J. Limnol., 63(1): 63-76, 2004

A consideration of potential confounding factors limiting chemical and biological recovery at Lochnagar, a remote mountain loch in Scotland

Neil ROSE*, Don MONTEITH, Helen KETTLE[1)], Roy THOMPSON[1)], Handong YANG and Derek MUIR[2)]

Environmental Change Research Centre, University College London, 26 Bedford Way, London WC1H 0AP, UK.
[1)]Department of Geology and Geophysics, University of Edinburgh, West Mains Road, Edinburgh EH9 3JW, UK
[2)]National Water Research Institute, Environment Canada, Burlington, Ontario L7R 4A6, Canada
*e-mail corresponding author: nrose@geog.ucl.ac.uk

ABSTRACT

Lochnagar, a remote loch in the Grampian Mountains of Scotland is one of the most studied freshwater bodies in the UK. It represents a key site in a number of monitoring programmes and has become the UK's 'flag-ship' mountain lake in various EU funded projects over the last 15 years. Palaeolimnological studies have revealed the extent and diverse provenance of atmospherically deposited pollution at the site and show that the loch began to acidify in the mid-19th century. However, despite abatement strategies dramatically reducing the emission and deposition of non-marine sulphate and trace metals since the 1970s, the loch pH shows little sign of recovery and full basin fluxes of, for example, Pb and Hg show no decline or even a continued increase. It is suggested that the lack of recovery from acidification over the last 15 years of monitoring results from the balancing of the decline in sulphate by increased nitrate, and that this increase is related to winter duration and severity. The lack of response by the sediment record to declines in metal deposition is thought to be due to a continuing input of previously deposited metals from the catchment. Hypotheses for these enhanced catchment inputs involve responses to a changing climate. Site specific climate reconstructions and predictions for the 21st century suggest an accelerated increase in temperature rise and increased winter precipitation and storminess. These predicted changes are likely to exacerbate the input of metals (and other stored pollutants) from the catchment but higher temperatures may also help to promote recovery from acidification.

Key words: acidification, atmospheric deposition, climate change, persistent organic pollutants, trace metals

1. INTRODUCTION

Sensitive geologies, sparse soils and extreme meteorology in many mountain areas combine to produce fragile ecosystems. Hence, despite their isolation from direct contamination, the stress of atmospheric pollutant deposition to remote lakes in upland areas often results in detectable chemical and/or biological change. Remote mountain lakes can therefore act as 'early warning' indicators for less sensitive sites, and the wider environment, and they have become a useful tool in monitoring the impacts of atmospherically deposited pollutants. Recent studies in Europe have shown that these pollutants have been impacting remote lakes for hundreds of years (Bindler *et al*. 2001; Yang *et al*. 2002a) and that this deposition can result in the accumulation of both trace metals and organochlorine compounds in biota sometimes to significant levels (Rognerud *et al*. 2002), whilst critical load exceedence, resulting from both sulphur and nitrogen deposition, is known to be widespread in mountain lakes throughout Europe (Curtis *et al*. 2002).

Since 1979 when the Convention on Long-Range Transboundary Air Pollution was adopted, there have been a number of Conventions and Protocols to reduce the deposition of acidifying and toxic substances including heavy metals and persistent organic pollutants. As a result of the earlier sulphur protocols, European emissions fell by 50% in 2000 (against a 1980 baseline) and are expected to have fallen by 58% in 2010. Monitoring the impacts of these treaties is fundamental and, here again, mountain lakes are important as evidence that these fragile sites are protected implies greater protection of less-sensitive sites.

Lochnagar, a remote mountain loch in the Grampian Mountains of Scotland, has become the UK's 'flag-ship' mountain lake and is now one of the most intensely studied freshwater bodies in the country. Water sampling, for the analysis of major ions, has been ongoing at the site since 1980 whilst the first palaeolimnological study was undertaken in 1986 (Jones *et al*. 1993; Battarbee *et al*. 1995). The site has been part of the UK Acid Waters Monitoring Network (UKAWMN) since its inception in 1988 and as a consequence annual surveys of epilithic diatoms, aquatic macro-invertebrates and aquatic macrophytes in the loch, and fish in the outflow stream have been made for over 15 years.

The UKAWMN also initiated a regime of quarterly water sampling and analysis, a frequency which was briefly increased to weekly, before becoming fortnightly, as a result of the inclusion of Lochnagar in the EU funded research projects MOLAR (1996 - 1999) and EMERGE (2000 - 2003). The inclusion of Lochnagar as a site in these EU projects also resulted in the sampling and analysis of trace metals in bulk deposition and lake water on a weekly (later fortnightly) basis and a full lake and catchment survey of trace metals and fly-ash particles in the soils and sediments (Yang *et al.* 2001, 2002a, b & c; Rose *et al.* 2002). This trace metal work has been extended to include annual sampling of a suite of ecological compartments ranging from catchment plants to aquatic flora, fauna and sediment trap material.

A small solar-powered automatic weather station (AWS) was installed close to the loch shoreline, as part of MOLAR, in August 1996 and half-hourly measurements of a range of parameters (air temperature and pressure, wind speed and direction, relative humidity) and daily rainfall, has continued to the present albeit with short periods of lost data resulting from power failures. This vast array of accumulated meteorological data has been used in climate models (Agustí-Panareda & Thompson 2002) and is supported by a chain of thermistors at eight depths in the loch recording water temperatures at 1-2 hour intervals. More recently, programmes monitoring stable isotopes and the chemistry of cloud water have begun whilst palaeolimnological studies on trends in persistent organic pollutants (POPs) (e.g. Rose *et al.* 2001) and climate change (Dalton *et al.* 2001) have also been undertaken.

Early palaeolimnological studies indicate that Lochnagar began to acidify in the mid-nineteenth century as a result of acid deposition, and lake water pH is thought to have declined from 5.6 at this time to around pH 5.0 by the 1940s (Jones *et al.* 1993; Battarbee *et al.* 1995). However, since the 1970s national emissions of sulphur have been declining in response to abatement strategies and although data from the nearby wet deposition monitoring station at Glen Dye, over the period 1986-1997, was found to show little evidence of a decline in non-marine sulphate (xSO_4) (Fowler & Smith 2000), the most recent assessment (Cooper & Jenkins 2003) suggests a substantial post-1995 decline, a trend which is also reflected in the Lochnagar outflow xSO_4 concentration. National trends for trace metals also show that Pb, Zn and Cu deposition has reduced by more than 75%, 55% and 30% respectively since the mid-1970s (Baker 2001) and that Hg emissions have fallen by more than 80% since 1970 (NAEI 1999). These national trace metal declining trends are reflected in local measurements of trace metal deposition at the Banchory monitoring station just 40 km to the east of Lochnagar (Oslo and Paris Commisions 1994; Playford & Baker 2000). However, despite the wealth of evidence for a considerable decline in pollutant deposition, continued monitoring and a new palaeolimnological assessment provide little substantive indication of ecological improvements in the aquatic environment of Lochnagar. pH reconstruction of a sediment core, taken in 1991, provided tentative signs of a slight pH increase from a minimum of 5.1 in 1960 but since then temporal variability in inferred pH has been too large to discern any clear upward trend. The most recent available data (March 2003) shows there has been neither increase in loch pH and alkalinity, nor decline in labile aluminium concentration, nor shift to a less acidic epilithic (rock dwelling) flora relative to a 1988 baseline. Furthermore, fluxes of trace metals to the sediment basin remain elevated, providing no indication of the recent decline detected in deposition.

Here, we discuss the evidence for both chemical and biological impacts caused by atmospherically deposited contaminants, assess the possible confounding factors which may have restricted biological recovery at Lochnagar and speculate on the potential impact on these by future climate change.

2. THE SITE

Lochnagar (56°57'N; 3°13'W) is a corrie loch lying at an altitude of 785 m below a north-east facing, steep back-wall (Fig. 1). The catchment geology is biotite granite overlain in places by blanket peats interspersed with areas of screes and large boulders. The peats are significantly eroded on the north-eastern side, especially near the loch-shore, but elsewhere support a sparse catchment vegetation of *Calluna*, *Vaccinium* and a number of mosses and lichens. In recent years winter snow cover has become more intermittent and whilst significant snow fields can accumulate over the main period (December – March) there is often less than 100% cover in the catchment. Nevertheless, patches of snow remain in sheltered cracks and gullies on the back-wall through to May or June. Similarly, winter ice cover has been extremely variable over the last decade, never achieving any great thickness and liable to break up during winter storms. The loch has an area of 9.8 ha, giving a catchment to lake area ratio of 9.4. It has a maximum depth of 26 m, the deep point being offset towards the southern end. Inflow streams are generally small and ephemeral whilst the outflow drains through a series of small pools and ultimately to the River Dee.

