DATA ANALYSIS AT THE JENA ^{14}C LABORATORY

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ABSTRACT. The Jena Analysis Code (JAC) was developed at the Jena radiocarbon laboratory for the analysis of all ^{14}C accelerator mass spectrometry (AMS) data measured there. The fundamental principles and algorithms of JAC are presented here, along with the equally important checking procedures. JAC places emphasis on the uncertainty due to background subtraction and other contributions to the statistical uncertainty of ^{14}C events.

INTRODUCTION

Data analysis, including calculating uncertainties in radiocarbon data, differs from lab to lab; however, little information is available about the full procedures applied, from the raw data to the final results. Exceptions are Donahue et al. (1990a), Rom et al. (1998), McNichol et al. (2001), and Wacker et al. (2010). A more detailed discussion of other approaches is given in the section "Comparison with Other Data Analysis Approaches." It should be stated that many publications dealing with data analysis emphasize single aspects, and some of the more general publications (Stuiver and Polach 1977; Mook and van der Plicht 1999) are in the tradition of radioactive decay counting and discuss the many conventions for expressing ^{14}C results essential to the different fields of application. In contrast, the present contribution focuses solely on the analysis of accelerator mass spectrometry (AMS) data and uses only fraction modern ($F^{14}C$; Reimer et al. 2004) for reporting ^{14}C isotope ratios. Without limiting the generality of the present approach, the equations are given for NOX (New Oxalic Acid or OxII or SRM-4990C, NIST) as the reference standard and ^{13}C as the reference isotope. The modifications required for using other reference standards or for using ^{12}C as the reference isotope are described elsewhere (e.g. Donahue et al. 1990a; McNichol et al. 2001).

The Jena AMS system, for which the Jena Analysis Code (JAC) was developed, is based on a 3MV Tandetron accelerator with simultaneous injection of all 3 carbon isotopes. It was delivered by High Voltage Engineering Europe and modified during the years of operation (see Steinhof et al. 2011 and references therein). In typical measurements at the Jena AMS lab, the target wheel of the ion source is filled with 30–50 targets of unknown samples. Additionally, targets made from the C1, C5, C6, and C7 IAEA reference material (Le Clercq et al. 1998) and from the OXI (SRM-4990B) and NOX (SRM-4990C) standards from NIST are included. For all these materials, 2 targets are put in a wheel, except for C1 (6 targets) and NOX (10 targets). The measurement of all targets in the wheel is called a "batch." The analysis of data by JAC is performed batch by batch. Note that in Jena, NOX is used as a "reference standard" based on the well-measured ratio NOX to OXI. In contrast to these, both the IAEA reference materials are "secondary standards." In the following, the term "standard" covers the secondary standards from IAEA as well as OXI and NOX from NIST.

Previously, we moved the target under the Cs beam, measured 12 different spots for 30 s each, and called this group of measurements a "run." Now, we do not move the targets, except for an initial run to clean the surface of the target, but we retain the term "run." A batch consists of these cleaning runs, which do not contribute to the results, and 10–14 subsequent runs on every target. The subdivision of the measurements in runs does not influence the results, but check 5 (see below) of the checking procedures is based on single-run results.

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Proceedings of the 21st International Radiocarbon Conference edited by A J T Jull & C Hatté
RADIOCARBON, Vol 55, Nr 2–3, 2013, p 282–293

https://doi.org/10.1017/S0033822200057386 Published online by Cambridge University Press

CALCULATION OF THE ISOTOPE RATIOS

For the derivations of the equation for the ^{14}C isotope ratios of unknown targets, first a "modern reference level" *MRL* (Seguin et al. 1994; Palonen and Tikkanen 2007) is determined from the measurements of the standards in the same batch. *MRL* with ^{13}C as the reference isotope and for the case of no background is given by

$$MRL = \frac{\frac{0.975}{1+\delta^{13}C_{std}} \cdot \frac{N_{14,\,std}}{N_{13,\,std}}}{R_{nom}} \quad (1)$$

where the subscript *std* refers to the parameter of a standard target (Table A2). The factor 0.975, equal to (1–25‰), results from normalization of the standards to $\delta^{13}C = -25‰$. R_{nom} is the nominal value of the respective standard normalized to $\delta^{13}C = -25‰$. In the case of NOX, $R_{nom} = 1.3407$ $F^{14}C$; for OXI, $R_{nom} = 1.0398$ $F^{14}C$. Note, this is different from the value normally cited for OXI ($R_{nom} = 1/0.95$ $F^{14}C$) because the latter value is normalized to $\delta^{13}C = -19‰$. The $N_{14,std}$ and $N_{13,std}$ give the number of ^{14}C and ^{13}C events measured with the standard. The units of $\delta^{13}C$ and the *N* are dimensionless; therefore, *MRL* has the inverse unit of R_{nom}. JAC would also operate if the values for the standards were given in pMC (percent modern carbon) as a unit for R_{nom}, then *MRL* would be in 1/pMC and the results would be in pMC.

