INVESTIGATIONS ON FISSION PRODUCTS IN THE ACCUMULATION AREA OF AN ALPINE GLACIER (KESSELWANDFERNER, OETZTAL ALPS)

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SUMMARY

In the summer of 1964 six firn pits were dug at various altitudes in the accumulation area of the Kesselwandferner (Oetztal Alps, Austria) covering the annual layers of net accumulation from 1960/61 to 1963/64. The total β-activity and the gamma-ray spectra of continuous sets of samples were measured. From the 0.66 MeV peak in the spectra the Cs-137 contribution to the fission product mixture was determined quantitatively. The results show that Cs-137 was essentially preserved within the annual layers of net accumulation from 1961/62 to 1963/64. Thus the late summer horizons of 1961 and 1964 present on the average characteristic changes in the Cs-137 activity profile, and can be used for dating purposes.

The results of the total β-activity measurements show, in addition, firn layers of relatively high total β-activity between the late summer horizons of 1961 and 1964. In this case, a displacement of individual fission products stronger than that of Cs-137 is possible by meltwater percolation into deeper firn layers. Recent measurements of the vertical distribution of the total β-activity show a significant decrease in firn layers above the late summer horizon of 1964. This may be also regarded as a criterion for dating purposes.

RÉSUMÉ

Pendant les mois d'été de 1964 six puits verticaux ont été creusés dans la zone d'accumulation du Kesselwandferner (Alpes de l'Ötztal), à différentes altitudes, afin de prélever les couches annuelles d'accumulation nette de névé de 1960/61 à 1963/64.

L'activité β totale de ces suites d'échantillons a été mesurée. La spectrométrie gamma a permis l'examen qualitatif et quantitatif des radioéléments artificiels des échantillons.

De ce pic de 0.66 MeV des spectres, la contribution de Cs-137 fut évaluée quantitivement. Les résultats montrent que Cs-137 particulièrement a été décelé dans les couches annuelles de 1961/62 à 1963/64. Par conséquent, ce sont les surfaces de fin été de 1961 et de 1964 qui représentent en moyenne un changement caractéristique dans le profil de Cs-137 et qui peuvent servir à dater des couches annuelles.

Aussi, les résultats de l'étude de l'activité β totale montrent une haute activité β totale entre les surfaces de fin été de 1961 et de 1964. Dans ce cas-là, un déplacement de certains produits de fission dans des couches inférieures de névé est possible, déplacement qui est plus considérable que celui de Cs-137. Des études récentes de la distribution verticale de l'activité β totale montrent un retrait significatif dans les couches de névé au-dessus de la surface de fin été de 1964, qui peut aussi servir à dater des couches annuelles.

INTRODUCTION

The aim of the investigations was to find characteristic features of the fission product activity in the firn layers of a typical Alpine glacier, and, furthermore, to study the influence of various parameters, like melt- and rainwater percolation, dust content and firn stratification, on the radioactive deposits.

In the summer of 1964 six vertical firn pits were dug at various locations and altitudes in the accumulation area of the Kesselwandferner (Oetztal Alps, Austria), covering the annual layers of net accumulation from 1963/64 to 1960/61. The depths of the pits were between 5.34-6.56 m. The sites of the pits can be seen in figure 1 of W. Ambach *et al.* (1968b).

Continuous vertical sets of samples were collected from the pit wall using a corer of 42 cm^2 in cross section. Samples were taken with great care in relation to the firn stratification, such that the samples ended at the late summer horizons. About 1-2 litres of melted firn were available from each sample. Counting was performed with a methane flow counter measuring the total β-activity, and with a multichannel gamma-ray spectrometer recording the spectra.

Counting techniques

Total β-activity measurements

The samples were acidified by adding 0.1 n HCl to bring the pH-value to about 3 soon after sampling. Then they were evaporated to dryness and the residues transferred into aluminium counting-trays of 30 mm in radius. A methane flow counter of an average background value of 26 cpm and 40% counting efficiency was used for the determination of the total β-activity. The measurements were performed at the Reaktorzentrum Seibersdorf (Austria) in March 1965. K-40 standards were used for calibration.