The epilithic diatom flora is relatively diverse and, over the UKAWMN monitoring period, has been dominated by acidophilous species, with *Achnanthes marginulata* generally the most abundant, except in 1991 and 1997-98 when *Tabellaria flocculosa* dominated. *A. marginulata* also dominates the diatom assemblage of sediment trap samples retrieved annually (1991 to present) from the deep-water area of the loch. The planktonic taxa *Aulacoseira distans* var. *nivalis* is also a major constituent in the trap samples. Macroinvertebrate and aquatic macrophyte diversity is poor and typical of acidic, high altitude lakes subject to winter freezing, wind-stress and relatively low year-round temperatures. The macroinvertebrate community is dominated by chironomids and the stonefly species *Capnia* spp, *Diura bicaudata* and *Siphonoperla torrentium*. Water beetles (*Oreodytes davisii*), caddisflies, acid tolerant mayflies (Limnephilidae) and craneflies (Tipulidae) are also common. The aquatic macrophyte flora is dominated by

Fig. 1. Lochnagar viewed from the east and map showing location (Photo: Neil Rose).

liverworts, especially *Nardia compressa* whilst the pteridophyte *Isoetes lacustris* and rush *Juncus bulbosus* var. *fluitans* are the only vascular species. The loch also supports a small brown trout population.

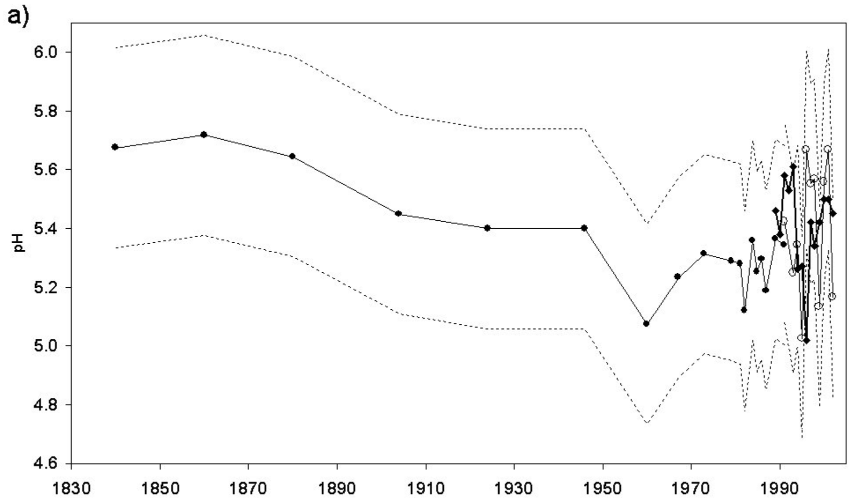
3. PALAEOLIMNOLOGICAL DATA

Visual comparisons of the catchment today with earlier paintings (Giles 1849) and sketches (Geikle 1887) provide no indication of change in the physical nature of the Lochnagar catchment since at least the mid-nineteenth century. Therefore any chemical and biological change within the loch over this period cannot be attributable to any changes in land-use and must result from atmospheric transport and deposition from regional or long-range sources or from climatic effects.

A range of palaeolimnological techniques have been applied to sediment cores taken from Lochnagar and show that, although remote, the site has received measurable levels of atmospherically deposited contamination since at least the mid-nineteenth century. Fossil diatoms provide evidence for an early biological response to this pollution. An increase in *Achnanthes marginulata* and contemporaneous declines in the abundances of *Achnanthes scotica*, *Frustulia rhomboides* var. *viridula* and *Brachysira vitrea* indicate a major change in the flora towards one favouring more acidic conditions (Jones *et al*. 1993; Battarbee *et al*. 1995). Reconstruction of pH from ^{210}Pb-dated sediment diatom assemblages (Weighted Averaging Partial Least Squares), using a mountain lake diatom-pH training set (Cameron *et al*. 1999), indicate a decline from about 5.7 to 5.1 over the last 150 years (Fig. 2a) with the lowest inferred pH value recorded in *ca* 1960. Between this point and the core top (1991) there is faint evidence for an increase in inferred pH, although inter-sample variability is large and the trend ceases to be significant if the 1960 sample is excluded.

pH reconstruction of diatoms in annual sediment trap samples collected since 1991 also provide no evidence of a linear response to the post-1995 declines in xSO_4 deposition or surface water xSO_4 concentration and inter-annual variability is large (Fig. 2b). Although five post-1995 samples reconstruct at over pH 5.6, samples collected in 1999 and 2002 reconstruct at less than pH 5.2. While the mean and range of sediment trap diatom pH values are very similar to that pH measured in early June over the same period, neither shows any trend. Between-year agreement for diatom inferred pH and measured pH is at best weak but this is not surprising. The diatom assemblage of sediment trap samples might be expected to reflect chemical status over the middle part of the year when productivity is likely to be highest. However the precise period of environmental integration represented by each sediment trap diatom sample is not known and is also likely to vary between years depending on the timing of ice melt, occurrence and intensity of turbulent storms, etc.

Supporting evidence for atmospheric deposition being the cause of the acidification of Lochnagar comes from the analysis of deposited contaminants stored in the lake sediments. Increases in concentration of trace metals and spheroidal carbonaceous fly-ash particles (SCPs) (Fig. 3a) the product of high temperature fossil-fuel combustion, are found to be coincident with the decline in diatom-reconstructed pH. However, contamination by Pb (Fig. 3a) and Zn occurred well before the



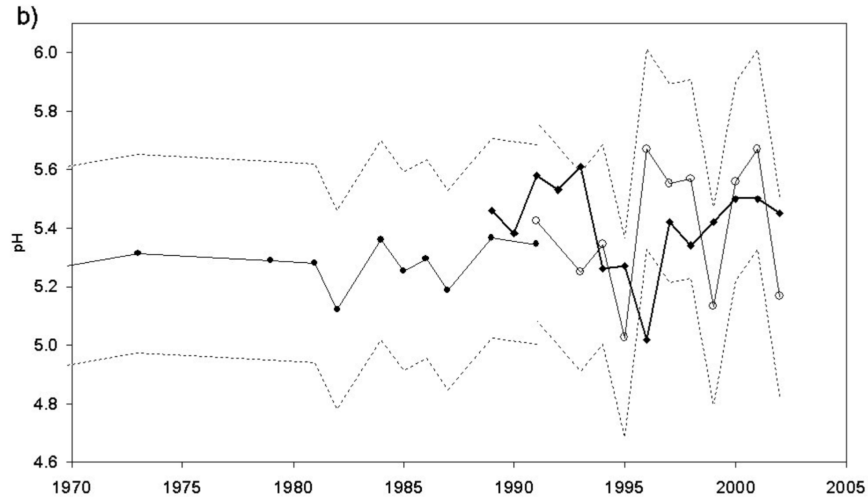


Fig. 2. **a**) Diatom-inferred pH reconstruction (●) for Lochnagar and recent sediment trap diatom-inferred pH (O) compared with June measured pH of the loch water since 1988. Standard error of diatom inferred pH as dotted lines. ^{210}Pb chronology: +/- 2 years for the period 1991-1945; 3 years for 1935-1924; 4-7 years for period 1924-1886. Post-1886 dates determined by extrapolation. **b**) As for a) but for the post-1970 period only to allow more detailed comparison of measured pH with sediment trap diatom inferred pH.

start of acidification and probably resulted from regional and European mining or smelting activities whilst similar temporal trends for Hg and SCPs show that the main source of Hg deposited at the site is from coal combustion. The sediment record also shows that atmospherically deposited contamination has a diverse provenance. Apart from fossil-fuel combustion, a major source of trace metals and SCPs, the sediment record contains an archive of persistent organic pollutants (POPs) from both industrial (e.g. PCBs) and agricultural (e.g. organochlorine pesticides) sources (Fig. 3b and Rose *et al.* 2001) showing profiles consistent with their differing origins and production histories.

Measured lake sediment profiles of contaminant concentration are often used to infer trends in the deposition of contaminants to a lake from the atmosphere, but true trends in the total inputs of contaminant can only be determined from full basin sediment fluxes. At Lochnagar, the analysis of 25 sediment cores from across the whole basin for a range of trace metals (Yang *et al.* 2002b) has shown that despite a recent decline in metal concentration the 'whole basin' trace metal flux has plateaued indicating continued contaminant inputs despite dramatic decreases in the emission and deposition of trace metals across the UK.