The *MRL* can be determined only during the times when targets of any of the non-blank standards are measured. However, it cannot be determined during the times when the unknown targets are on. Even if all the power supplies of the accelerator are well stabilized, it is not guaranteed that the *MRL* is stable during the whole batch. For the times of the measurements of the unknown samples, the approach for *MRL* is therefore a mean value based on the measurements of all standards in a batch plus a small time-dependent deviation $DMRL_{unk}$

$$MRL_{unk} = \overline{MRL} + DMRL_{unk} \quad (2)$$

where the subscript *unk* indicates parameters of an unknown target. The ^{14}C isotope ratios of an unknown target R_{meas} are therefore given by

$$R_{meas} = \frac{\frac{0.975}{1+\delta^{13}C_{unk}} \cdot \frac{N_{14,\,unk}}{N_{13,\,unk}}}{\overline{MRL} + DMRL_{unk}} \quad (3)$$

Since all ^{14}C standards have only non-zero $\delta^{13}C$, the standard formula for $\delta^{13}C_{unk} = R_{13,unk}/R_{13,std} - 1$ (e.g. Brand 1996) is not applicable, where R_{13} is the ratio of ^{13}C over ^{12}C currents, i.e. $R_{13} = I_{13} / I_{12}$. In principle, $\delta^{13}C_{unk}$ could be deduced according to Equation 7 of Rom et al. (1998), but as can be seen in Figure 1, the fractionation is so strong that the values do not reflect the original $\delta^{13}C$ of the sample. Therefore, the effort in deducing $\delta^{13}C$ is not done in JAC. If the following are done,

- replacing the ratio of the $\delta^{13}C$ by a ratio $R_{13,unk}/R_{13,std}$;
- replacing N_{13} by $\Delta t \cdot I_{13} / (q \cdot e)$, where *q* is the charge state of the ions, *e* the elementary charge (1.602×10^{-19} C), and Δt the measurement time;
- using a series expansion for the nominator, neglecting higher-order terms;

one obtains

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$$R_{meas} = \frac{R_{13,\,std}}{R_{13,\,unk}} \cdot \frac{N_{14,\,unk}}{\Delta t \cdot I_{13,\,unk}/(e \cdot q)} \cdot \left(1 - \frac{DMRL_{unk}}{\overline{MRL}}\right) \cdot \frac{0.975/(1 + \delta^{13}C_{std})}{\overline{MRL}} \qquad (4)$$

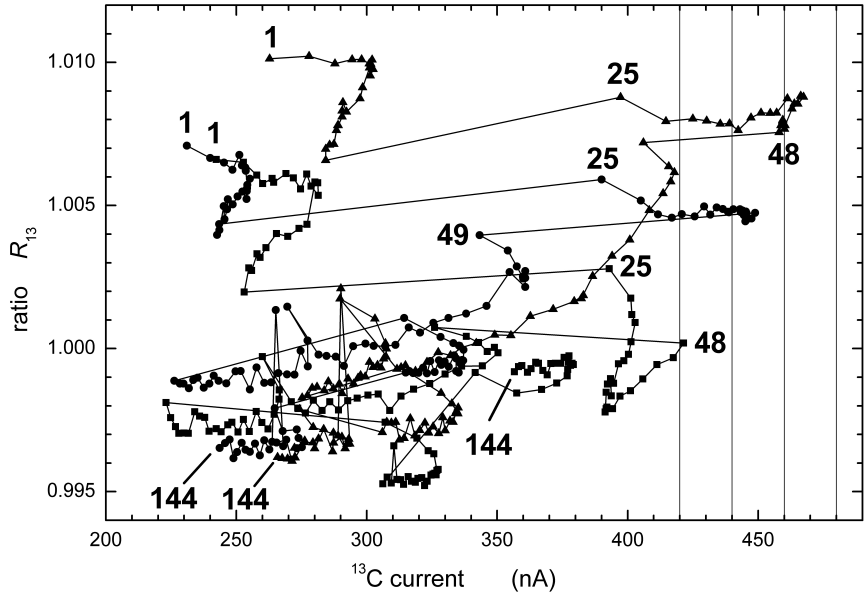


Figure 1 The ratios $R_{13} = I_{13} / I_{12}$ vs. ^{13}C current for 3 NOX targets measured during a batch. For the sake of clarity, only the values of 3 of the 10 NOX targets of the batch were plotted. The values of the different targets are indicated by different symbols. The numbers indicate the number of single measurements. Between data points 24 and 25, the Cs temperature of the ion source was increased. The data of the same targets are also shown in Figures 3 and 4. The vertical lines show 3 of the ^{13}C current intervals used in Figure 3. Note that during combustion, the 3 NOX samples gave the same $\delta^{13}C$ values within 0.05‰, which is 1/300 of the displayed range.

