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British and Scandinavian lake sediment records of carbonaceous particles from fossil-fuel combustion

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Spheroidal carbonaceous particles are emitted to the atmosphere during oil and coal combustion. The sedimentary record of these particles has been analysed for six Scottish, two Norwegian and one Swedish lake. Concentration profiles in the sediments parallel fuel-consumption trends. There are also large differences in carbonaceous particulate concentrations indicating geographical differences in loading of air pollutants from fossil-fuel combustion.

Introduction

During oil and coal combustion, particulate matter together with SO_2, NO_x, polycyclic aromatic hydrocarbons (PAH) and metals are emitted into the atmosphere. Most of the particulate matter emitted from oil combustion, and some of the particles from coal combustion, are carbonaceous particles. The carbonaceous particles analysed in this investigation are more than 5–10 μm in diameter, black, porous and spheroidal (figure 1*a*). They are formed when fuel drops or particles are incompletely burnt and the volatile components vaporized, leaving a spheroidal skeleton of nonvolatile elemental carbon (McCrone & Delly 1973). As these particles are mainly composed of elemental carbon, they are chemically very resistant, well preserved in sediments, and not affected by diagenesis as are many other pollutant indicators e.g. sulphur (Holdren *et al.* 1984) and zinc (Carignan & Tessier 1985). The sedimentary spheroidal carbonaceous particle (SCP) record can therefore serve as an indirect record of other air pollutants originating from fossil-fuel combustion. The particle record in lake sediment cores reflects the history of particle deposition. Such stratigraphic investigations have been done in the U.S.A. (Griffin & Goldberg 1983), Sweden (Wik *et al.* 1986) and Great Britain (Battarbee *et al.* 1988). Surface sediment samples (Renberg & Wik 1985*a*) and soil samples (Wik & Renberg 1987) have been used to detect geographical differences in deposition.

This investigation aimed to study the historical deposition pattern of SCPs in nine Surface Water Acidification Project (SWAP) lakes, and assess loading differences between these lakes. The lakes are: Loch Chon, Loch Doilet, Loch Tinker, Lochan Uaine, Lochan Dubh and Round Loch of Glenhead in Scotland, Verevatn and Röyrtjörna in Norway and Lilla Öresjön in Sweden (figure 1*b*). See Battarbee & Renberg (this symposium) for further site details.

Methods

Cores from the three Scandinavian lakes were sampled with a freeze-corer and cut in contiguous 0.5 cm subsamples (Renberg 1981). The Scottish cores were taken with a Mackereth mini-corer and 0.5 cm, occasionally 1.0 cm, subsamples taken at varying, but mostly 1 cm, intervals. Sediment preparation and counting procedures followed the method described by Renberg & Wik (1985*b*) with the samples analysed at times 50 magnification for the Scandinavian sites and times 40 for the Scottish.

Results and discussion

Spheroidal carbonaceous particle profiles have been analysed for 26 British lakes. Common features are the presence of few particles in sediments older than 1900, and a notable sharp increase in concentrations at about 1940, continuing to the surface sediment (Battarbee *et al.* 1988). The six Scottish SWAP lakes show reasonable agreement with these general features, although there is some variability between sites (figure 1*c–h*). The three northernmost lakes show a slightly reduced surface concentration, a feature recorded for the first time. This decrease indicates that these lakes may now be less exposed to pollutants from fossil-fuel combustion than about ten years ago.