Figure 4 shows the case for Pb. Farmer *et al.* (1999) demonstrate that as coal combustion was the major contributor to lead emissions in the UK between 1830 and 1930 (especially in Scotland where Pb content of coal was double that of English and Welsh coal), the coal consumption curve can be a useful surrogate for historical Pb emissions in the UK. These show an increase from earliest estimates through to the 1960s followed by a dramatic decline to the present, such that

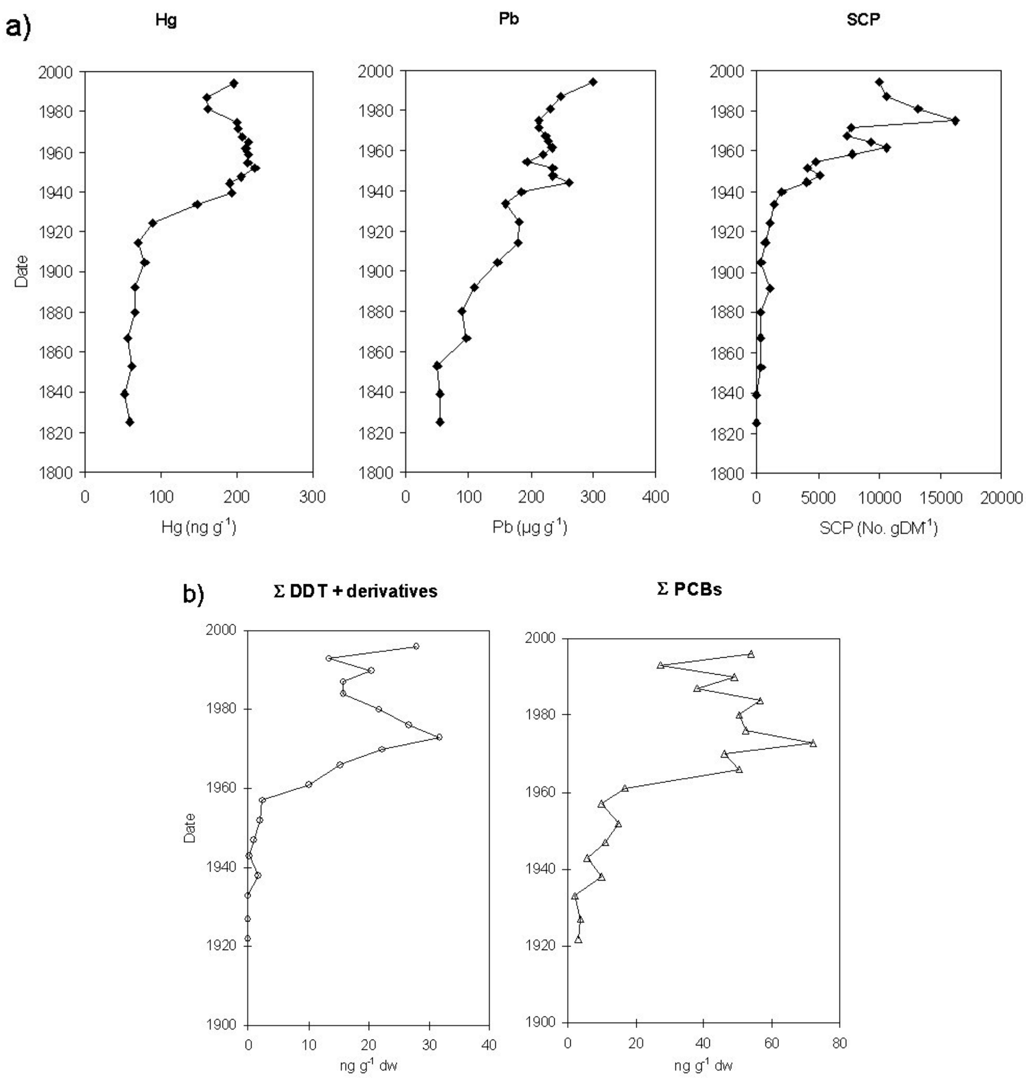


Fig. 3. The sediment record of atmospherically deposited contamination in Lochnagar. (**a**) selected trace metals and SCPs and (**b**) ΣDDT + derivatives and ΣPCBs.

UK coal consumption in the 1990s was equivalent to that in the mid-19th century. The full-basin flux for Pb in Lochnagar mirrors these trends showing a major increase from the mid-19th century through to the 1950s/1960s and then a small decline. However, from the 1970s, this relationship breaks down and whilst there is a major decline in Pb emission and deposition across the UK, full basin sediment fluxes at Lochnagar have remained stable or have even increased slightly.

From the mid-1970s onwards, evidence for the major reduction in Pb emissions and national and local Pb deposition are also provided by direct measurements (Fig. 4). UK Pb emissions show a considerable decline since the mid-1970s (Baker 2001) principally as a result of the removal of Pb from vehicle fuels. This decline is reflected in deposition measurement at both the national and local scale. The UK Rural Monitoring Network, since 1972 the longest metals deposition dataset available for the UK (here represented by the Wraymires site), and the annual volume weighted Pb data from Banchory, both show this major decline. However, although UK measured data do not extend back as far as the inferred peak in Pb emission and deposition, European emissions data do. These show an increase from the earliest estimates in 1955 to a peak in the mid-1970s and then a decline to recent times (Olendrzyński *et al.* 1996; Pacyna & Tørseth 1997) and it is not unreasonable to expect that UK patterns would be similar, in agreement with estimates from coal combustion (Fig. 4).

There is no reason to believe that atmospheric deposition at Lochnagar follows anything other than national trends, especially given the concurrence of Banchory data, so the absence of a recent decline in the flux of Pb to the loch sediment points to mediating processes oc-

