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# CONTENT OF $^{14}C$ IN MARINE MAMMALS FROM NORTHERN EUROPE

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ABSTRACT. The reservoir effect of Scandinavian sea water has been determined by dating seals and whales killed well before man's impact on the natural $^{14}C$ concentration became significant. The samples were collected at different places along the Swedish coasts and in the Gulf of Finland. They derive from AD 1657 or 1658, 1868, 1875, 1894, and 1906. The EDTA treatment of bones was used to obtain collagen free from contaminants.

An elk, originating from AD 1881 was selected for comparison. A seal from AD 1899 from the Caspian Sea was also included in the investigation.

All results have been normalized to $\delta^{13}C = -25‰$ vs PDB. The determinations yield values of the reservoir effect in agreement with earlier results obtained from shells and mammals. The final results are discussed in light of previous variations of $^{14}C$ content in the atmosphere. Using a smoother curve, the reservoir effect is slightly smaller than was hitherto believed.

An event thought to be of cosmic origin caused the count rate of both proportional and Geiger counters to increase significantly around December 4, 1978. The correction for this has been studied. The statistics for background, oxalic acid, and unknown samples, measured repeatedly after this correction, were as good as usual.

## INTRODUCTION

Although the reservoir effect of marine samples has long been known, and efforts always have been made to correct our results for this effect (eg, Feyling-Hanssen and Olsson, 1959-1960; Olsson and Blake, 1961-1962; Eriksson and Olsson, 1963; 1967; Olsson and Eriksson, 1965; Birkenmajer and Olsson, 1971), precise information was lacking. This study was made to increase our knowledge of the reservoir effect of the waters that is of special interest to Scandinavian geologists.

Apart from a few mollusk shells (Olsson, 1960; Olsson, El-Gammal, and Göksu, 1969), some recent bones affected by the nuclear bomb (species selected for differences in reservoir effect due to diet) (Olsson and others, 1974; Olsson and El-Daoushy, 1978), and one bone vertebra assumed to be recent (Olsson, Klasson, and Abd-el-Mageed, 1972; Olsson and others, 1974), samples were unavailable until recently.

### *Samples*

Details of most samples have been given by Olsson and El-Daoushy (1978). Two samples from the western coast of Sweden were submitted by C Fredén, who obtained them from Naturhistoriska Museet, Göteborg, by courtesy of J Lepiskaar. One sample *(Balaenoptera physalus)*, collected in 1875 at Morup, was dated as collagen, but the water extract from the initial step of the treatment was also dated. The second sample *(Orcinus orca)*, collected in 1868 at Lysekil, was dated as collagen. Details of this sample have been given by Malm (1871, p 79).

One sample, from the eastern coast of Sweden, a whale seen alive in 1657 but found dead in 1658, submitted by C Fredén and Å Hörnsten, was separated into two fractions during combustion. The sample was collected in Nätrafjärden, far from the natural waters for this species. Details are given by Nordlander (1934).

One sample *(Phoca foetida),* submitted by J J Donner, Helsinki, was collected in the Gulf of Finland in 1894. Another *(Phoca hispida botnica)* was submitted by G Westergren, Stockholm, and came from the small islands of Källskären in the Gulf of Bothnia in 1906.

One sample *(Phoca caspica),* submitted by Westergren from the collection in Naturhistoriska Riksmuseet, Stockholm, is included in this study, although it is from the Caspian Sea. It was from Kulalai, collected in 1899 and submitted by Lönnberg.

One sample *(Alces alces)* collected in 1881 near Trosa, was selected as a reference sample for terrestrial grazing animals and submitted by Westergren.

### *Chemical treatment*

The collagen fraction of bones should be used for bone-dating. Other fractions are also used as summarized by Olsson and others (1974) and El-Daoushy, Olsson, and Oro (1978). In the Uppsala laboratory, a method using EDTA for pretreatment as well as a version of the HCl treatment have been tested. These are described in detail by Olsson and others (1974) and El-Daoushy, Olsson, and Oro (1978). Several other methods were also tried in Uppsala, but only reliable treatments were chosen for this study. Because of the high quality of the samples and the small risk of contamination, details are not included here. The essential step at the end of the treatment is that the collagen is dissolved in slightly acidic water.

