially creating 'ecological inertia' within the food web by slowing the return of the latter as pH rises (Ledger & Hildrew 2005). The effect of acidity on this redundancy of feeding pathways within the whole food web has not been investigated previously using network-based approaches. As acidified systems tend to be species poor, and dominated by generalist consumer species and few specialists (e.g. Hämäläinen & Huttunen 1996; Ledger & Hildrew 2005), there should be greater trophic redundancy (i.e. species of acid streams should have more similar diets and share more predators than more speciose webs of relative specialists), which could make them resistant to perturbations and hence more robust to the loss of food resources than those at higher pH (Naeem 1998; Solé *et al.* 2003; Peralta *et al.* 2014).

As with other features of the whole network (such as connectance) or parts of the network (e.g. generality) the so-called 'small-world' properties of food webs have also been linked to stability (Watts & Strogatz 1998; Montoya & Solé 2002) and to the rate at which perturbations propagate (Montoya, Pimm & Solé 2006). Essentially, if species are highly connected to the rest of the food web in a 'small-world' network, then perturbations may spread (and dissipate) rapidly, but if there are less well connected (more degrees of separation) this may lead to longer-lived oscillatory dynamics and feedbacks that require a long time to reach equilibrium. Thus, it is not simply the linkage density or strength of connections that are important for determining stability or food-web inertia, but their particular configuration. Even large food webs from circumneutral or higher pH systems can exhibit these properties, with most species being only 1-2 degrees of separation from the rest of the web (Thompson *et al.* 2015). Network efficiency (see Text Box) is a measure of how well connected a network is, as well as the distribution of those connections across a network (Latora & Marchiori 2001), and can enable inferences to be made about the small-world properties of food webs. Although rarely applied to date in food web studies, this metric derived from the wider field of network science could provide new insights into how these small-world aspects of food web topology itself might shape the trajectory of biological recovery (Monteith *et al.* 2005; Layer *et al.* 2010b). The lengthening of food chains associated with the re-invasion of consumers might be expected to increase the overall efficiency of the network, as the wider breadth of diet of new, large, top predators effectively reduces the distance (in terms of number of links) between resources.

D. The Upland Waters Monitoring Network

The data analysed here (from the UWMN) consist of repeated observations on the same 23 sites (Figure 1) over 24 years (1988 – 2012). Spatially, the sites encompass a wide range of pH (from 3.71 to 7.49), and include some that were strongly acidified at the onset of monitoring (e.g. Old Lodge) and others that were circumneutral and have changed little (e.g. Allt na Coire nan Con). The gradual long-term chemical recovery of many of these sites, particularly among those formerly the most acidic, provides a unique and large-scale picture of the chemical drivers and biological responses in the network over both space and time. Characterizing the 'baseline' variation in food web structure in the near absence of changes in environmental stress is crucial for our understanding of how a community recovers from that stressor and, more broadly, for gauging its potential responses to future environmental changes. The food webs compiled from these data provide the replication and statistical power that has previously been insufficient for rigorous analysis food web responses to acidification and chemical recovery.

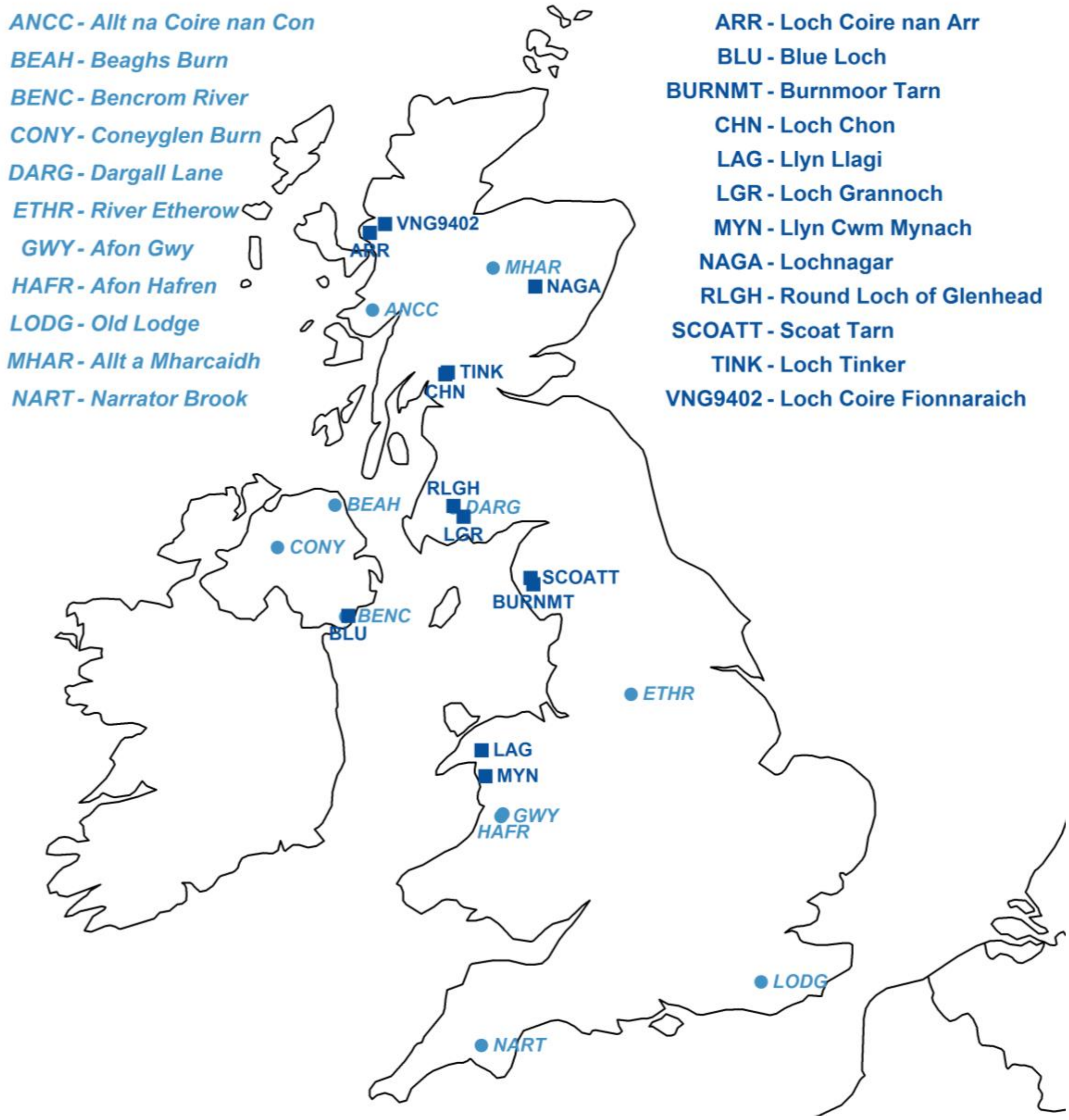


Figure 1. The Upland Waters Monitoring sites, consisting of 11 lakes (dark blue squares) and 12 streams (light blue circles).

We aimed to describe changes in food web structure as water chemistry recovered over three decades of chemical and biological monitoring. Our emphasis was on structural changes in binary networks of the presence/absence of nodes and links, rather than the effects of acidity on the dynamical stability of species populations *per se*. To determine the effect of environmental variables on food web structure, we tested the following hypotheses, using data gathered across all sites and years:

1. When analysed across the entire acidity gradient, food web structure will be directly affected by acidity. Food webs of less-acidified systems should exhibit higher linkage density, food chain length and network efficiency but lower connectance consumer generality and redundancy (Table 1).
2. There will be directional and predictable changes in food web structure at each site through time as acidity decreases (Table 1).
3. If indirect food-web effects arising from biotic interaction are unimportant, changes in network structure through time will match recovery from acidification; i.e. those sites which experience change in their chemistry will also change at a comparable pace in their food web attributes. (Figure 2).

Additionally, in order to investigate if the rates of recovery were different between stream and lake sites, the two ecosystem types were analysed both together and separately.

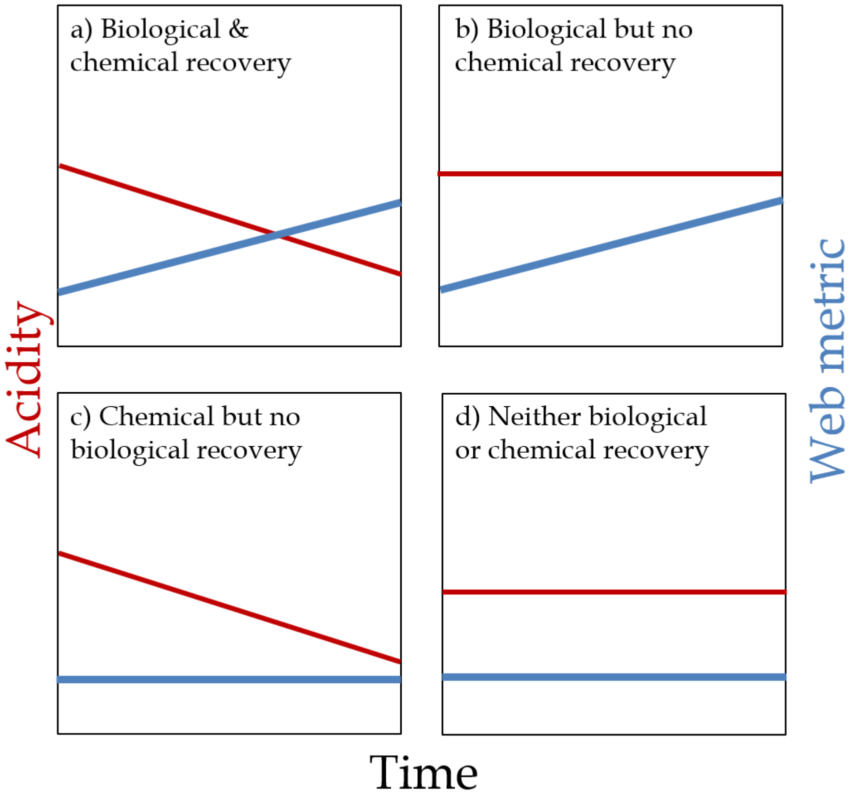


Figure 2. A conceptual figure of the possible outcomes for biological (thick, blue line) and chemical (thin, red line) recovery at the UWMN sites over time.

Table 1. Summary of predicted relationships between acidity and food web structure.

Food web metric	Predicted relationship with decreasing acidity
Connectance	↓
Linkage density	↑
Mean trophic height	↑
Maximum trophic height	↑
Resource vulnerability	↑
Consumer generality	↓
Standard deviation in vulnerability	no change
Standard deviation in generality	no change
Network efficiency	↑
Redundancy	↓

II. METHODS

A. Sites

The UWMN consists of 11 streams and 12 lakes from across the UK (Figure 1): full site descriptions and sampling methodologies are provided in Patrick, Monteith & Jenkins (1995) and Kernan *et al.* (2010). Water chemistry, epilithic diatom, macroinvertebrate and fish sampling began in spring 1988 and has continued annually at most sites up to 2012, except for a few exceptions when access was occasionally restricted (see Kernan *et al.* 2010). The sites are distributed along a latitudinal gradient across the UK, which can be interpreted as a proxy for the degree of acid deposition at the outset: sites at high latitude were exposed to relatively little acid deposition, whilst those sites at lower latitudes were generally more acidified (Patrick *et al.* 1991). One lake site, Loch Coire nan Arr, was affected by damming that increased the water level, so it was replaced in 2001 by the nearby Loch Coire Fionnaraich, which has comparable characteristics (Figure 1).

B. Chemistry

Water samples for chemical analysis were taken in acid-rinsed bottles monthly from streams and quarterly from lakes. A large number of chemical determinands were recorded at each site: for more details see Kernan *et al.* (2010) and Monteith *et al.* (2014). In total 13 variables that are considered key drivers or indicators of acidification (Monteith *et al.* 2014) were used here: pH, alkalinity, H^+, conductivity, nitrate (NO_3), non-labile aluminium, soluble aluminium, labile aluminium, Dissolved Organic Carbon (DOC), sodium (Na+), sulphate (SO4^{2-}), calcium (Ca^{2+}) and chloride (Cl^-). With the exception of pH, we used the annual arithmetic mean of all chemical data as summary statistics for each site. Annual average pH was calculated by first converting pH to H^+ concentration, calculating the annual arithmetic mean, and then converting back to pH.

C. Biota

Benthic diatoms, macroinvertebrates and fish were sampled annually from 1988-2012. Benthic diatoms were sampled according to standard UWMN protocols (Patrick *et al.* 1991), by selecting five cobble-sized stones from the streambed, or from the permanently submerged littoral zone in lakes. Three samples were taken from each site, in streams at the top, middle and bottom of fixed 50m reaches, and in the lakes from three discrete locations on the shoreline (away from inflow or outflow streams). The biofilm on the surface of the stone was scrubbed into a funnel, washed into a plastic vial with stream or lake water, and immediately preserved in Lugol's iodine solution. In the laboratory, samples were prepared by digestion in hydrogen peroxide (H_2O_2) and diluted with distilled water. To enable subsequent examination by light microscopy at 1000x magnification, a sub-sample was pipetted onto a coverslip, dried and mounted onto a slide using Naphrax (Battarbee *et al.* 2001). Three hundred diatom valves from each sample were identified to species to give the diatom species assemblage per site per year.

Macroinvertebrates were sampled according to standard UWMN protocols (Patrick *et al.* 1991), by taking five separate one-minute kick samples with a standard hand net (330μm mesh) from riffle sections of streams and from the dominant littoral habitat of lakes. Macroinvertebrates were subsequently sorted and preserved with 70% Industrial Methylated Spirit. Oligochaeta, Diptera and Bivalvia were identified to class, family and genus, respectively, while all others were identified to species. All taxa were counted and the counts from the five samples summed to represent the macroinvertebrate assemblage per site per year.

Annual electrofishing surveys were conducted according to standard UWMN protocols (Patrick *et al.* 1991). Surveys were undertaken between mid-September and mid-October at each stream site or, for lakes, in the outflow stream immediately downstream. It was assumed that composition of the fish assemblage in lake outflows could serve as a proxy for that of fish in the lake itself. Three 50m reaches, distributed across 500m of the stream or lake outflow, were isolated using stop nets and electrofished. Depletion electrofishing was employed and all salmonids were counted, while the presence of any other fishes was also recorded.

D. Food web construction

Species lists were compiled for each site in each year for which there was complete biological and chemical data, yielding 442 food webs. Binary food webs were constructed using the WebBuilder function in R (Gray *et al.* 2015) and the database of freshwater aquatic trophic interactions contained therein, based on the presence/absence of species (nodes) at each site in each year and the occurrence of a trophic linkage in the feeding link database. This method is based on the assumption that all feeding links between specific pairs of species that have been reported previously would be realized, wherever and whenever both species co-exist at a study site (Hall & Raffaelli 1991; Martinez 1991; Layer *et al.* 2010b; Pocock, Evans & Memmott 2012), although many of the feeding links within that database were, in fact, derived from direct observation from previous UWMN surveys (Ledger & Hildrew 2005; Layer *et al.* 2010b; a, 2011). When species-specific trophic interactions had not previously been described for some rare or understudied taxa (nodes), feeding links were assigned on the basis of taxonomic similarity; for instance, by assuming that different species within the same genus consumed and were consumed by the same species. This method is often used when constructing freshwater food webs as in these systems consumer diets tend to be highly generalist, and determined primarily by the size of their prey (e.g. Layer *et al.* 2010b; a; Layer, Hildrew & Woodward 2013). Food webs built in this manner are structurally comparable to those built solely through analysis of consumers gut contents; for instance, the method has predicted the links of four well documented freshwater food webs with 40-60% accuracy (on a scale from -100% to 100%, Gray *et al.* 2015).

E. Network metrics

Several commonly used metrics were calculated for each food web, including: connectance (C = L/S^2; where L= the number of trophic links, S = the number of species) and linkage density (LD = L/S), mean trophic height (after Levine 1980: defined as 1 plus the mean trophic level of a consumer's resources, averaged across all consumers) and maximum trophic height (defined in the same way, except that the maximum value across all consumers was taken). Mean generality (G; number of resources per consumer, see Text Box) and vulnerability (V; number of consumers per resource, see Text Box) was calculated, as was normalised G and V for each taxon *k*, as:

$$G_k = \frac{1}{L/S}\sum_{i=1}^{S} a_{ik} \tag{1}$$

$$V_k = \frac{1}{L/S}\sum_{j=1}^{S} a_{jk} \tag{2}$$

Where S is the number of nodes and L the number of links in a food web. $a_{ik} = 1$ if taxon k consumes taxon i (otherwise $a_{ik} = 0$), and $a_{jk} = 1$ if taxon k is being consumed by taxon j (otherwise $a_{jk} = 0$). Mean G_k and V_k in any given food web equal 1, making their standard deviations, which give an indication of the variability in G and V respectively across a network, comparable across networks of different size. These metrics were all calculated using the R package cheddar (Hudson *et al.* 2013).

Network efficiency (Latora & Marchiori 2001, see Text Box) describes the 'reachability' of each node by any other node, and is a measure of overall connectivity, and was calculated using the sna R package (Butts 2013) as:

$$E = \frac{1}{S(S-1)} \sum_{i \neq j \in G} \frac{1}{d_{ij}} \quad (3)$$

Where d_{ij} is the shortest path length between node i and j.

The proportional node redundancy (see Text Box) of each network was calculated by grouping nodes into trophic species (i.e. nodes with common resources and consumers) and then calculated as:

$$Redundancy = 1 - \frac{T}{S} \quad (4)$$

Where T is the number of trophic species within the network. Redundancy was calculated using functions from the cheddar package (Hudson *et al.* 2013) in R.

