Geophys. J. Int. (1994) **116**, 683–687

Influence of climate on the radioactivity of lake and sea sediments: first results

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Accepted 1993 August 2. Received 1993 August 2; in original form 1992 June 2

SUMMARY

Low-level nuclear counting has been carried out on sediment cores from Lake Bouchet in France and from the Black Sea. The cores span the past 20 Ka. The variation of α and β particle counting rates with core depth, i.e. with time, shows features which can reasonably be associated with climatic change. The discussion is augmented by some high resolution γ ray spectrometry measurements of the specific activity of particular component radionuclides (^{214}Pb, ^{212}Pb and ^{40}K) in the Black Sea cores.

Key words: climate, radioactivity, sediments.

INTRODUCTION

To facilitate the study of long-term variations in climate (Lamb 1977), physical measurements which can be interpreted as proxy indicators of past climate are of importance. A few examples of such measurements applicable in particular contexts are: the variation of the relative abundance of ^{18}O (Shackleton, Imbrie & Hall 1983), the variation of magnetic susceptibility (Kent 1982; Robinson 1986); and the variation of the relative abundance of ^{14}C (Sonett & Suess 1984). In turn, the possible influences on past climate (or perhaps strictly the influence on the proxy indicators) can be considered. For many decades astronomers and meteorologists have looked for clues for a causal connection between solar activity and climate (Roberts & Olson 1973; Centre National d'Etudes Spatiales 1980; Runcorn *et al* 1990). Links between climate and geomagnetism have also been sought (Gnevyshev & Ol' 1971; Flohn 1974; Doake 1977; Abrahamsen 1986). A sound relationship has been established between solar activity, ^{14}C relative abundance and climate over the past 4 Ka (Lamb 1977; Stuiver & Quay 1980; Sonett & Suess 1984; Rampino *et al.* 1987).

Here we consider the depth dependence of the radioactivity of a lake sediment as a possible alternative proxy climate indicator. The leaching and transport of radioisotope-bearing minerals or clays and their deposition in the lake basin might well depend on rainfall and temperature—that is, on climate. Hence the radioactivity of a sample of sediment could be dependent on the climate at the time of deposition, especially if the catchment area of the lake has not changed with time. A similar argument has been presented (Creer, Thouvey & Blunk 1990) in relation to the concentration of magnetic minerals in a lake sediment sample which is responsible for the sample's magnetic susceptibility and which may in turn be taken as an indicator of climate, magnetic susceptibility having earlier been related to climate in marine sediments (e.g. Robinson 1986) and in loess deposits (e.g. Kukla *et al.* 1988).

Measurements of sediment radioactivity were made to test this idea of their use as a proxy climate indicator. The principal body of measurements was made on sediment from a core (B52) from Lake Bouchet in France and kindly provided by Professor K. Creer. Additional measurements were also made on sediments from two Black Sea cores.

After completion of this work, an interesting account by deMenocal, Bristow & Stein (1992) became available, relating the γ-ray log from Hole 798B in the Sea of Japan to palaeoclimatic information.

Lake Bouchet sediment measurements

Sediments from this lake have been the subject of a comprehensive multidisciplinary research programme involving sedimentological, geochemical, palynological, radiocarbon and palaeomagnetic studies (Bonifay *et al.* 1987; Creer *et al.* 1990). The 62 samples available for investigation had previously been used for palaeomagnetic measurements and each weighed about 8 g.

The anticipated dominant radioactive isotopes in the sediment are the members of the ^{238}U series, ^{232}Th series and ^{40}K; all other naturally occurring radioactive isotopes having a negligible abundance by comparison. The ^{238}U and ^{232}Th series contain both α and β particle emitters, whereas

^{40}K is a β emitter. Both α particle counting, detecting members of the ^{238}U and ^{232}Th series, and β particle counting, detecting both these series and ^{40}K in addition, were undertaken.