For further data evaluation, the terms not referring to a certain target are omitted; therefore, the ^{14}C isotope ratios are no longer given in $F^{14}C$ but in an arbitrary unit (a.u.). The ^{14}C isotope ratio of a single measurement R_i is then given by

$$R_i/\text{a.u.} = \frac{N_{14,\,i}}{R_{13,\,i} \cdot I_{13,\,i} \cdot \Delta t} \cdot \left(1 - \frac{DMRL_i}{\overline{MRL}}\right) \cdot 4.964 \text{ nA s} \qquad (5)$$

where i indicates the parameters referring to the single measurements (i.e. with Δt = 30 s). This equation is applied to all targets and standards, as well as unknown sample targets; therefore, R_i has a single subscript (Table A2). The factor 4.964 is used in JAC, but it is not of general importance since R_i is in a.u. anyway. It just should be noted that it includes an arbitrary factor 10^{12} to avoid small numbers. The units $F^{14}C$ as well as a.u. are dimensionless quantities and differ only in a constant; a.u. in JAC is approximately 0.01 $F^{14}C$.

Furthermore, it is assumed that *DMRL* is zero, i.e. that the *MRL* is constant during the batch. This is a nontrivial step and is discussed further below. Note that after setting *DMRL* = 0, all remaining parameters in Equation 5 are measured directly in each single measurement. Based on these R_i and their uncertainties ΔR_i (see next section), an isotope ratio R_{raw} for every target is calculated according to Equation A2. (Equation numbers with an "A" refer to the Appendix.)

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$$R_{raw} = \frac{\sum_i R_i/(\Delta R_i)^2}{\sum_i (\Delta R_i)^{-2}} \quad (6)$$

The term *raw* indicates that these values are still in a.u. Applying Equation 6 to the measured standard targets, a set of corresponding data pairs ($R_{nom,}$ R_{raw}) is received. From these data pairs, the parameters A and B are determined by a line fit according to Equation 7. A and B are used to transform the isotope ratios from arbitrary units to $F^{14}C$ including the background subtraction (see below).

$$R_{raw,std}/\text{a.u.} \;\; A \cdot R_{nom} + B \quad (7)$$

The units of A and B are a.u./$F^{14}C$ and a.u., respectively. The line fit is performed using the subroutine "fit" of Press and Teukolsky (1986). At present, the fit is mostly performed with the values of C1 and NOX only (16 data points), but it can also be performed with e.g. only the C5 and C6 standards. Originally, all standard targets were used in the fit, but the C6 values often had outliers. The problem with the C6 values was solved later with the revised value of Xu et al. (2010). As long as 10 targets of NOX per batch are used, there is no large gain in precision when using the other standards too, but its inclusion is an option for the future, allowing the reduction of the number of standards. The ^{14}C isotope ratio R_{meas} of an unknown target in $F^{14}C$ is then calculated from its measured value R_{raw} according to Equation 8. This transformation without the background correction is often called "standard normalization," e.g. Wacker et al. (2010).

$$R_{meas}/F^{14}C = \frac{R_{raw} - B}{A} \quad (8)$$

CALCULATION OF THE UNCERTAINTIES

The uncertainty of R_i, calculated according to Equations 5 and A1, is given by

$$\Delta R_i^2 = \underbrace{R_i^2\left(\frac{\sqrt{N_{14,i}}}{N_{14,i}}\right)^2}_{SU^2} + \underbrace{R_i^2\left[\left(\frac{\Delta(DMRL)}{DMRL}\right)^2 + \left(\frac{\Delta I_{13,i}}{I_{13,i}}\right)^2 + \left(\frac{\Delta R_{13,i}}{R_{13,i}}\right)^2\right]}_{AU^2} \quad (9)$$

where the uncertainty contribution of $\overline{MRL}$ is neglected. For values $N_{14,i} < 10$, instead of the square root, the values in Table A1 are inserted. As indicated, ΔR_i has 2 parts: the statistical uncertainty SU from the stochastic nature of the measurement (Poisson error) and the so-called additional uncertainty AU. AU takes into account the uncertainties due to the instability in transmission and in the current measurements. The determination of AU will be explained further below.

The uncertainty of the mean value of all measurements of a certain target R_{raw} (Equation 6) can be calculated either from the ΔR_i uncertainties by error propagation (Equation A3), or from the scattering of the R_i values (Equation A4), denoted by ΔR_{wm} and $\Delta R_{sc,}$ respectively. In JAC, the maximum of both is taken for ΔR_{raw}:

$$\Delta R_{raw} = \max[\Delta R_{wm}, \Delta R_{sc}] = \max\left[\left(\sum_i \Delta R_i^{-2}\right)^{-1/2}, \sigma/(\sqrt{n})\right] \quad (10)$$

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where σ is the standard deviation of the *n* single measurements.