F. Statistical analyses

All statistical analyses were done in R version 3.1.1 (R Core Team 2013), PRIMER-E with PERMANOVA + (2006). To simplify the chemical data, Principal Component Analysis (PCA) was performed on all 13 water chemistry variables across all sites and years, using a resemblance matrix constructed from Euclidean distances. As some of the variables were measured on different scales (i.e. NO_3 vs pH), and to reduce the influence of extremely large or small values, each variable were centred to zero and scaled by their standard deviations (van den Berg *et al.* 2006). Sample scores on the first PC axis (PC1) were extracted for use as a proxy for a general gradient in overall acidity in further analysis.

Effects of acidity on food web structure

For data-visualisation purposes only, as we were unable to fully account for both temporal and spatial pseudoreplication of our data simultaneously using multivariate analysis, principal coordinates analysis was used on all data across all sites and time points. The resemblance matrix of food web metrics was constructed from square root transformed variables, using Bray-Curtis distances. More rigorous statistical inferences were drawn from univariate approaches, in which pseudoreplication was addressed within the variance structure of the relevant model(s), as explained below.

To assess the effect of acidity on food web structure (our first hypothesis), each network metric was regressed against the derived acidity gradient (PC1), and any trend assessed with Generalised Linear Mixed Effects models. Alongside acidity (PC1), site type (lake or stream) was fitted as a fixed effect, and any potential interactions with acidity (PC1) were assessed on the basis of stepwise model simplification and model AIC (Crawley 2013). For each model, site was fitted as having a random effect on the intercept of the model, and year was fitted to have a random effect on the slope and intercept of the model.

Directional change in food web structure

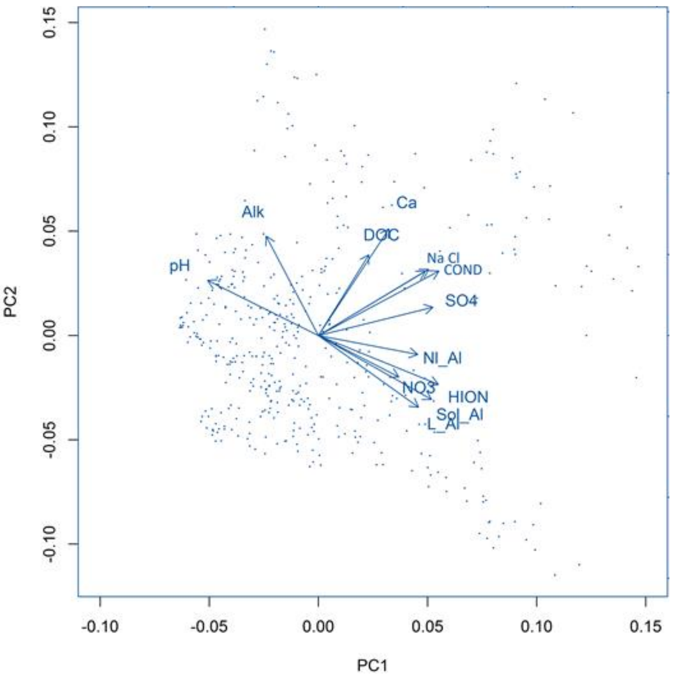
Mann–Kendall trend tests were used to determine if there were significant monotonic trends in the acidity and food web structure over time at each site (our second hypothesis). The acidity gradient (PC1) extracted from the PCA above, and all the network metrics described above, were calculated for each site in each year. These variables were then assessed for monotonic trends over time at each site.

Food web recovery from acidification

To test our third hypothesis we used χ^2 contingency tests to assess the extent to which sites that exhibited clear decreases in acidity also showed evidence of directional change in their food web structure (as in Murphy *et al.* 2014). For acidity (PC1), and for each network metric, we counted the number of sites (out of 23) that exhibited (a) a biological and a chemical trend, (b) a biological but not a chemical trend, (c) a chemical but not a biological trend (i.e., evidence for ecological inertia), and (d) neither a biological nor a chemical trend (Figure 2). The χ^2 test assessed whether the distribution of sites across these four categories was due to chance.

III. RESULTS

The PCA of the chemical variables revealed that the first axis was strongly correlated with pH, H^+ ions and SO_4. It was therefore was used a proxy for the acidity-related stress to which each food web was exposed (Figure *3*), as it encompassed the variation in these pH related chemistry variables. From here on PC1 is called 'acidity' and refers not only to pH, but to the chemical stress associated with low pH.



Variable	Correlation with PC1
pH	0.311
Alk	0.147
HION	-0.336
Cond	-0.337
NO3	-0.225
Nl_Al	-0.278
Sol_Al	-0.317
DOC	-0.141
L_Al	-0.281
Na	-0.309
Ca	-0.198
Cl	-0.299
SO4	-0.321

Figure 3. *PCA ordination of chemical variables. The correlation between each variable and acidity (PC1) is given in the accompanying table. The first axis, PC1, is strongly related to pH, SO_4 and aluminium such that PC1 can be interpreted as an 'acidity gradient'.*

A. Effects of acidity on food web structure

When the food web data were analysed at the regional (UK) scale, and modelled against the acidity gradient extracted from Figure 3, some clear trends in food web structure emerged, although several did not match or even ran counter to our initial hypotheses. Contrary to our expectations (Table 1) connectance, and trophic height were unrelated to the acidity gradient (

Table 2; more acid sites are to the right in Figure 5). As predicted, linkage density increased with decreasing acidity (

Table 2; Figure 5), lake food webs had lower linkage density than stream food webs (Figure 5). Normalised consumer generality and normalised vulnerability did not change (

Table 2; Figure 6), nor did their standard deviations, across the acidity gradient. As predicted, however, network efficiency was lower in more acidified conditions (

Table 2; Figure 7), suggesting that more acidified food webs were connected such that the average path length between nodes was greater than for circumneutral food webs. Node redundancy was highest in more acidified food webs (

Table 2; Figure 7), suggesting that circumneutral food webs had more unique feeding pathways, confirming our prediction. Overall, lake food webs had lower network efficiency and higher node redundancy than streams (

Table 2; Figure 7). Figure 4 indicates more variation in lake food web structure along PCO1, contrasting with our expectation that stream food webs might be more dynamic and variable than lake food webs.

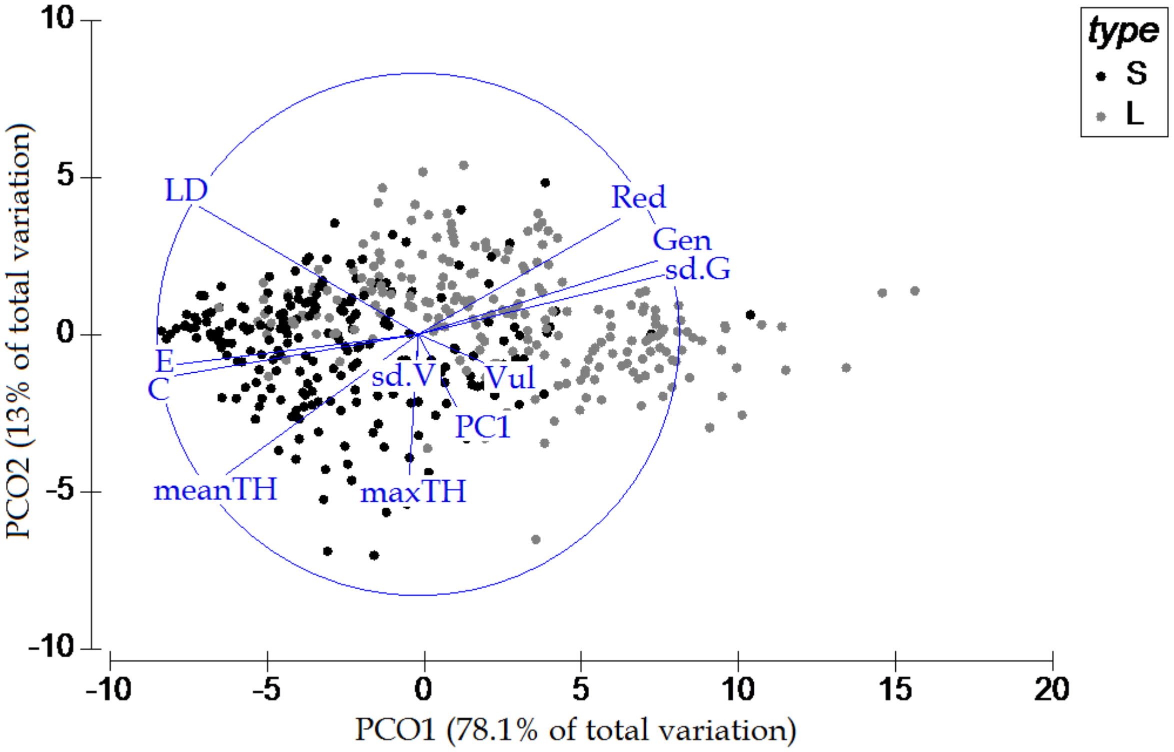


Figure 4. Ordination of food webs (points) based on the resemblance matrix created from food web metrics using Bray-Curtis distances, which was analysed with PERMANOVA. Site type is shown as either black points (streams) or grey points (lakes). This plot allows comparison of food web metrics (blue vectors and names) with the predictor variable PC1 extracted from Figure 3, in multivariate space. Longer vectors indicate a stronger correlation. The blue circle indicates the boundary for a correlation of 1. C = connectance, LD = linkage density, meanTH = mean trophic height, maxTH = maximum trophic height, Vul = resource vulnerability, Gen = consumer generality, E = network efficiency, Red = node redundancy

Table 2. Statistics of fit for the multiple mixed effects models. All models include a random effect of site on the intercept of the linear relationship, and year on the slope of the linear relationship. Bold p-values indicate significance at α = 0.05.

Response variable	Predictor variable	d.f.	F-value	P-value
Connectance	PC1	99	-1.537	0.1270
	type	1	7.991	**<0.0001**
Linkage density	PC1	72	-3.902	**<0.0001**
	type	1	2.686	**0.0130**
Mean trophic height	PC1	76	-0.017	0.9864
	type	1	4.294	**0.0003**
Maximum trophic height	PC1	74	1.407	0.1640
	type	1	-0.068	0.9460
Network efficiency	PC1	101	-2.306	**0.0231**
	type	1	8.288	**<0.0001**
Normalised vulnerability	PC1	54	-0.264	0.7929
	type	1	-2.478	**0.0208**
Normalised generality	PC1	67	1.122	0.2666
	type	1	-6.531	**<0.0001**
sd(Vulnerability)	PC1	115	-0.395	0.6940
	type	1	-0.450	0.6570
sd(Generality)	PC1	112	1.952	0.0534
	type	1	-7.228	<0.0001
Redundancy	PC1	91	3.577	0.0005
	type	1	-5.269	<0.0001

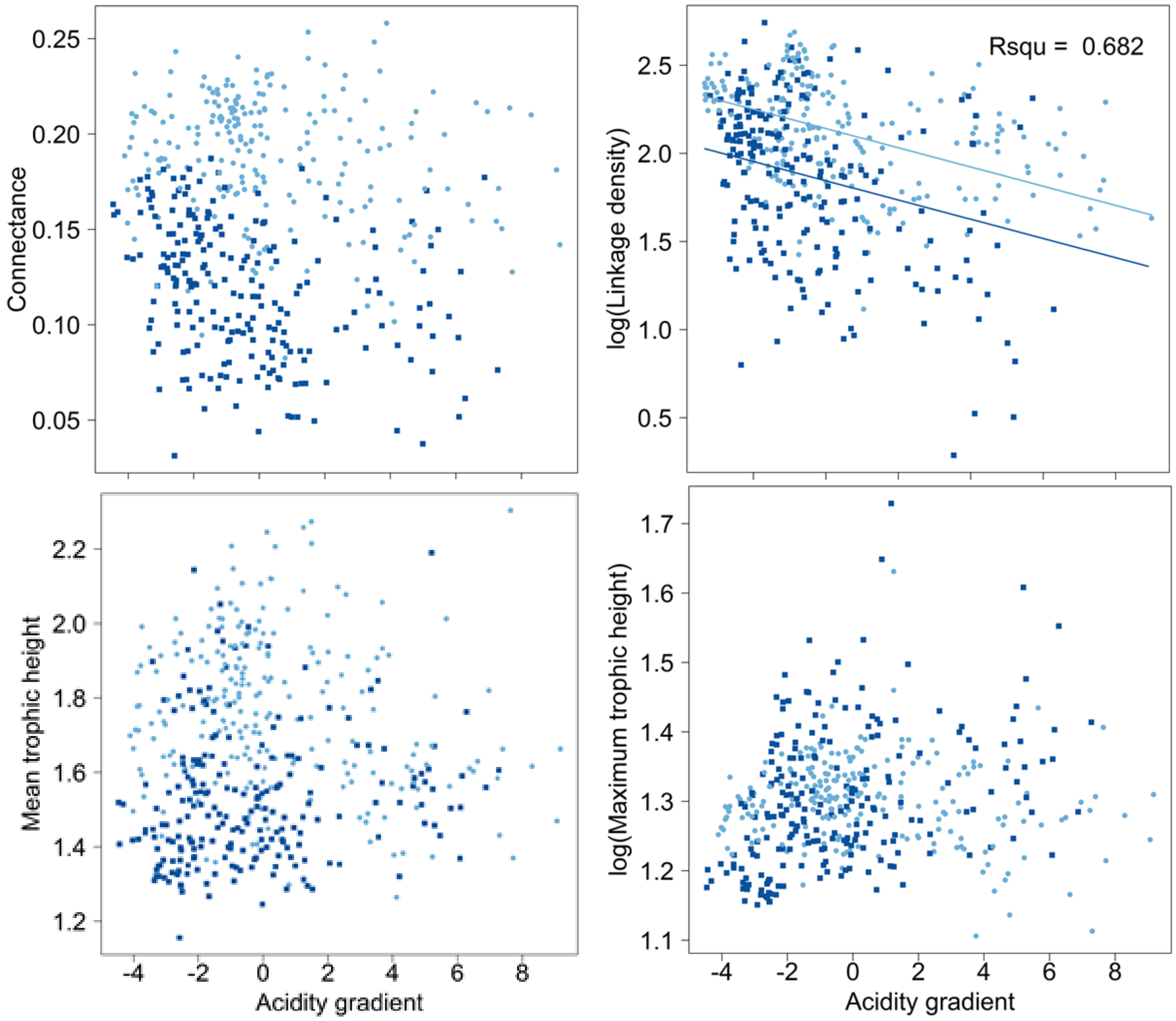


Figure 5. The relationship between connectance, linkage density, mean and maximum trophic height, and environmental stress. The acidity gradient is PC1 extracted from Figure 3, and is strongly related to pH, SO_4 & labile aluminium, with increasing environmental stress (acidity) from left to right, such that the most acidified food webs are to the right of each plot. Lines indicate fitted values from GLMM; where $p < 0.05$, the conditional R^2 as an indication of overall model explanatory power is shown (Johnson 2014). Where site type (lake or stream) was found to be a significant predictor variable, separate lines are given for each site type. Lake food webs are indicated by dark blue squares, while streams are light blue circles.

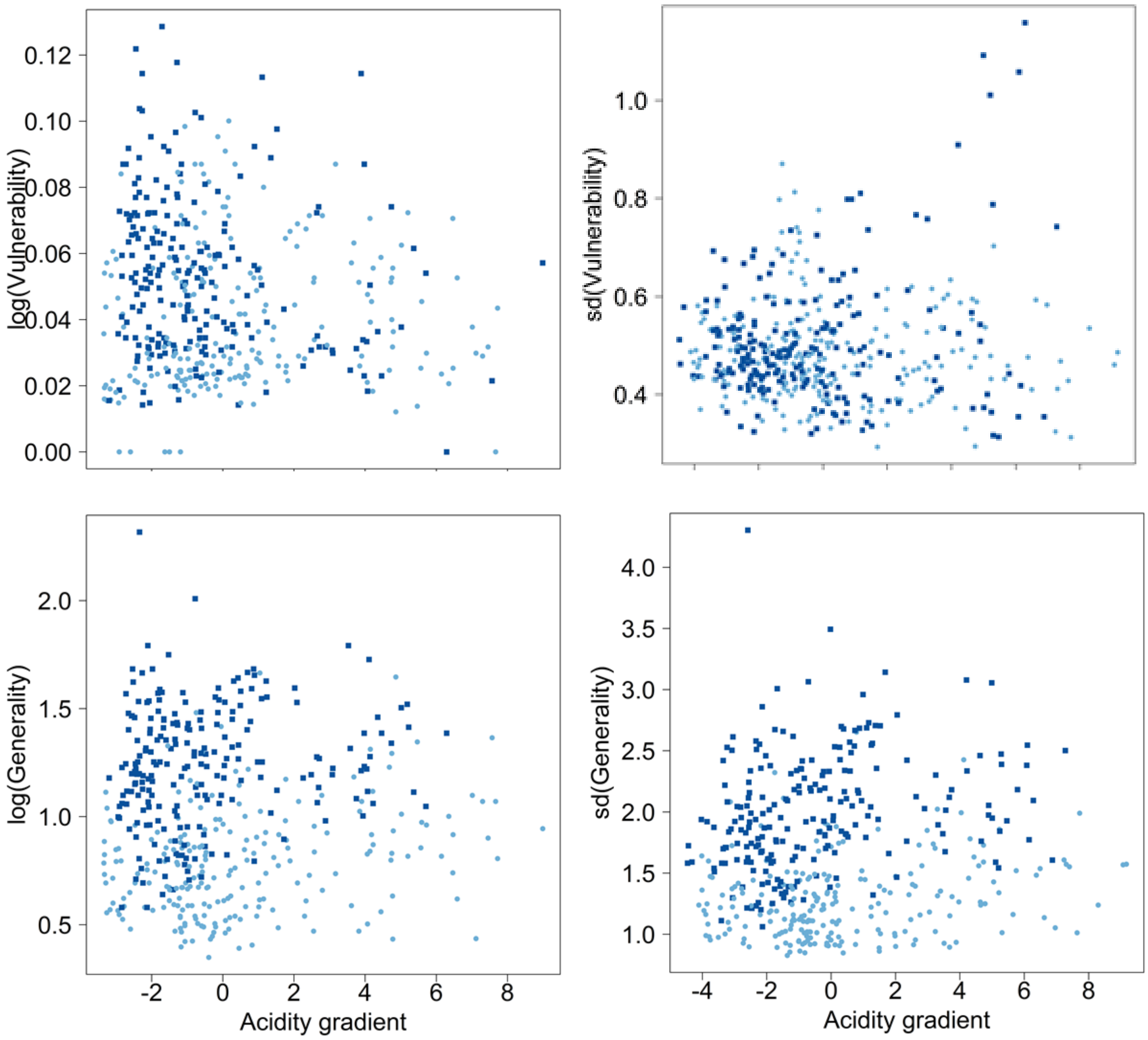


Figure 6. Variation in resource vulnerability and consumer generality (both normalised to species richness) across the stress (acidity) gradient (greater acidity to the right, see Figure 5). Lake food webs are indicated by dark blue squares, while streams are light blue circles.