### *Activity and age calculations*

The net activity of each sample was normalized to the accepted standard $\delta^{13}C$ value of $-25‰$ on the PDB scale, to compensate for isotopic fractionation (Olsson and Osadebe, 1974). From this normalized activity, the radiocarbon age is calculated and reported (Olsson and El-Daoushy, 1978). The normal procedure is then to compare the activity of the sample when alive with the standard activity (95 percent of the activity of the NBS oxalic acid in AD 1950, with a $\delta^{13}C$ value of $-19‰$ on the PDB scale). This is done by correcting the activity of the standard sample for the decay from 1950 to the year of measurement and that of the sample from the year of death to the year of measurement. The two activities are compared to yield the $\Delta$ value ($\delta^{13}C$ and age normalized) and this value is translated into the reservoir age.

The formula used for the $\delta^{13}C$ normalization is that recommended by the editorial statement in *Radiocarbon,* although that formula is slightly incorrect (Olsson and Osadebe, 1974; Stuiver and Robinson, 1974). For this study, the formula is accurate enough since normal age determinations involve calculations using the same equation. Moreover, the error is smaller for bone samples than for shells. The procedure for the calculations is given by Olsson (1966). The changes made later in the program do not affect the basic principles of the calculations.

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tion in the atmosphere and the smaller and slower variations in the oceans. Very recent samples, eg, those collected after AD 1955, should be avoided, since the nuclear bomb effect is very pronounced in the atmosphere and it is difficult to correct for this in sea water. Since the beginning of this century, the industrial effect has been important. Mangerud and Gulliksen (1975) presented a diagram of great value for the correction of marine samples. But because of the natural variations of the $^{14}C/^{12}C$ ratio during the last four centuries, the picture is more complicated. A simple age correction is not accurate enough for samples older than 60 years. Stuiver (1978) published a diagram giving the radiocarbon age as a function of the real age for a period of 440 years. His measurement had a high degree of precision. A smoothed curve must be used for discussing marine samples. Suggested values are given below as a first step towards an improvement on the simple correction for the industrial effect (Mangerud and Gulliksen, 1975) and also applied by Tauber (1979). Mangerud and Gulliksen (1975) called attention to the possibility of correcting for $^{14}C$ fluctuations by using a modified, dendrochronologic calibration curve.

Since Stuiver's curve indicates a radiocarbon age of about 110 years within ca $\pm 40$ years from 1810 to 1920, and even within ca $\pm 20$ years from 1810 to 1890, and the activity of sea water should show smaller fluctuations than that of the atmosphere, a sample from 1875 might be dated at 110 years if no reservoir effect is present. Thus, the sample dated as U-4142 (table 1) is ascribed a reservoir effect of $415 - 110$ or $305 \pm 50$ years. Similarly, other marine animals from 1860 to 1930 would be dated at an age of ca 110 years if no reservoir effect were present. Samples about 320 to 400 years old, however, would probably be dated at ca 320 years. Samples from AD 1720 would probably be dated much older than Stuiver's curve indicates, because the ocean does not keep track of the rapid increase of the $^{14}C$ activity of the atmosphere in the 100 years from AD 1620 to 1720. The increase in $\Delta$ is ca 2 percent, *ie,* similar to the decrease on account of the industrial effect. Instead of a radiocarbon age of ca 80 years for a terrestrial sample, an age of ca 160 years seems realistic if no reservoir effect is present. Thus, the expected radiocarbon age, excluding the reservoir effect, for marine samples from 1720 to 1800 would be ca 200 years.

In using marine mammals for the determination of the reservoir effect, a small error is introduced, because of the age of the animal at death.