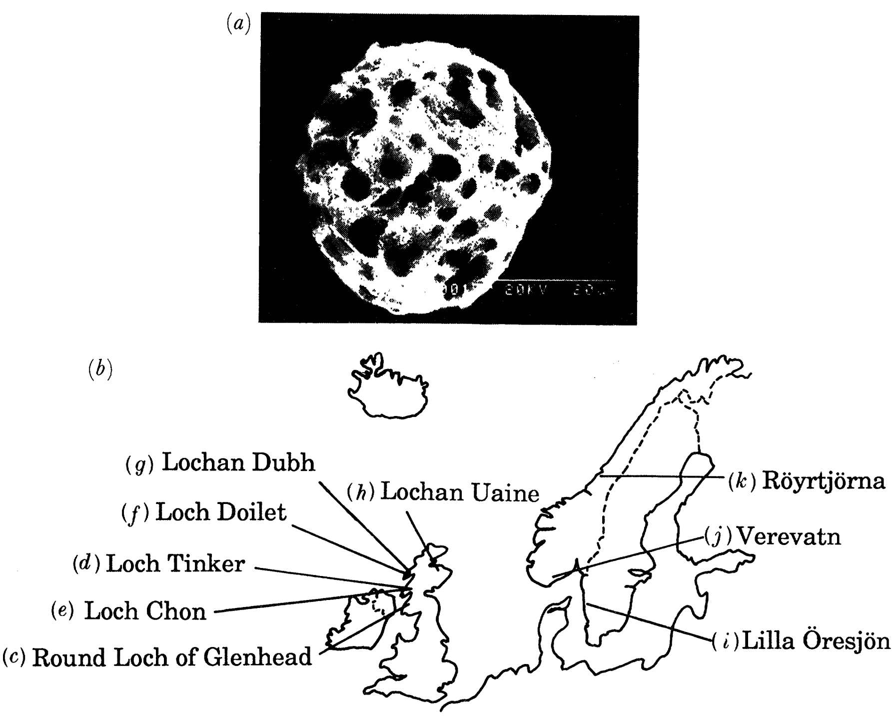

Figure 1. (*a*) A spheroidal carbonaceous particle (SCP). (*b*) Map showing the location of investigated lakes; SCP concentration (particles gds^{-1}) plotted against depth (cm) with ^{210}Pb dates (Appleby and El-Dauoshy, this symposium) for; (*c*) Round Loch of Glenhead (cored 1984), (*d*) Loch Tinker (cored 1985), (*e*) Loch Chon (cored 1987), (*f*) Loch Doilet (cored 1986), (*g*) Lochan Dubh (cored 1986), (*h*) Lochan Uaine (cored 1986), (*i*) Lilla Öresjön (cored 1986), (*j*) Verevatn (cored 1986) and (*k*) Röyrtjörna (cored 1987). Profiles indicating trends in consumption of; (*l*) oil (○) and coal (●) in U.K. power stations (Electrical Council 1988), (*m*) fuel oils (○) and coal (●) in Sweden (Statistiska Centralbyrån 1972, Statistical papers series J 1971–1978, Energy statistics yearbook 1983–1986, S. Lundberg, Swedish Petroleum Institute, personal communication) and (*n*) fuel oils (○) and coal (●) in Norway (A. G. Furuset Central Bureau of Statistics Norway, personal communication).

