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ROUTINE CHECKS IN THE UPPSALA CONVENTIONAL ^{14}C LABORATORY TO ACHIEVE RELIABLE RESULTS

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ABSTRACT. I describe here a series of routine self-checks that the Uppsala ^{14}C laboratory performs with all measurements. We estimate all uncertainties in the physical measurement of a sample. We study long-term stability, calculate mean values for oxalic acid and background and compare expected and real statistical distributions of uncertainties. To reduce the risk of bias, the samples from each series are almost exclusively run on the same counter. Some samples are, however, run on two or more counters to check the possible bias to achieve reliable activity comparisons with other laboratories. It is always possible to trace which counter is used, since different number series are used for different counters.

INTRODUCTION

In the Uppsala conventional ^{14}C laboratory we try to reach a realistic estimate of the statistical uncertainty. Thus, all the uncertainties in the physical measurement are estimated - not only the statistical uncertainty of the decay. When the laboratory was moved from the tower of the old building of the Department of Physics to another building with three rooms and an attic above the laboratory, the barometric-pressure dependence of the background disappeared. An uncertainty for this zero value is included in the calculations. We study long-term stability and, at times, calculate mean values for oxalic-acid activity and background for series of measurements spanning one year. The real statistical distribution is compared with expected distribution estimated from all possible uncertainties in the measurements. A few background and oxalic-acid measurements should be evaluated before a result is released. Thus, final calculation of results includes measurements for a period of several months.

Because of the risk of bias, we always record the results from one counter under one number (U-number) and those from another counter under a second number, if a sample is measured in two counters. Similarly, results are reported separately from different periods when the standard values are different, even if only one counter is used. This facilitates later corrections if it is necessary to change oxalic-acid activity, background or any correction factor.

Small differences between series of samples, such as atmospheric carbon-dioxide samples from two localities can be studied without including the uncertainty of the standard, if measured in one laboratory using only one counter for a limited period. The sequence between the samples should then be chosen to alternate with the series.

Pretreatment of samples is also essential. Shell samples should be treated with special care since they are usually affected when stored for a long time in the atmosphere. This is especially apparent for shells close to the detection limit. Groundwater contamination of shells also occurs easily. Wet storage of organic samples without freezing can cause contamination by bacteria. After mechanical and chemical treatment, a suitable fraction for each sample should be chosen and the possible influence of contaminants *in situ* and later added to the sample during storage in nature should be considered.

STATISTICAL ANALYSES OF RECENT OXALIC-ACID MEASUREMENTS

We always check statistically, over periods of months or even years, long-term stability of background and oxalic-acid measurements. This test was earlier performed by merely plotting the results with error bars, calculating mean values for short periods to check any trend in the values

with time, and simply counting the values within ± 1σ; between −2σ and −1σ; between +1σ and +2σ; and outside ± 2σ. Now a simple statistical analysis is performed according to Stuiver (1982). This yields a ratio between realistic and expected sigma values (σ_R/σ_{exp}) - sometimes called the "error multiplier." Our computer program allows convenient plotting of a histogram with the measured values and curves, assuming a Gaussian distribution of expected and actual values. The sigma ratio for one counter for the greater part of 1986 was 0.96 (Olsson 1988: 203, Fig 7), for 1987, 1.04 (Olsson 1989), for 1988, 0.79, and for 1989 (until the end of August), 1.08. If 1988 and 1989 are combined, the sigma value is 0.97 (42 values measured on oxalic acid from 7 combustions). The results, based on measurements early in 1988, will be slightly adjusted because of the statistical distribution and the mean values for the last two years.

BACKGROUND MEASUREMENTS

Olsson (1989) reports typical values for background measurements. The values for 1988 yield a sigma ratio of 1.22. The first eight months of 1989 gave 0.97, after a minor data selection. Data selection is justified when treating results from high or low voltages on the plateau of the characteristics of a counter. The periods with a stable background are usually shorter than one year but exceptions are documented.

One of our basic corrections for samples and background is normalization to a chosen standard working voltage determined as a certain voltage over that for which half of the count rate for muons on the plateau is received (Olsson 1958, 1966). We always measure and normalize to a standard barometric pressure the muon count rates in the proportional and Geiger counters. The background measurements are routinely compared with these values and we normally see no correlation. Exceptions occurred in the autumns of 1977 and 1978 (Follestad & Olsson 1979; Olsson 1980) when muon and background count rates rose. Consequently, we specially treated the results and included in the calculations increased uncertainties for the background, over the statistical ones derived from the count rate.

Before AD 1985, background had to be corrected for varying barometric pressure, but increased shielding from building material was sufficient to remove barometric-pressure dependence when the laboratory was moved from the tower of a building to a house with three laboratory floors and an attic above. An uncertainty for this missing correlation between background and barometric pressure is still applied.