Alpha particle counting was by the long established ZnS(Ag) scintillation counter (Turner, Radley & Meyneord 1958; Linitzis & Galloway, 1982). Three α counters were used, intercalibrated with standard uranium ores. The background counting rate, in fact negligible, was measured using an inactive quartz sample. The sample counting times were in the range 10–60 ks to provide a standard deviation of about ±5 per cent from counting statistics

Beta particle counting was by an NE102A plastic scintillation counter, operated to count all β particles above a threshold of 116 keV, as found to be appropriate for β dosimetry (Galloway & Liritzis 1991). The sample counting times were in the range 10–30 ks to provide a standard deviation of about ±2 per cent from counting statistics.

The α and β counting rate variations with depth are shown in Figs 1(a) and (b), respectively. The two are similar to each other apart from the one point at 375 cm depth where there is a particularly high α counting rate. This point apart for the moment, both curves show a generally lower counting rate down to a depth of 100 cm, followed by an increase within 50 cm to about double the counting rate, followed by rather smaller fluctuations thereafter. That there should be a similarity between the two curves is not surprising, as decays within the ^{238}U and ^{232}Th series contribute to both, whereas the β curve also records the presence of ^{40}K.

If there were no ^{40}K present and both the ^{238}U and ^{232}Th series were in secular equilibrium, then the β counting rate for each sample should be proportional to its α counting rate. Fig. 2 shows a plot of the β counting rate against the α counting rate for the sediment samples, along with the line of proportionality determined from ores containing no ^{40}K and with the ^{238}U and ^{232}Th series in secular equilibrium. If the secular equilibrium condition applies, no point can lie below the line and the β counting rate above the line must be due to ^{40}K. All points but one lie clearly above the line and this one is the 375 cm depth measurement already noted as standing out from all the others among the α counting rate points in Fig. 1(a). Although not wishing to labour the discussion of one point, it can be noted that this 375 cm depth point in Fig. 2 is not significantly below the line (by only twice the standard deviation due to counting statistics)