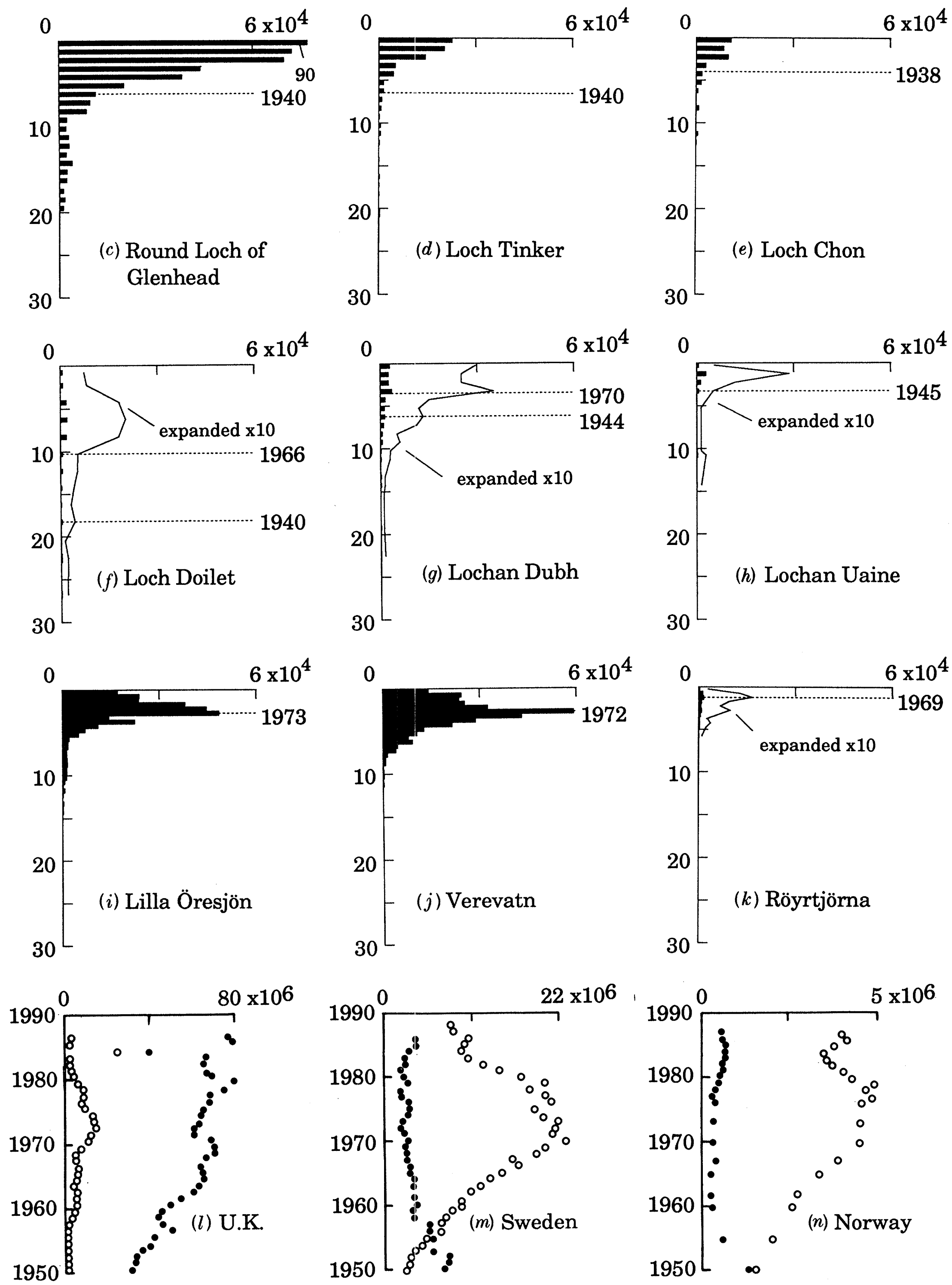

FIGURE 1. For description see opposite.

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Characteristic features of SCP profiles in Swedish lakes are low concentrations, during the second half of the 19th century, which then increase slowly until a marked rise takes place after the end of World War II, associated with increased industrial activity. A maximum is reached about 1970; later values decline towards the surface sediment (Renberg & Wik 1985*a*). The SCP profile from Lilla Öresjön shows these features, although the marked upswing in concentration has been dated to about 1960 (figure 1*i*). No SCP profiles from Norwegian lakes have been published before. The profile from Verevatn (figure 1*j*), the southern Norwegian lake, resembles the Lilla Öresjön profile. Both concentration levels and profile features are similar, with a clear concentration maximum at about 1970. The only real difference is the date for the marked concentration increase, which in Verevatn dates to about 1930. Röyrtjörna in central Norway has a very short and compressed profile because of the slow sediment accumulation rate, but even here a clear maximum concentration occurs about 1970 (figure 1*k*).

