Reassessment of Uncertainty Expansion by Linear Addition of Long-term Components from Top-down Information.

**APPENDIX**

$$F^{14}C\_{sample}=F^{14}C\_{cal} \frac{\left〈\left(-\right)\left(\frac{0.975}{1+δ^{13}C\_{sample}}\right)^{2}\right〉}{\left〈\left(-\right) \left(\frac{0.975}{1+δ^{13}C\_{cal}}\right)^{2}\right〉} (A1)$$

$$dF^{14}C\_{sample}=\left\{\left(\frac{σ\_{counts}}{\left〈-\right〉}\right)^{2}+\left(\frac{σ\_{cal}}{\left〈-\right〉}\right)^{2}+\left(\frac{σ\_{bg-long term} \left〈-\right〉}{\left〈-\right〉\left〈-\right〉}\right)^{2}+\left(\frac{2σ\_{δ^{13}C\_{sample}}}{1+\left〈δ^{13}C\_{sample}\right〉}\right)^{2}+\left(\frac{2σ\_{δ^{13}C\_{cal}}}{1+\left〈δ^{13}C\_{cal}\right〉}\right)^{2}\right\}^{0.5}F^{14}C\_{sample} (A2)$$

Symbols

SE: standard error cts: counts sd: standard deviation *p*: total passes s: sampling size 〈⋅〉1 to *p*: mean

$σ\_{counts}= \left〈-\right〉 σ\_{counts, rel} $; σbg-long term is the long-term repeatability of the blank and the other σ parameters are parameters standard errors corresponding to a single measurement batch. *Rcal* and *Rsample* are the means over the cycles of each *pass*. The numerator and the denominator of Eq. A1 are weighted averages over all the *p* passes, thus they are mean of means. *Rbg* is the overall mean of the blank and it is treated as a constant in Eq. A1. For each *pass*, the blank-corrected ratio is corrected with the corresponding value of the stable isotope.

Short demonstration of Eq. A2

The final F14C (Eq. A1) can be seen as a mean of passes and its variance, leaving aside the weights, is Var(1/p ΣF) = 1/p2 Var(*F**1*+ *F**2*+… *F**p*) = 1/p2 [Var(*F 1*)+Var(*F 2*)+…Var(*F p*)] as shown in Eq. S3. The individual variances are independent to each other. The factor *p*2 comes from $Var\left(\frac{1}{p}Σ\right)=\frac{1}{p^{2}}Var\left(Σ\right)$.

 $SE(F^{14}C,ps)^{2}=\frac{1}{p^{2}}\left[SE(F^{14}C,s)\_{pass1}^{2}+SE(F^{14}C,s)\_{pass2}^{2}+SE(F^{14}C,s)\_{pass3}^{2}+…\right]$ (S3)

The partial derivatives of Eq. A1 without the operators 〈⋅〉 leads to Eq. A2 which is the variance for only one *pass* and can be simplified as Eq. S4. We were able to reproduce the derivation of Eq. A2 by calculus with the assistance of the mathematics software WxMaxima (Vodopivec et al. 2019).

*SE*(*F14C,s*)2 *= K1⋅SE*(*cts, s*)2 *+ K2⋅SE*(*cal, s*)2 *+ K3⋅SE*(*bg, s*)2*+K4⋅SE*(*d13Csam, s*)2 *+ K5⋅SE*(*d13Ccal, s*)2 (S4) where SE(*X*, *s*) is the standard error of the X parameter within one pass composed of *s* cycles.

The overall standard error is calculated by replacing Eq. S4 into each individual variance of Eq. S3. Because the squared components of Eq. S4 are linearly combined then the individual variance for each variable are linearly grouped and the overall variance is the p2-factored sum of the individual SE(X,s)2 as shown in Eq. S5

$SE(F^{14}C,ps)^{2}=\frac{1}{p^{2}}\left[K\_{1}\left\{SE(cts,s)\_{1}^{2}+SE(cts,s)\_{2}^{2}+…\right\}+K\_{2}\left\{SE(cal,s)\_{1}^{2}+SE(cal,s)\_{2}^{2}+…\right\}+K\_{3}\left\{SE(bg,s)\_{1}^{2}+SE(bg,s)\_{2}^{2}+…\right\}+K\_{4}\left\{SE(d^{13}C,s)\_{1}^{2}+SE(d^{13}C,s)\_{2}^{2}+…\right\}+K\_{5}\left\{\right\} \right]$ (S5)

In Eq. S5, each group defined by curly brackets can be replaced by the mean variance: $\frac{K}{p}\frac{\sum\_{1}^{p}Var(X,s)}{p}=\frac{K}{p}\left〈Var(X,s)\right〉=K\left[{SE\left(X,s\right)}/{\sqrt{p}}\right]^{2}=K∙SE(X, ps)^{2}$ which is the squared standard error for all the cycles *s* and passes *p* of each parameter *X*. Finally, we arrive into Eq. S6.

*SE*(*F14C,ps*)2*=K1⋅SE*(*cts, ps*)2*+K2⋅SE*(*cal, ps*)2*+K3⋅SE*(*bg, ps*)2*+K4⋅SE*(*d13Csam, ps*)2*+K5⋅SE*(*d13Ccal, ps*)2 (S6). Eq. S6 is comparable to Eq. A2 if the variables defining *K* are replaced by their respective means and the uncertainties σ are taken as the standard errors representing the variability of each parameter along the cycles and passes. In conclusion, for Eq. A1 to work as a weighted mean, every σ uncertainty in Eq. A2 has to be an overall standard error of the corresponding variable. If the final F14C is seen as a numerator mean and a denominator mean then the same conclusion is reached because the error propagation for a division is still a linear combination of SE(X,s)2.