Gamma-ray spectra

Gamma-ray spectra of the same samples on the same trays were recorded with a 256 channel LABEN gamma-ray spectrometer at the Reaktorzentrum Seibersdorf in May 1965. The detector used was a $3 \times 3''$ NaJ(Tl) crystal. The counting efficiency at an energy of 0.66 MeV was 9.4%. 128 channels were available for the data recording of each sample during 273 minutes. Because of the low intensity of the gamma-emitting radionuclides in the fission product mixture of the samples, the longest counting time was used during which temperature constancy of the spectrometer could be guaranteed.

The methods used for the qualitative and quantitative gamma-ray spectra analyses have been described by F.A. Prantl (1968).

Experimental results and discussion

Total β-activity measurements

Figure 1*a* represents the behaviour of the mean values of the total β-activity of all six pits P1-P6 in the annual firn layers from 1963/64 to 1960/61. Within each annual firn layer, moreover, the particular values of the total β-activities in the six individual pits are plotted differently. An increase of the total β-activity towards the uppermost annual firn layer of 1963/64 will be noticed. The particular values of the total β-activity show great differences within the same annual firn layer. These differences originate mainly from varying local accumulation and ablation conditions, from melt- and rainwater transport through the capillary system of the firn, and the varying retention of fission products in the layers. The latter was found to be determined by coagulation and adsorption processes, and by the incorporation of fission product compounds into micro-organisms. So, the dust content and the chemical composition of the layers influence the vertical distribution of the total β-activity (R. Reiter, 1964; F.A. Prantl, 1968).

The increase in the total β-activity at the late summer horizons of 1961 and 1962 is related to a period of intense nuclear bomb testing with high explosion energies. This period started in the fall of 1961 and stopped at the end of 1962.

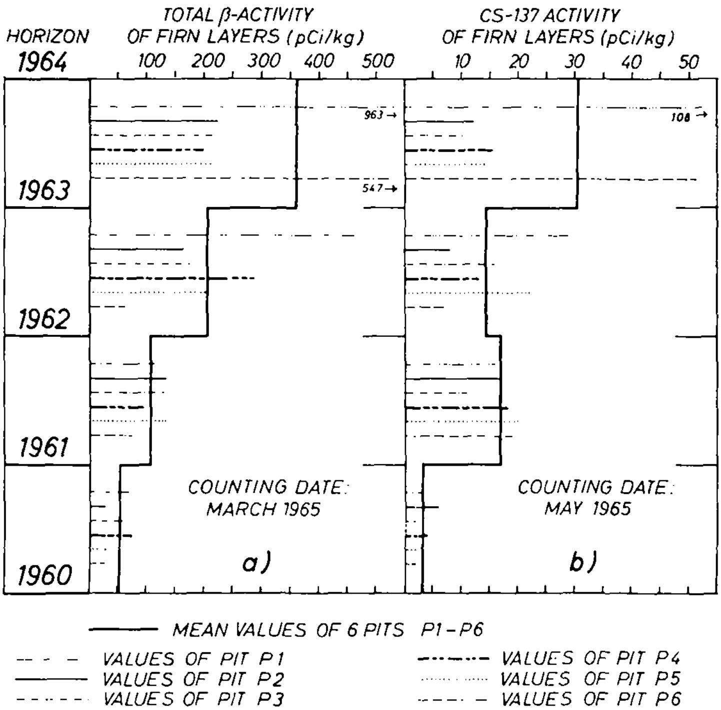


Fig. 1 — Annual fission product activities in the firn layers from 1963/64 to 1960/61 of six pits P 1 – P 6 on the Kesselwandferner (Oetztal Alps):
a) total β-activity;
b) Cs-137 activity;
firn sampling date: August 1964; the annual horizons correspond to the late summer seasons.

Figure 2*a* shows the annual mean values of the total β-activity of the air in Vienna (Hohe Warte) (*) for the periods from 1963/64 to 1960/61. For direct comparisons between the air activity and the firn activity it becomes necessary to convert the latter into the activity at the time of the deposition of the fission products on the glacier, i.e. to correct for the radioactive decay. This has been done using the Way-Wigner formula (K. Way and E.P. Wigner, 1948), and making assumptions concerning the time interval between fission and counting of the radioactive fallout. The total β-activity profile of the firn, thus corrected, is plotted in figure 2*b*. The qualitative agreement of the air activity and the firn activity profile is satisfactory. The lowest annual firn layer of 1960/61 shows too high a value for the total β-activity, mainly because of meltwater percolation from superimposed layers.

