Chapter 1: Modelling Past Environmental Changes Using Lake Sediment Records

1.0 Introduction

Anthropogenic pollution of the environment by trace metals has occurred for 2000 years or more whilst levels of pollution have greatly intensified during the past 200 years of industrial revolution. As a result of these activities the magnitude of the anthropogenic fluxes of many potentially toxic trace metals from the lithosphere to the atmosphere clearly exceed the natural flux between 2-14 times. Furthermore, the total toxicity of metals released into the atmosphere exceeds the combined toxicity of radioactive and organic species generated each year (Nriagu and Pacyna, 1988). A variety of techniques have been employed to measure the atmospheric deposition of trace metals. These include lake sediments (Haworth and Lund, 1984), peat bogs (Shotyk *et al*., 1998), ice sheets (Hong *et al*., 1994) and more directly, rainfall collectors (Foundation for Water Research, 1995).

Unfortunately, an atmospheric sampling programme based on rainfall collectors requires several years of data since the concentrations in precipitation may vary widely depending on the season, wind direction and rainfall (Evans *et al*., 1986). Precipitation measurements must include both the wet and dry component because the latter may contribute significantly to the total deposition (AEA, *pers. comm.*).

The study of metal concentrations along profiles has been extensively used to elucidate the pollution history of lake sediments and peat bogs (Dörr *et al*., 1991; Evans *et al*., 1986; Farmer *et al*., 1997; Hamilton-Taylor, 1988; Livett, 1988). In particular, the use of particulate sedimentation rates calculated from radionuclide profiles, in concert with metal concentration data, allows for calculation of an atmospheric pollutant flux history. In an ideal situation these natural archives have an advantage over rainfall collectors in that they can provide both a time-averaged and a time-specific record of trace metal fallout.

The major criticism of using lake sediments to measure atmospheric deposition arises from two problems (i) that some areas of the lake accumulate sediments, and their associated chemical species, faster than other parts of the lake. The result of this redistribution or ‘focussing’ of the sediments is that one or two cores from the lake may provide an unreliable estimate of the atmospheric flux (Evans *et al*., 1986), and (ii) when investigating lake-catchment systems, as a general rule they are often over simplified with many authors making the implicit assumption that the relationship between the atmospheric flux and the lake sediment record is one-to-one. That is to say that any trace metal deposition which falls on the catchment is immobile and therefore does not affect the atmospheric flux calculated from the sediment record (Borg and Johansson, 1989; Dillon and Evans, 1982; Evans *et al*., 1986; Lindburg and Turner, 1988; Blais and Kalff, 1993). On the other hand peat bogs are often referred to as collectors of solely atmospheric deposition and are not influenced by as many outside factors as lake sediments. The type of bog is very important (Shotyk, 1995) and due to their scarcity they are not as commonly investigated.

Crucial to the interpretation of these archived records are reliable dating methods, a method of correcting for sediment focussing and/or catchment inputs and a way of determining the source of pollution. Dating is commonly achieved via radioactive decay of the naturally occurring isotope of lead (^{210}Pb, $t_{1/2}$ = 22.3 yrs). Artificial radionuclides such as caesium-137 (^{137}Cs) and americium-241 (^{241}Am) from the atmospheric testing of nuclear weapons can provide invaluable supporting evidence. Since the geochemistry of ^{210}Pb is identical to that of lead it has been suggested that ^{210}Pb is a useful environmental tracer of atmospheric lead (Benninger *et al*., 1975; Dominik *et al*., 1984). The use of Pb/^{210}Pb ratios in lake sediments for correcting for post-depositional processes has been shown to provide a reasonable estimate of lead deposition (Dörr *et al*., 1991; Evans *et al*., 1986). The source of lead pollution is normally attributed using the unique chemical property of isotope ratios (Chow *et al*., 1975). Each lead ore deposit has its own characteristic isotopic composition which is fixed during mineral genesis, *e.g.* petrol originating from different countries shows a variety of signatures. Lead produced by automobile exhaust (^{206}Pb/^{207}Pb = 1.06-1.12) is clearly different from lead coming from other anthropogenic (^{206}Pb/^{207}Pb = 1.18-1.19) and natural sources (^{206}Pb/^{207}Pb = 1.20-1.30) (Petit *et al*., 1984).

In this study, lake sediments and peat bog archives (termed indirect archives of deposition as opposed to direct measurements from rainfall deposition) from a localised site in Cumbria's Lake District known to be affected by catchment inputs, have been used firstly to investigate and reconstruct past changes in lake sediment lead, zinc and copper (Pb, Zn, Cu) depositional fluxes using a combination of stable and radioactive lead isotopes.

Secondly, the data collected is used to investigate through numerical models the relationship between lake sediments and atmospheric flux which is hoped will achieve a greater understanding of the limits of atmospheric flux history calculated from trace metal sediment profiles.

1.1 Data Collection

The site chosen for this study is a relatively well-researched lake in the English Lake District, Blelham Tarn. This site has been extensively worked on by the University of Liverpool's Environmental Radioactivity Research Centre (ERRC) and this study's collaborative partner the Institute of Freshwater Ecology (IFE). Measurements of trace metals, radionuclides and stable lead isotopes were carried out on lake sediments, peat bogs, soils and rainwater. These are supplemented by previous (AEA, *pers. comm.*; Hilton *et al*., 1985; Smith, 1993; Smith *et al*., 1997) and continuing (Appleby *et al.*, 1999) work in this area. The results were used to calculate anthropogenic atmospheric fluxes to the lake. An understanding of the processes affecting the lake archive and flux record necessitated a comprehensive study of the history of catchment inputs to the lake.

Catchment inputs were investigated using a variety of techniques including lead tracers, lead isotope ratios, soil metal concentration profiles and direct deposition of both trace metals and radionuclides.

Monitoring of direct deposition involved the establishment of a year long ^{210}Pb rainwater sampling program operated just outside the Blelham basin.

1.2 Modelling

The simple model of the lake catchment system as proposed by Appleby (1997) is used to assess atmospheric fluxes calculated from lake sediment records. The model is based on the morphometric characteristics of the lake and on the fraction of contaminant which is attached to the particulate phase. In the model, a certain fraction of an input of pollution to the lake is transferred to the lake sediments via settling of suspended particles, the rest is lost to the outflow.