In this discussion, it has been tacitly assumed that the aim is to arrive at a reservoir effect that is not affected by short-term fluctuations of radiocarbon in the atmosphere. Rather, it is a measure of the ex-

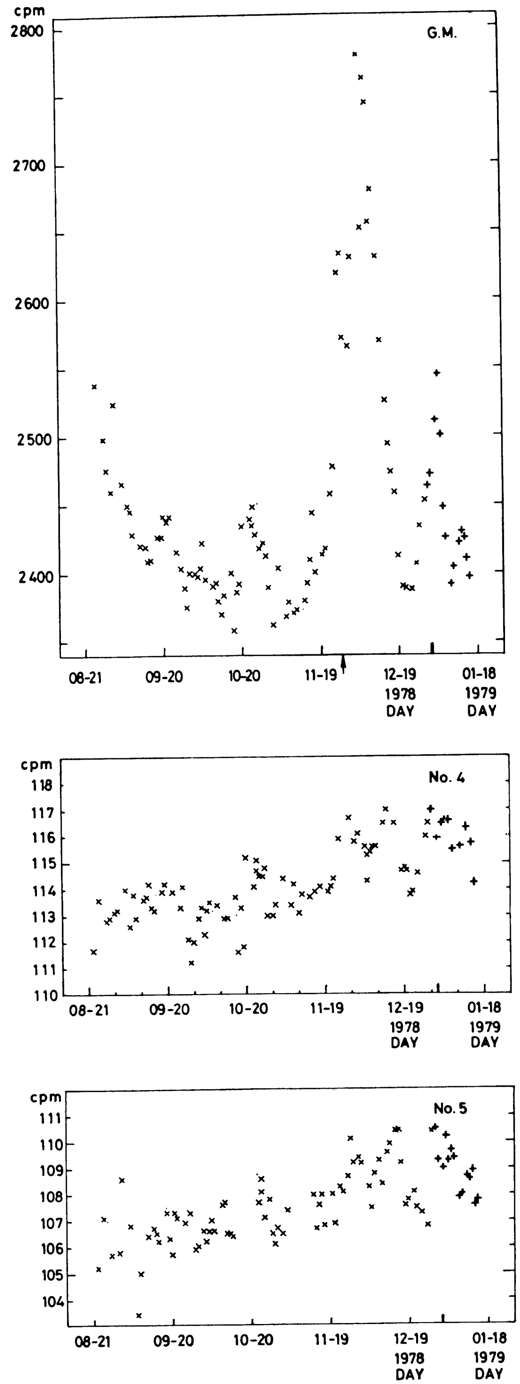


Fig 1. The muon-count rate of the Geigers and of the proportional counters (Fall 1978).