Determination of the additional uncertainty *AU* (Equation 9) is based on the following idea: calculation of the uncertainty by error propagation and by scattering are 2 equivalent ways. Both values can differ for single targets, but if all uncertainties in the calculation of ΔR_i are taken into account correctly, then in the limit of many targets, in exactly half the cases ΔR_{wm} should be larger and in half the cases smaller. To find the appropriate value for *AU*, the data analysis for a selected number of targets was repeated with different values of *AU* to determine the break-even point (Figure 2). As can be seen from Equation 9, *AU* is a factor times R_i; the input in JAC is therefore the factor AU/R_i. Because the number of targets required is large, not one but a series of batches with normal and stable conditions is selected. To have a smaller statistical uncertainty, only targets with a ^{14}C isotope ratio >0.9 $F^{14}C$ are considered. For determination of the present value of *AU*, 5 batches from 2012 were selected, which yielded 178 suitable targets. The criteria for the selection were 1) each were from a different month, and 2) that the reduced chi-square (χ^2_{red}) is smaller than 2 without applying an additional error factor (see check 7 in next section). The value found is $AU/R_i = 3.8 \times 10^{-3}$ (Figure 2). An estimate of the term $\Delta R_{13}/\Delta R_{13}$ alone (Equation 9) based on the short-term fluctuations gave only 10% of this value, which is reasonable because I_{12} and I_{13} have (due to the chopper) nearly equal values.

Due to the selection of batches with sufficiently small χ^2_{red}, the value of *AU* represents only such batches. This choice was made with the aim of learning more about the limitations of the AMS system with respect to high precision. By choosing only one kind of target material, e.g. NOX, the reproducibility of the sample preparation could be determined with the same method, but would require more batches to get the same number of targets (in the present selection only 50 NOX targets were included). The values of *AU* obtained from earlier approaches are consistent but cannot be compared directly because they were based on ^{14}C isotope ratios from runs (instead of single measurements). Note that the value determined for *AU* is a mean value with respect to the time period of the chosen batches. Because *AU* is found to be much smaller than the statistical uncertainty, a check every year seems sufficient as long as there are no major modifications of the AMS system.

With the values of *AU* the uncertainties of the single measurements ΔR_i (Equation 9) can be calculated. Subsequently the isotope ratio R_{raw} of every target (Equation 6) and its uncertainty ΔR_{raw} (Equation 10) can be calculated in a.u. With these values, the intensive checks are performed (up to check 5, see next section). Also, the line fit to determine the transformation constant *A* and *B* (Equation 7) can be done. What is missing are the uncertainties ΔR_{meas} of the isotope ratios (in $F^{14}C$), which follow from Equations 8 and A1:

$$\Delta R_{meas} = \sqrt{R_{meas}^2 \Delta A^2 + A^2 \Delta R_{raw}^2 + \Delta B^2} \tag{11}$$

ΔA is calculated in 3 different ways. One value for ΔA is given by the line fit, denoted ΔA_{fit}. The 2 additional ways start from an alternative way calculating *A* and *B* from the mean values of the reference and the blank standard targets $\overline{R_{raw,\,std}}$ and $\overline{R_{raw,\,blk}}$, averaged according to Equation A2 over all respective targets of the batch:

$$B = \overline{R_{raw,\,blk}} \qquad A = \frac{\overline{R_{raw,\,std}} - \overline{R_{raw,\,blk}}}{R_{nom}} \tag{12}$$

From these, an equation for ΔA can be deduced by applying Equation A1. A second value of ΔA, denoted ΔA_{wm}, results from this equation by inserting the uncertainties of $\overline{R_{raw,\,std}}$ and $\overline{R_{raw,\,blk}}$ cal-

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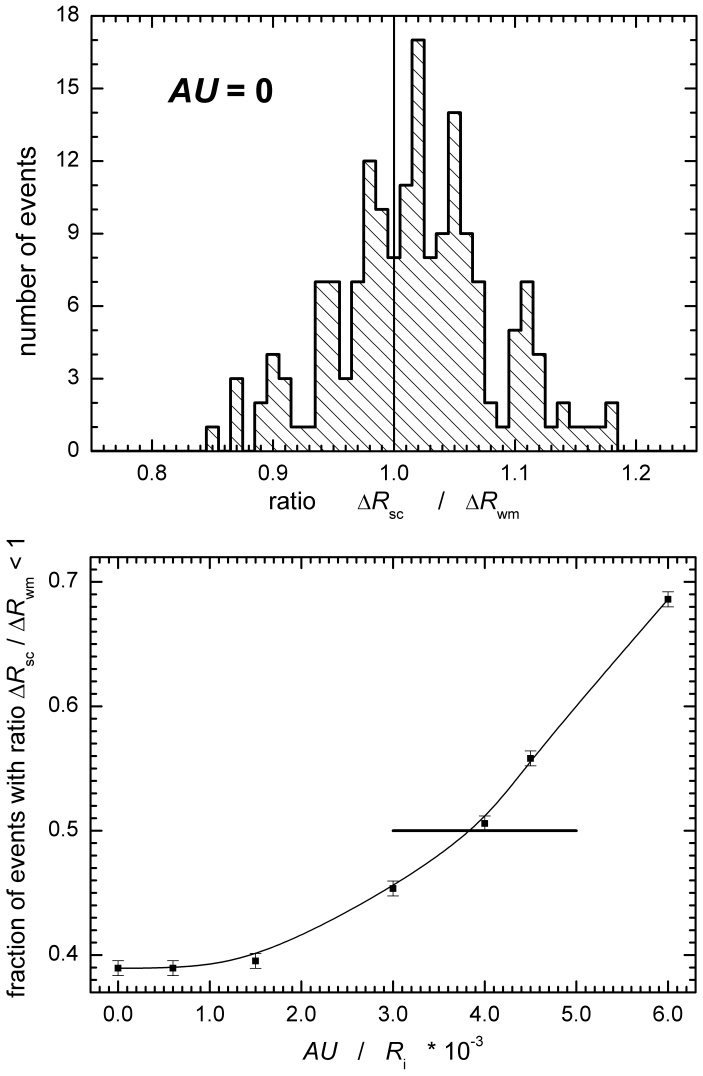