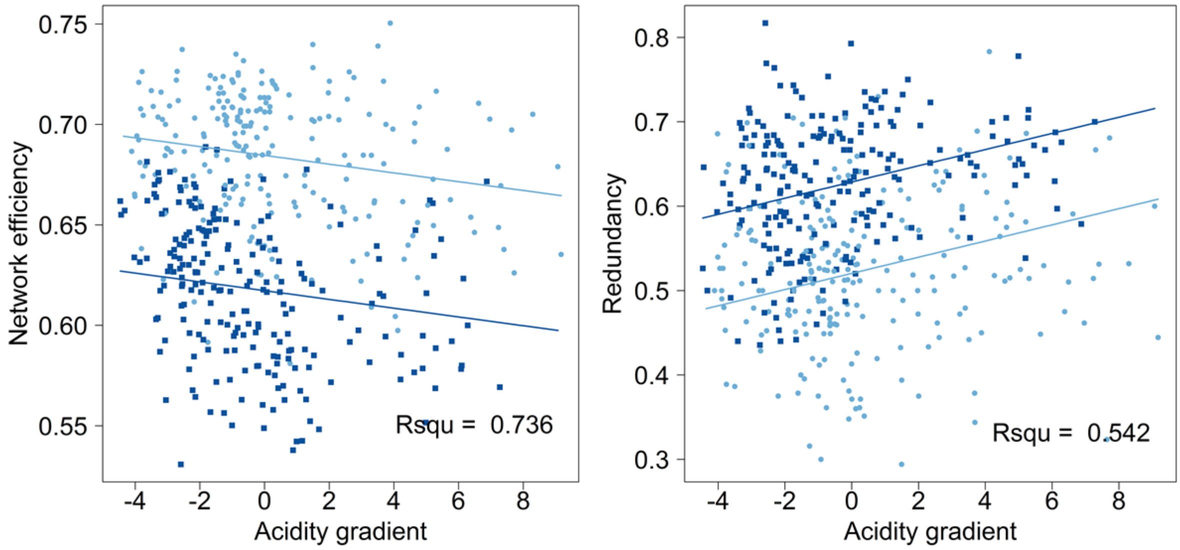


Figure 7. Network efficiency increases with decreasing environmental stress (acidity, greater acidity to the right, see Figure 5), node redundancy decreases with decreasing environmental stress. Lines indicate fitted values from GLMM; where p < 0.05 the conditional R² as an indication of overall model explanatory power is shown (Johnson 2014). Where site type (lake or stream) was found to be a significant predictor variable, separate lines (light blues streams; dark blue lakes) are given for each site type. Lake food webs are indicated by dark blue squares, while streams are light blue circles.

B. Directional change in food web structure

There was overall a clear directional trend in chemical recovery: eighteen of the 23 sites exhibited a monotonic declining trend in their PC1 axis scores (i.e. decreasing acidity) over time (Figure 8;Figure 9). Three of the five sites which showed no trend in their PC1 scores (i.e. no directional change in acidity over time) were located in the north of Scotland, which always experienced less acid deposition and so were not highly acidified at the outset of monitoring (Figure 1)(Patrick, Monteith & Jenkins 1995). This suggests that at least partial chemical recovery from acidification has occurred at most sites at which it was expected (Monteith *et al.* 2014).

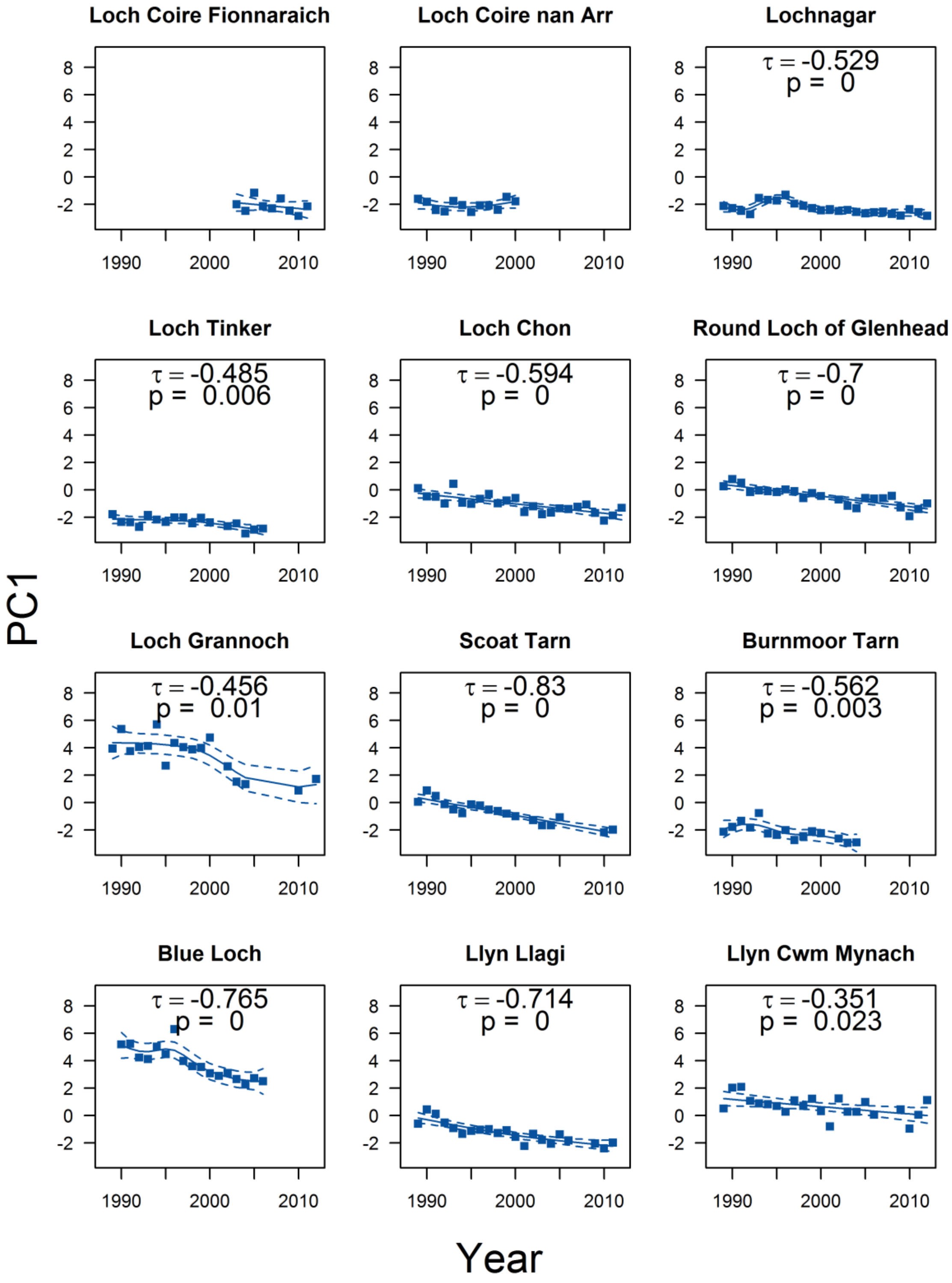


Figure 8. Trends in overall acidity (PC1 extracted from Figure 3) at each of the UWMN lake sites. For those sites showing significant monotonic temporal trends (as determined through Mann–Kendall trend tests, see Methods) the test statistic and associated p-value are shown. Sites are arranged in order of their decreasing latitude, which can be used as a proxy for their initial acidified state, more acidified sites were generally in the south (bottom panels), while the least acidified sites were more northern (top panels).

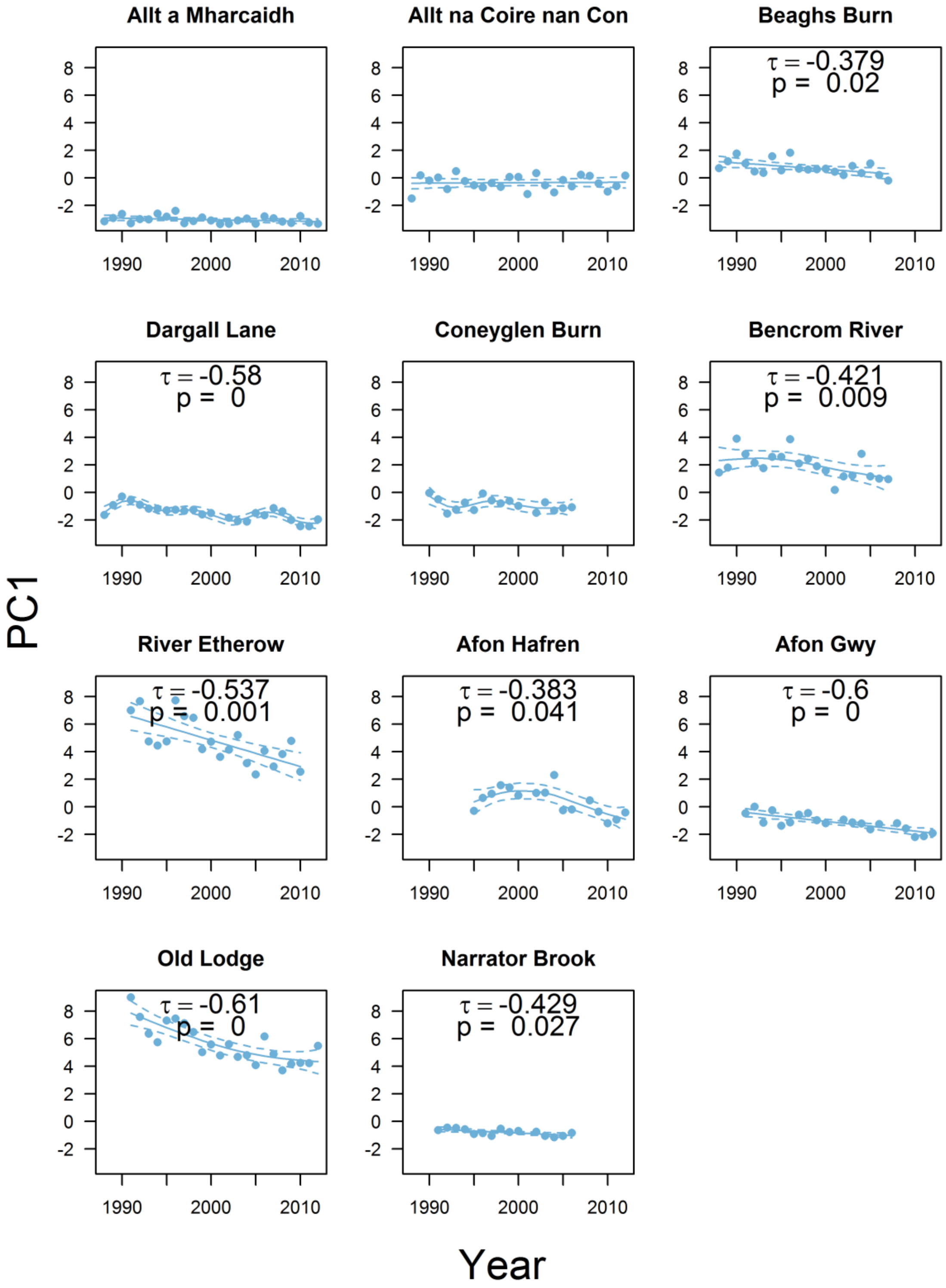


Figure 9. Trends in overall acidity (PC1 extracted from Figure 3) at each of the UWMN stream sites. For those sites showing significant monotonic temporal trends (as determined through Mann–Kendall trend tests, see Methods) the test statistic and associated p-value are shown. Sites are arranged in order of their decreasing latitude, which can be used as a proxy for their initial acidified state, more acidified sites were generally in the south (bottom panels), while the least acidified sites were more northern (top panels).

Directional change in food web structure was also evident across many of the UWMN sites, in line with the chemical trends and with our second main hypothesis, although it was far from ubiquitous. Of the eighteen sites showing chemical recovery, around half also showed significant increasing trends in connectance (9 sites; Table 3; Table 4), linkage density (7 sites; Table 3; Table 4), mean trophic height (8 sites; Table 3; Table 4), resource *Vulnerability* (6 sites; ; Table 3; Table 4), standard deviation in resource *Vulnerability* (5 sites, Figure 10; Figure 11) and network efficiency (7 sites; Table 3; Table 4). Of the eighteen showing chemical recovery, significant declines were evident in consumer *Generality* (10 sites; Table 3; Table 4), redundancy (10 sites; Table 3) and the standard deviation of *Generality* (10 sites; Figure 10; Figure 11). Maximum trophic height increased in one site, decreased in two, and showed no trend in the other 20 (Figure 10; Figure 11). See Appendices for more detailed plots of each trend over time.

There was evidence for a delay in food web recovery after chemical recovery at the UWMN sites; most sites occupied the 'both biological and chemical recovery' or 'chemical but no biological recovery' portions of the conceptual recovery figure (Figure 2) for each of their food web metrics (Table 3; Table 4). Very few sites exhibited change in their food web structure in the absence of directional change in their acidity (PC1); the food webs of Loch Coire Fionnaraich and Allt na Coire nan Con both showed increasing linkage density and resource vulnerability over time, in the absence of a significant temporal trend in acidity (Figure 10, Figure 11). Similarly, the food web of Coneyglen Burn decreased in redundancy over time, despite no significant temporal trend in acidity (Figure 10, Figure 11).

Table 3. The number of the twelve lake sites showing a significant temporal trend in their food web metrics over time, as determined by Mann–Kendall trend tests (see Methods). Chemical recovery here is indicated by a significant temporal trend in acidity (PC1). C = connectance, LD = linkage density, meanTH = mean trophic height, maxTH = maximum trophic height, Vul = resource vulnerability, Gen = consumer generality, E = network efficiency, Red = node redundancy.

Biological & chemical recovery		Biological but no chemical recovery	
C = 6	LD = 2	C = 0	LD = 1
meanTH = 4	maxTH = 0	meanTH = 0	maxTH = 0
Vul = 3	Gen = 6	Vul = 0	Gen = 1
E = 4	Red = 4	E = 0	Red = 0
Chemical but no biological recovery		No biological or chemical recovery	
C = 4	LD = 8	C = 2	LD = 1
meanTH = 6	maxTH = 8	meanTH = 2	maxTH = 2
Vul = 6	Gen = 4	Vul = 2	Gen = 1
E = 6	Red = 6	E = 2	Red = 2

Table 4. The number of the eleven stream sites showing a significant temporal trend (in the direction predicted in hypotheses; recovery) in their food web metrics over time, as determined by Mann–Kendall trend tests (see Methods). Chemical recovery here is indicated by a significant temporal trend in acidity (PC1). C = connectance, LD = linkage density, mean.TH = mean trophic height, max.TH = maximum trophic height, Vul = resource vulnerability, Gen = consumer generality, E = network efficiency, Red = node redundancy.