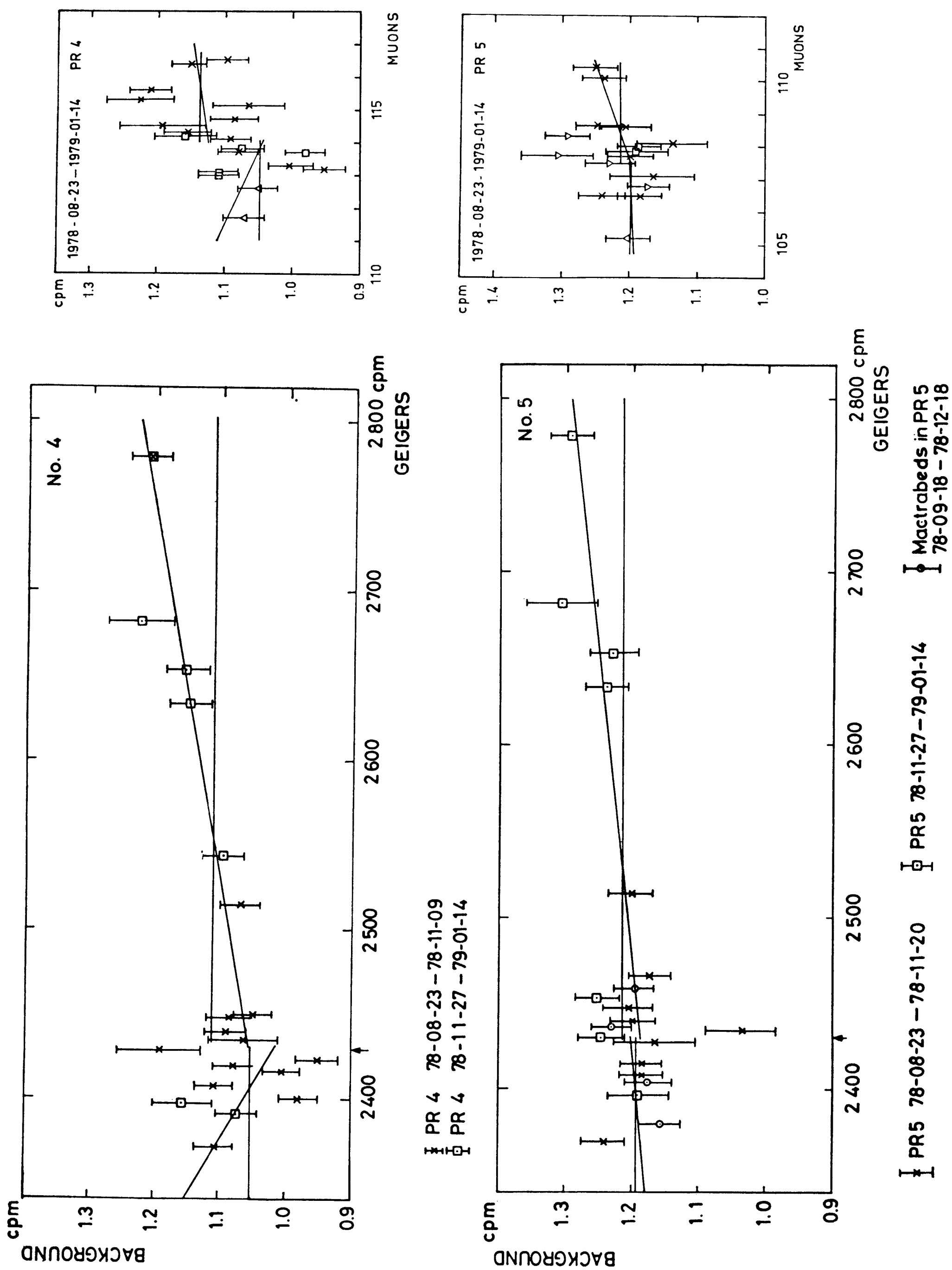


Fig 2. The background variations (Fall 1978).

### *Activity measurements*

The procedure for the measurements is, essentially, given by Olsson (1958), but some complications arose in this study. Shortly after the laboratory was completed, variations of the background outside the expected limits were noticed after making due corrections, including changes of barometric pressure (Olsson and Blake, 1961-62). A slight improvement could have been achieved by using a neutron monitor, but this desideratum could never be realized. The muon and Geiger count rates were monitored and the background plotted and analyzed, period after period. Since the barometric-pressure dependence is now well known in this laboratory, all three variables were normalized for barometric pressure before the analysis, to allow for detection of special events. Normally, no clear dependence of the background on the muons can be detected for different periods. The total picture for several periods taken together is now being studied. In the autumn of 1977, however, the Geigers indicated an unexpected peak coinciding with peaks in the muon channels and an increase of the background (Follestad and Olsson, 1979). A year later, there were similar disturbances, although the Geiger count rate increased by a much higher percentage of the count rate than the muon rate did in the two proportional counters (fig 1). There were significant background increases also (fig 2). Peaks occurred in the Geiger count rate earlier, but the background was not significantly affected. These phenomena will be discussed later. It should be mentioned that the background was corrected by using the excess Geiger count rate as well as the excess muon count rate. The four measurements agreed within statistics (fig 3).

The results of the present measurements are given in table 1 and figure 4. The mean value for marine mammals normally living to the west of Sweden is $320 \pm 35$ and, for those to the east of Sweden, $310 \pm 50$. The value for the elk, $115 \pm 95$, is not too far from zero, statistically.