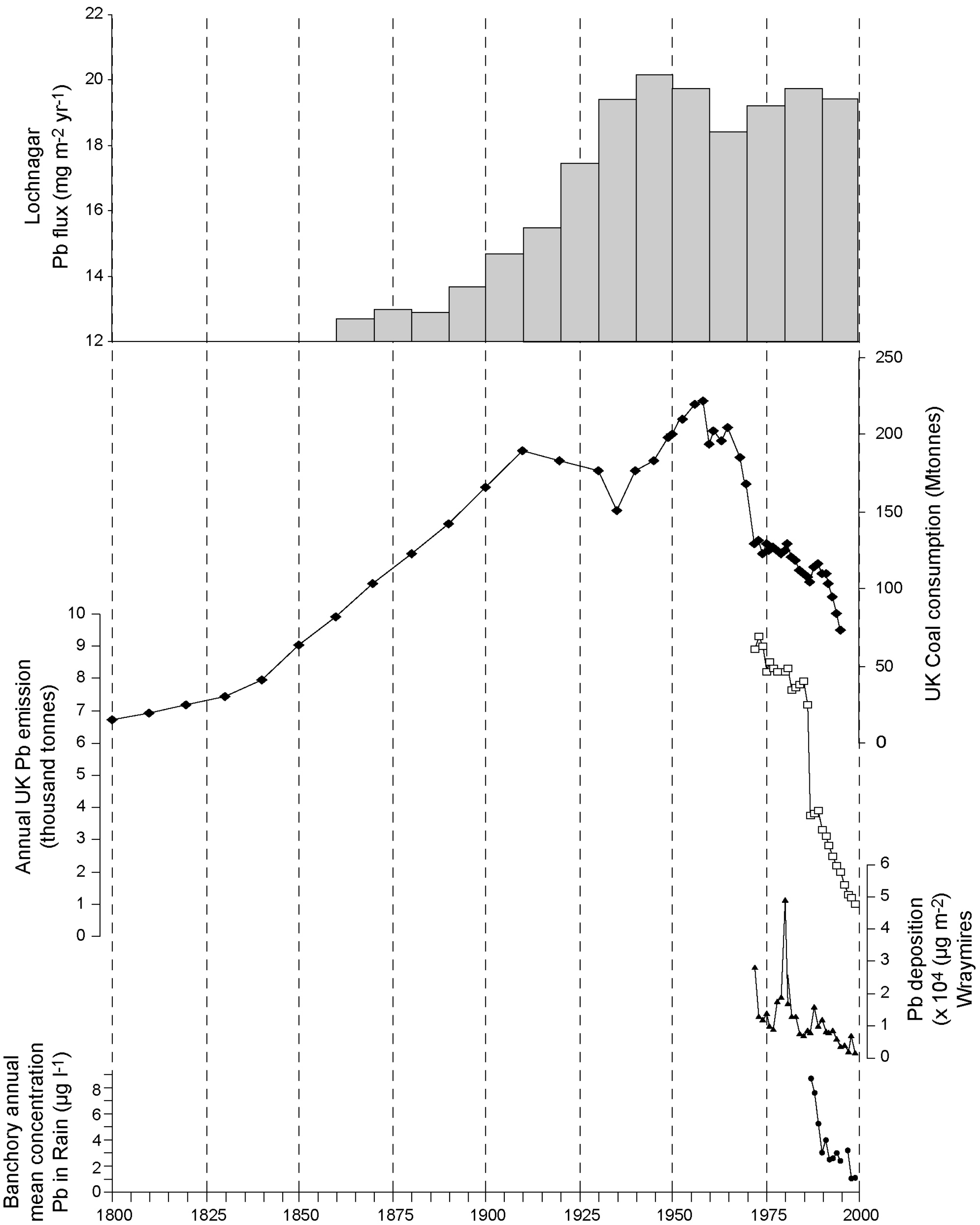


Fig. 4. The Lochnagar full basin sediment flux for Pb (histogram columns are decadal); UK coal consumption since 1800 (♦; data taken from Farmer *et al.* 1999); annual UK Pb emission data (□); measured Pb deposition data (1972 – 1999) from the UK rural network site at Wraymires (▲; data from Baker 2001); annual mean volume weighted Pb in rain from the North Sea Network Banchory site (●; data from Oslo and Paris Commissions 1994; Playford & Baker 2000).

curring within the catchment. Yang *et al.* (2002b) showed that the catchment soils at Lochnagar, although sparse, contain the equivalent of more than 400 years of Pb deposition at current (2000) levels. They proposed that Pb released from this store is now a significant contributor to the total flux to the loch and is negating the expected decline resulting from deposition reductions. A similar situation has also been observed for Hg at Lochnagar (Yang *et al.* 2002b) although the full basin flux of Hg continues to increase through time, and more importantly across the period of Hg emission reduction, rather than plateau as is observed for Pb. Further evidence for catchment inputs of previously deposited pollutants comes from a preliminary study on Pb isotopes in the Lochnagar sediment record (Yang, unpublished data). These data show a decline in $^{206}Pb/^{207}Pb$

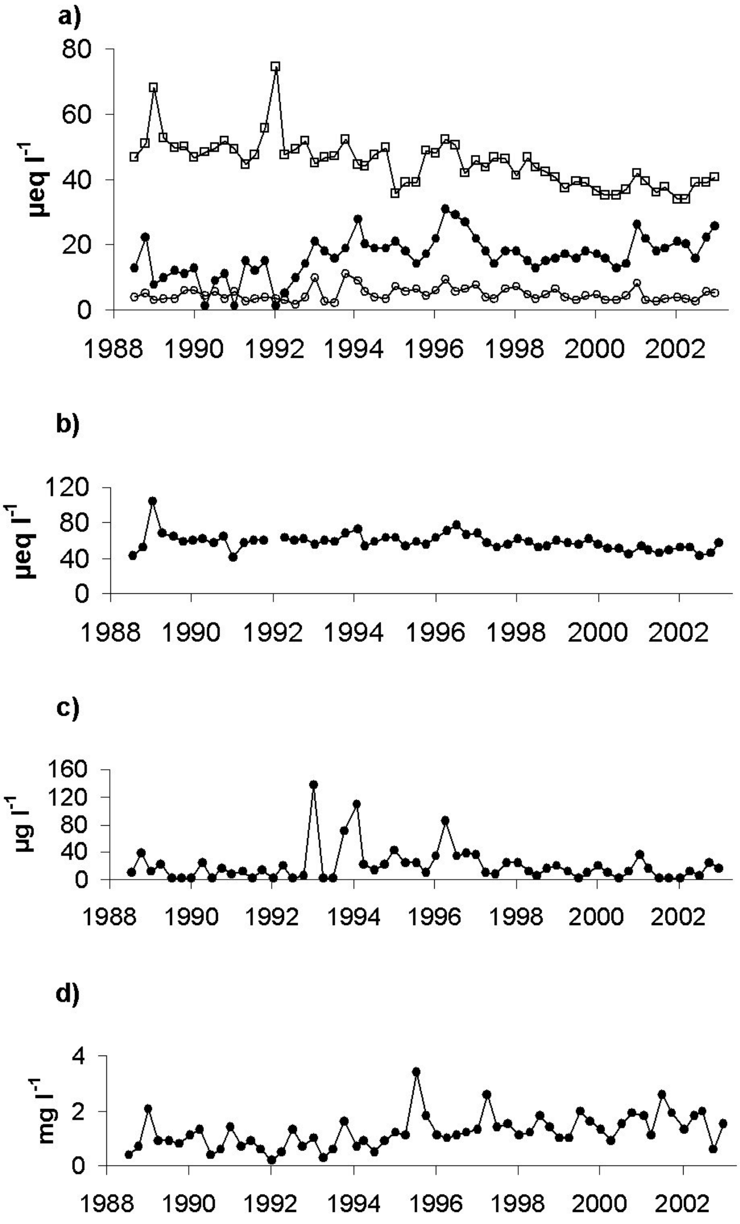


Fig. 5. Water chemistry for Lochnagar 1988 – 2003. Concentrations are shown for (**a**) non-marine sulphate □, nitrate ●, and hydrogen ion O, (**b**) calcium + magnesium, (**c**) labile aluminium and (**d**) dissolved organic carbon (DOC). Note change in units.

ratio as a result of increased industrial and vehicular Pb, but no corresponding increase of the ratio in the surface sediment layers in response to vehicular Pb reduction.

4. MONITORING DATA

In an assessment of national trends in sulphur deposition over the period 1986-1997 Fowler & Smith (2000) found a strong geographical pattern, with the largest decreasing trends, of around 3 µeq l^{-1} y^{-1} xSO_4 occurring in south-east and central England. Lochnagar is located in an area where the gradient of deposition reduction was found to change rapidly, with significant regional declines of around 1 µeq l^{-1} y^{-1} observed to the east but no significant change immediately to the west. Data from the nearby wet deposition monitoring station at Glen Dye, near Banchory, showed no significant change in xSO_4, or NO_3^-, in precipitation in either absolute or volume weighted concentrations. Despite this, Monteith & Evans (2000) found that for the first ten year period of the UKAWMN, xSO_4 in Lochnagar waters showed a slight but significant decline (0.67 µeq l^{-1} y^{-1}), while NO_3^- concentration increased by 1.13 µeq l^{-1} y^{-1} (Fig. 5a). This has important implications for loch chemistry as the ratio of NO_3^- to xSO_4 is particularly high, in a UK context, and NO_3^- therefore makes a major contribution to loch acidity.

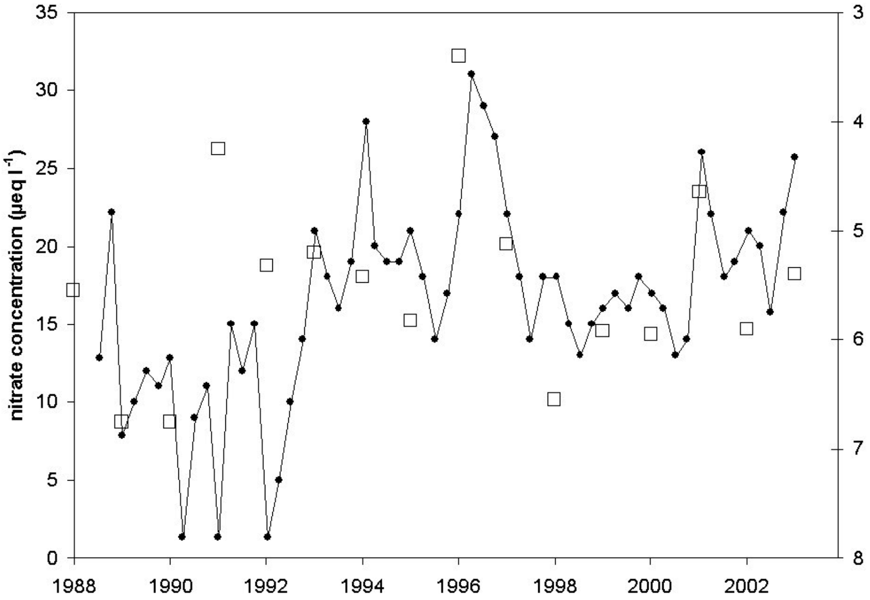


Fig. 6. Lochnagar nitrate concentrations (●) and temperature of coldest month (□) from the central England temperature series, 1988 – 2002. Temperatures are shown on an inverted axis.