FILLING PRESSURE

About a decade ago, I discovered that the oxalic-acid values were difficult to foresee when normal filling pressure was changed ca 100mbars or more from one period to another (Olsson 1982). An analysis showed that, besides the normal correction because of the number of CO_2 molecules in the counter, another minor correction had to be made. Oxalic-acid samples, then, measured from 1972 to 1980, showed that the statistical distribution of the normalized ^{14}C activities, also adjusted for this extra pressure dependence, was of the same quality as for the measurements since 1986.

CALCULATIONS

Olsson (1966) published an early version of our computer program for the calculations and defined the principles behind the program (Olsson 1958). Olsson *et al* (1962) discussed the uncertainties in detail and Olsson (1988) described the efforts to find two correction factors for one counter after moving the laboratory to a new building.

MEASUREMENTS TO DETECT SMALL VARIATIONS OF ACTIVITY

We always calculate and report separately results obtained by measurements in two or more counters or at different times in one counter. This facilitates later corrections that may be necessary because of a shift in the background or oxalic-acid values. For slight oscillations in activities, as for tree-ring measurements for wiggle matching or for differences in atmospheric activity over the Arctic and Sweden (Olsson 1989), it is advisable to eliminate the standard-activity uncertainty. This is possible if samples from the two series are measured exclusively in one counter, and alternately. When the results are compared with results from other laboratories, the oxalic-acid measurements should be included.

Some samples are measured in two or more counters for internal checks. For example, we measured water from the Swedish YMER–80 expedition in two counters (Figs 1 & 2), studied the distribution of the differences between the results for each sample (Fig 3), together with the distribution of the oxalic-acid samples (Figs 4 & 5), and finally the mean value of the results for each sample (Fig 6). We also studied the distribution of the differences between the results from the two counters for air samples (Fig 7), searching for further evidence because there were rather few values in each statistical analysis. The international study of eight tree-ring samples (International Study Group 1982) produced another set of results, allowing a comparison between the two counters in use at that time (Fig 8). In all the cases reported here, the weighted mean value for the differences was zero within the limit of errors.

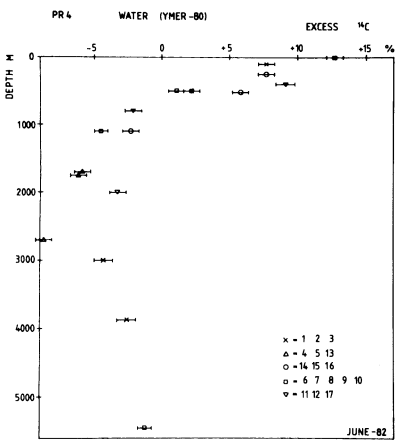

Fig 1. ^{14}C excess over the international standard for water samples collected at the YMER–80 expedition, measured with proportional counter no. 4

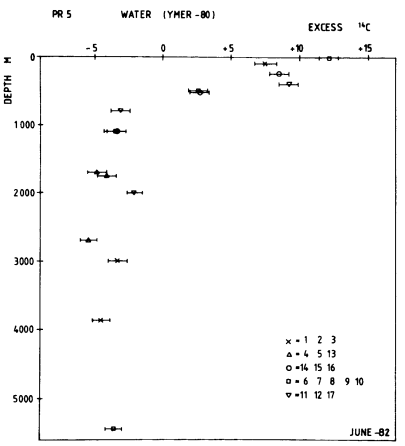

Fig 2. ^{14}C excess over the international standard for water samples collected at the YMER–80 expedition, measured with proportional counter no. 5

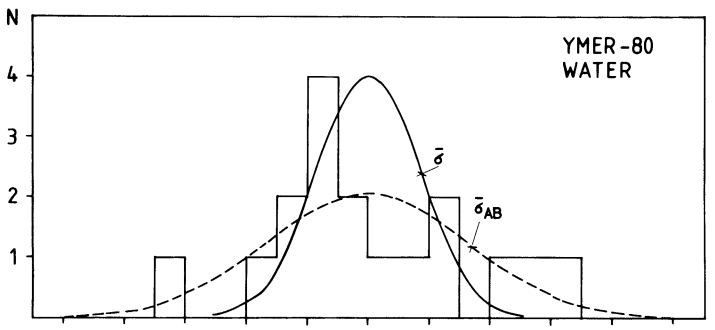

Fig 3. Distribution of the differences between the activities as measured with proportional counters 4 and 5 for 17 water samples. $\sigma_{AB}/\sigma_{exp} = 1.88$.