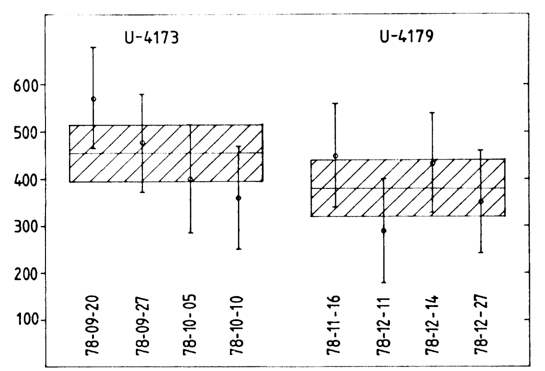


Fig 3. The age determinations for two samples (Fall 1978). The starting day for the measurement is indicated in all cases.

TABLE 1

The reservoir effect of some marine mammals, according to measurements made in Uppsala and corrections based on Stuiver's values for the activity in the atmosphere (1978), as given in fig 4, *ie,* the expected age if there is no reservoir effect. The collagen was used in all cases, except for U-2650, for which the water extract was used.

| Dating no. | Sample | Place where collected | Area | Year | True age BP | $\delta^{13}C$ ‰ (PDB) | $^{14}C$ age BP ($T_{1/2} = 5570$) | Expected $^{14}C$ age | Reservoir age |
|---|---|---|---|---|---|---|---|---|---|
| U-4142 | *Balaenoptera physalus* | Morup, Halland | W of Sweden | 1875 | 75 | −12.9 | 415 ± 45 | 110 ± 20 | 305 ± 50 |
| U-2650 | Same as U-4142 but water extract | | | | | −17.0* | 200 ± 380 | 110 ± 20 | 90 ± 380 |
| U-4168 | *Orcinus ocra* | Lysekil, Bohuslän | W of Sweden | 1868 | 82 | −12.5 | 360 ± 100 | 110 ± 20 | 250 ± 105 |
| U-4211 | Whale | Nätrafjärden Älgön Ängermanland | E of Sweden but natural area W of Sweden | 1657 or 1658 | 293 | −18.2 | 650 ± 55 | 300 ± 20 | 350 ± 60 |
| U-4167 | Same as U-4211 | | | 1657 or 1658 | 293 | −17.0 | 640 ± 55 | 300 ± 20 | 340 ± 60 |
| U-4173 | *Phoca hispida* | Källskären The Gulf of Bothnia | E of Sweden | 1906 | 44 | −18.0 | 455 ± 60 | 110 ± 20 | 345 ± 65 |
| U-4179 | *Phoca foetida* | The Gulf of Finland | E of Sweden | 1894 | 56 | −16.2 | 380 ± 60 | 110 ± 20 | 270 ± 65 |
| U-4113 | *Phoca caspica* | The Capsian Sea | | 1899 | 51 | −15.4 | 455 ± 50 | 110 ± 20 | 345 ± 55 |
| U-4219 | *Alces alces* | Trosa | Terrestrial | 1881 | 69 | −22.0 | 220 ± 90 | 105 ± 20 | 115 ± 95 |

* Assumed value.

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TABLE 2
The reservoir effect of arctic mammals re-evaluated from measurements published by Håkansson (1974) and Tauber (1979). The corrections are based on Stuiver's values for the activity in the atmosphere (1978), but a smoothed curve is used (fig 4), *ie,* the expected age if there is no reservoir effect.