Figure 2 Determination of the additional uncertainty AU (defined in Equation 9) using the ratio $\Delta R_{sc}/\Delta R_{wm}$ (see text). For this determination, AU is treated as a variable. The top figure shows the distributions for $AU = 0$ as an example. In the bottom figure, each symbol shows the fraction of $\Delta R_{sc}/\Delta R_{wm} < 1$ for varied values of AU/R_i; the error bars show the effect of ±1 target. The correct value of AU follows from the condition that the fraction of $\Delta R_{sc}/\Delta R_{wm} < 1$ is equal to 0.5 (horizontal line, bottom figure). The value found for AU/R_i is 3.8×10^{-3}.

culated according to Equation A3 (weighted mean). A third value, denoted ΔA_{sc} results if these uncertainties are calculated according to Equation A4 (scattering) instead. The differences in A and B calculated by the line fit and by Equation 12 are not significant, but the differences in the 3 ΔA are, especially the one based on scattering. For the calculation of ΔA, the maximum of these 3 values is used:

$$\Delta A = \max\left[\Delta A_{fit}, \Delta A_{wm}, \Delta A_{sc}\right] \quad (13)$$

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In earlier versions of JAC, analog parameters ΔB_{fit}, ΔB_{wm}, and ΔB_{sc} were calculated and an equation analog to Equation 13 was applied. This yielded a large number of outliers for the blank standard C1 (10–20%). The conclusion is that in the case of the blank standards, the underlying condition for Equation A4 is not fulfilled. Applying Equation A4 implies that the fluctuations of the single measurements reflect only their measurement uncertainty and, therefore, the uncertainty of the mean is smaller by a factor of $\sqrt{n}$. Alternatively, it is assumed that the standard deviation σ_{blk} of the blank standard targets reflects only the fluctuations of the blank samples themselves and set

$$\Delta B = \sigma_{blk} \tag{14}$$

ΔB calculated according to Equation 14 was found to be always larger than ΔB_{fit} or ΔB_{wm}, and so no maximum analog to Equation 13 has to be taken.

To get an idea of the contributions of the different uncertainties, $F^{14}C = 1.0$ is chosen as an example:

- Typical for such an isotope ratio are 2150 ^{14}C events in a single measurement (with 30-s measurement intervals). Therefore, Equation 9 gives $\Delta R_i / R_i = (1/2150 + (3.8 \times 10^{-3})^2)^{1/2}$, i.e. the statistical part of the uncertainty dominates AU.
- For checking the uncertainties of the transformation to $F^{14}C$, typical uncertainties and values are $\Delta R_{unk} = 2 \times 10^{-3}$ a.u.; $\Delta A = 0.9 \times 10^{-3}$ a.u./$F^{14}C$; $\Delta B = 8 \times 10^{-4}$ a.u.; $R_{unk} = 100$ a.u.; and $A = 0.01$ a.u./$F^{14}C$. As can be seen by substituting these values in Equation 11, the uncertainty ΔR_{raw} dominates the contributions from the line fit. This means that our number of reference standards is sufficiently high.

CHECKS DURING THE DATA ANALYSIS

A series of checks is performed during the analysis of every batch. In the first round of data analysis of a batch, checks 2 to 6 (see below) are performed only on the NOX standards, which give the strongest statements because of their large number and high ^{14}C isotope ratios. Check 3 is performed for all targets and check 7 for all standards. If irregularities are evident, checks 4 and 5 can be done for all targets.

In checks 4 and 5, reduced deviations are used for graphical presentation. For their generation, first a mean value is calculated from all NOX targets in the batch. Then, the isotope ratios are calculated with respect to the parameter that should be investigated. In the case of check 3, these are the isotope ratios of the ^{13}C current intervals; in the case of check 4, the ones of the runs. In the next step, the "reduced deviations" are calculated by dividing the deviations of these ^{14}C isotope ratios from the respective mean by their uncertainty. For the reduced deviation presentations, the ^{14}C isotope ratios in a.u. (Equations 5 and 6) are used, so that the uncertainties ΔA and ΔB do not conceal a possible problem. The advantage of reduced deviation presentations is a better overview; in the case of correctly calculated uncertainties, around 68% of the data points lie within ±1, and 99.7% within ±3.