Nitrate and xSO_4 are often considered together in representing the net anthropogenic acid anion ('mineral acidity') contribution to the charge-balance of run-off. However, at Lochnagar they appear to show different relationships with cations. While the decline in xSO_4 is largely paralleled by a decline in non-marine base-cations (Fig. 5b), the step-change in NO_3^- (Figure 5a) is reflected by increases in hydrogen ion and soluble aluminium concentrations (Figure 5a & c). The net result is that although the equivalence sum of NO_3^- and xSO_4 has remained relatively constant over the monitoring period, hydrogen ions and soluble aluminium appear, if anything, to have risen slightly (Fig. 5a & c). This seems to result from the strongly seasonal variation in NO_3^- which, at Lochnagar, appears to be closely associated with winter duration, whilst xSO_4 inputs show far less seasonality. Similar observations on an apparently disproportionate importance of NO_3^- in episodic depression of pH during snow-melt have been made in the north-eastern United States (Murdoch & Stoddard 1992; Sullivan *et al*. 1997) and this seasonality has important implications for the biota in Lochnagar and its recovery from acidification, in particular for the spring emergence of macroinvertebrates and for spring or early summer blooming algal species.

The reason for the upward trend in loch NO_3^-, over a period for which there is no evidence for any trend in deposition (Fowler & Smith 2000), is not entirely clear. The NO_3^- time series is consistent with a shift from seasonal to year round leaching as described by the nitrogen saturation hypothesis (Stoddard 1994) but while the timing of any nitrogen saturation process should be site-specific, three other UKAWMN lochs, Loch Chon, Round Loch of Glenhead and Loch Grannoch all demonstrate very similar temporal patterns.

Regionally coherent variation in NO_3^- concentrations across a range of UKAWMN lakes has previously been described by Monteith *et al*. (2000) and linked to the winter North Atlantic Oscillation (NAO) Index. The NAO Index is a measure of the average Iceland-Azores pressure difference which has a dominant influence on meteorological conditions in the UK during winter and spring. Years when the average December to March NAO Index is negative are associated with cold, dry conditions dominated by north-easterly air-masses and these years coincide with the highest spring concentrations of NO_3^-. Monteith *et al*. (2000) proposed that NO_3^- leaching is enhanced during negative NAO winters due to the greater duration and intensity of soil freezing. This may increase both biocidal effects on the soil microbial population and frost damage to plant roots and other tissue, thereby increasing the availability of nitrogenous cell lysis products for mineralization. Low soil temperatures may also reduce assimilation of nitrogen by soil biota. The net effect of these processes would be greater export of NO_3^- from the catchment. We do not have a sufficiently long air temperature record for the site to allow a statistical comparison of NO_3^- concentration and air temperature. However, data from the Central England Temperature Series (CET), which correlate strongly with local air temperature records across the UK provide supporting evidence for a climatic effect. With the exception of one year, 1991, NO_3^- concentrations show an inverse relationship with December to March average CET (Fig. 6). Available water temperature data provide no indication of a direct link with NO_3^- concentration, the latter tending to peak around the time of ice melt. It is also unlikely that within lake under-ice processes are an important factor in determining between-year variability since similar temporal patterns are also seen in streams and in lakes at much lower altitude.

In summary, although a decrease in xSO_4 and an increase in NO_3^- have roughly balanced the concentration of strong acid anions in the loch, NO_3^- appears to have had a disproportionate effect on hydrogen ion and aluminium concentration due to its more concentrated delivery from the catchment at the end of the winter. The absence of any improvement in acidity over the past 15 years can therefore be linked to colder winter temperatures over this period, whilst a reduction in calcium appears to be driven by a decline in xSO_4. The combination of no improvement in acidity and decreased base cations may have had a particularly detrimental influence on the aquatic biota.

In common with nearly all sites on the UKAWMN, Lochnagar has undergone a substantial increase in dissolved organic carbon (Fig. 5d). Whilst Freeman *et al.* (2001) attributed this increase to a gradual rise in air temperature over the past decade, several other mechanisms have also been proposed and debated, particularly with respect to the possible effects of changing hydrological patterns (e.g. Tranvik & Jansson 2002; Evans *et al.* 2002). As the UKAWMN dataset has continued to grow there is increasing evidence that DOC increases across the Network may, in part, relate to declines in sulphate (Monteith 2004), possibly through its influence on soil acidity and/or ionic strength, although this cannot fully account for the observed trends. The inverse linkage between sulphate and DOC has long been recognised (e.g. Krug & Frink 1983) and attributed to the effect that increasing pH has on decreasing DOC solubility. At Lochnagar a slight trend of around 0.2 mg l^{-1} y^{-1} DOC over 15 years remains even when potential influences of sulphate have been controlled for, and this can be linked with smoothed annual temperature data over the same period.

The implications of these findings are that DOC concentrations have probably been increasing gradually since air temperatures began to increase and sulphate deposition began to decline in the 1970s. The combination of these two effects could have raised the flux of pollutants, with a high DOC affinity, into the loch as a result of organic complexation but would not necessarily have had an impact on the rate of deposition of these pollutants to the sediment. However, it is interesting to note that the timing of this DOC increase is contemporaneous with the breakdown in the relationship between whole basin trace metal flux and atmospheric deposition implying an increase in the influence of catchment inputs from this time.

Populations of epilithic diatoms, macroinvertebrates and aquatic macrophytes have been monitored annually since 1988, while annual sediment traps have been deployed since 1993 and these data have recently been subjected to time trend analysis as part of the 15 year interpretative exercise for the UKAWMN (Monteith 2004). In contrast to many other sites in the UKAWMN (where, unlike Lochnagar, there is evidence for a recent decline in acidity) there is no evidence for any change in the assemblages over the last 15 years and no "new" species have been recorded since the start of the large reduction in sulphate concentration around 1996. Given, the absence of improvement in acidity, the lack of biological change for these groups is perhaps not surprising, although it is interesting to note that this also implies that there has been no clear response to the considerable increase in dissolved organic carbon over the monitoring period (Fig. 5d).

The aquatic macrophytes species list has remained unchanged over the monitoring period. However there has been a very substantial increase in the abundance of the rush, *Juncus bulbosus* var. *fluitans*. This species was largely confined to a small patch in the southern end of the loch but has since spread to form a dense sward, at around 1 m water depth along much of the eastern edge and is also abundant elsewhere. It is particularly well adapted to the low availability of dissolved inorganic carbon characteristic of acidic oligotrophic lakes although it may show particularly prolific growth if water column CO_2 is increased slightly in acidified lakes as a result, for example, of re-acidification following the cessation of liming. *J. bulbosus* var. *fluitans* also shows strong nitrogen limitation, preferring organic nitrogen as a source (Schuurkes *et al.* 1986). Monteith & Evans (2000) proposed that the expansion of this species may be linked to the increased NO_3^- concentration in the loch water.

5. DISCUSSION

5.1. Confounding factors

A number of hypotheses have been proposed to explain the absence of a decline in the trace metal sediment record of lakes where deposition has significantly declined. First, that this is due to a simple time-lag effect, i.e. metals deposited onto a lake catchment take a number of years to work through to the water body and the enhanced catchment inputs we are observing now are the result of high metal deposition decades ago. This hypothesis implies that it will be at least several decades before the benefit of current emissions reductions are observed as a reduction in the metals inputs to Lochnagar sediments. Second, increased erosion, of the upper contaminated levels of catchment soils (possibly resulting from the effects of increased drought or episodes of high rainfall) are bringing the catchment-stored metals into the lake. At Lochnagar there is significant peat erosion, but the faces of the main eroding peats near the north-east loch shore are over a metre high covering hundreds, if not thousands, of years. Therefore, erosion of these faces would be expected to result in a substantial amount of uncontaminated material entering the loch resulting in similar, or lower, sediment metal concentrations, but greater input fluxes. Third, a gradual increase in average air temperatures and a reduction in sulphate deposition have led to enhanced decomposition and solubility of soil organic matter which is then increasingly leached as dissolved organic carbon (DOC) in wet periods. Metals are known to have a strong affinity for DOC and hence elevated DOC input might result in enhanced metal input from the catchment. Certainly, the DOC levels in Lochnagar waters have increased in recent years (Fig. 5d) and positive correlations for both Hg and methyl Hg with DOC have been observed in high altitude lakes in the United States (Krabbenhoft *et al*. 2002). The same could be occurring at Lochnagar. POPs are also known to have a strong affinity for DOC (Gao *et al*. 1998; Winch *et al*. 2002) and this mechanism would therefore also be applicable for catchment stored organic contaminants. Fourth, warmer winters resulting in longer ice-free periods would provide longer periods in which algae could scavenge metals from the lake water. These algae, when they eventually become part of the sediment, would then take more incorporated metal into the record. At Lochnagar, frequent ice observations have only been made over the last decade and it is now established that ice break-up can occur at any time through the winter following high temperature periods or storms. Anecdotal evidence from walkers and climbers suggests that if this winter ice-fragmentation occurred in the past, then it was certainly less frequent. Modelling studies also indicate that the period of winter ice cover at Lochnagar has also reduced by about two months over the period 1960 to 2000.