| Dating no. | Sample | Place where collected | Area | Year | True age BP | $\delta^{13}C$ ‰ (PDB) | $^{14}C$ age BP ($T_{1/2} = 5570$) | Expected $^{14}C$ age | Reservoir age |
|---|---|---|---|---|---|---|---|---|---|
| K-347 | *Odobenus rosmarus* | Thule | Northern Greenland | 1915 | 35 | −10.5 | 645 | 110 ± 20 | 535 ± 50 |
| K-346 | *Pagophilus groenl* | | Western Greenland | 1886 | 64 | −16.1 | 545 | 110 ± 20 | 435 ± 55 |
| K-348 | *Ursus maritimus* | Kap Rink | Eastern Greenland | 1932 | 18 | −14.5 | 585 | 110 ± 20 | 475 ± 55 |
| Lu-779 | *Ursus maritimus* | Kap Stephensen | Eastern Greenland | 1932 | 18 | −16.2 | 515 | 110 ± 20 | 405 ± 50 |
| K-350 | *Homo sapiens* | Angmagssalik | Eastern Greenland | 1750 ± 50 | 200 | −13.0 | 715 | 200 ± 20 | 515 ± 75 |
| K-351 | *Homo sapiens* | Skjoldungen | Eastern Greenland | 1750 ± 50 | 200 | −12.6 | 705 | 200 ± 20 | 505 ± 75 |
| Lu-715 | *Ursus maritimus* | Kapp Wijk | Svalbard | 1900 ± 50 | 50 | −14.6 | 530 | 110 ± 20 | 420 ± 75 |
| K-349 | *Sibbaldus muscul* | Aaro Sund | Southeastern Jutland, Denmark | 1931 | 19 | −17.2 | 510 | 110 ± 20 | 400 ± 55 |

### *Other measurements of collagen of marine origin*

The reservoir effect, using marine animals, was studied by Tauber (1979). Also included in the discussion were fishermen and polar bears, which should have a lower activity than contemporaneous terrestrial samples (Olsson, 1979). The activity in two polar bears was measured by Håkansson (1974). Tauber used the values for the industrial effect in sea water suggested by Mangerud and Gulliksen (1975). The values given by Tauber are reproduced in table 2 and used for a re-evaluation of the reservoir effect. Marine mammals are expected to exhibit smaller differences between different areas than shells. A weighted mean value (455±25 years) may thus be of interest in further discussions and may be calculated.

### *Measurements on plant material*

An interesting series of five samples from the southern Baltic, submitted by B Berglund, Lund, has been dated in the Stockholm Laboratory (Engstrand, 1965). Four of these samples were collected between 1888 and ca 1917 and one between 1936 and 1939. All samples are thus expected to have a radiocarbon age of ca 110 years, excluding the reservoir effect. The results were age-corrected but not $^{13}C$-normalized in the original publication. A re-evaluation is given in table 3.

A series of seven samples from Kristineberg (Östlund and Engstrand, 1963) was collected between 1905 and 1959. Because of the risk of influence from the atomic bomb tests, only two results are included in this discussion (table 3). The results were age-corrected and apparently $^{13}C$-normalized in the original publication.

### *Measurements on shell samples*

The shell samples discussed by Broecker and Olson (1961), Håkansson (1969; 1970), Mangerud and Gulliksen (1975), Olsson (1960), and

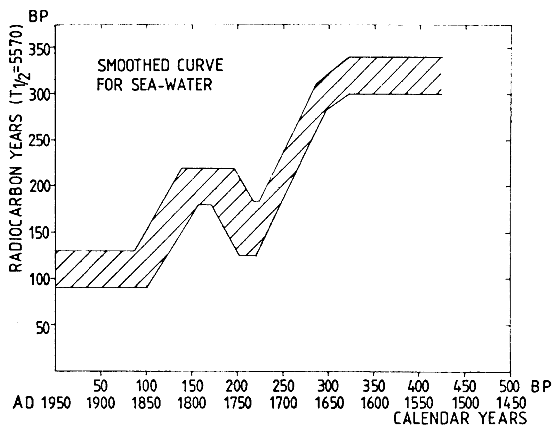


Fig 4. Suggested corrections for sea water for decay and industrial effect, taking into consideration the variations in the ratio $^{14}C/^{12}C$ given by Stuiver (1978) for the atmosphere.