Biological & chemical recovery		Biological but no chemical recovery	
C = 3	LD = 5	C = 0	LD = 1
meanTH = 4	maxTH = 1	meanTH = 0	maxTH = 0
Vul = 3	Gen = 4	Vul = 1	Gen = 0
E = 3	Red = 6	E = 0	Red = 1
Chemical but no biological recovery		No biological or chemical recovery	
C = 5	LD = 3	C = 3	LD = 2
meanTH = 4	maxTH = 7	meanTH = 3	maxTH = 3
Vul = 5	Gen = 4	Vul = 2	Gen = 3
E = 5	Red = 2	E = 3	Red = 2

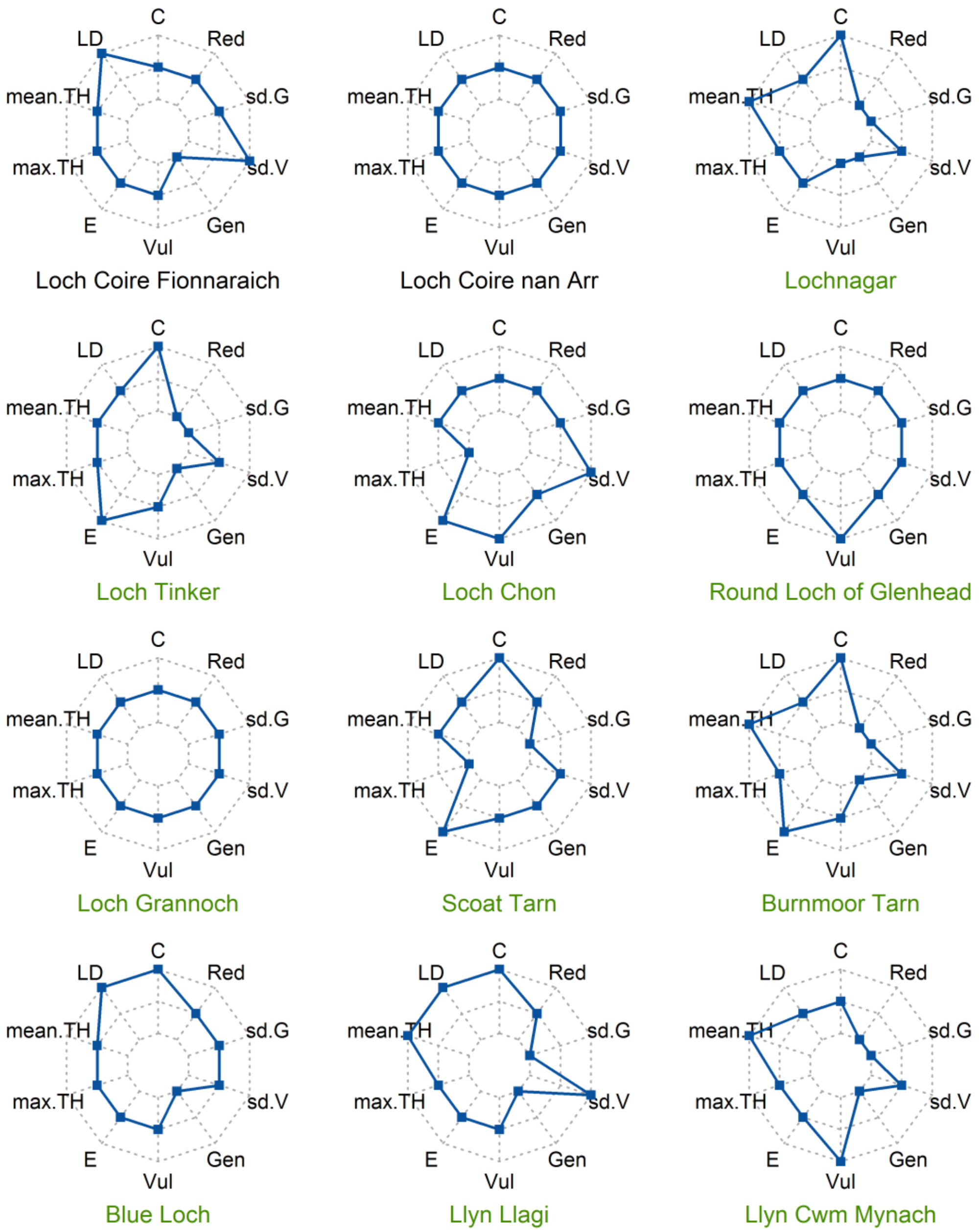


Figure 10. Trends in food web metrics at each of the UWMN lake sites. Sites are arranged in order of their decreasing latitude (top left to bottom right; see Figure 1), which can be used as a proxy for their initial acidified state (generally least acid to the top left, more acidified sites at the bottom). Site names in green indicate a monotonic decreasing trend in acidity over time at that site, site names in black indicate no trend in acidity. Points on the inner ring of each radial plot indicated a negative trend in that variable at that site over time, points on the middle ring indicate no significant trend while points on the outer ring indicate a positive trend. C = connectance, LD = linkage density, mean.TH = mean trophic height, max.TH = maximum trophic height, E = network efficiency, Vul = resource vulnerability, Gen = consumer generality, sd.V = standard deviation in resource vulnerability, sd.G = standard deviation in consumer generality, Red = node redundancy. See Appendices for detailed plots of each trend over time.

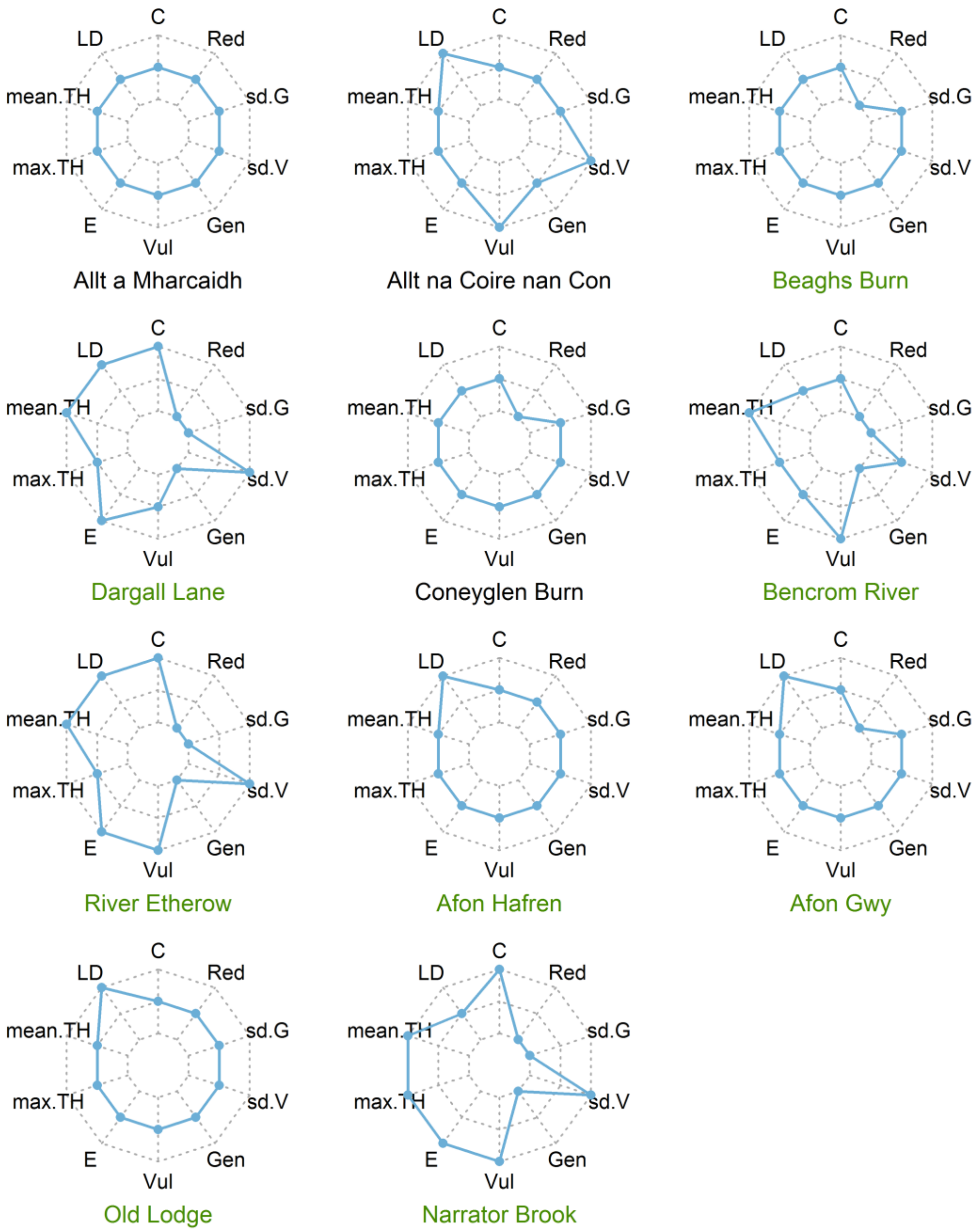


Figure 11. Trends in food web metrics at each of the UWMN stream sites. Sites are arranged in order of their decreasing latitude (top left to bottom right; see Figure 1), which can be used as a proxy for their initial acidified state (generally least acid to the top left, more acidified sites at the bottom). Site names in green indicate a monotonic decreasing trend in acidity over time at that site, site names in black indicate no trend in acidity. Points on the inner ring of each radial plot indicated a negative trend in that variable at that site over time, points on the middle ring indicate no significant trend while points on the outer ring indicate a positive trend. C = connectance, LD = linkage density, mean.TH = mean trophic height, max.TH = maximum trophic height, E = network efficiency, Vul = resource vulnerability, Gen = consumer generality, sd.V = standard deviation in resource vulnerability, sd.G = standard deviation in consumer generality, Red = node redundancy. See Appendices for detailed plots of each trend over time.

C. Food web recovery from acidification

Trends in chemistry over time were not linearly related to shifts in food web structure, or at least have not yet related to the latter, strengthening the evidence for inertia in food web recovery. The χ^2 tests revealed that there was little congruence between those sites exhibiting chemical and biological recovery (Table 5). However, the χ^2 tests did reveal that those sites showing a trend in standard deviation in consumer *Generality* also tended to show a trend in acidity (Table 5). This generally refutes our third hypothesis, and provides more evidence for a lag or inertia in food web recovery; those sites recovering from acidification over time showed little systematic change in their food web structure, suggesting that biological recovery (in terms of food web structure) does not directly track chemical recovery at these sites.

Table 5. Results from the χ^2 contingency test (see main text). Bold p-values indicate significance at $\alpha = 0.05$. See the legend of Table 3 for abbreviations.

	PC1
C	0.113
LD	1
mean.TH	0.123
max.TH	0.574
E	0.146
Vulnerability	1
Generality	1
sd.V	1
sd.G	**0.046**
Redundancy	0.325

IV. Discussion

Our analyses reveals fundamental structural changes occurring in food web structure in response to decreasing acidity over the three decades of the study. These structural changes could have profound implications for the stability of the systems' food webs, and could be hindering biological recovery. Confirming our hypotheses, when analysed at the regional (UK) scale, acidified food webs had lower linkage density, and network efficiency but had more redundancy within their feeding pathways. Contrary to our other hypotheses, we found no effect on connectance, trophic height, nor on resource *Vulnerability* and consumer *Generality* or the standard deviations of both. When analysed at the site scale some further trends in network metrics over time became clearer, but overall these were mixed and often harder to associate with decreasing acidity *per se*. There was strong evidence for a lag in biological recovery, as those sites showing a recovery in both their biology and their chemistry tended to be a nested subset of those that only showed a chemical trend.

A. Food web recovery across the UWMN sites and acidity gradient

The general increasing mean trophic height of food webs over time at each site (see also Figure D Appendices) reflected the reverse of the typical responses to acidification, where species are lost throughout the food web, but top predators such as fish (Henriksen, Fjeld & Hesthagen 1999) and many predatory macroinvertebrates are especially vulnerable (Layer *et al.* 2011). The return of these acid-sensitive species over time causes food chain lengths and the trophic height of the web as a whole to increase (Woodward & Hildrew 2001; Layer et al 2011) – in our dataset all of the sites which experienced this lengthening of food chains were also decreasing in their acidity. However, not all sites with falling acidity also exhibited increases in trophic height, which is again suggestive of food-web inertia. Often site-specific trends were not evident, but were when the data were analysed across the full acidity gradient, also suggesting that other environmental drivers might be modulating the relationship with the food web. For instance DOC, which was closely related to PC2, is known to limit secondary production in lakes (Finstad *et al.* 2014).

Along the derived acidity gradient, normalised *Vulnerability* and *Generality* were unchanged. Over time at individual sites, however, the latter tended to decrease and the former to increase, but across the 23 sites as a whole only seven showed increasing *Vulnerability* and most showed complex and non-linear patterns over time (see Appendices). Decreasing *Generality* and increasing *Vulnerability* with decreasing acidity is consistent with the proposition that specialist consumers and also larger top predators re-colonise communities following chemical recovery (Woodward & Hildrew 2001; Layer, Hildrew & Woodward 2013). This should result in increased *Vulnerability* (more consumers per resource due to greater consumer species richness) and reduced *Generality* (fewer resources per consumer due to increased specialism). The reappearance of acid-sensitive consumers including both invertebrates (such as species of the mayflies *Baetis spp.* and *Caenis spp.*, or the snail *Radix balthica*) and salmonid fish at high pH should lead to both a general elongation and greater compartmentalisation in the web and specialism becomes more prevalent both within and across trophic levels.

The connectivity of the food webs as a whole changed across the derived acidity gradient: network efficiency, which describes how 'reachable' each node is from every other, increased with decreasing acidity. If pockets of species are poorly connected to other species, the average shortest path length between all pairs of nodes will increase. Thus, species within more acidified food webs were less well-connected on average across the whole network. The increased species richness and addition of top predators such as salmonid fish to the system (Woodward & Hildrew 2001) may explain the increased efficiency of these less-acidified food webs. The top predator of these freshwater systems, the brown trout (*Salmo trutta*) is a highly generalist engulfing predator which will consume anything within a given size range of prey. The addition of these (acid-sensitive) generalist interactions between top predators and those macroinvertebrates within its prey-size range may well increase the reachability between those resource nodes, as well as ultimately linking together different feeding pathways (e.g., the allochthonous vs autochthonous resource base of the food web), even though these may be becoming more compartmentalised horizontally among their increasingly specialist primary consumers.

Acidified food webs contained proportionally more redundant feeding pathways than their circumneutral counterparts, the proportion of 'trophic species', nodes feeding on and being fed on by the same species, is larger in the smaller, more acidified food webs. This is consistent with the increase is specialist consumers as acidity decreases. Additionally, acidified food webs tend to have few species and few links (Layer *et al.* 2010b), making the scope for unique feeding pathways small.

Contrasting trends emerged when our data were analysed at the site or regional scale. When our data were analysed at the individual site scale, trends were mixed and were not necessarily always related to decreasing acidity, while clearer trends often emerged from the regional scale analysis. This could arise if communities are highly variable when released from a stressor, and other drivers (e.g. nutrients) that were previously uninfluential start to shape local habitat filtering (e.g. Micheli *et al.* 1999). Additionally, site scale sources of variation, such as potentially powerful contingent site characteristics, might have swamped potential underlying trends in food web structure over time. Indeed, site identity was a necessary variable in our models that encompassed a range of site specific variables, such as latitude. Additionally, weather conditions were uncontrolled and extreme events close to the small sampling widow for each site, has caused some sites to lose and regain their significant trends in biotic recovery over time (Monteith & Evans 1998, 2005; Kernan *et al.* 2010). Additionally, the portion of the acidity gradient that each site is exposed to is small relative to that of the whole dataset.

B. The recovery of freshwater food webs from acidification

Although some clear responses were evident, the food web metrics used here might not be the most appropriate for detecting recovery from acidification. There were considerable inter-site differences in food web structure, but not all were sensitive to changes in acidity and there was still considerable unexplained variation in the models. It seems likely that, as our understanding grows, more sensitive measures of food web structure will emerge, perhaps through analysis of substructure rather than 'whole network' properties, and that these might be better at capturing ecological responses to environmental change.

That acidified ecosystems might exhibit 'ecological inertia' has increasingly been suggested as mechanism to explain the delay in biological recovery (Lundberg, Ranta & Kaitala 2000; Ledger & Hildrew 2005; Kernan *et al.* 2010; Layer *et al.* 2010b). Various lines of evidence are increasingly suggesting that acidified food webs are dynamically stable and resistant to re-colonisation by acid-sensitive species, even as chemical conditions start to improve. Townsend *et al.* (1987) measured the persistence of 27 stream invertebrate communities across a pH gradient, and found that those communities from the most acidified sites were indeed the most persistent, although data on species interactions and network structure were not available at the time. Later, Layer *et al.* (2010b) used dynamic modelling to determine the robustness of stream food webs to species extinctions, and found that food webs from more acidified conditions were more robust, but the long-term temporal data were not available to test this prediction empirically. Here we provide the largest scale evidence to test these ideas, which broadly support the general notion that redundancy is an important component of stability that could confer robustness on the system. In ecosystems redundancy can increase the reliability of process rates and buffer the effects of species loss (Naeem 1998; Peralta *et al.* 2014): we found that food webs from acidified waters had higher redundancy, suggesting that they might be more robust, and might therefore provide more stable (albeit often slower) process rates (Naeem 1998; Peralta *et al.* 2014). As acidity decreases in fresh waters, decomposition of leaf-litter, which fuels much of the food web, does indeed accelerate (Jenkins, Woodward & Hildrew 2013), although the extent to which species richness modulates this relationship is still largely unknown (but see Jonsson *et al.* 2002). Additionally, we found that more acidified food webs had lower global efficiency, which is associated with reduced small-world properties. Ecological networks that are small worlds are often relatively stable (Solé & Montoya 2001; Dunne, Williams & Martinez 2002), as they offer many alternative pathways of interaction. These apparently contrasting responses to different dimensions of stability warrant further investigation to reveal if acidified food webs are indeed more (or less) stable in some regards and not others (e.g. Donohue *et al.* 2013).

C. Caveats and future directions

The use of inferred feeding links in food web studies has been criticised on the basis that they might over estimate diet breadth, and fail to detect behavioural differences between sites (Hall & Raffaelli 1997; Raffaelli 2007), yet to build complete food webs *de novo* from replicated natural systems is simply logistically unfeasible, so a trade-off between replication and realism is inevitable. The use of 'summary' food webs, which include the full complement of known possible tropic interactions can still be a useful tool for understanding community structure, especially as in freshwaters most species are highly generalist and their diets are largely size-driven and consistent among systems when presented with the same potential prey species (Woodward *et al.* 2010b; Layer, Hildrew & Woodward 2013; Gray *et al.* 2015). Indeed, given the nature of building summary food webs, that they tend to overestimate interactions between species, they are more likely to be insensitive to environmental change rather than reveal erroneous trends (i.e. it is more likely that the structure of summary food webs is conserved given that any changes will be entirely driven by changes in species composition rather than feeding behaviour). Hence, we contend that the trends revealed here are broadly realistic, and warrant further examination, especially as the feeding links described in many of our webs had been observed in the same system, albeit only for a snapshot of the full set of sampling occasions. Future work could involve a more formal validation of randomly selected portions of the network via direct analysis of gut contents (as in Woodward, Speirs & Hildrew 2005; Layer *et al.* 2010b) and also the application of new molecular approaches that could potentially capture a more complete picture of the entire food web with a fraction of the current effort required (Gray *et al.* 2014).