In reality, it is likely that the increase in metal inputs from the catchment at Lochnagar is a combination of several of the above effects. However, if the temperature and precipitation driven processes are important then climate scenarios that infer increased summer or winter temperatures and/or increased rainfall and/or increased storminess would point to a continuation of enhanced metal inputs as a result of the second, third and fourth hypotheses.

Climate change could also have a significant effect on the recovery of the loch from acidification. Data from the UKAWMN demonstrate the importance of NO_3^- for the acidity of Lochnagar, due to its relatively high contribution to the total acid anionic charge and its seasonally restricted delivery to the loch. NO_3^- concentration is linked to low winter temperatures, and the increased frequency of relatively cold winters between 1995 and 2002 (specifically 1996, 1997 and 2001) (Fig. 6) may account for the recent increase in this anion. Conversely, xSO_4 would appear to be having relatively little impact on acidity as its decline appears to be largely balanced by declining base-cations.

Palaeoecological pH reconstruction demonstrates a substantial anthropogenically driven acidification of Lochnagar over the past 150 years but it is not possible to estimate the relative contributions of xSO_4 and NO_3^- to this decline. The pH reconstruction hints at a minor recovery since an exceptionally acidic period *ca* 1960 and it is interesting to note that this corresponds to the coldest winter in the past 40 years and a colder winter period more generally. It is therefore possible that the apparently exceptionally acidic conditions at this time are at least in part a climatic feature. However, prior to 1960 no relationship is evident between diatom inferred pH and winter temperature and NO_3^- may have had a much smaller role with regard to total acid anion strength. These relationships suggest that future winter warming should further lower the acidity of the loch by reducing the leaching of NO_3^-. As nitrogen deposition is forecast to fall in response to further emission controls this should also have a beneficial long-term effect, but since there is no evidence of a short term relationship between deposition and surface water concentration, the time-scale is difficult to anticipate.

5.2. Future recovery or continued contamination?

Annual mean air temperatures for Lochnagar between 1781 and 1997 have been reconstructed as described in Agustí-Panereda & Thompson (2002). These data are presented in figure 7 and show increases equivalent to 0.3 and 0.5 °C/100 y for the periods 1801–1900 and 1901–1997 respectively. Assuming no future changes in the link between the large-scale circulation (as predicted by climate models) and the local climate at Lochnagar it is possible to predict future changes in air temperature at the site. This is done using an empirical model relating air temperatures from the NCAR Department of the Environment Parallel Climate Model (DOE PCM) to the Lochnagar AWS data. Using an empirical model derived by the method described in Kettle *et al*. (2003), lake surface temperatures may also be predicted. Figure 7 also shows both of these for the period to 2100. The DOE PCM uses emissions controlled by the Intergovernmental Panel on Climate Change (IPCC) A2 scenario which is reasonably conservative, and so the 1.5 – 2.5 °C increase in both air and lake water temperature predicted for the end of the 21st century could be considered moderate although still significantly higher than the increases reconstructed over the previous 200 years. Mean annual air temperature increase at Lochnagar is therefore predicted to accelerate over the next century.

The UK Climate Impacts Programme (UKCIP), using the IPCC Special Report on Emissions and a series of modelling experiments using the Hadley Centre HadCM2 model (Hulme *et al*. 2002), have also predicted a range of climatic changes for the UK under various scenarios from low (B1) to high (A1FI). For north-east Scotland under all scenarios, mean annual temperature increases from between 1.5 and 3.5 °C, over the 1961 – 1990 mean, by the 2080s,

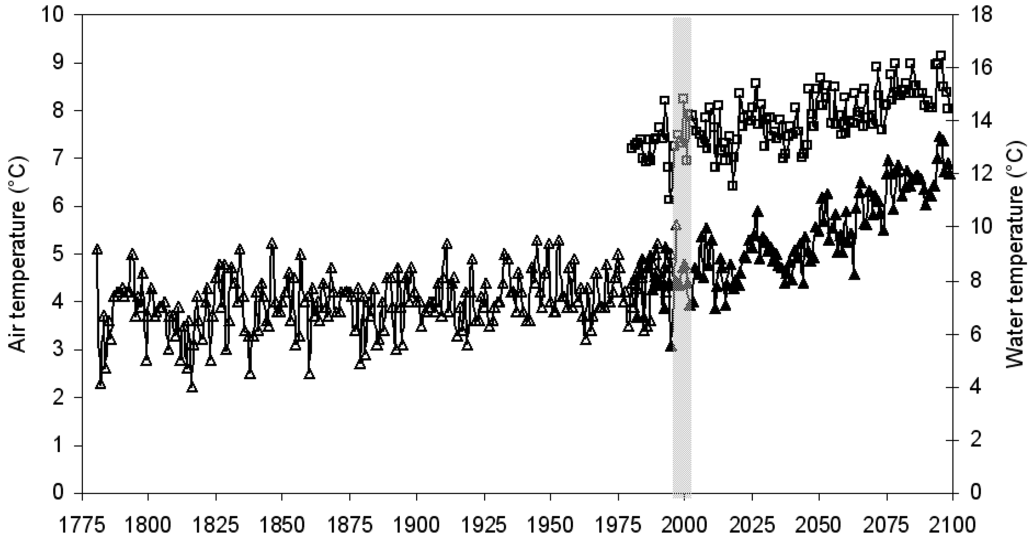


Fig. 7. Reconstructed mean annual air temperature at Lochnagar for 1781 – 1997 (∆) and predicted annual mean air (▲) and annual maximum monthly mean surface water (□) temperatures for 1980 – 2100 based on the mean 1997 – 1999 measured data. Shaded area shows the period of measured data at the loch-side Automatic Weather Station (1996 – present).

winter (December, January and February mean) temperature increases by 1.0 – 2.5 °C and summer (June, July and August mean) temperature by 1.5–4.0 °C. These predicted changes are in reasonable agreement with the site specific modelled predictions outlined under the A2 scenario described above. Similarly, under all scenarios, winter precipitation is predicted to increase by 10 to 25% and summer precipitation to decrease by -10 to -40% and whilst wind speed predictions remain very uncertain more depressions are likely to cross the UK leading to more frequent severe winds in the winter (Hulme *et al*. 2002).

Recent measured and modelled data therefore show that mean annual temperatures have increased at Lochnagar since the late-18th century whilst all available climate predictions for this area of the UK suggest that annual, summer and winter temperatures will continue to increase, possibly at an accelerated rate, until at least the end of the 21st century. The lake waters and thin catchment soils will track the changes in air temperature showing the same accelerated warming. Changes to ice-cover duration will be even more marked, with ice-cover becoming a rare event as winter temperatures warm and rarely drop below 0 °C for any extended periods of time. A reduction in summer precipitation could lead to unprecedented prolonged dry periods, although annual and winter precipitation are both predicted to increase. Finally, whilst mean winter wind speed is predicted to remain fairly constant over the period, predicted increases in the NAO Index over the next century (Hulme *et al*. 2002) suggest an increase in the frequency of winter storms. The conditions required for all the hypotheses leading to the exacerbation of contaminant inputs to Lochnagar from those stored in the catchment are thus fulfilled over the remainder of the 21st century and therefore, given these predictions and the estimated catchment storage of contaminants, it could reasonably be expected that catchment inputs will remain a major source of contamination to Lochnagar for many decades to come.