The particles analysed (greater than 5–10 μm) are considered to have rather short atmospheric lifetimes and their concentration pattern in lake sediments can be expected to reflect mainly emissions from more regional sources. However, considerable amounts of these particles have been found in very remote places in both Britain and Sweden. Studies have shown that under certain weather conditions these particles can be transported over long distances (Davies *et al.* 1984). Therefore, for the Swedish and southern Norwegian lakes, some influences from nearby European countries cannot be totally excluded.

To permit comparisons between SCP sedimentary records with combustion figures, statistics indicating trends in the consumption of fuel oils and coal since 1950 in Britain, Sweden and Norway are summarized in figure 1*l–n*. Similar statistics covering the period before 1950 have been presented elsewhere for Britain (Darley 1985) and Sweden (Renberg & Wik 1984). Energy statistics are presented in different ways in different countries and therefore it is not advisable to compare the actual values presented in figure 1*l–n*. The consumption trends, however, are comparable.

The dominant energy source in Britain has always been coal, whereas Scandinavia shifted from coal to oil after World War II. In all European countries a rapid increase in fuel consumption began after World War II. In Britain the post-war increase continued more or less until the present, but in Norway, and particularly in Sweden, there has been a dramatic drop in the consumption of fuel oils since about 1970. The Norwegian statistics presented in figure 1*n* include several products. However, detailed statistics from 1970, indicate that the recent decrease in oil burning in Norway is even more pronounced than figure 1*n* suggests.

There is a remarkable resemblance between sedimentary SCP concentration profiles and fuel consumption trends. All SCP profiles parallel the increased energy consumption after the war, and the recent decrease in use in Scandinavia is also reflected in the Scandinavian lake sediments. Particle fluxes have also been calculated and they exhibit similar trends as the concentration profiles. When comparing fuel statistics and the particulate sedimentary records it is, however, important to remember that growing environmental concern first led to the building of taller stacks, which decreased local fallout but spread emissions more widely. More recently control devices have been installed to reduce emission levels and improved combustion techniques have had a similar effect.

The SCP concentration values for the lakes vary, indicating regional differences in the amount of air pollutants deposited from combustion sources. Among the Scottish lakes the concentrations for Lochan Uaine, Loch Doilet and Lochan Dubh in the north are significantly lower than for Loch Chon and Loch Tinker, which are in turn lower than for the Round Loch of Glenhead, the southernmost lake. The maximum value in the Round Loch of Glenhead (93×10^3 particles g^{-1} dry sediment (gds^{-1})) is about 45-times higher than in Lochan Uaine, the lake with the lowest maximum value (2.1×10^3 particles gds^{-1}). The concentration for Round Loch of Glenhead is the highest recorded so far in Britain. In Scotland the geographical concentration pattern found for the SWAP lakes generally agrees with other work carried out (Battarbee *et al.* 1988), with the exception of Lochan Uaine, which has a low particle concentration compared with other lakes from the same area. However, the trace metal concentrations (Rippey, this symposium) are also very low for this lake and the fact that it is a very remote, high-altitude lake might explain these results.

Among the Scandinavian lakes the particle concentration levels for Lilla Öresjön and Verevatn are similar although the maximum concentration is a little higher in Verevatn (59×10^3 compared with 48×10^3 particles gds^{-1}). Both lakes have high SCP concentrations compared with other sites previously studied in Scandinavia. The maximum concentration in Röyrtjörna is low (1.6×10^3 particles gds^{-1}), about 35 times lower than in Verevatn. The SCP concentration of the surface sediment from Lilla Öresjön fits well into a SCP map for Sweden (Wik & Renberg (unpublished)). The map is based on analyses of more than 100 lakes and shows a good agreement with maps of atmospheric sulphate deposition. The concentration in Röyrtjörna is comparable to lakes from low deposition areas in N. Sweden and N. Scotland.