TABLE 3

The reservoir effect of marine plants re-evaluated from measurements performed in Stockholm (Engstrand, 1965, and Östlund and Engstrand, 1963). The corrections are based on Stuiver's values for the activity in the atmosphere (1978), but a smoothed curve is used (fig 4), *ie*, the expected age if there is no reservoir effect.

| Dating no. | Sample | Place where collected | Area | Year | True age BP | $\delta^{13}C$ ‰ (PDB) | $^{14}C$ age BP ($T_{1/2} = 5570$) | Expected $^{14}C$ age | Reservoir age |
|---|---|---|---|---|---|---|---|---|---|
| St-1467 | *Chara horrida* | Valjeviken Sölveborgsviken | Blekinge, southern Baltic | 1888 to 1896 | 58 | −10 | 430 | 110 ± 20 | 320 ± 90 |
| St-1468 | *Chara aspera* | Same as St-1467 | | | 58 | −10 | 335 | 110 ± 20 | 225 ± 65 |
| St-1469 | *Ulva lactuca* | Southwestern Skåne | Southern Baltic | 1910 to ca 1917 | 37 | −10 | 355 | 110 ± 20 | 245 ± 130 |
| St-1474 | *Fucus vesiculosus* | Karlshamn Ronneby Tjurkö | Blekinge, southern Baltic | 1936 to 1939 | 13 | −16 | 330 | 120 ± 20 | 210 ± 75 |
| St-1517 | *Zostera marina* | Torhamns-fjärden | Blekinge, southern Baltic | 1900 | 50 | −13 | 360 | 110 ± 20 | 250 ± 65 |
| St-365 | *Fucus serratus* | Kristineberg Gullmars-fjorden | W of Sweden | 1905 | 45 | −13 | 300 | 110 ± 20 | 190 ± 60 |
| St-363 | *Fucus serratus* | Same as St-365 | | 1950 | 0 | −13 | 385 | 110 ± 20 | 275 ± 60 |

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Olsson, El-Gammal, and Göksu (1969) have been re-evaluated in a similar way as mammal and plant samples. The results are given in figure 6 for samples from Nordic waters. The 11 samples (Krog and Tauber, 1974) collected before the nuclear tests and measured in the Copenhagen laboratory were never subjected to a $^{13}C$ determination. For this summary, a value of $(-1\pm2)‰$ was assumed. The results are also given figure 6.

## CONCLUSION

Table 1 and figure 5 show no significant difference between values of the reservoir age of the water west and east of Sweden, as judged from the determinations on marine mammals made in Uppsala. The weighted mean value is $315 \pm 30$ years. Shell samples were used earlier for similar determinations for the eastern coast of Jutland and the western coast of Sweden, and the results agree within the limits of error (fig 6). The weighted mean value of these nine samples is $335 \pm 20$. Two plant samples from Kristineberg indicate a slightly lower value of the reservoir age, as do five samples from the southern Baltic, shown in table 3. The mean ages for these two groups are $245 \pm 35$ and $235 \pm 45$ years, respectively.

Shells from Norway are included in figure 6. The trend of the reservoir age stated by Mangerud and Gulliksen (1975) is confirmed even more strongly after the present re-evaluation. The values of the reservoir age are higher for samples from northern Norway than for samples from southern Norway, which, in turn, seem to be higher than those for the Swedish west coast.

The samples from the Faroe Islands and Iceland are too few to allow a final conclusion. The samples from Svalbard (Spitsbergen) need further analysis. For the time being, they are grouped in the samples from Vestspitsbergen and Nordaustlandet, with mean values for the two laboratories. Altogether (425±25), they give a value close to that for northern Norway.

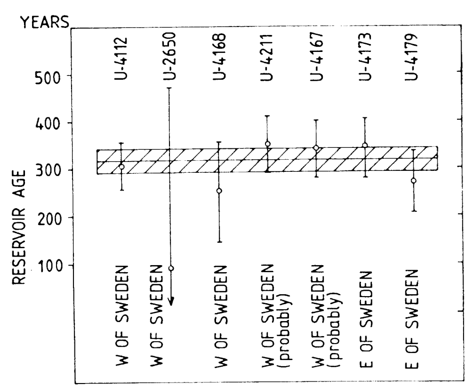


Fig 5. The reservoir age, as determined in Uppsala, using marine animals. The bars indicate $\pm$ 1$\sigma$.