The sediment record at Lochnagar over recent years has shown that the flux to the lake basin has remained fairly constant for Pb and increased slightly for Hg (Yang *et al*. 2002b). However, this has been over a period of widespread and, in some cases dramatic, reductions in the emission and deposition of trace metals and other atmospherically transported pollutants. Contamination by directly deposited pollutants is therefore currently at its lowest levels for many years and the question remains whether further significant reductions are possible. If not, then the continued, or possible increase in, metal inputs from the catchment will result, not in a constant input of contaminants as has been the case in recent years, but an increase in input over current levels. In this case, there are implications for metal availability for sediment dwelling biota, especially detritivores, and the food chain above. However, the rate of climate enhanced catchment inputs in the short to medium term, and their ultimate decline in the long term, is open to speculation and significant uncertainties remain over the continuation of catchment contaminant loss. Neither chemical nor physical removal processes will extract all stored contaminants and therefore inputs from this source will also eventually decline, as long as future direct deposition remains low and the catchment store unreplenished. The time-scale for this is currently uncertain.

Chemical recovery from acidification has been observed at sites across Scotland (e.g. Ferrier *et al.* 2001) but Lochnagar does not currently show this trend. In the future, while sulphur deposition is likely to continue to decline in response to continued reductions in national and international emissions, the implications of these climate scenarios for recovery from acidification at Lochnagar are mixed.

It would appear that NO_3^- is having the dominant effect on mineral acidity at Lochnagar and, providing there is no further accumulation of nitrogen within the catchment, an increase in winter temperatures would be expected to result in a decline in winter NO_3^- concentrations and consequently the delivery of hydrogen ions and biologically available aluminium to the loch. On the other hand, an increase in precipitation would be expected to increase the ratio of hydrogen and aluminium ions to acid anions by influencing the dominant flow paths, with a larger proportion of sulphur and nitrogen reaching the loch directly by surficial routes and hence a smaller proportion being buffered by the underlying geology. This would therefore partly offset the reduction in acidity expected due to decreased fluxes of pollutants. In addition, the predicted rise in air temperature and the decline in sulphur deposition would be expected to increase the delivery of organic acidity to the site. The effect of the latter should simply be the converse of the acidification process where it is likely that DOC concentrations declined, so this may be seen as a part of the recovery process. However, the effect of the former would be to increase levels of organic acidity to unprecedented levels. Although DOC is often described as a 'weak acid', since it only partly dissociates in solution, the net effect of an increase would also counter the effects of a decline in mineral acidity to an as yet unknown extent.

The net effects of predicted climate changes for recovery from acidification are also difficult to estimate at present, although continued monitoring is beginning to allow the quantification of the influence of the various drivers. It is likely that were the sources of anthropogenic acid deposition excluded entirely, the combination of increased temperature and precipitation would prevent the full recovery of pH and biologically available aluminium to pre-industrial conditions.

Finally, the depositional history and driving forces impacting upon Lochnagar are not unique. Many upland lakes in the UK, and beyond, are showing increased DOC levels (Freeman *et al.* 2001) and catchment erosion, whilst the impacts of climate change are, of course, global. Currently, it is unknown how widespread the problems identified at Lochnagar are and this must now be a research priority.

6. CONCLUSIONS

- Despite the remote location of Lochnagar, atmospherically deposited trace metals, persistent organic pollutants and fly-ash particles are recorded in the sediment record. The sediments also record a significant acidification since the mid-19th century as a result of non-marine sulphate and nitrate deposition.
- There is a good agreement between historical trends in contamination as recorded in the Lochnagar sediment and increases in UK emissions over the period 1850 – 1970, but the sediments do not show decreases associated with recent, dramatic reductions in emissions or deposition. This discrepancy is thought to relate to the release of previously deposited pollutants stored in the catchment. Of the four hypotheses which can be invoked to explain this lag between deposition on the catchment and transport to the lake, three involve, or are modified by, climatic processes.
- Further, despite major reductions in the deposition of non-marine sulphate leading to reductions in loch water non-marine sulphate concentrations, loch pH and associated biological recovery from acidification are not unequivocally observed. This is thought to be due to the enhanced role of nitrate, balancing the decline in non-marine sulphate, and related to the duration and severity of winters.
- Future climate change is expected to result in warmer temperatures at Lochnagar, especially during the winter months, enhanced winter precipitation, but reduced summer precipitation, and possibly a higher frequency of extreme events including winter storms. We speculate that these conditions will lead to continued inputs of pollutants from the catchment over the next century.
- Predicted climate influence on the recovery from acidification at Lochnagar is more complex. Whilst an increase in winter temperatures may be expected to alleviate the delivery of nitrate and labile aluminium to the loch, higher precipitation would result in a smaller proportion of deposited acidity being buffered before reaching the loch water. Furthermore, increased air temperature and decline in non-marine sulphate deposition is expected to increase the delivery of DOC to the loch. Not only might this increase the levels of pollutants, with a high DOC affinity, entering the loch from the catchment but as a weak acid, it would counter the effects of nitrate and non-marine sulphate decline to an as yet unknown extent.
- While any future reductions in the emission of atmospheric pollutants will undoubtedly help reduce the total flux of pollutants to remote lakes, especially in the long-term by not replenishing the catchment store, the pollution of catchment soils already caused by long-range atmospheric transport over the last 150 years will remain as a legacy of human profligacy and will impact the chemistry and biota of these sites at least into the 22nd century.

ACKNOWLEDGMENTS

We would like to acknowledge the ongoing support for studies at Lochnagar from the the UK Department of Environment, Food and Rural Affairs (via the UK AWMN and 'Freshwater Umbrella' programmes) and from the EU via the MOLAR (Measuring and modelling the dynamic response of remote mountain lake ecosystems to environmental change. A programme of mountain lake research, 1996 - 1999 EU contract ENV4-CT95-0007) and EMERGE (European mountain lake ecosystems: Regionalisation, diagnostics and socio-economic evaluation, 2000 - 2003 EU contract EVK1-CT-1999-00032) research projects. We are particularly grateful to Jo Porter for many years of field support and to the Balmoral Estate Office for facilitation of our research efforts. We also thank Roger Flower and Annette Kreiser for the use of their diatom data, Ron Harriman (SOAFED) for the water chemistry data, and Anders Wilander for useful comments on the manuscript. Elanor McBay of the UCL Geography Department Cartographic Office produced some of the figures.

REFERENCES

Agustí-Panareda, A. & R. Thompson (2002) Reconstructing air temperature at eleven remote alpine and arctic lakes in Europe from 1781 to 1997 AD. *J. Paleolimnol*., 28: 7-23.

Baker, S.J. 2001. *Trace and major elements in the atmsophere at rural locations in the UK: Summary data for 1999*. National Environmental Technology Centre Report number AEAT/R/ENV/0264: 80 pp.

Battarbee, R.W., V.J. Jones, R.J. Flower, P.G. Appleby, N.L. Rose & B. Rippey. 1995. Palaeolimnological evidence for the atmospheric contamination and acidification of high Cairngorm lochs, with special reference to Lochnagar. *Bot. J. Scotl*., 48: 79-87.

Bindler. R., I. Renberg, N.J. Anderson, P.G. Appleby & N.L. Rose 2001. Mercury accumulation rates and spatial trends in lake sediments from West Greenland. *Env. Sci. Tech*., 35: 1736-1741

Cameron, N.G., H.J.B. Birks, V.J. Jones, F. Berge, J. Catalan, R.J. Flower, J. Garcia, B. Kawecka, K.A. Koinig, A. Marchetto, P. Sánchez-Castillo, R. Schmidt, M. Šiško, N. Solovieva, E. Štefkovál & M. Toro. 1999. Surface-sediment and epilithic diatom pH calibration sets for remote European mountain lakes (AL:PE Project) and their comparison with the Surface Waters Acidification Programme (SWAP) calibration set. *J. Paleolimnol*., 22: 291-317.

Cooper, D.M. & A. Jenkins 2003. Response of acid lakes in the UK to reductions in atmospheric deposition of sulfur. *Sci. Tot. Env*., 313: 91 – 100.

Curtis, C.J., A. Barbieri, L. Camarero, M. Gabathuler, J. Galas, K. Hanselmann, J. Kopacek, R. Mosello, U. Nickus, N.L. Rose, E. Stuchlik, H. Thies, M. Ventura & R. Wright 2002. Application of static critical load models for acidity to high mountain lakes in Europe. *Wat. Air Soil Pollut. Focus*, 2: 115-126.