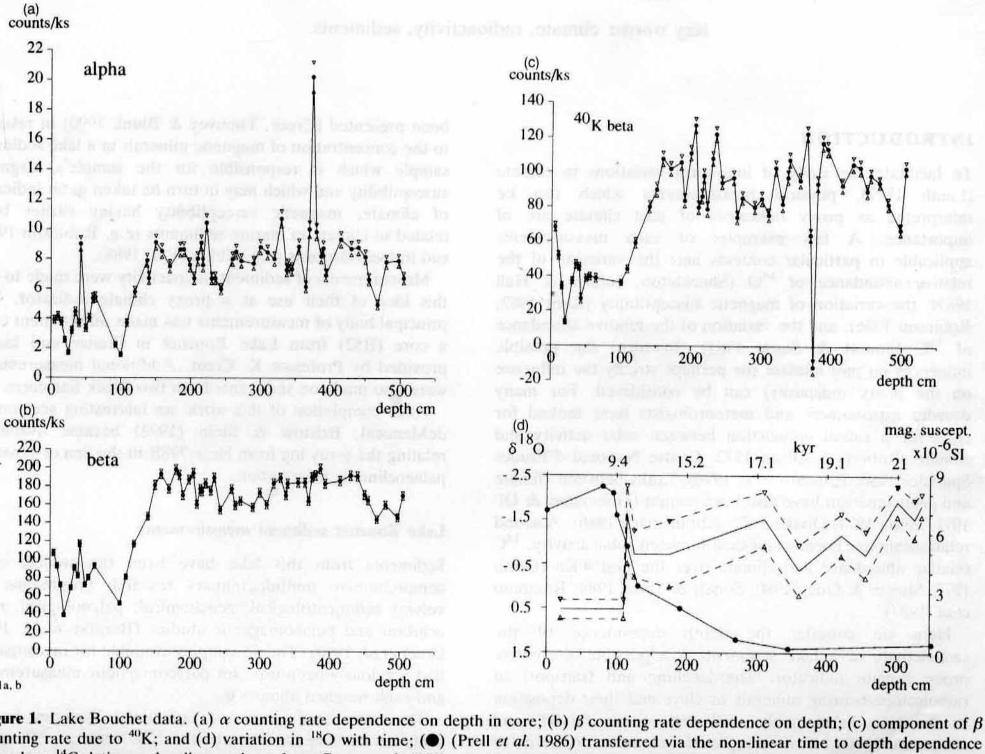

Figure 1. Lake Bouchet data. (a) α counting rate dependence on depth in core; (b) β counting rate dependence on depth; (c) component of β counting rate due to ^{40}K; and (d) variation in ^{18}O with time; (●) (Prell *et al.* 1986) transferred via the non-linear time to depth dependence (based on ^{14}C dating and pollen analyses from Creer *et al.* 1990) to relate to the above curves. Also in (d) is the trend of the variation of magnetic susceptibility (Creer *et al.* 1990); the trend is shown by the line without points whereas the broken lines indicate the range of the associated Gaussian standard errors.

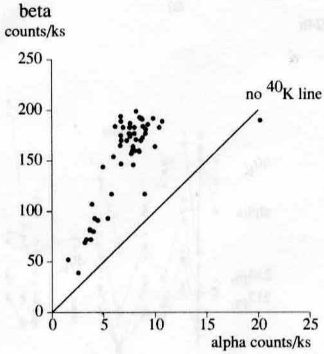

Figure 2. Plot of β counting rate against α counting rate for each of the Lake Bouchet samples. The straight line indicates the relationship for material with no ^{40}K.

and so its position could be consistent with an absence of ^{40}K rather than necessarily significant ^{238}U or ^{232}Th series disequilibrium. It should also be realized in making the comparison of β and α counting rates, whether as in Fig. 1 or Fig. 2, that as the range of α particles in the sediment is about two orders of magnitude less than the range of the β particles the α counting inevitably samples a volume of sediment which is only about 1 per cent of the volume sampled by the β counting. Counting of α particles may thus be more sensitive than β counting to inhomogeneities in the material being counted. Bearing these reservations in mind, Fig. 2 illustrates that

β rate due to $^{40}\mathrm{K} = (\beta \text{ rate}) - m\ (\alpha \text{ rate})$,

where m is the slope of the 'no ^{40}K' line. The component of the β counting rate due to ^{40}K evaluated in this way (with $m = 10$) is shown in Fig. 1(c). It shows a similar depth dependence, apart from the 375 cm point, to the α and the total β curves in Figs. 1(a) and (b).

The depth to time transform for Lake Bouchet sediments, based on ^{14}C dating and on pollen analyses, is provided by Creer *et al.* (1990) to a depth of 20 m. The transform is not linear but over the 5 m with which we are concerned it is approximated well by two linear parts, with rates of 0.1 mm yr^{-1} down to 1.4 m and thereafter 0.5 mm yr^{-1} down to 5 m. From these data age is indicated in Fig. 1(d).

The variation in the ^{18}O abundance along an ice core or deep-sea sediment core has a long history of use as an indicator of past climate (Dansgaard *et al.* 1971; Imbrie *et al.* 1984; Kukla *et al.* 1988). Following this approach, the average oxygen isotope record of Prell *et al.* (1986) for the period covered by these data is plotted in Fig. 1(d) to the same time-scale as Figs 1(a–c). It is immediately clear that the marked change in ^{18}O abundance centred at 11 Ka and associated with the end of glaciation occurs at the same time as the marked change in α, β and ^{40}K β counting rates. The major climatic event within the time span covered thus appears to be reflected in the radioactivity measurements.

Magnetic susceptibility has also been shown to relate to climate (Kent 1982; Robinson 1986; Kukla *et al.* 1988) and data for Lake Bouchet down to 20 m is provided by Creer *et al.* (1990). The trend of the Creer *et al.* (1990) magnetic susceptibility data for the 5 m depth with which we are concerned is illustrated in Fig. 1(d). Again, the major change in the level of radioactivity at a depth of between 1.0 and 1.5 m is associated with a dramatic increase in susceptibility at a depth of about 1.2 m.