(*) We are grateful to the Zentralanstalt für Meteorologie und Geodynamik, Vienna, Hohe Warte for making the data available.

A more detailed study of the vertical distribution of the total β-activity in the individual pits shows that samples collected underneath late summer horizons generally have higher values of the total β-activity than those collected above late summer horizons. This result is to be seen from figure 3. These variations in the total β-activity correspond to seasonal effects. Only in the lowest annual firn layer of 1960/61 were they caused by a displacement of fission products in meltwater from superimposed layers and by adsorption processes in the dust layers. The seasonal effects have a two-fold origin. They are caused partly by seasonal variations of the atmospheric fallout activity, and partly by the melting conditions at the glacier surface. Melting of layers near the glacier surface leads to an enrichment of fission products (W. Ambach *et al.*, 1968a). In a similar way the annual dust layers or the pollen horizons are formed (W. Ambach and H. Eisner, 1966).

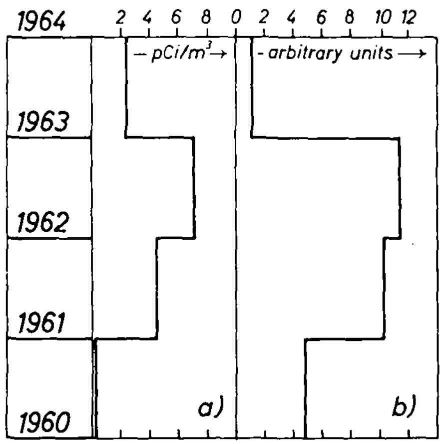


Fig. 2 — Annual mean values of the total β-activity from the late summer of 1960 to the late summer of 1964:
a) of the air in Vienna (Hohe Warte);
b) of six firn pits P 1 – P 6 on the Kesselwandferner (Oetztal Alps) corrected for the radioactive decay.

In figure 4, for example, the relation between the vertical distribution of the total β-activity and the specific residue is shown for pit P1. The specific residue is the dust content of the sample in mg per kg firn. In the lowest annual firn layer of 1960/61 the significant influence of the specific residue will be noticed. This is, because here the total β-activity was introduced chiefly by meltwater percolation from superimposed layers of comparably high total β-activity, and was retained by the dust layers. Thus, the maxima of the total β-activity are clearly attributed to the maxima of the specific residue in the annual firn layer of 1960/61.

Furthermore, a comparison between the variations in the total β-activity and the firn stratification showed that a distinct influence of ice layers could not be established. Yet, it was found that layers of depth hoar are related mainly to low values of total β-activity. The influence of dust layers and layers of depth hoar in all pits is shown by the simplified plot of the firn stratification in figure 3. The detailed firn stratification of all six pits has been given by H. Queck (1967).

Gamma-ray spectra analyses

In almost all spectra peaks were found at the energies of 0.13, 0.17, 0.43 and 0.66 MeV. The peaks at 0.13 and 0.66 MeV were complex. The qualitative analyses of the spectra showed that the 0.13 MeV peak mostly contained two lines from different nuclides, the 0.13 MeV Ce-Pr-144 line and the 0.11 MeV Sb-Te-125 line. The peaks at 0.17 MeV and 0.43 MeV presented pure lines and could be attributed to two more lines of the Sb-125 spectrum.

The 0.66 MeV peak is a characteristic feature of all spectra. It contains mainly the Cs-137 line and only at the sides of the peak comparably weak lines of Sb-125 at an energy of 0.60 MeV, and of Ce-Pr-144 at an energy of 0.70 MeV, may contribute partly to the complex 0.66 MeV peak.

From the spring of 1962 to the spring of 1964 a peak at an energy of 0.84 MeV appeared in some of the spectra indicating the presence of Mn-54. However, the contribution of this nuclide to the total β-activity remained relatively weak.

In a few spectra only lines of Ru-Rh-106 could be identified. In all cases the Ru-Rh-106 content was much lower than expected from calculations (N. Harley *et al.*, 1965). This proves that great differences may occur in the retention of various nuclides in the layers. A relatively very small retention of Sr-90 was found (G. Crozaz, 1967).