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Several mammals were determined previously (table 2). The whale from Denmark indicates a similar reservoir age to that indicated by other samples (shells and mammals) from the same large area. The polar bear from Svalbard has, within the limits of error, the same reservoir age as the shell samples. The reservoir ages of the mammals from Greenland (table 2) agree fairly well with those of the shells from Greenland (fig 6), but the reservoir ages seem to vary along the coast. No significant difference is seen between polar bears, fishermen, and other samples.

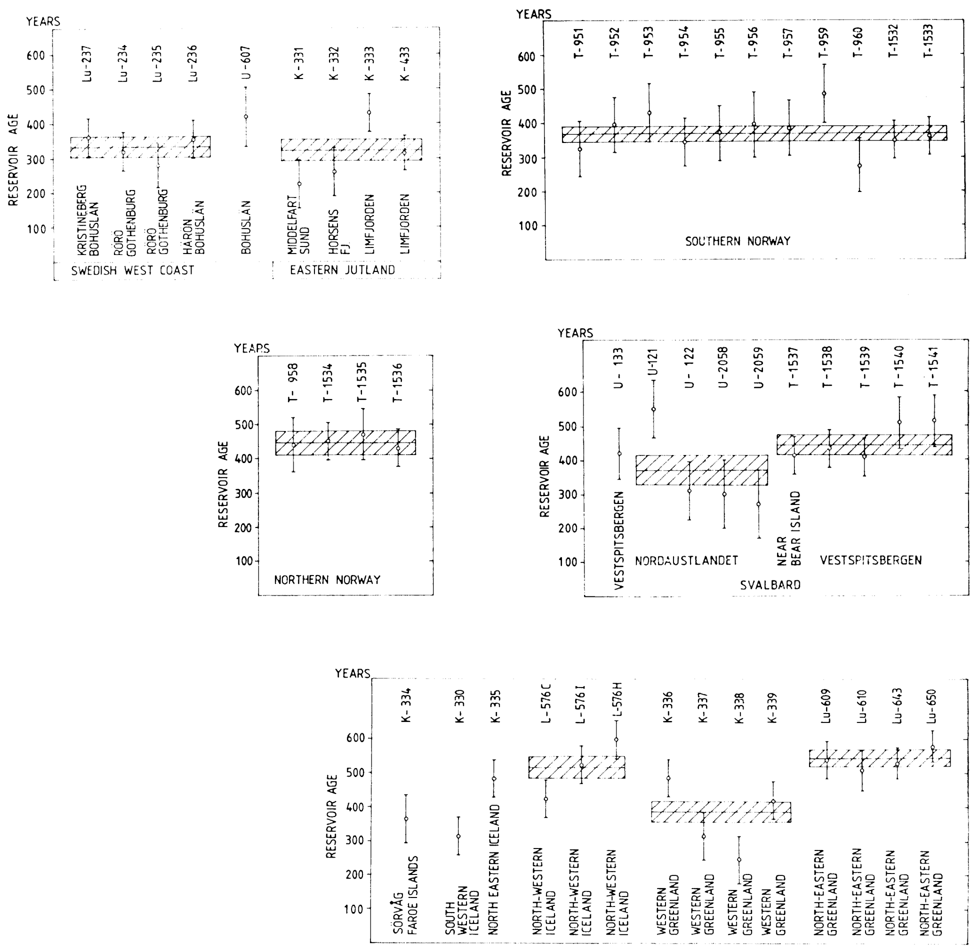


Fig 6. The revised reservoir age, as determined in five laboratories, using shells from areas near the Nordic countries. The mean value for eastern Jutland is $320 \pm 30$, the Swedish west coast $340 \pm 30$, southern Norway $370 \pm 25$, northern Norway $445 \pm 35$, Iceland $470 \pm 25$ (the statistical spread may indicate variations), Svalbard $425 \pm 25$, and Greenland $480 \pm 20$ (the statistical spread may indicate variations also for Svalbard and Greenland). The bars indicate $\pm 1\sigma$.

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