Interpretation of the smaller fluctuations in radioactivity at depths between 1.5 and 5.0 m is less clear. Both the α and the β counting data have been analysed for periodic variation and periods associated with solar and terrestrial phenomena found (Xanthakis, Liritzis & Galloway 1992). The average ^{18}O data (Prell *et al* 1986) in Fig. 1(d) does not contain such short-term detail, precluding the search for finely detailed correlation. Comparison of this less varying 1.5–5.0 m region with the magnetic susceptibility data (Creer *et al.* 1990) suggests the possibility of anti-correlation, in that, for example, locally lower values of β counting are observed at about 2.8 and 4.8 m depth where the susceptibility attains locally higher values. However, this possibility does not detract from the clear correlation with the dramatic change at about 1.1 m depth noted earlier. It may be significant that the magnetic susceptibility data (Creer *et al.* 1990) is the combination of measurements on five cores covering the same depth range, so averaging out core to core variations, and it may be that similar averaging would be required to obtain the full significance of the smaller radioactivity variations.

BLACK SEA SEDIMENT MEASUREMENTS

A few small samples previously used for magnetic studies were available from two sediment cores from the Black Sea. Eleven samples were from borehole 3, Aprelefskaplost, in the age range 13–23 Ka (Bozhilova *et al.* 1989) and 13 from borehole A-34 in the age range 12–16 Ka (Kalcheva *et al.* 1989, 1990).

These samples were subjected to α particle counting by ZnS(Ag) scintillation counter, as for the Lake Bouchet samples. In addition, high resolution Ge γ-ray spectrometry (Galloway 1991a, 1991b) was undertaken with a view to separating the ^{238}U series, the ^{232}Th series and the ^{40}K activities. Because of the small sample mass available, 8 g or less, even measuring for two days gave a statistical accuracy of better than 10 per cent for only three of the component isotopes, namely ^{214}Pb in the ^{238}U series, ^{212}Pb in the ^{232}Th series and ^{40}K. The results are shown in Fig. 3. In the borehole 3 data the highest α counting rate point is not supported by high specific activities in the γ-ray measurements (which average over two to three orders of magnitude more material) so that sample inhomogeneity may be responsible, as postulated, for the high α counting point in the Lake Bouchet data. Otherwise the Black Sea measurements in Fig. 3 show the α counting and the three specific activities to vary approximately in step with one another.

Magnetic susceptibility data on the Black Sea cores (Bozhilova *et al.* 1989; Kalcheva *et al.* 1990) is also shown in Fig. 3 for comparison with the radioactivity data. Considering Fig. 3(a) (borehole 3) and putting aside the high α counting rate point at about 2.3 m discussed earlier, there is no significant variation in radioactivity with depth, whereas in the susceptibility plot only three or perhaps four