For quantitative analyses of the gamma-ray spectra the contribution of the Cs-137 line (number of counts per channel and minute, n_p) to the complex 0.66 MeV peak was determined by using a special method (F.A. Prantl, 1968). The Cs-137 activity A, can then simply be calculated from the formula :

$$A = \frac{n_p}{R \times e \times a \times f_b \times f_c},$$

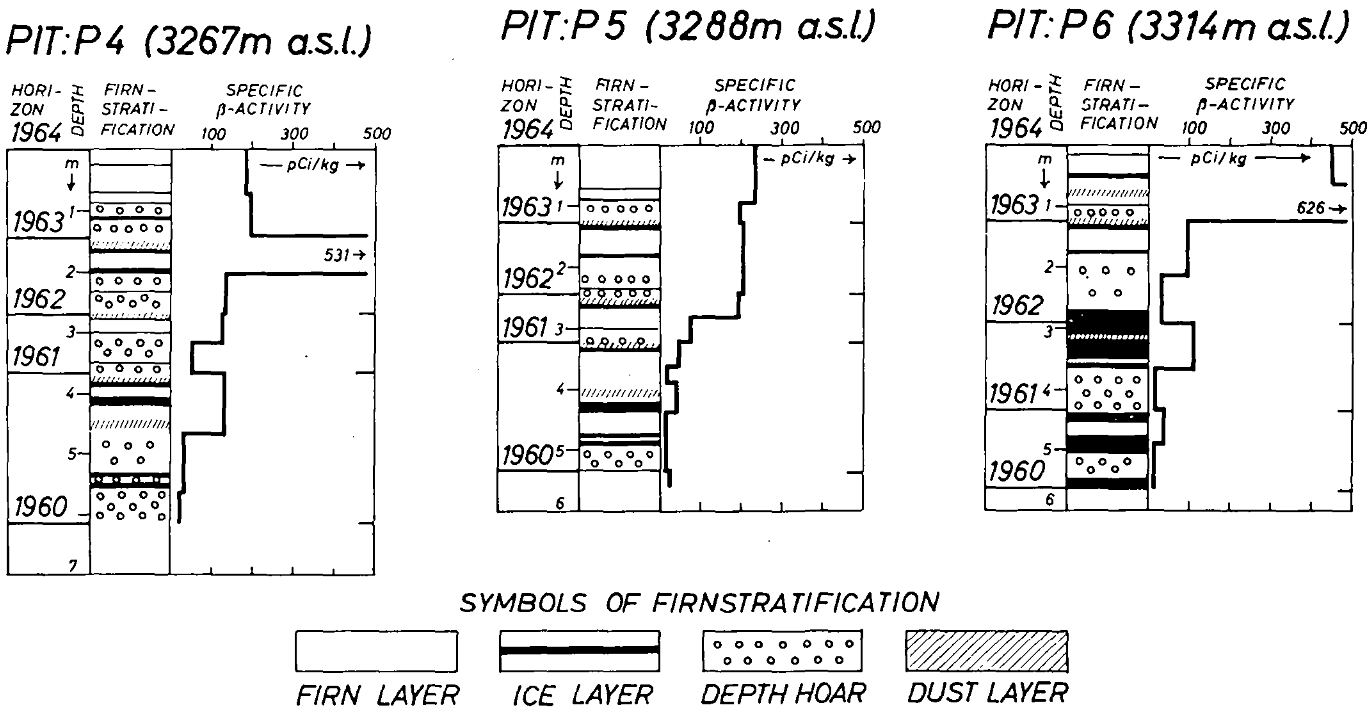
where R is the peak-to-total ratio, e the detector efficiency, a the correction for the tray size used, f_b the branching ratio and f_c a conversion factor (R.L. Heath, 1964; C. E. Crouthamel, 1960). For our experimental conditions the Cs-137 activity was $A = n_p \times 5.51$ pCi.

Fig. 1b gives the results of the Cs-137 activity measurements. The differences in the values within each annual firn layer at individual pits originate from the factors already discussed for the total β-activity.