The routinely performed checks are:

Check 1 Check the limits of the ^{14}C integration limits in the energy spectra of the detector.

Check 2 Ratios I_{13} over I_{12} are checked graphically, similar to Figure 1 but for all NOX targets of the batch.

Check 3 During the calculation of the ^{14}C isotope ratios in a.u., the ratio of the uncertainties according to the weighted mean and the fluctuation (Equation 10) are checked.

Check 4 The dependence of the ^{14}C isotope ratios from the ion current is investigated to find the usable current range (Figure 3). Based on this, a minimum and maximum for the ion current can be set for the subsequent analysis. In Figure 3, the maximum currents do not exceed the usable current range very much. What this could look like can be seen in Figure 6 of Steinhof et al. (2011).

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Check 5 The single-run ^{14}C isotope ratios are checked by a plot of the reduced deviations (Figure 4). The single-run ^{14}C isotope ratios are calculated according to Equation 6, but the summation is performed only over the single measurements belonging to the respective run. The check versus the run number is a check versus the operation time on the respective target. Based on this check, single runs of a target can be excluded from further analysis. When this happens, it is mostly with small targets, if they are exhausted at the end. But contaminated targets would also be easily identified by having all runs (in case of NOX) at too-low values.

Check 6 After performing the line fit (Equation 7), the values of the different NOX targets are checked for outliers, i.e. checked for a reduced deviation outside of ±3. With 10 NOX targets, 1 outlier in a batch is accepted.

Check 7 For the final ^{14}C isotope ratios of all the standards except the blanks, a reduced chi-square χ^2_{red} is calculated. The blanks are excluded from the χ^2_{red} calculation because they systematically lead to its reduction, indicating that ΔB, according to Equation 11, might be too large. If $\chi^2_{red} > 2.0$, all uncertainties ΔR_{meas} (Equation 10) are multiplied by an additional error factor to provide a $\chi^2_{red} \leq 2$. This additional error factor is then also applied to the uncertainties of the unknown samples in the batch.

For a case of real troubleshooting, several other output options are available, e.g. single-measurement ^{14}C isotope ratios R_i versus I_{13}, R_{13}, or time.

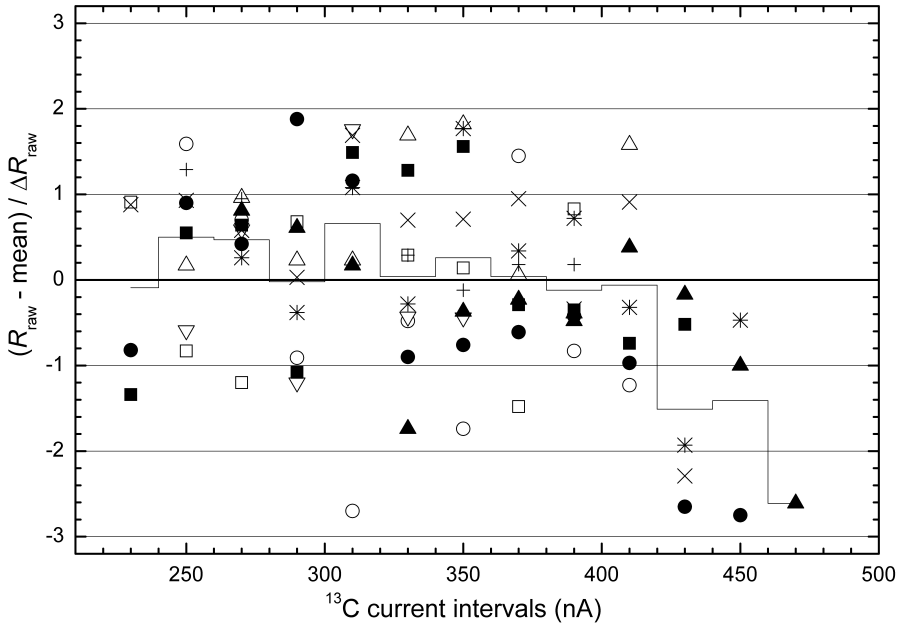


Figure 3 Reduced deviation of R_{raw} in ^{13}C current intervals vs. ^{13}C current. The symbols indicate different NOX targets. The 3 targets also shown in Figure 1 are indicated here with the same (full) symbols. The step-function line shows the mean values of the respective I_{13} current intervals. The ^{13}C current intervals are 20 nA wide. Because of the bending down of the deviations for $I_{13} > 420$ nA, this is taken as the limit of the usable current range, and the data points with $I_{13} > 420$ nA are excluded from further analysis.