Another potential limitation to the food webs produced here is that they do not include the full freshwater community, in particular the meiofauna and microfauna (e.g. Schmid-Araya *et al.* 2002) and true apex predators such as the European Dipper (*Cinclus cinclus)* or Otter (*Lutra lutra)*. Top predators can have varying effects on food web structure in these systems (Woodward & Hildrew 2001; Layer *et al.* 2011), and so their exclusion may be omitting an important source of variation in this data. However, this was unavoidable in this study, as in almost all other food webs described to date, because the presence of these cryptic or very rare species has not been systematically recorded. Additionally, although the fish assemblage of the lakes were sampled from the lake outflows, all these low-productivity upland sites are typically dominated by brown trout (*Salmo trutta*) and the occasional European eel (*Anguilla anguilla*) in both the running and standing waters across the acidity gradient: of the 434 sampling occasions on which fish were present at a site, brown trout were always present, reflecting its dominance in these systems. The next most common species was the European eel (*Anguilla anguilla*), was found on 136 occasions and all other species (*Esox lucius, Gasterosteus aculeatus, Lampetra spp., Phoxinus phoxinus* and *Salmo salar*) were found on <60 sampling occasions.

D. Conclusion

It is clear from this study that both spatial and temporal scales are important considerations when assessing food web responses to environmental change in real time (Chave 2013). When our data were analysed at the individual site scale, trends were mixed and were not necessarily always related to decreasing acidity. When the data were analysed at the regional (UK) scale, some clear and significant trends emerged, highlighting the need for large, replicated collections of food webs as well as the need for caution when extrapolating from small collections or individual food webs. Identifying the effects of individual chemical drivers was often challenging given the range of potential drivers in a nationwide dataset that also spans several decades.

To the best of our knowledge this is the largest collection of food webs that span both large temporal and spatial gradients: the next largest set of empirical food webs from natural systems of which we are aware is less than half the size (the 170 soil webs described by Mulder *et al.* 2011) and the remainder are far smaller still, with most studies being conducted on unreplicated singletons (Ings et al 2009). Our study is thus one of the first to address macroecological questions relating to the structure of food webs across time and a broad environmental gradient in a (relatively) standardised manner. Our analysis reveals fundamental structural changes occurring in the food webs as they respond to changes in acidity, these structural changes could have profound implications for the stability of the system, and may be limiting biological recovery. It would be instructive to investigate further the stability of these food webs, in order to explore more fully whether intrinsic inertia is indeed limiting their recovery, and how that might possibly be manipulated to accelerate the rate of recovery.

V. Acknowledgements

This paper is a contribution to Imperial College's Grand Challenges in Ecosystems and the Environment initiative. CG was supported by a Queen Mary University of London studentship and the Freshwater Biology Association. The UK UWMN is supported by the UK Department for Environment Food and Rural Affairs (DEFRA), NERC through the Centre for Ecology & Hydrology (CEH), the Department of the Environment (Northern Ireland), the Environment Agency (EA), the Forestry Commission (FC), Natural Resources Wales (NRW), the Scottish Environmental Protection Agency (SEPA), Scottish Natural Heritage (SNH) and the Welsh Government, the Scottish Government through Marine Scotland Science Pitlochry, Queen Mary University of London and ENSIS Ltd. at the Environmental Change Research Centre, University College London.

VI. References

Battarbee, R.W., Anderson, N.J., Appleby, P.G., Flower, R.J., Fritz, S.C., Haworth, E.Y., Higgitt, S., Jones, V.J., Kreiser, A., Munro, M.A.R., Natkanski, J., Oldfield, F., Patrick, S.T., Richardson, N.G., Rippey, B. & Stevenson, A.C.L.E. (1988) *Lake Acidification in the United Kingdom 1800–1986: Evidence from Analysis of Lake Sediments*. ENSIS Ltd, London.

Battarbee, R.W., Jones, V.J., Flower, R.J., Cameron, N.G., Bennion, H., Carvalho, L. & Juggins, S. (2001) Diatoms. *Tracking Environmental Change Using Lake Sediments. Volume 3: Terrestrial, Algal and Siliceous Indicators* pp. 151–202. Kluwer Academic Publishers, Dordrecht, The Netherlands.

Battarbee, R.W., Simpson, G.L., Shilland, E.M., Flower, R.J., Kreiser, A., Yang, H. & Clarke, G. (2014) Recovery of UK lakes from acidification: An assessment using combined palaeoecological and contemporary diatom assemblage data. *Ecological Indicators*, **37, Part B**, 365–380.

van den Berg, R. a, Hoefsloot, H.C.J., Westerhuis, J. a, Smilde, A.K. & van der Werf, M.J. (2006) Centering, scaling, and transformations: improving the biological information content of metabolomics data. *BMC genomics*, **7**, 142.

Briand, F. & Cohen, J.E. (1984) Community food webs have scale-invariant structure. *Nature*, **307**, 264–267.

Butts, C. (2013) sna: Tools for Social Network Analysis. R package version 2.3-2. http://CRAN.R-project.org/package=sna.

Chave, J. (2013) The problem of pattern and scale in ecology: What have we learned in 20 years? *Ecology Letters*, **16**, 4–16.

Cohen, J.E. & Briand, F. (1984) Trophic links of community food webs. *Proceedings of the National Academy of Sciences of the United States of America*, **81**, 4105–4109.

Crawley, M.J. (2013) *The R Book*, second.

Dillon, P.J., Yan, N.D., Harvey, H.H. & Schindler, D.W. (1984) Acidic deposition: effects on aquatic ecosystems. *CRC Critical reviews in Environmental Control*, **13**, 167–194.

Donohue, I., Petchey, O.L., Montoya, J.M., Jackson, A.L., Mcnally, L., Viana, M., Healy, K., Lurgi, M., O'Connor, N.E. & Emmerson, M.C. (2013) On the dimensionality of ecological stability. *Ecology Letters*, **16**, 421–9.

Driscoll, C.T., Lawrence, G.B., Bulger, A.J., Butler, T.J., Cronan, C.S., Eagar, C., Lambert, K.F., Likens, G.E., Stoddard, J.L. & Weathers, K.C. (2001) Acidic Deposition in the Northeastern United States: Sources and Inputs, Ecosystem Effects, and Management Strategies. *BioScience*, **51**, 180.

Dunne, J.A., Williams, R.J. & Martinez, N.D. (2002) Food-web structure and network theory: The role of connectance and size. *Proceedings of the National Academy of Sciences of the United States of America*, **99**, 12917–12922.

Durance, I. & Ormerod, S.J. (2007) Climate change effects on upland stream macroinvertebrates over a 25-year period. *Global Change Biology*, **13**, 942–957.

Evans, C.D., Monteith, D.T. & Harriman, R. (2001) Long-term variability in the deposition of marine ions at west coast sites in the UK Acid Waters Monitoring Network: Impacts on surface water chemistry and significance for trend determination. *Science of the Total Environment*, **265**, 115–129.

Evans, C.D., Monteith, D.T., Reynolds, B. & Clark, J.M. (2008) Buffering of recovery from acidification by organic acids. *Science of the Total Environment*, **404**, 316–325.

Feld, C.K., Birk, S., Bradley, D.C., Hering, D., Kail, J., Marzin, A., Melcher, A., Nemitz, D., Pedersen, M.L., Pletterbauer, F., Pont, D., Verdonschot, P.F.M. & Friberg, N. (2011) From Natural to Degraded Rivers and Back Again: A Test of Restoration Ecology Theory and Practice. *Advances in Ecological Research*, **44**, 119–209.

Findlay, D., Kasian, S., Turner, M. & Stainton, M. (1999) Responses of phytoplankton and epilithon during acidification and early recovery of a lake. *Freshwater Biology*, **42**, 159–175.

Finstad, A.G., Helland, I.P., Ugedal, O., Hesthagen, T. & Hessen, D.O. (2014) Unimodal response of fish yield to dissolved organic carbon. *Ecology Letters*, **17**, 36–43.

Friberg, N., Bonada, N., Bradley, D.C., Dunbar, M.J., Edwards, F.K., Grey, J., Hayes, R.B., Hildrew, A.G., Lamouroux, N., Trimmer, M. & Woodward, G. (2011) Biomonitoring of Human Impacts in Freshwater Ecosystems: The Good, the Bad and the Ugly. *Advances in Ecological Research*, **44**, 1–68.

Frost, T.M., Montz, P.K. & Kratz, T.K. (1998) Zooplankton community responses during recovery from acidification in Little Rock Lake, Wisconsin. *Restoration Ecology*, **6**, 336–342.

Gerson, J.R., Driscoll, C.T. & Roy, K.M. Patterns of nutrient dynamics in Adirondack lakes recovering from acid deposition. *Ecological Applications*.

Grahn, O., Hultberg, H. & Landner, L. (1974) Oligotrophication: a self-accelerating process in lakes subjected to excessive supply of acid substances. *Ambio*, **3**, 93–94.

Gray, D.K. & Arnott, S.E. (2011) Does dispersal limitation impact the recovery of zooplankton communities damaged by a regional stressor? *Ecological Applications*, **21**, 1241–1256.

Gray, C., Baird, D.J., Baumgartner, S., Jacob, U., Jenkins, G.B., O'Gorman, E.J., Lu, X., Ma, A., Pocock, M.J.O., Schuwirth, N., Thompson, M. & Woodward, G. (2014) Ecological networks: the missing links in biomonitoring science. *Journal of Applied Ecology*, **51**, 1444–1449.

Gray, C., Figueroa, D.H., Hudson, L.N., Ma, A., Perkins, D. & Woodward, G. (2015) Joining the dots: an automated method for constructing food webs from compendia of published interactions. *Food Webs*, **5**, 11–20.

Hall, R.J., Likens, G.E., Fiance, S.B. & Hendrey, G.R. (1980) Experimental Acidification of a Stream in the Hubbard Brook Experimental Forest, New Hampshire. *Ecology*, **61**, 976–989.

Hall, S.J. & Raffaelli, D. (1991) Food-Web Patterns: Lessons from a Species-Rich Web. *Journal of Animal Ecology*, **60**, 823–841.

Hall, S.J. & Raffaelli, D.G. (1997) Food web patterns: What do we really know? *Multitrophic Interactions* (eds A.C. Gange), & V.K. Brown), pp. 395–417. Blackwells Scientific Publications, Oxford, UK.

Hämäläinen, H. & Huttunen, P. (1996) Inferring the minimum pH of streams from macroinvertebrates using weighted averaging regression and calibration. *Freshwater Biology*, **36**, 697–709.

Henriksen, A., Fjeld, E. & Hesthagen, T. (1999) Critical load exceedance and damage to fish populations. *Ambio*, **28**, 583–586.

Henriksen, A., Lien, L., Traan, T.S., Rosseland, B.O. & Sevaldrud, I.S. (1990) The 1000-lake survey in Norway 1986. *The Surface Waters Acidification Programme* (ed B.J. Mason), pp. 199–212. Cambridge University Press, Cambridge, UK.

Hildrew, A.G. (2009) Sustained Research on Stream Communities: A Model System and The Comparative Approach. *Advances in Ecological Research*, **41**, 175–312.

Hildrew, A. & Ormerod, S. (1995) Acidification: causes, consequences and solutions. *The ecological basis for river management* (eds D.M. Harper), & A.J.D. Ferguson), pp. 147–160. J. Wiley.

Hildrew, A.G., Townsend, C.R. & Hasham, A. (1985) The predatory chironomidae of an iron-rich stream - feeding ecology and food web structure. *Ecological Entomology*, **10**, 403–413.

Hudson, L.N., Emerson, R., Jenkins, G.B., Layer, K., Ledger, M.E., Pichler, D.E., Thompson, M.S.A., O'Gorman, E.J., Woodward, G. & Reuman, D.C. (2013) Cheddar: analysis and visualisation of ecological communities in R. *Methods in Ecology and Evolution*, **4**, 99–104.

Ings, T.C., Montoya, J.M., Bascompte, J., Bluthgen, N., Brown, L., Dormann, C.F., Edwards, F., Figueroa, D., Jacob, U., Jones, J.I., Lauridsen, R.B., Ledger, M.E., Lewis, H.M., Olesen, J.M., van Veen, F.J.F., Warren, P.H. & Woodward, G. (2009) Ecological networks - beyond food webs. *Journal of Animal Ecology*, **78**, 253–269.

Jenkins, A., Whitehead, P.G., Cosby, B.J. & Birks, H.J.B. (1990) Modelling long-term acidification: A comparison with diatom reconstructions and the implications for reversibility. *Philosophical Transactions of the Royal Society B: Biological Sciences*, **327**, 435–440.

Jenkins, G.B., Woodward, G. & Hildrew, A.G. (2013) Long-term amelioration of acidity accelerates decomposition in headwater streams. *Global Change Biology*, **19**, 1100–1106.

Johnson, P.C.D. (2014) Extension of Nakagawa & Schielzeth's R 2 GLMM to random slopes models. *Methods in Ecology and Evolution*, **5**, 944–946.

Johnson, R.K. & Angeler, D.G. (2010) Tracing recovery under changing climate: response of phytoplankton and invertebrate assemblages to decreased acidification. *Journal of the North American Benthological Society*, **29**, 1472–1490.

Jonsson, M., Dangles, O., Malmqvist, B. & Guérold, F. (2002) Simulating species loss following perturbation: assessing the effects on process rates. *Proceedings. Biological sciences / The Royal Society*, **269**, 1047–52.

Keller, W., Yan, N.D., Gunn, J.M. & Heneberry, J. (2007) Recovery of Acidified Lakes: Lessons From Sudbury, Ontario, Canada. *Acid Rain - Deposition and Recovery* p. pp 317–322.

Keller, W., Yan, N.D., Somers, K.M. & Heneberry, J.H. (2002) Crustacean zooplankton communities in lakes recovering from acidification. *Canadian Journal of Fisheries and Aquatic Sciences*, **59**, 726–735.

Kernan, M., Battarbee, R.W., Curtis, C., Monteith, D.T. & Shillands, E.M. (2010) *Recovery of Lakes and Streams in the UK from Acid Rain. The United Kingdom Acid Waters Monitoring Network 20 Year Interpretative Report*. Report to the Department for Environment, Food and Rural Affairs.

Kowalik, R.A., Cooper, D.M., Evans, C.D. & Ormerod, S.J. (2007) Acidic episodes retard the biological recovery of upland British streams from chronic acidification. *Global Change Biology*, **13**, 2439–2452.

Latora, V. & Marchiori, M. (2001) Efficient Behavior of Small-World Networks. *Physical Review Letters*, **87**, 198701.

Lawton, J.H. (1989) Food webs. *Ecological concepts* (ed J.M. Cherrett), pp. 43–78. Blackwell Scientific, Oxford, UK.

Layer, K., Hildrew, A.G., Jenkins, G.B., Riede, J.O., Rossiter, S.J., Townsend, C.R. & Woodward, G. (2011) Long-Term Dynamics of a Well-Characterised Food Web: Four Decades of Acidification and Recovery in the Broadstone Stream Model System (ed G Woodward). *Advances in Ecological Research*, **44**, 69–117.

Layer, K., Hildrew, A., Monteith, D. & Woodward, G. (2010a) Long-term variation in the littoral food web of an acidified mountain lake. *Global Change Biology*, **16**, 3133–3143.

Layer, K., Hildrew, A.G. & Woodward, G. (2013) Grazing and detritivory in 20 stream food webs across a broad pH gradient. *Oecologia*, **171**, 459–471.

Layer, K., Riede, J.O., Hildrew, A.G. & Woodward, G. (2010b) Food Web Structure and Stability in 20 Streams Across a Wide pH Gradient. *Advances in Ecological Research*, **42**, 265–299.

Ledger, M.E., Brown, L.E., Edwards, F.K., Milner, A.M. & Woodward, G. (2012) Drought alters the structure and functioning of complex food webs. *Nature Climate Change*, **3**, 223–227.

Ledger, M.E. & Hildrew, A.G. (2005) The ecology of acidification and recovery: changes in herbivore-algal food web linkages across a stream pH gradient. *Environmental Pollution*, **137**, 103–118.

Lepori, F. & Ormerod, S.J. (2005) Effects of spring acid episodes on macroinvertebrates revealed by population data and in situ toxicity tests. *Freshwater Biology*, **50**, 1568–1577.

Levine, S. (1980) Several measures of trophic structure applicable to complex food webs. *Journal of Theoretical Biology*, **83**, 195–207.

Likens, G.E. & Bormann, F.H. (1974) Acid rain: A serious regional environmental problem. *Science*, **184**, 1176–1179.

Lundberg, P., Ranta, E. & Kaitala, V. (2000) Species loss leads to community closure. *Ecology Letters*, **3**, 465–468.

Martinez, N.D. (1991) Artifacts or attributes? Effects of resolution on the Little Rock Lake food web. *Ecological Monographs*, **61**, 367–392.

Masters, Z.O.E., Peteresen, I., Hildrew, A.G. & Ormerod, S.J. (2007) Insect dispersal does not limit the biological recovery of streams from acidification. *Aquatic Conservation: Marine and Freshwater Ecosystems*, **17**, 375–383.

McLaughlin, Ó.B., Emmerson, M.C. & O'Gorman, E.J. (2013) *Habitat Isolation Reduces the Temporal Stability of Island Ecosystems in the Face of Flood Disturbance*, 1st ed. Elsevier Ltd.

Micheli, F., Cottingham, Kathryn L., Bascompte, J., Bjørnstad, O.N., Eckert, G.L., Fischer, J.M., Keitt, T.H., Kendall, B.E., Klug, J.L. & Rusak, J.A. (1999) The Dual Nature of Community Variability. *Oikos*, **85**, 161–169.

Monteith, D.T. & Evans, C.D. (1998) *United Kingdom Acid Waters Monitoring Network 10 Year Report. Analysis and Interpretation of Results, April 1988 - March 1998. Report to the Department for Environment, Food and Rural Affairs (Contract EPG 1/3/160).*

Monteith, D.T. & Evans, C.D. (2005) The United Kingdom Acid Waters Monitoring Network: a review of the first 15 years and introduction to the special issue. *Environmental Pollution*, **137**, 3–13.