The variations in the mean annual values of the Cs-137 activity from all six pits no longer correspond to those of the total β-activity of the air in Vienna. These variations are due only partly to Cs-137 changes in the air and the precipitation. They originate mainly from the degree of Cs-137 retention in the layers. In dust layers the adsorption and the incorporation of Cs-137 by micro-organisms is essentially high. Therefore, from 1961 to 1964 the characteristic spring maxima of Cs-137 were particularly well retained in the dust layers of the late summer horizons of those years. This was proved by the detailed Cs-137 activity profiles in each pit (F.A. Prantl, 1968). In the annual firn layer, especially of 1960/61, the Cs-137 activity showed a rapid decrease in contrast to the variations of the total β-activity in this layer. This indicates the high Cs-137 retention of the dust layer of the late summer horizon of 1961. The high Cs-137 retention of dust layers is confirmed by the fact that on the average the Cs-137 contribution to the total β-activity is in agreement with the expected values.

Conclusion

The mean annual values of the total β-activity and the Cs-137 activity show a tendency to increase from the lowest layer of annual net accumulation of 1960/61 to the uppermost layer of 1963/64. The activity measurements in the individual pits



PIT: P4 (3267m a.s.l.)
PIT: P5 (3288m a.s.l.)
PIT: P6 (3314m a.s.l.)
HORIZON 1964
DEPTH
FIRN-STRATIFICATION
SPECIFIC β-ACTIVITY
100 300 500
pCi/kg
m
1963
1962
1961
1960
531
626
SYMBOLS OF FIRNSTRATIFICATION
FIRN LAYER
ICE LAYER
DEPTH HOAR
DUST LAYER

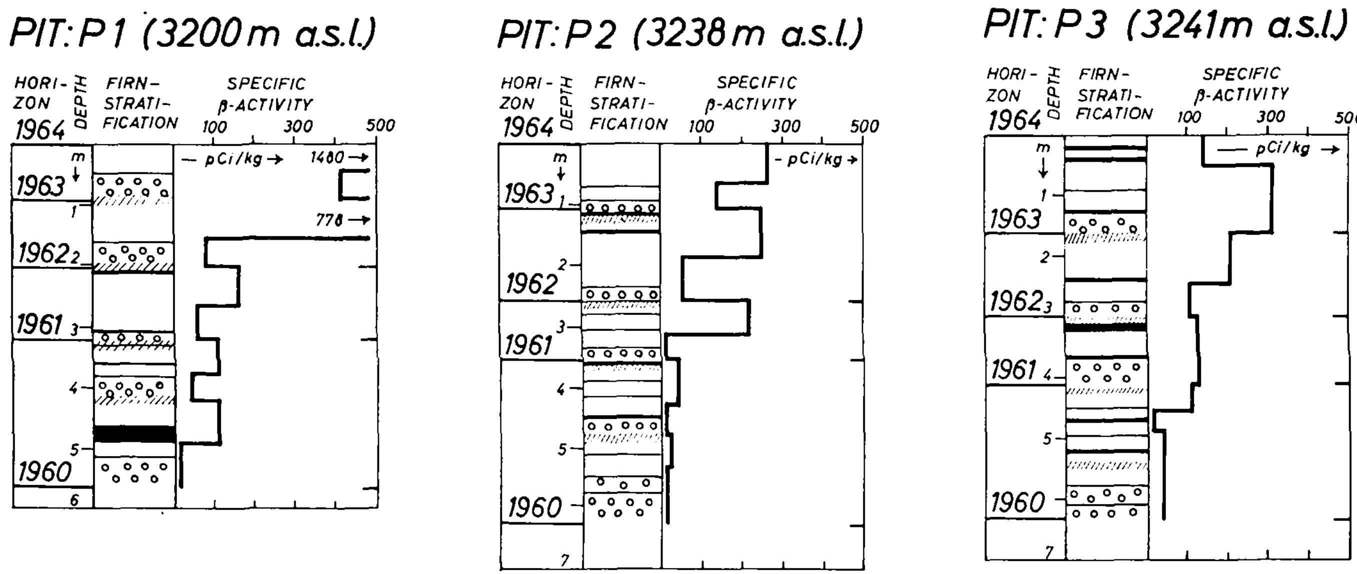


Fig. 3 — Vertical distribution of the total β-activitiy of individual samples from the pits P 1 — P 6 on the Kesselwandferner (Ötztal Alps) during the periods from 1963/64 to 1960/61:

— the annual horizons correspond to the late summer seasons;
— firn sampling date: August 1964;
— counting date: March 1965.