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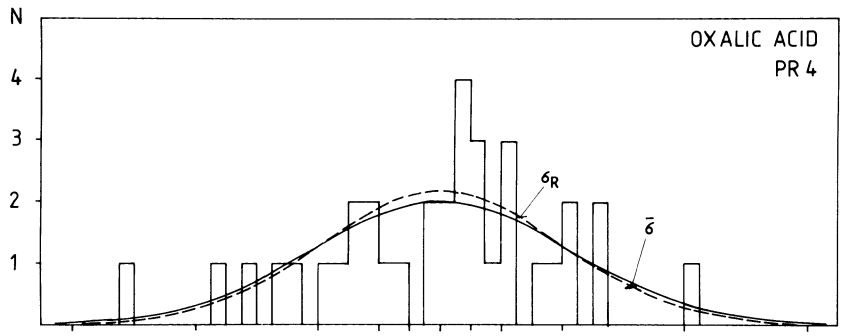

Fig 4. Distribution of the oxalic-acid measurements from 6.11.80 to 7.4.82 with proportional counter no. 4. $\sigma_R/\sigma_{exp} = 0.92$.

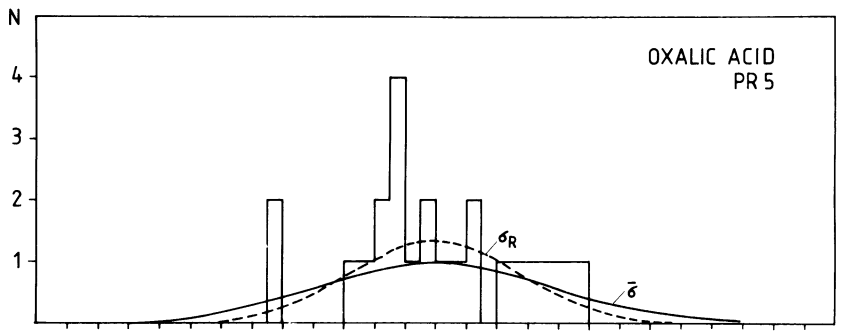

Fig 5. Distribution of the oxalic-acid measurements from 19.11.80 to 11.2.82 with proportional counter no. 5. $\sigma_R/\sigma_{exp} = 0.72$.

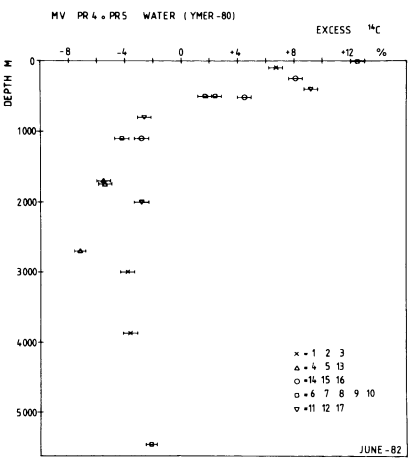

Fig 6. ^{14}C excess over the international standard for water samples collected at the YMER-80 expedition, measured with proportional counters 4 and 5. Weighted mean values are used.

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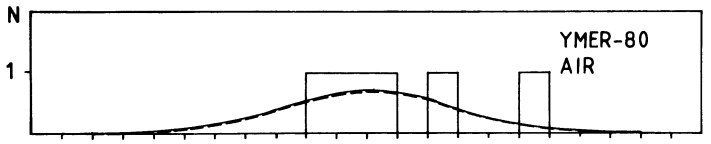

Fig 7. Distribution of the differences between the activities as measured with proportional counters 4 and 5 for 7 air samples. $\sigma_{AB}/\sigma_{exp} = 1.04$.

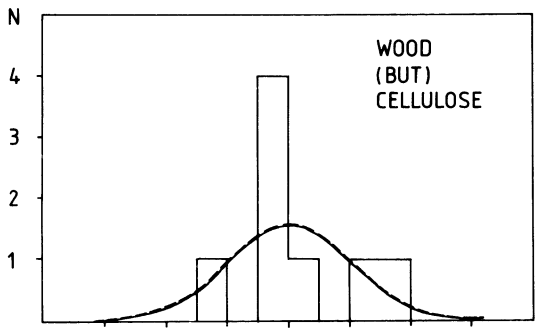

Fig 8. Distribution of the differences between the activities as measured with proportional counters 4 and 5 for 8 tree-ring samples. $\sigma_{AB}/\sigma_{exp} = 1.01$.

COLLECTION AND PRETREATMENT OF SAMPLES

Care must be taken not only with the measurement, but also with the collection of samples, choice of suitable samples, description of the context and storage and pretreatment of samples. It is advisable to remove the outer part of shells and fractionate the rest in, layer after layer, and to store shell samples in sealed vessels from the time of collection until dating (Olsson *et al* 1968). Groundwater and a humid environment may contaminate shells and long storage of old shells is especially dangerous unless special precautions are taken. Similarly, bones are easily contaminated. It is difficult to extract amino acids through conventional measurement techniques because of the lack of large-sized samples. Many pretreatment techniques are used in our laboratory. The EDTA method has yielded good results (Olsson *et al* 1974; El-Daoushy *et al* 1978). Sediments, especially those with little carbon content, are easily contaminated by old material introduced with the inorganic fraction; I have preferred a fraction soluble in NaOH (Olsson 1973, 1979, 1983, 1985, 1988, 1989b). Despite this, the reservoir effect usually causes radiocarbon ages to be significantly too old.

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