Monteith, D.T., Evans, C.D., Henrys, P.A., Simpson, G.L. & Malcolm, I.A. (2014) Trends in the hydrochemistry of acid-sensitive surface waters in the UK 1988–2008. *Ecological Indicators*, **37**, 287–303.

Monteith, D.T., Hildrew, A.G., Flower, R.J., Raven, P.J., Beaumont, W.R.B., Collen, P., Kreiser, A.M., Shilland, E.M. & Winterbottom, J.H. (2005) Biological responses to the chemical recovery of acidified fresh waters in the UK.

Environmental Pollution, **137**, 83–101.

Montoya, J.M., Pimm, S.L. & Solé, R. V. (2006) Ecological networks and their fragility. *Nature*, **442**, 259–264.

Montoya, J.M. & Solé, R. V. (2002) Small world patterns in food webs. *Journal of Theoretical Biology*, **214**, 405–412.

Mulder, C., Boit, A., Bonkowski, M., De Ruiter, P.C., Mancinelli, G., Van der Heijden, M.G.A., Van Wijnen, H.J., Vonk, J.A. & Rutgers, M. (2011) A Belowground Perspective on Dutch Agroecosystems: How Soil Organisms Interact to Support Ecosystem Services. *Advances in Ecological Research*, **44**, 277–357.

Murphy, J.F., Winterbottom, J.H., Orton, S., Simpson, G.L., Shilland, E.M. & Hildrew, A.G. (2014) Evidence of recovery from acidification in the macroinvertebrate assemblages of UK fresh waters: A 20-year time series. *Ecological Indicators*, **37**, 330–340.

Naeem, S. (1998) Species redundancy and ecosystem reliability. *Conservation Biology*, **12**, 39–45.

Nedbalová, L., Vrba, J., Fott, J., Kohout, L., Kopáček, J., Macek, M. & Soldán, T. (2006) Biological recovery of the Bohemian Forest lakes from acidification. *Biologia*, **61**, 453–465.

O'Gorman, E.J. & Emmerson, M.C. (2010) *Manipulating Interaction Strengths and the Consequences for Trivariate Patterns in a Marine Food Web*, 1st ed. Elsevier Ltd.

O'Neill, R. V. (1998) Recovery in complex ecosystems. *Journal of Aquatic Ecosystem Stress and Recovery*, **6**, 181–187.

Patrick, S., Monteith, D.T. & Jenkins, A. (Eds.). (1995) *UK Acid Waters Monitoring Network: The First Five Years. Analysis and Interpretation of Results, April 1988 - March 1993*. London.

Patrick, S., Waters, D., Juggins, S. & Jenkins, A. (1991) *The United Kingdom Acid Waters Monitoring Network: Site Descriptions and Methodology Report. Report to the Department of the Environment and Department of the Environment Northern Ireland*. London.

Peralta, G., Frost, C.M., Rand, T.A., Didham, R.K. & Tylianakis, J.M. (2014) Complementarity and redundancy of interactions enhance attack rates and spatial stability in host-parasitoid food webs. *Ecology*, **95**, 1888–1896.

Pimm, S.L. (1980) Properties of Food Webs. *Ecology*, **61**, 219–255.

Pimm, S.L., Russell, G.J., Gittleman, J.L. & Brooks, T.M. (1995) The Future of Biodiversity. *Science*, **269**, 347–350.

Pocock, M.J.O., Evans, D.M. & Memmott, J. (2012) The Robustness and Restoration of a Network of Ecological Networks. *Science*, **335**, 973–977.

R Core Team. (2013) R: A language and environment for statistical computing.

Raffaelli, D. (2007) Food Webs, Body Size and the Curse of the Latin Binomial. *From Energetics to Ecosystems: The Dynamics and Structure of Ecological Systems* (eds N. Rooney),, K.S. McCann), & D.L.G. Noakes), pp. 53–64. Springer Netherlands.

RoTAP. (2012) *Review of Transboundary Air Pollution. Acidification, Eutrophication, Ground Level Ozone and Heavy Metals in the UK. Contract Report to the Dept of the Environment, Food and Rural Affairs*.

Scheffer, M. & Carpenter, S.R. (2003) Catastrophic regime shifts in ecosystems: linking theory to observation. *Trends in Ecology and Evolution*, **18**, 648–656.

Schindler, D.W. (1988) Effects of Acid rain on freshwater ecosystems. *Science*, **239**, 149–157.

Schmid-Araya, J.M., Hildrew, a. G., Robertson, a., Schmid, P.E. & Winterbottom, J. (2002) The importance of meiofauna in food webs: Evidence from an acid stream. *Ecology*, **83**, 1271–1285.

Schoener, W. & Schoenerz, T.W. (1989) Food Webs From the Small To the Large. *Ecology*, **70**, 1559–1589.

Smith, B.D. & Zeder, M.A. (2013) The onset of the Anthropocene. *Anthropocene*, **4**, 8–13.

Solé, R. V., Ferrer-Cancho, R., Montoya, J.M. & Valverde, S. (2003) Selection, tinkering, and emergence in complex networks. *Complexity*, **8**, 20–33.

Solé, R. V. & Montoya, J.M. (2001) Complexity and fragility in ecological networks. *Proceedings of the Royal Society B: Biological Sciences*, **268**, 2039–2045.

Steffen, W., Grinevald, J., Crutzen, P. & McNeill, J. (2011) The Anthropocene: conceptual and historical perspectives. *Philosophical Transactions of the Royal Society A*, **369**, 842–867.

Stoddard, J.L., Jeffries, D.S., Lukewille, A., Clair, T.A., Dillon, P.J., Driscoll, C.T., Forsius, M., Johannessen, M., Kahl, J.S., Kellogg, J.H., Kemp, A., Mannio, J., Monteith, D.T., Murdoch, P.S., Patrick, S., Rebsdorf, A., Skjelkvale, B.L., Stainton, M.P., Traaen, T., van Dam, H., Webster, K.E., Wieting, J. & Wilander, A. (1999) Regional trends in aquatic recovery from acidification in North America and Europe. *Nature*, **401**, 575–578.

Sutcliffe, D.W. & Hildrew, A.G. (1989) Invertebrate communities in acid streams. *Acid Toxicity and Aquatic Animals* (eds R. Morris),, E.W. Taylor),, D.J.A. Brown), & J.A. Brown), pp. 13–29. Seminar Series of the Society for Experimental Biology, Cambridge University Press.

Sutherland, W.J., Broad, S., Caine, J., Clout, M., Dicks, L. V., Doran, H., Entwistle, A.C., Fleishman, E., Gibbons, D.W., Keim, B., LeAnstey, B., Lickorish, F.A., Markillie, P., Monk, K.A., Mortimer, D., Ockendon, N., Pearce-Higgins, J.W., Peck, L.S., Pretty, J., Rockström, J., Spalding, M.D., Tonneijck, F.H., Wintle, B.C., Wright, K.E., William J. Sutherland, Steven Broad, Jacqueline Caine, Mick Clout, Lynn V. Dicks, Helen Doran, Abigail C. Entwistle, Erica Fleishman, David W.

Gibbons, Brandon Keim, Becky LeAnstey, Fiona A. Lickorish, Paul Markillie, Kathryn A. Monk, Diana Mortimer, Nancy Ockendon, James W. Pearce-Higgins, Lloyd S. Peck, Jules Pretty, Johan Rockström, Mark D. Spalding, Femke H. Tonneijck, Bonnie C. Wintle & Katherine E. Wright. (2016) A Horizon Scan of Global Conservation Issues for 2016. *Trends in Ecology and Evolution*, **31**, 44–53.

Thébault, E. & Fontaine, C. (2010) Stability of Ecological Communities and the Architecture of Mutualistic and Trophic Networks. *Science*, **329**, 853–856.

Thompson, M.S.A., Bankier, C., Bell, T., Dumbrell, A.J., Gray, C., Ledger, M.E., Lehman, K., McKew, B.A., Sayer, C.D., Shelley, F., Trimmer, M., Warren, S.L. & Woodward, G. (2015) Gene-to-ecosystem impacts of a catastrophic pesticide spill: testing a multilevel bioassessment approach in a river ecosystem. *Freshwater Biology*.

Thompson, R.M., Brose, U., Dunne, J.A., Hall, R.O., Hladyz, S., Kitching, R.L., Martinez, N.D., Rantala, H., Romanuk, T.N., Stouffer, D.B. & Tylianakis, J.M. (2012) Food webs: reconciling the structure and function of biodiversity. *Trends in Ecology and Evolution*, **27**, 689–697.

Townsend, C.R., Hildrew, A.G. & Francis, J. (1983) Community structure in some southern English streams: the influence of physicochemical factors. *Freshwater Biology*, **13**, 521–544.

Townsend, C.R., Hildrew, A.G. & Schofield, K. (1987) Persistence of Stream Invertebrate Communities in Relation to Environmental Variability. *Journal of Animal Ecology*, **56**, 597–613.

Tylianakis, J.M., Tscharntke, T. & Lewis, O.T. (2007) Habitat modification alters the structure of tropical host-parasitoid food webs. *Nature*, **445**, 202–205.

Warren, P.H. (1994) Making connections in food webs. *Trends in Ecology & Evolution*, **9**, 136–141.

Watts, D.J. & Strogatz, S.H. (1998) Collective dynamics of ‘small-world’ networks. *Nature*, **393**, 440–2.

Webster, K.E., Frost, T.M., Watras, C.J., Swenson, W.A., Gonzalez, M. & Garrison, P.J. (1992) Complex biological responses to the experimental acidification of Little Rock Lake, Wisconsin, USA. *Environmental Pollution*, **78**, 73–78.

Williams, R.J. & Martinez, N.D. (2000) Simple rules yield complex food webs. *Nature*, **404**, 180–183.

Williams, R.J. & Martinez, N.D. (2004) Limits to trophic levels and omnivory in complex food webs: theory and data. *The American Naturalist*, **163**, 458–68.

Woodward, G., Benstead, J.P., Beveridge, O.S., Blanchard, J., Brey, T., Brown, L.E., Cross, W.F., Friberg, N., Ings, T.C., Jacob, U., Jennings, S., Ledger, M.E., Milner, A.M., Montoya, J.M., O’Gorman, E.J., Olesen, J.M., Petchey, O.L., Pichler, D.E., Reuman, D.C., Thompson, M.S.A., Van Veen, F.J.F. & Yvon-Durocher, G. (2010a) Ecological Networks in a Changing Climate. *Advances in Ecological Research*, **42**, 71–138.

Woodward, G., Blanchard, J., Lauridsen, R.B., Edwards, F.K., Jones, J.I., Figueroa, D., Warren, P.H. & Petchey, O.L. (2010b) Individual-Based Food Webs: Species Identity, Body Size and Sampling Effects. *Advances in Ecological Research*, **43**, 211–266.

Woodward, G., Brown, L.E., Edwards, F.K., Hudson, L.N., Milner, A.M., Reuman, D.C. & Ledger, M.E. (2012) Climate change impacts in multispecies systems: drought alters food web size structure in a field experiment. *Philosophical Transactions of the Royal Society B: Biological Sciences*, **367**, 2990–2997.

Woodward, G. & Hildrew, A.G. (2001) Invasion of a stream food web by a new top predator. *Journal of Animal Ecology*, **70**, 273–288.

Woodward, G. & Hildrew, A.G. (2002) Body-size determinants of niche overlap and intraguild predation within a complex food web. *Journal of Animal Ecology*, **71**, 1063–1074.

Woodward, G., Speirs, D.C. & Hildrew, A.G. (2005) Quantification and resolution of a complex, size-structured food web. *Advances in Ecological Research*, **36**, 85–135.

Woodward, G., Thompson, R., Townsend, C.R. & Hildrew, A.G. (2005) Pattern and process in food webs: evidence from running waters. *Aquatic Food Webs: An Ecosystem Approach* (eds A. Belgrano),, U.M. Scharler),, J. Dunne), & R.E. Ulanowicz), pp. 51–66. Oxford University Press, Oxford, UK.

Yan, N.D., Girard, R., Heneberry, J.H., Keller, W.B., Gunn, J.M. & Dillon, P.J. (2004) Recovery of copepod, but not cladoceran, zooplankton from severe and chronic effects of multiple stressors. *Ecology Letters*, **7**, 452–460.

Yan, N.D., Leung, B., Keller, W., Arnott, S.E., Gunn, J.M. & Raddum, G.G. (2003) Developing conceptual frameworks for the recovery of aquatic biota from acidification. *Ambio*, **32**, 165–169.

Yodzis, P. (1989) *Introduction to Theoretical Ecology*. Harper & Row, Cambridge, UK.

VII. Text Box

Definitions of food web metrics used in this study

Food Web metric	**Definition**
Connectance (C)	Number of links (L) / Number of species $(S)^2$. The proportion of potential trophic links that do occur (Warren 1994).
Linkage density	L/S. Number of links per taxon. A measure of average diet specialisation across the food web (Tylianakis, Tscharntke & Lewis 2007).
Generality (G)	The mean number of prey per consumer (Schoener & Schoenerz 1989).
Vulnerability (V)	The mean number of consumers per prey (Schoener & Schoenerz 1989).
Mean food chain length	Average number of links found in a food chain across a food web (Levine 1980; Williams & Martinez 2000).
Maximum food chain length	The maximum number of links found in any food chain in a food web (Levine 1980; Williams & Martinez 2000).
Efficiency	How well connected a network is, as well as the distribution of those connections across a network. High efficiency indicates that the species of a food web are all closely connected to one another (Latora & Marchiori 2001)
Redundancy	The trophic similarity among species within a web, high redundancy indicates that many of the species in a food webs are the same resources and consumers; many of the feeding pathways are the same (Briand & Cohen 1984; Cohen & Briand 1984).

II. Appendices

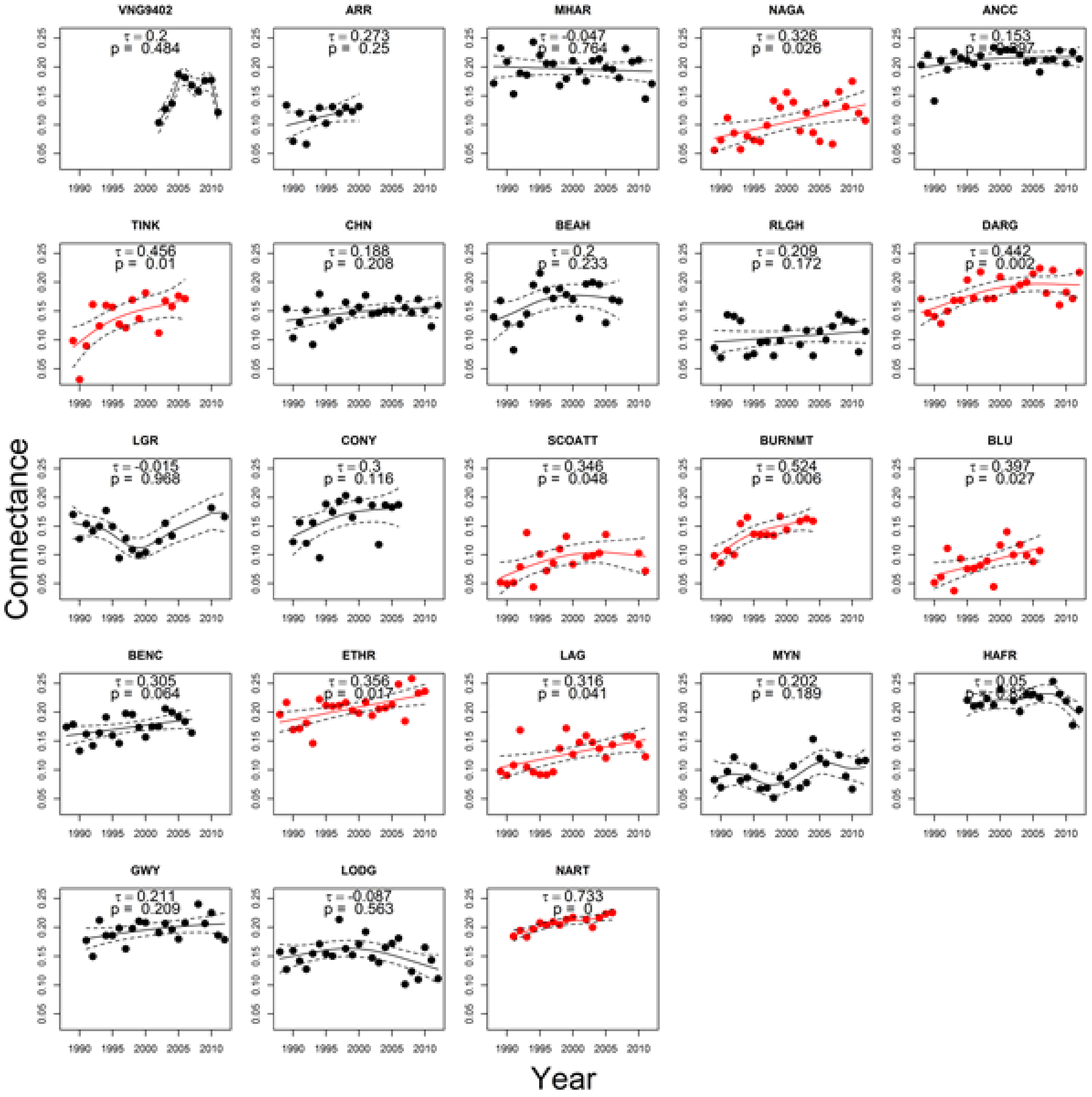


Figure A. Trends in connectance at each of the UWMN sites. Sites are arranged in order of their decreasing latitude, which can be used as a proxy for their initial acidified state, more acidified sites were generally in the south (bottom of plot), while the least acidified sites were more northern (top of plot). See Figure 1 for site name abbreviations.