For some of these checks, the ideas resulted from difficulties already solved, e.g. check 1 was introduced during a period when the ionization chamber detector was not completely gas-tight and check 4 from the time when we tested the useful current range on purpose. But they are still useful. Consider, for example, at the limitations of our AMS system with respect to the maximum ion current I_{13}, which are known, but which differ from batch to batch between 380 and 450 nA. Without check 4, we would have to stick a safety margin below the lower value of this limit, e.g. 380 nA. The

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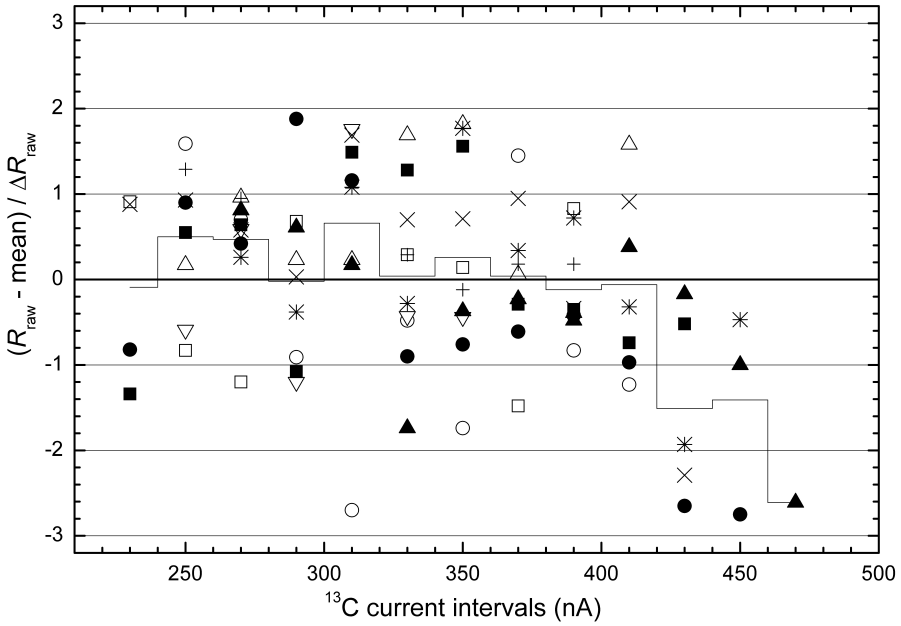


Figure 4 Reduced deviation vs. run number. The symbols indicate the same targets as in Figure 3. Run 1 is the cleaning run, which does not contribute to the results. In this batch, no further runs are excluded from further analysis.

safety margin accounts mainly for variations in the ion currents of different targets. With check 4, the ion source can operate with a mean current I_{13} of 400 nA (instead of say 340 nA) in exchange for excluding only a few single measurements.

COMPARISON WITH OTHER DATA ANALYSIS APPROACHES

Transmission

The most nontrivial aspect in JAC is the assumption that the *MRL* is constant, i.e. *DMRL* is zero (see Equation 2). The underlying problem is that the standards are measured at different times than the unknown samples (in all AMS systems) and that the stability can be controlled only by measuring standards, especially because *MRL* also includes instabilities in the ion source. To lessen the effect of this weak point of AMS, several sophisticated approaches have been published:

- Seguin et al. (1994) proposed a digital filtering technique for the time-dependence of the transmission;
- Tumey et al. (2004) discussed a time-dependent correction to compensate for machine drifts;
- Palonen and Tikkanen (2007) and Palonen (2008) developed a data analysis model based on Bayesian analysis to calculate the time variations of *MRL* during sample measurements, and;
- Zoppi (2010) introduced 2 efficiency functions whose ratio is equivalent to *MRL,* and performed a multidimensional surface fitting for both.

In contrast to these approaches, JAC implies the constancy of the *MRL*, and based on the checks during the data analysis, restricts the considered data to the useful current and time (given by the run number) ranges. For our AMS system, we found no other useful parameters than current and time. At least for batches without problems, the small value of *AU* demonstrates that the simple approach

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of JAC is sufficient. In many batches with problems, often large ratios ΔR_{sc} over ΔR_{wm} are observed within 1 run, so it is doubtful whether one of the sophisticated approaches would help. Therefore, there are no plans to modify JAC in this respect.

Background

Regarding the differences in treating the background correction, one has to distinguish different aspects. One aspect is machine-dependent corrections applied by Wacker et al. (2010) for their compact AMS systems. If applicable, such a correction would be also implemented in JAC. Another aspect reflects the mode of applying the blank standards. Currently in Jena, only identically sized blanks are used. If in the context of very small samples this were no longer possible in the future, other algorithms will have to be implemented (see e.g. Donahue et al. 1990b; Brown and Southon 1997; McNichol et al. 2001). This might then require the inclusion of information from previous batches into the analysis of the present one.

Another difference is the step of the data reduction at which the background correction is performed. For example, Wacker et al. (2010) perform this step before the fractionation correction during the calculations of the isotope ratios of the single measurements. However, because the background correction is based on values averaged over the entire batch (or several batches), this should not matter.