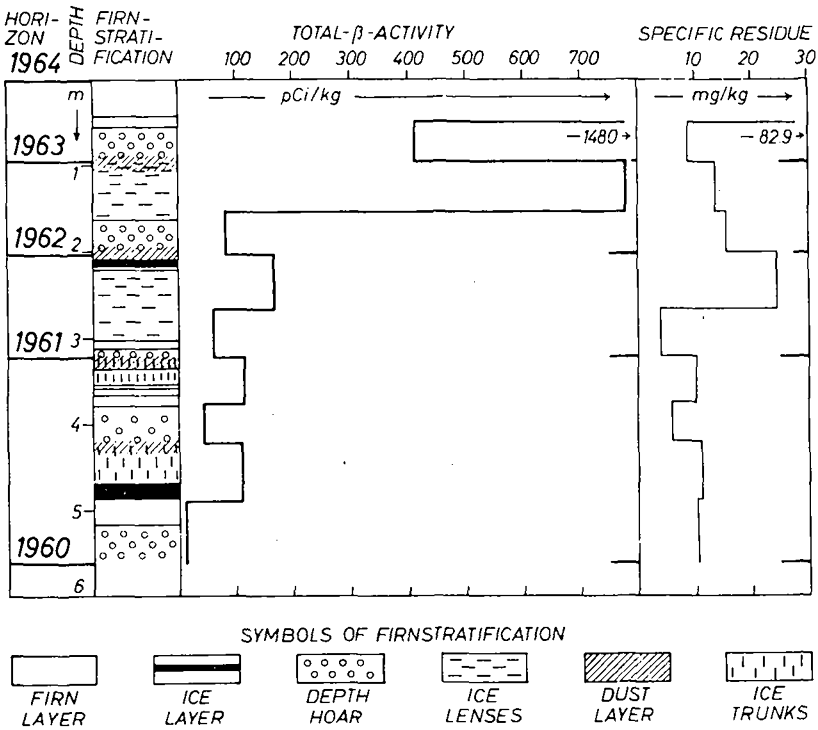


Fig. 4 — Relation of the vertical distribution of the total β-activity of individual samples from the pit P 1 on the Kesselwandferner (Oetztal Alps) to the specific dry residue and to the firnstratification:
— the annual horizons correspond to the late summer seasons;
— firn sampling date: August 1964;
— counting date: March 1965.

show variations originating from seasonal effects. In addition, especially in the annual firn layer of 1960/61, secondary processes due to meltwater percolation could be observed. Great differences in the local activity distribution were found. Therefore, generally, it is not possible to use one single activity profile for dating the layers of an Alpine glacier. The most important reasons are local differences in the accumulation and ablation conditions, melt- and rainwater transport effects, and the specific local retention in the layers. Because of the differences in the retention of various fission products in the layers, investigation of the vertical distribution of individual isotope activities becomes useful. Dust layers show a high retention for many fission products, layers of depth hoar are related mostly to comparably low activity values. No distinct influence of ice layers could be found.

In spite of all the difficulties mentioned, the method works well on Alpine glaciers under certain circumstances, namely, in regions of relatively low or high melting conditions, and if activity profiles from several pits are averaged, as for the pits on the Kesselwandferner.

Because of the good Cs-137 retention of dust layers a more distinct activity change than that for the total β-activity could be observed in the late summer horizon of 1961. Due to the high retention and the relatively long half life of Cs-137 this characteristic feature is preserved for a relatively long period, and can be used for dating purposes.

Recent activity measurements in still younger layers of annual net accumulation in pits on the Kesselwandferner and in a 12 m pit on the Stubacher Sonnblickkees (Hohe Tauern, Austria) show a distinct decrease in activity above the late summer horizon of 1964 (W. Ambach *et al.*, 1968). This is in agreement to the measurements of the air and the precipitation activity. Therefore, it can be assumed that a characteristic activity change occurs at the late summer horizon of 1964 which can be used also for dating purposes. Thus, the late summer horizons of 1961 and of 1964 mark a period of high firn activity. Moreover, the profile of the 12 m pit on the Stubacher Sonnblickkees proves that mass budget years with relatively high melting conditions lead to a strong enrichment of fission products during periods of high fission product pollution. The resulting maximum of the total β-activity can be regarded as a characteristic feature of the activity profile even in neighbouring glaciers, and can be used for dating purposes.

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