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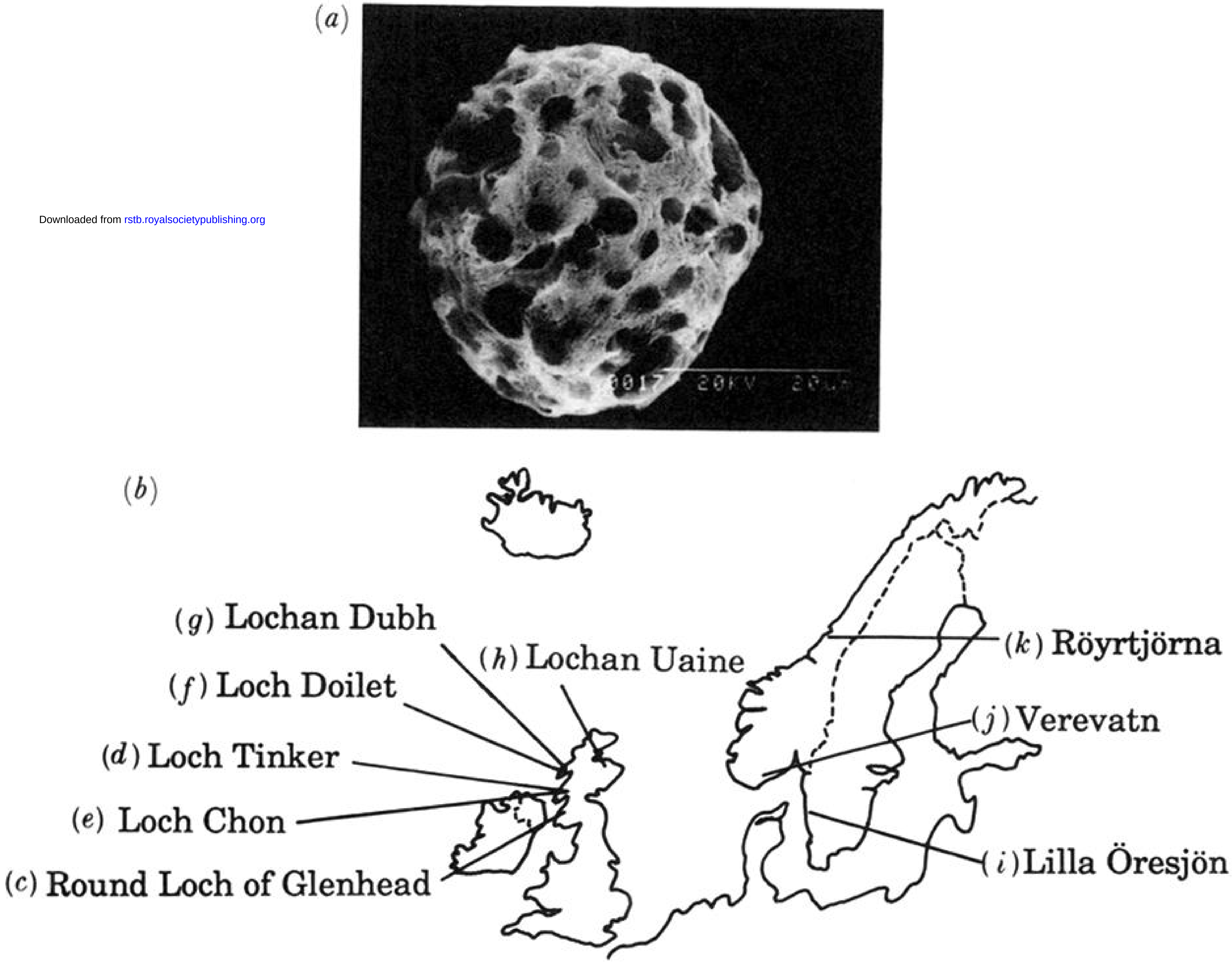

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