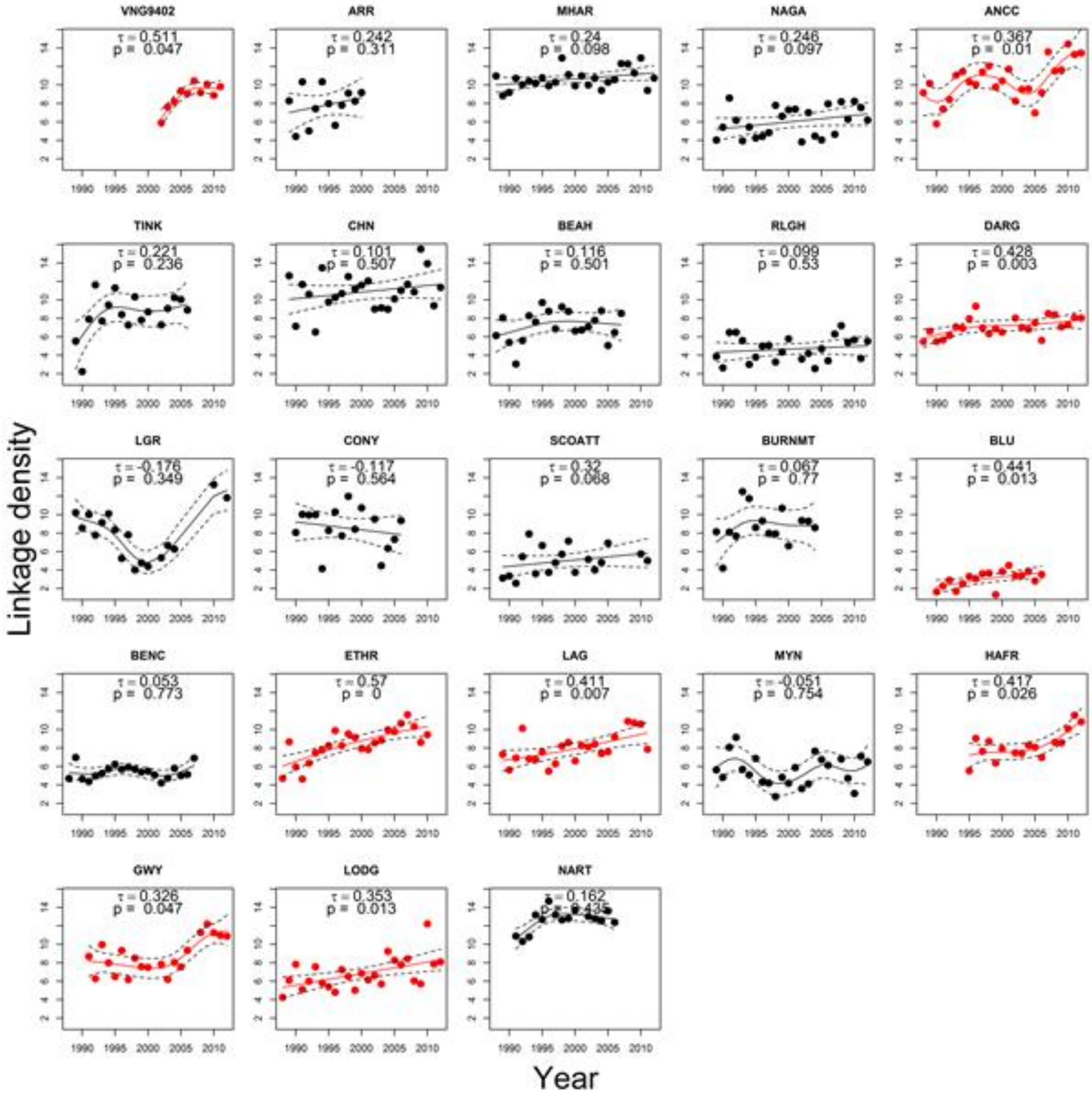


Figure B. Trends in linkage density at each of the UWMN sites. Site ordering is explained in the legend of Figure A. See Figure 1 for site name abbreviations.

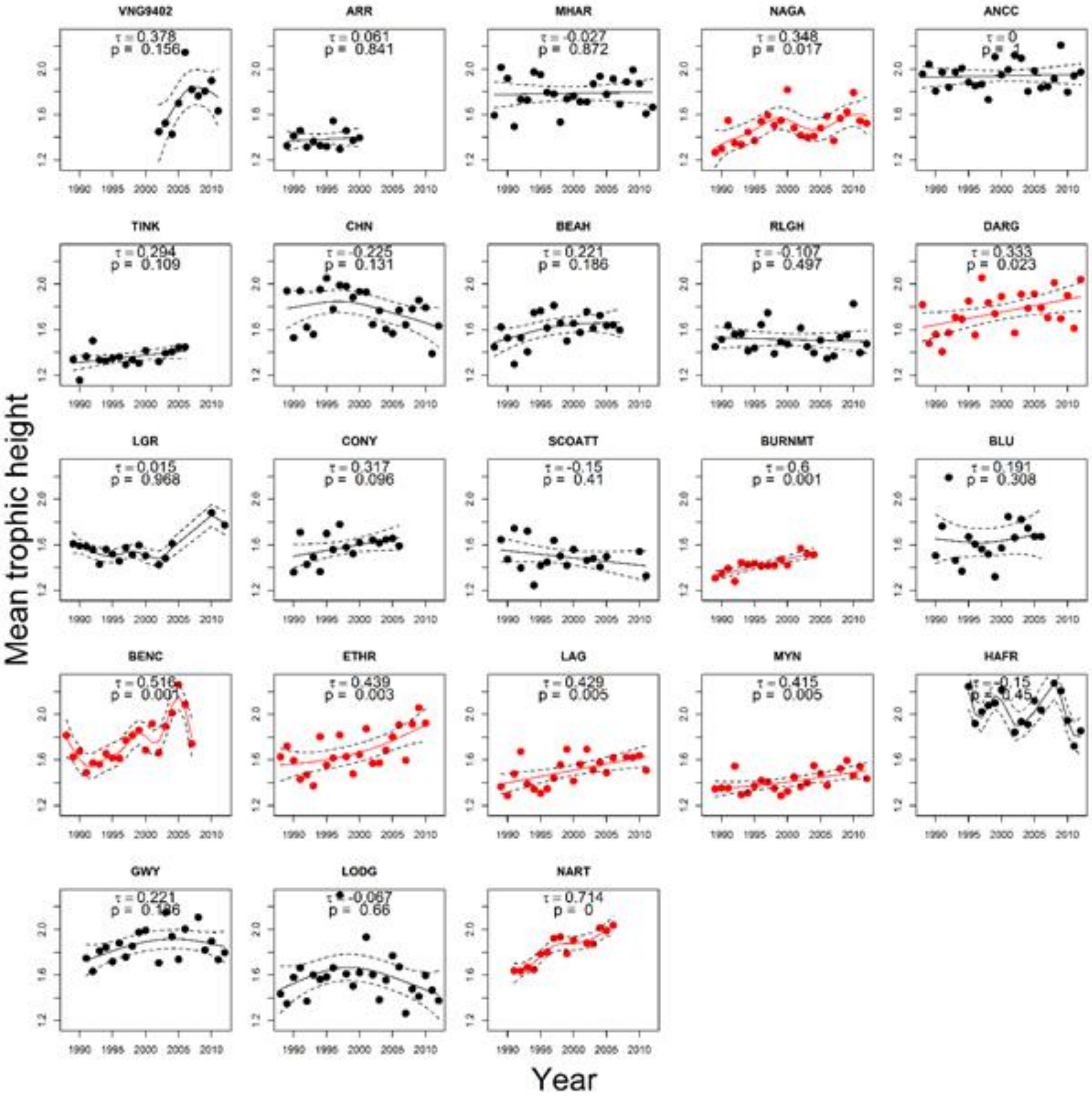


Figure C. Trends in mean trophic height at each of the UWMN sites. Site ordering is explained in the legend of Figure A. See Figure 1 for site name abbreviations.

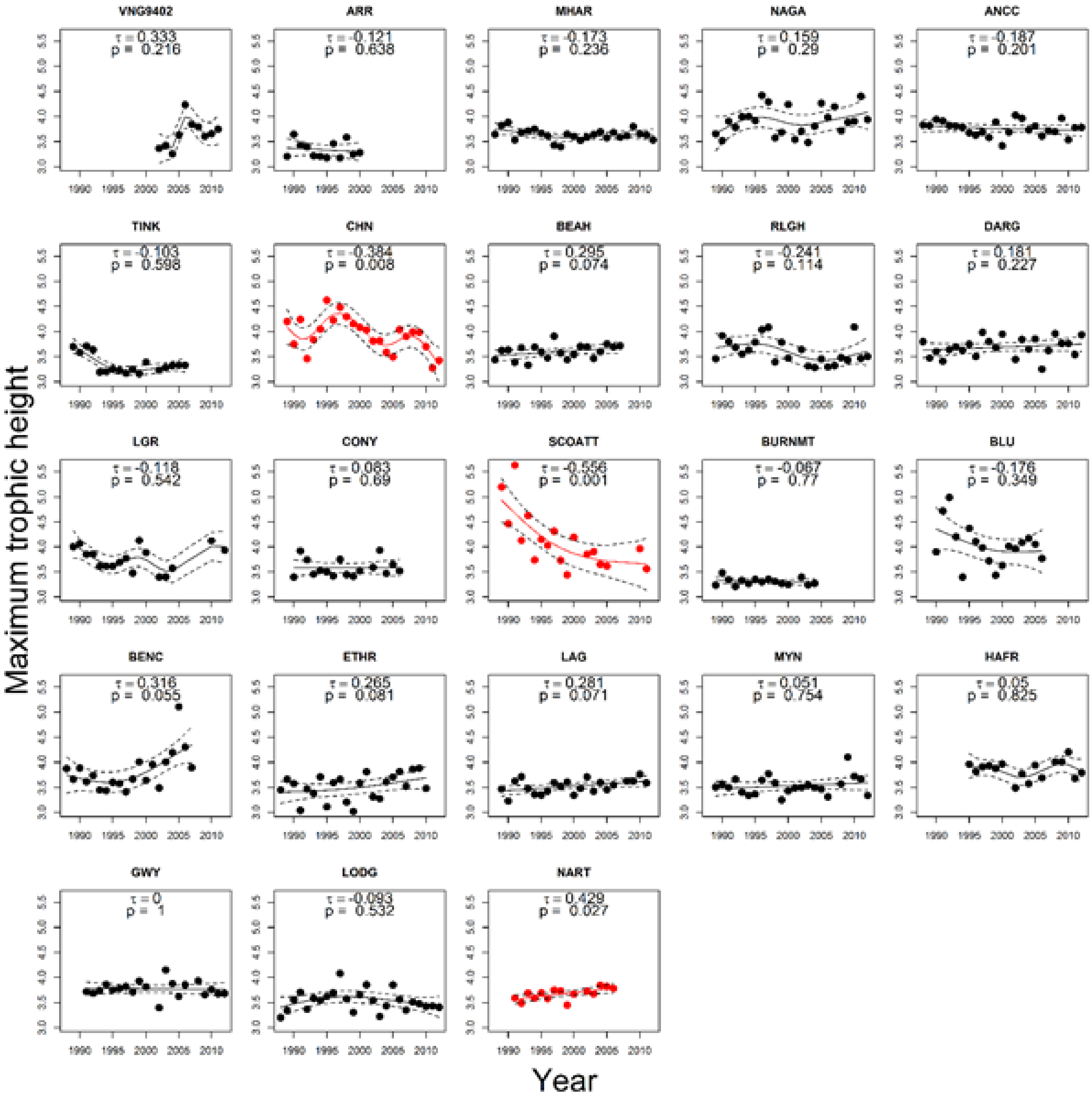


Figure D. Trends in maximum trophic height at each of the UWMN sites. Site ordering is explained in the legend of Figure A. See Figure 1 for site name abbreviations.

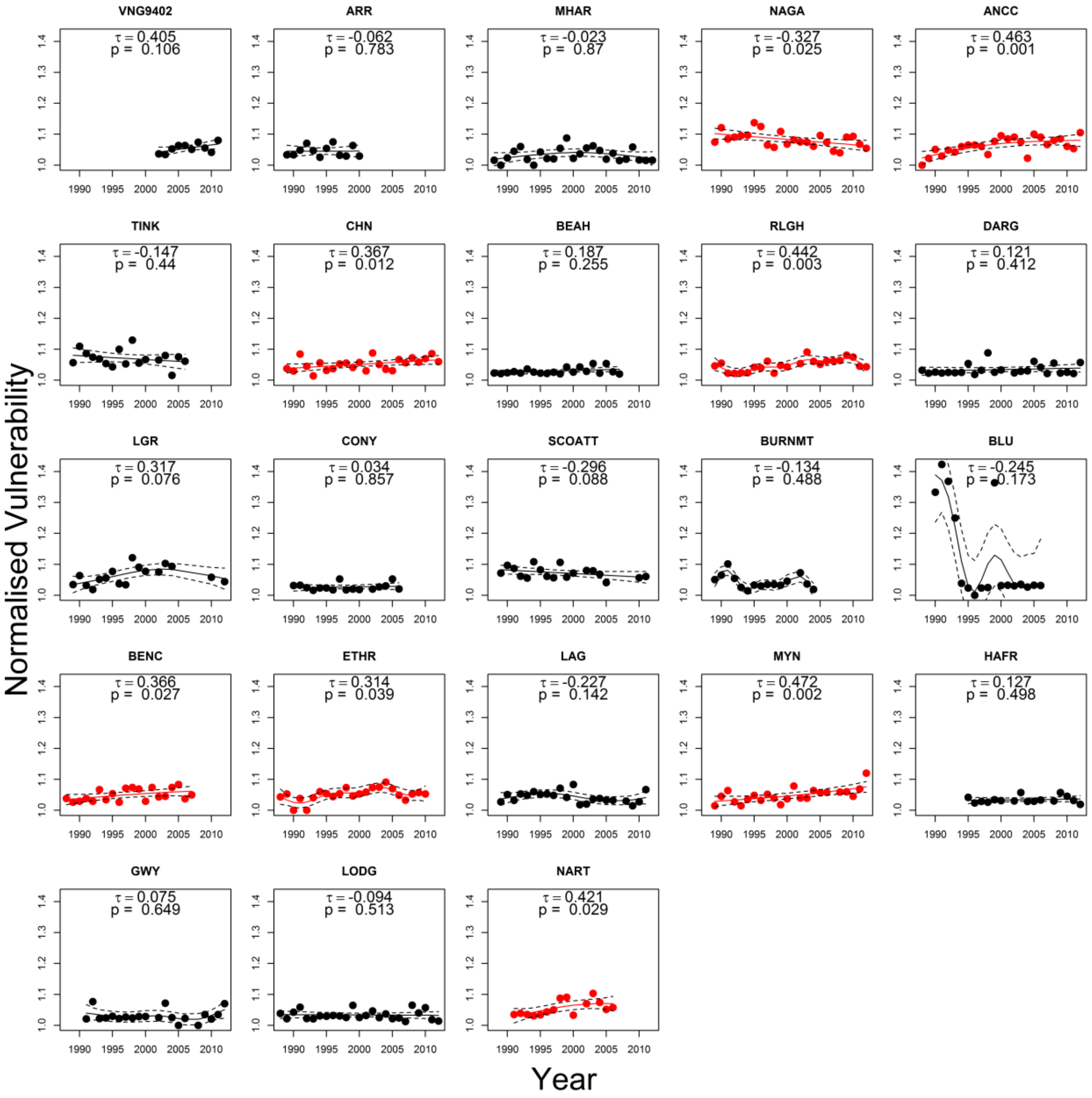


Figure E. Trends in average food web vulnerability at each of the UWMN sites. Site ordering is explained in the legend of Figure A. See Figure 1 for site name abbreviations.