An interesting question would be whether the uncertainty for the background is calculated according to Equation 14, A3, or A4. Wacker et al. (2010) and Donahue et al. (1987) apply formulas equivalent to Equation 14 but no published information concerning this aspect has been obtained from other labs.

Non-Statistical Uncertainty

As the performance of the ^{14}C AMS systems with respect to ion current and transmission increases, the contribution of the uncertainty additional to the counting statistic part becomes more important. In JAC, this is taken into account by the term AU in Equation 9. In the literature, only Burr et al. (2007) and Wacker et al. (2010) have described similar contributions, which they call "random machine error" and "additional external error," respectively. Similar to what is done in JAC, its value is estimated from long-term experience. In contrast to the "one-time" procedure used in JAC, Burr et al. (2007) build up their long-term average on daily values resulting from their OXI and NOX standards.

Taking the Maximum of the Uncertainty

Taking the maximum of the uncertainties based on the counting statistic and scattering, as in Equations 10 and 13, might overestimate the uncertainty in the mean but reduce the chance of underestimating the uncertainty for a selected result. An equivalent procedure was found only in Nadeau et al. (1998) and McNichol et al. (2001).

SUMMARY

The JAC program has been developed in recent years at the Jena AMS lab. It has proven to be a reliable tool for data analysis. It is well suited for performing high-precision measurements because the non-statistical uncertainty is taken into account. Variations in the *MRL* are under control due to the checking procedures and, therefore, no modifications of JAC's treatment of the *MRL* are planned.

https://doi.org/10.1017/S0033822200057386 Published online by Cambridge University Press

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APPENDIX

The underlying equations for error analysis can be found in many textbooks (e.g. Bevington 1992). Nevertheless, for better traceability, the equations used are given here. The uncertainty Δy of a result calculated from several uncorrelated parameters u, v, is given by

$$\Delta y = \sqrt{\sum_{u,\,v,\,\ldots} \left(\frac{\partial y}{\partial u}\right)^2 \Delta u^2 + \left(\frac{\partial y}{\partial v}\right)^2 + \Delta v^2 + \ldots} \qquad \text{(A1)}$$

For a series of single measurements y_i with uncertainties Δy_i, the weighted mean value $\bar{y}$ is given by Equation A2 and its uncertainty Δy_{wm} is given by Equation A3.

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$$\bar{y} = \frac{\sum_i y_i / \Delta y_i^2}{\sum_i \Delta y_i^{-2}} \tag{A2}$$

$$\Delta y_{wm} = \left(\sum_i \Delta y_i^{-2} \right)^{-1/2} \tag{A3}$$

Alternatively to Equation A3, an uncertainty Δy_{sc} of a parameter measured n times can be deduced from the scattering of the values using the standard deviation σ

$$\Delta y_{sc} = \frac{\sigma}{\sqrt{n}} = \sqrt{\frac{1}{n(n-1)} \sum_i (y_i - \bar{y}_{am})^2} \tag{A4}$$

where $\bar{y}_{am}$ is the arithmetic mean value. Δy_{sc} is also called the standard deviation of the mean.

The statistical uncertainty of a signal based on n counted ions is usually taken as $\sqrt{n}$. But for signals based on a very small number, e.g. the $\Delta^{14}R_i$ for a blank sample, the uncertainties according to Table A1 are used instead of $\sqrt{n}$.

Table A1 Statistical uncertainties for a small number n of events. The values correspond to the upper limits of the non-symmetric uncertainties (Schmidt et al. 1984).

Number of events n	Statistical uncertainty
0	1.84
1	3.30
2	4.64
$2 < n < 10$	$\sqrt{n} + 1$
$n \geq 10$	$\sqrt{n}$

Table A2 Notations used for the most important parameters. For the ion current I and number of ions N, the former give the mass of the respective ion and the latter the kind of target, e.g. "std" for standard target (see table). This is still true for the ^{13}C current ratio $R_{13,m}$, but in the case of the analog ^{14}C isotope ratio the "14" is left out. Therefore, a subscript for the kind of ^{14}C isotope ratio is given ("h" in the table). For R (the ^{14}C isotope ratio) if it is obvious, the subscript "m" is left out, e.g. a nominal ^{14}C isotope ratio R_{nom} exists only for standards. The second subscript ("statt") of R is also left out if an equation is valid for all kinds of targets, like R_{meas} and R_i.

Symbol	Parameter	Subscripts	Meaning (possible values)
$I_{n,m}$	Ion current of carbon isotope	n	Isotopic mass of the ions: 12, 13, 14
$N_{n,m}$	Number of carbon ions	m	Kind of target: "std" = standard target; "unk" = target of an unknown sample; "blk" = blank target.
$R_{13,m}$	$^{13}C/^{12}C$ isotope or current ratio	h	Kind of ^{14}C isotope ratio: "raw" = raw values, i.e. in a.u.; "nom" = nominal value of a standard; "i" = single measurement; "meas" = measurement result of an unknown sample.
$R_{h,m}$	^{14}C isotope ratio		

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