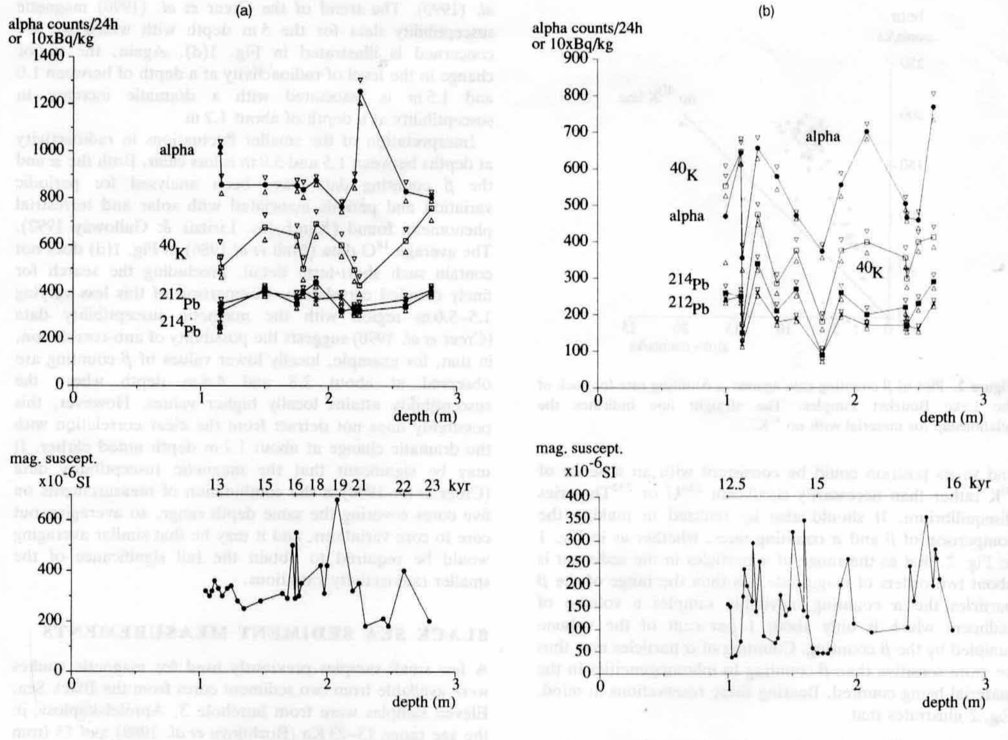

Figure 3. Black Sea data. (a) Borehole 3, α counting rate (●) and ^{214}Pb (■), ^{212}Pb (X) and ^{40}K (□) specific activities plotted as they depend on depth in the core and compared with magnetic susceptibility data from Bozhilova *et al.* (1989), which also provided the time-scale. (b) Similar data for borehole A-34 with susceptibility and dating from Kalcheva *et al.* (1990).

individual points are much higher than the general trend. In Fig. 3(b) (borehole A-34) there is some similarity between the variations in radioactivity with depth and the magnetic susceptibility variations, but with a much smaller 'peak-to-valley' ratio in the case of radioactivity.

Unfortunately, neither set of samples encompasses the end of glaciation, which was the clearly discernible feature in the Lake Bouchet data.

We may note in the context of marine sediments that Castagnoli and co-workers (1984, 1988a, 1988b, 1990) have discussed the presence of periodicities, reminiscent of solar periodicities, in the thermoluminescence signal from minerals extracted from sea sediment cores covering the past 2 Ka. As thermoluminescence is induced in crystals by ionizing radiation, predominantly from the nearby radioactivity, the relevant chain of events may involve solar influence on climate, which in turn influences the amount of weathering and deposition of radioactive material, which in turn induces the latent thermoluminescence. That is, the observed thermoluminescence variation may result from the kind of radioactivity variation reported here.

CONCLUSIONS

The idea which motivated this work, that the radioactive content of a sediment may be influenced by climatic factors controlling leaching, weathering and transport just before deposition, is supported over the limited time range of sediment available by the marked change at 11 Ka BP in accord with ^{18}O and magnetic susceptibility indications of the end of the last glaciation. Comparing measurements of α particle counting rate, β particle counting rate and γ-ray spectroscopically determined specific activities separating the ^{238}U series, the ^{232}Th series and ^{40}K gave no indication of greater sensitivity to past climate on the part of any particular radioactive isotope out of the set investigated. Sample inhomogeneity can be more of a problem with α particle counting.

ACKNOWLEDGMENTS

We are grateful to Professor K. Creer for providing the Lake Bouchet samples and for useful discussions, and to

Professor G. S. Boulton and Dr J. P. Lautridou for commenting on the results. We are grateful also to Mr H. J. Napier for valuable technical assistance. I. L. thanks Dr D. F. O. Russell, The Russell Trust and the Onassis Foundation for financial assistance.

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