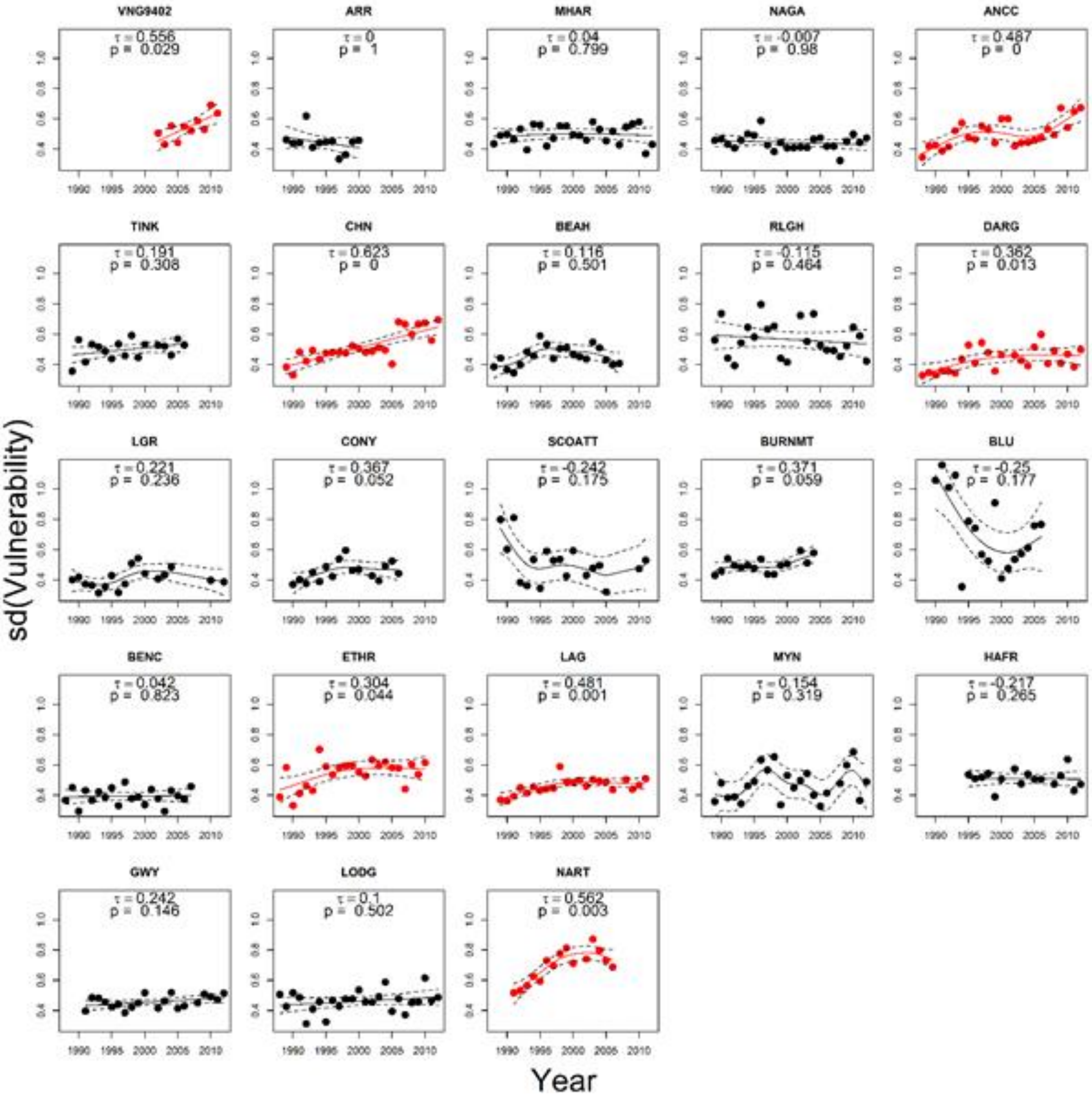


Figure F. Trends in the standard deviation of food web vulnerability at each of the UWMN sites. Site ordering is explained in the legend of Figure A. See Figure 1 for site name abbreviations.

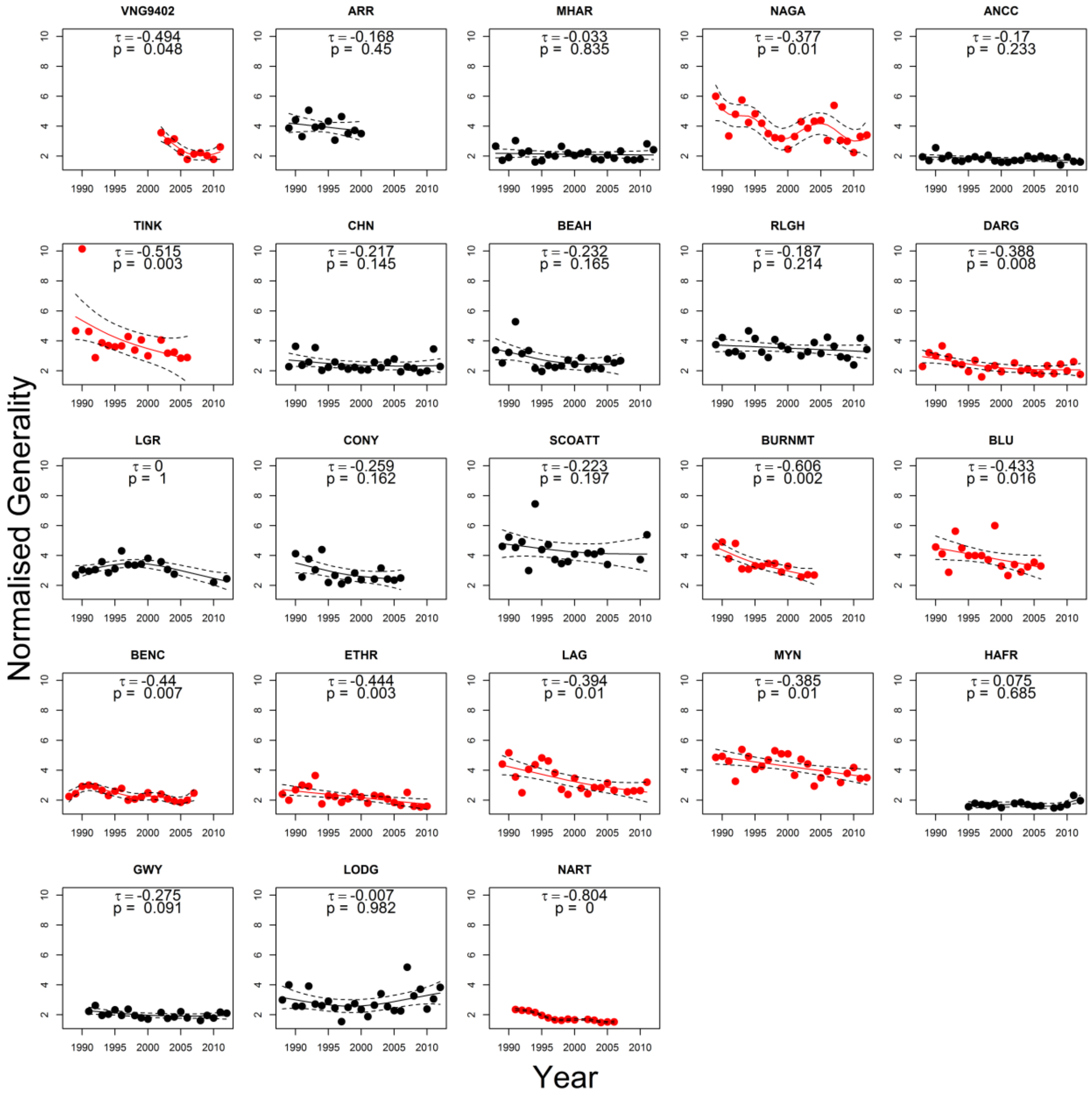


Figure G. Trends in food web generality at each of the UWMN sites. Site ordering is explained in the legend of Figure A. See Figure 1 for site name abbreviations.

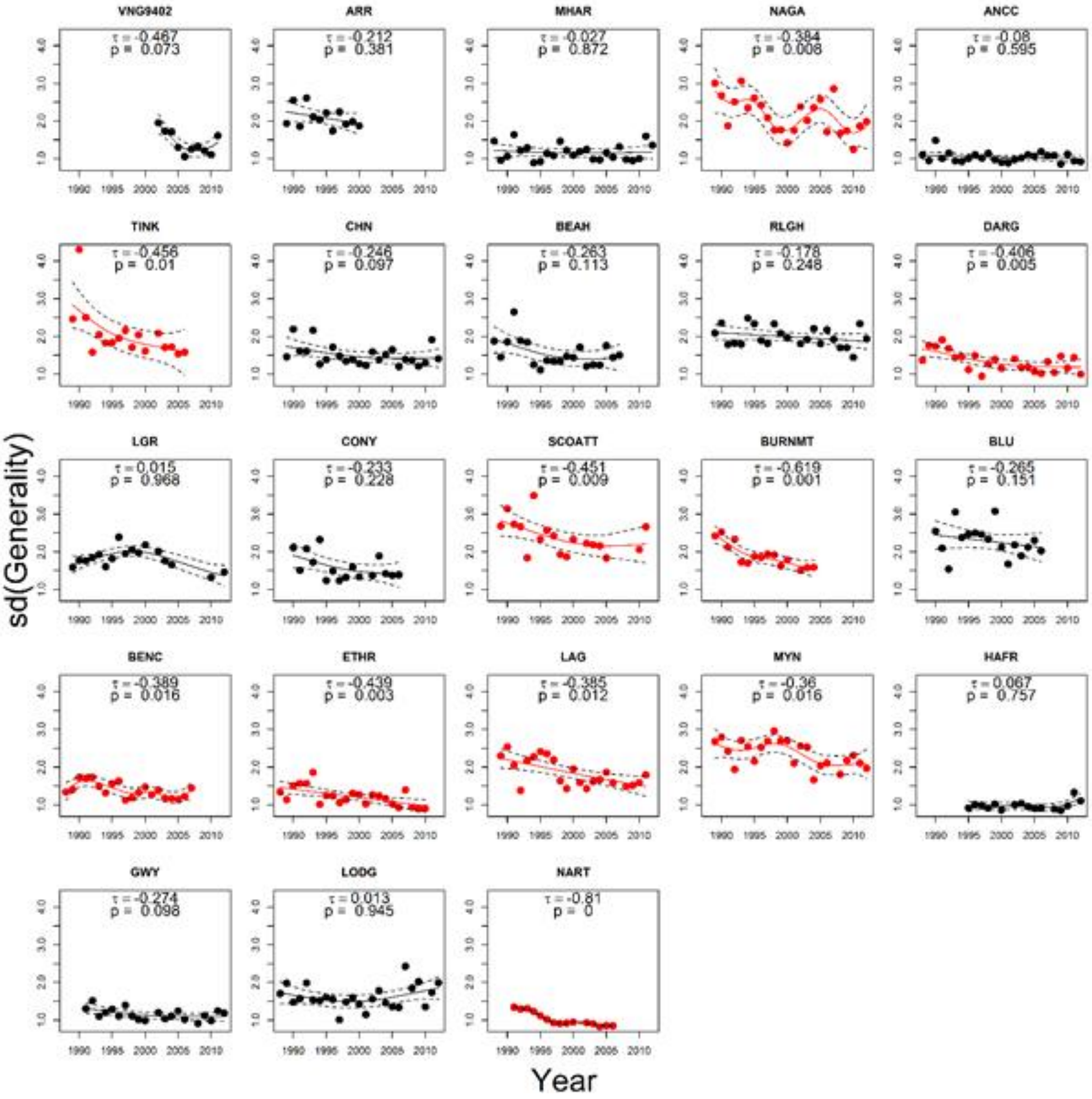


Figure H. Trends in the standard deviation of food web generality at each of the UWMN sites. Site ordering is explained in the legend of Figure A. See Figure 1 for site name abbreviations.

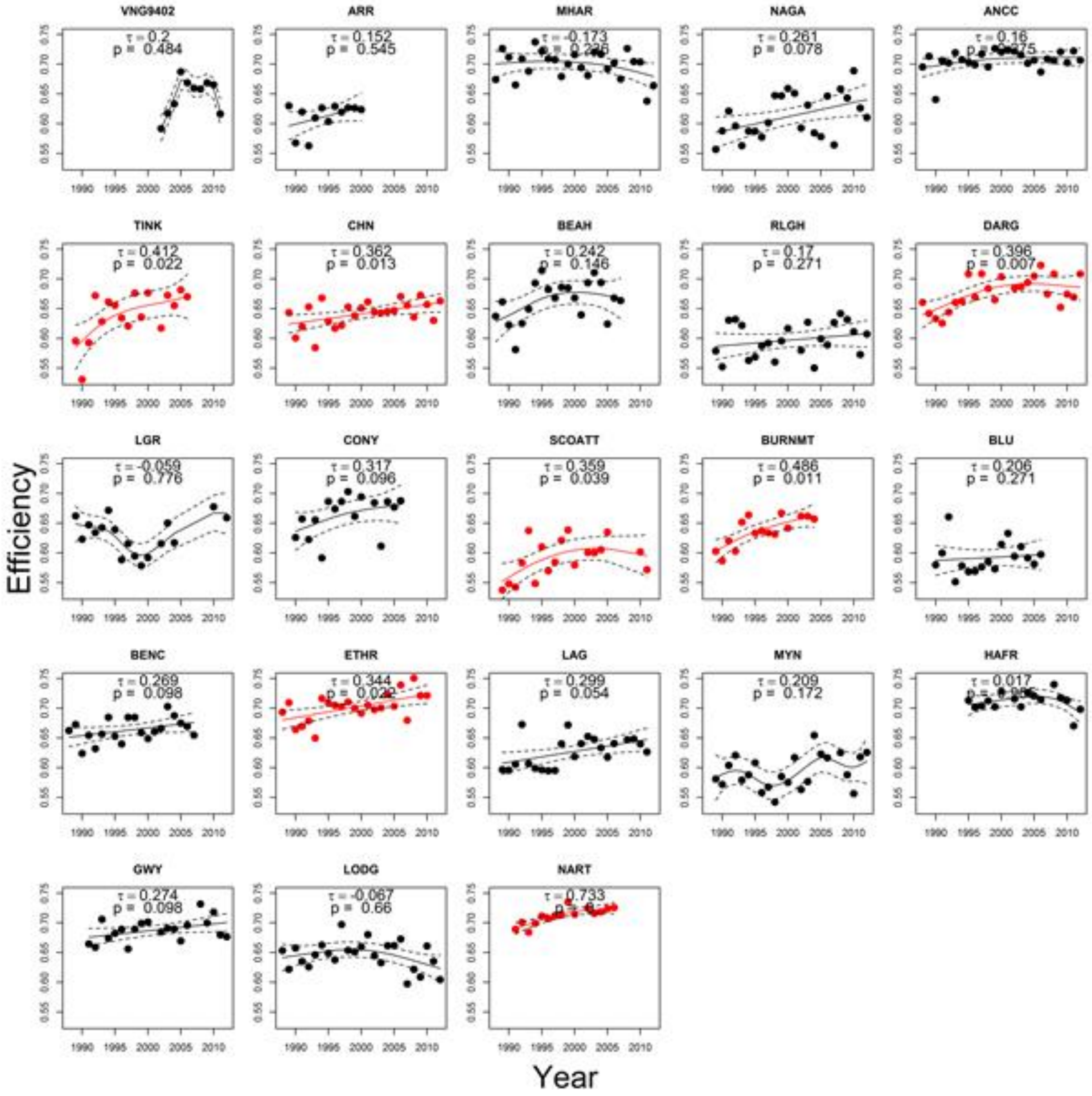


Figure I. Trends in food web efficiency at each of the UWMN sites. Site ordering is explained in the legend of Figure A. See Figure 1 for site name abbreviations.

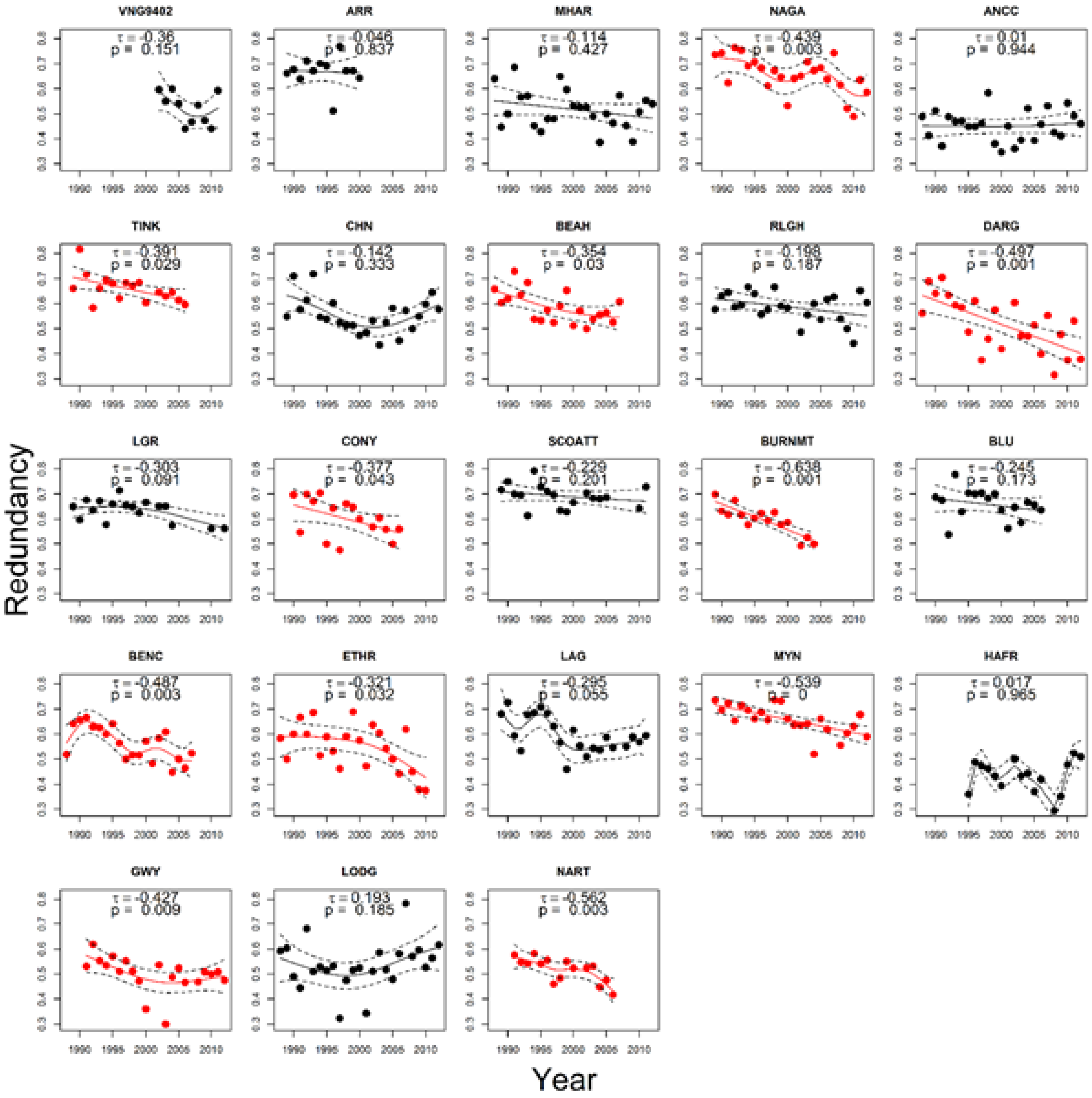


Figure J. Trends in food web redundancy at each of the UWMN sites. Site ordering is explained in the legend of Figure A. See Figure 1 for site name abbreviations.