Dalton, C.P., R.W. Battarbee, H.J.B. Birks, S.J. Brooks, N.G. Cameron, S. Derrick, R.P. Evershed, S.M. Peglar, J.A. Scott & R. Thompson. 2001. *Holocene lake sediment core sequences from Lochnagar, Cairngorm Mts, Scotland – UK.* Final report for CHILL-10,000. Environmental Change Research Centre, University College London. Research Report No. 77.

Evans, C.D., C. Freeman, D.T. Monteith, B. Reynolds & N. Fenner. 2002. Climate change - Terrestrial export of organic carbon – Reply. *Nature*, 415: 862.

Farmer, J.G., L.J. Eades & M.C. Graham. 1999. The lead content and isotopic composition of British coals and their implications for past and present releases of lead to the UK environment. *Env. Geochem. Health*, 21: 257-272.

Ferrier, R.C., R.C. Helliwell, B.J. Cosby, A. Jenkins & R.F. Wright 2001. Recovery from acidification of lochs in Galloway, south-west Scotland UK: 1979-1998. *Hydrol. Earth System Sci*., 5: 421-431.

Fowler, D. & R. Smith. 2000. Spatial and temporal variability in the deposition of acidifying species in the UK between 1986 and 1997. In: Monteith, D.T & C.D. Evans (Eds). *UK Acid Waters Monitoring Network: 10 year report*. ENSIS Publishing, London: 364 pp.

Freeman, C., C.D. Evans, D.T. Monteith, B. Reynolds & N. Fenner. 2001. Export of organic carbon from peat soils. *Nature*, 412: 785.

Gao, J.P., J. Maguhn, P. Spitzauer & A. Kettrup. 1998. Distribution of polycyclic aromatic hydrocarbons (PAHs) in pore water and sediment of a small aquatic ecosystem. *Int. J. Env. Anal. Chem*., 69: 227-242.

Geikle, A. 1887. *The scenery of Scotland: Viewed in connection with its physical geology*. Macmillan. London.

Giles, J. 1849. *Loch-na-Gar*. Watercolour. Mary Evans Picture Library.

Hulme, M., G.J. Jenkins, X. Lu, J.R. Turnpenny, T.D. Mitchell, R.G. Jones, J. Lowe, J.M. Murphy, D. Hassell, P. Boorman, R. McDonald & S. Hill. 2002. *Climate change scenarios for the United Kingdom: The UKCIP02 scientific report*. Tyndall Centre for Climate Change Research, School of Environmental Sciences, University of East Anglia, Norwich, UK: 120 pp.

Jones, V.J., R.J. Flower, P.G. Appleby, J. Natkanski, N. Richardson, B. Rippey, A.C. Stevenson & R.W. Battarbee. 1993. Palaeolimnological evidence for the acidification and atmospheric contamination of lochs in the Cairngorm and Lochnagar areas of Scotland. *J. Ecol*., 81: 3-24.

Kettle, H., R. Thompson, N.J. Anderson & D.M. Livingstone. 2003. Empirical modelling of summer lake surface water temperatures in south-west Greenland. *Limnol. Oceanogr*., 49: 271-282.

Krabbenhoft, D.P., M.L. Olson, J.F. Dewild, D.W. Clow, R.G. Striegl, M.M. Dornblaser & P. Vanmetre. 2002. Mercury loading and methylmercury production and cycling in high-altitude lakes from the western United States. *Wat. Air Soil Pollut.: Focus*, 2: 233-249.

Krug, E.C. & C.R. Frink. 1985. Acid rain on acid soil: a new perspective. *Science*, 221: 520-525.

Monteith, D.T. (Ed.). 2004. *UK Acid Waters Monitoring Network: 15 Year Report. Analysis and Interpretation of Results, April 1988-March 2003*. Ensis Ltd. London.

Monteith, D.T. & C.D. Evans (Eds). 2000. *UK Acid Waters Monitoring Network: 10 Year Report. Analysis and Interpretation of Results, April 1988-March 1998*. Ensis Ltd. London.

Monteith, D.T., C.D. Evans & B. Reynolds. 2000. Are temporal variations in the nitrate content of UK upland freshwaters linked to the North Atlantic Oscillation? *Hydrological Processes*, 14: 1745-1749.

Murdoch, P.S. & J.L. Stoddard. 1992. The role of nitrate in the acidification of streams in the Catskill Mountains of New York. *Wat. Res. Res*., 28: 2707-2720.

National Atmospheric Emissions Inventory (NAEI) 1999. *UK Emissions of Air Pollutants 1970 to 1999*. 13th Annual Report. Web page accessed 6th October 2003. http://www.aeat.co.uk/netcen/airqual/naei/annreport/annrep99/

Olendrzyński, K., S.Anderberg, J. Bartnicki, J. Pacyna & W. Stigliani. 1996. Atmospheric emissions and depositions of

cadmium, lead and zinc in Europe during the period 1955-1987. *Env. Rev.*, 4: 300-320.
Oslo & Paris Commissions 1994. *Calculation of atmospheric inputs of contaminants to the North Sea 1987 – 1992*. OSPARCOM: 27 pp.
Pacyna, J & K.Tørseth. 1997. Central European hot spots of air pollution. In: Gutkowski, R.M & T. Winnicki (Eds), *Restoration of forests*. Kluwer Academic Publishers: 15-35.
Playford, K. & S.J. Baker. 2000. *Atmospheric inputs of heavy metals to the North Sea: Results for 1999*. National Environment Technology Centre Report AEAT/ENV/336/ 47044102.
Rognerud, S., J.O. Grimalt, B.O. Rosseland, P. Fernandez, R. Hofer, R. Lackner, B. Lauritzen, L. Lien, J.C. Massabuau & A. Ribes. 2002. Mercury and organochlorine contamination in brown trout (*Salmo trutta*) and Arctic charr (*Salvelinus alpinus*) from high mountain lakes in Europe and the Svalbard archipelago. *Wat. Air Soil Pollut.: Focus*, 2: 209-232.
Rose, N.L., S. Backus, H. Karlsson, & D.C.G. Muir. 2001. An historical record of toxaphene and its congeners in a remote lake in western Europe. *Env. Sci.Tech*., 35: 1312-1319.
Rose, N.L., E. Shilland, H. Yang, T. Berg, L. Camarero, R. Harriman, K. Koinig, L. Lien, U. Nickus, E. Stuchlik, H. Thies & M. Ventura. 2002. Deposition and storage of spheroidal carbonaceous fly-ash particles in European mountain lake sediments and catchment soils. *Wat. Air Soil Pollut.: Focus*, 2: 251-260.
Schuurkes, J.A.A.R., C.J. Kok & C. Den Hartog. 1986. Ammonium and nitrate uptake by aquatic plants from poorly buffered and acidified waters. *Aquatic Botany*, 24: 131-146.
Stoddard, J.L. 1994. Long-term changes in watershed retention of nitrogen. In: Baker, L.A. (Ed.), *Environmental chemistry of lakes and reservoirs*. American Chemical Society, Washington DC: 223-284.
Sullivan, T.J., J.M. Eilers, B.J. Cosby & K.B.Vache. 1997. Increasing role of nitrogen in the acidification of surface waters in the Adirondack Mountains, New York. *Wat. Air Soil Pollut*., 95: 313-336.
Tranvik, L.J. & M. Jansson. 2002. Climate change - Terrestrial export of organic carbon. *Nature*, 415: 861.
Winch, S., J. Ridal & D. Lean. 2002. Increased metal bioavailability following alteration of freshwater dissolved organic carbon by ultraviolet-B radiation exposure. *Env.Toxicol*., 17: 267-274.
Yang, H., N.L. Rose, R.W. Battarbee & J. Boyle. 2001. Storage and distribution of trace metals and spheroidal carbonaceous particles (SCPs) from atmospheric deposition in the catchment peats of Lochnagar, Scotland. *Env. Pollut*., 115: 231-238.
Yang, H., N.L. Rose & R.W. Battarbee. 2002a. Distribution of some trace metals in Lochnagar, a Scottish mountain lake ecosystem and its catchment. *Sci. Tot. Env*., 285: 197-208.
Yang, H., N.L. Rose, R.W. Battarbee & J.F. Boyle. 2002b. Mercury and lead budgets for Lochnagar, a Scottish mountain lake and its catchment. *Env. Sci. Technol*., 36: 1383-1388.
Yang, H., N.L. Rose, R.W. Battarbee & D.T. Monteith. 2002c. Trace metal distribution in the sediments of the whole lake basin for Lochnagar, Scotland: a palaeolimnological assessment. *Hydrobiologia*, 479: 51-61.