Table A1. Number of selected data and rejected outliers

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|   |  NordTest, # accepted (rejected) | Δreplicates |  | Bias |
| N2 | nominal Fm | Pseudo *uRw* | *uRw* | Pseudo *uRw* combined | *uRw* combined | replicates *uRw* | replicates *uRw* combined |  | Pseudo bias | Pseudo bias combined |
|  | 0 | 288(97) | 171 | 17(3) | 9 | 113(58) | 8(1) |  |   |  |
|  | 0.2305 | 7(1) | 3 |  |  | 3 |  |  | 7(1) |  |
|  | 0.4114 | 66(5) | 20 |  |  | 24(1) |   |  |  66(5) |  |
|  | 0.4953 | 8 | 4 |  |  | 4 |   |  |  8 |  |
|  | 1.040\* | 16(2) | 4 |  |  | 4 |  |  | 16(2) |  |
|  | 1.34066 | 414(115) | 174 | 10 | 5 | 288(9) | 5 |  |  414(115) | 10 |
|  | 1.5061 | 6 | 3 |  |  | 3 |  |  | 6 |  |
| He | 0 | 68(19) | 41 | 8 | 4 | 35(6) | 4 |  |  |  |
|  | 0.2305 | 15 | 7 |  |  | 7 |   |  | 15 |  |
|  | 0.4114 | 13(3) | 4 |  |  | 4 |   |  |  13(3) |  |
|  | 0.4953 | 13(2) | 7 |  |  | 7 |   |  | 13(2) |  |
|  | 1.3407 | 131(12) | 38 | 10 | 5 | 35(3) | 5 |  |  131(12) | 10 |
|   | 1.5061 | 15 | 7 |   |   | 7 |   |  | 15 |  |

\*Oxa1 corrected to *δ13C=* -25‰



Fig. A1. Examples of Long-term repeatability component of the Top-down approach. Half-Gaussian histogram for Δreplicates and zero-centred histogram of blank F14C values. *uRw* is calculated from the bootstrap 1-σ uncertainty divided by the root of the sample size. Conventional *uRw* is not included.



Fig. A2. Age uncertainty vs radiocarbon age.

Table A2. Comparison of our expanded uncertainty with long-term repeatability uncertainties (standard deviation) from diverse laboratories.

|  |  |
| --- | --- |
| A2-a | F14C uncertainty  |
| F14C | this work He 10-3 *uexpand* | this work N2 10-3 *uexpand* | Ref. AWI 10-3 std. dev.  |
| blank | 0.8±0.4 | 1.0±0.6 | 0.8 |
| 0.2305 | 1.3±0.4 | 1.7±0.6 | 1.5 |
| 0.4114 | 1.7±0.4 | 2.3±0.6 | 2.7 |
| 0.4953 | 1.9±0.4 | 2.5±0.7 | 2.1 |
| 1.040 | 3.1±0.6 | 4.2±0.9 | 1.6 |
| 1.3407 | 3.8±0.7 | 5.1±0.9 | 2.7 |

|  |  |
| --- | --- |
| A2-b | F14C and 14C age uncertainty  |
| F14C | this work He 10-3 *uexpand* (yrs) | Ref. CIO 10-3 std. dev.(yrs)  |
| blank | 0.8±0.4 (2980) | 0.5 (3400) |
| 0.15 | 1.1±0.4 (62) | 0.9 (50) |
| 0.30 | 1.5±0.4 (40) | 1.2 (31) |
| 0.50 | 1.9±0.4 (31) | 1.5 (24) |
| 0.80 | 2.6±0.5 (26) | 2.0 (20) |
| 1.00 | 3.0±0.6 (24) | 2.3 (18) |

|  |  |
| --- | --- |
| A2-c | F14C Expanded uncertainty  |
| F14C | this work He 10-3 *uexpand*  | Ref. CHRO 10-3 std. dev. |
| 0.00180 | 0.8±0.4 | 0.11 |
| 0.00258 | 0.8±0.4 | 0.27 |
| 0.00316 | 0.8±0.4 | 0.76 |
| 0.1503 | 1.1±0.4 | 0.6 |
| 0.2742 | 1.4±0.4 | 1.2 |
| 1.3407 | 3.8±0.7 | 2.0 |

|  |  |
| --- | --- |
| A2-d | Expanded uncertainty  |
| F14C (yrs BP)  | this work He 10-3 *uexpand* (yrs) | Ref. FIRI 10-3 std. dev.(yrs)  |
| 0.100(18500) | 1.0±0.4(84) | 1.1(90)§ |
| 0.227(11910) | 1.3±0.4(47) | 2.1(75)§ |
| 0.565(4590) | 2.1±0.5(29) | 2.7(39)#,§ |
| 0.9906(76) | 3.0±0.6(24) | 3.9(32)\* |
| 0.100(18510) | 1.0±0.4(85) | 0.9(75)§ |
| 0.226(11940) | 1.3±0.4(47) | 2.0(70)§ |
| 0.985(120) | 3.0±0.6(24) | 4.0(33)\* |
| § Originally reported in yrs# Scott et al. 2007\* Originally reported in pMC |