Calculation and uncertainty of Pb-210 dates for PIRLA project lake sediment cores

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Calculation and uncertainty analysis of ^{210}Pb dates for PIRLA project lake sediment cores *

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Abstract

Lead-210 assay and dating are subject to several sources of error, including natural variation, the statistical nature of measuring radioactivity, and estimation of the supported fraction. These measurable errors are considered in calculating confidence intervals for ^{210}Pb dates. Several sources of error, including the effect of blunders or misapplication of the mathematical model, are not included in the quantitative analysis. First-order error analysis and Monte Carlo simulation (of cores from Florida PIRLA lakes) are used as independent estimates of dating uncertainty. CRS-model dates average less than 1% older than Monte Carlo median dates, but the difference increases non-linearly with age to a maximum of 11% at 160 years. First-order errors increase exponentially with calculated CRS-model dates, with the largest 95% confidence interval in the bottommost datable section being 155 ± 90 years, and the smallest being 128 ± 8 years. Monte Carlo intervals also increase exponentially with age, but the largest 95% occurrence interval is 152 ± 44 years. Confidence intervals calculated by first-order methods and ranges of Monte Carlo dates agree fairly well until the ^{210}Pb date is about 130 years old. Older dates are unreliable because of this divergence. Ninety-five per cent confidence intervals range from about 1–2 years at 10 years of age, 10–20 at 100 years, and 8–90 at 150 years old.

Introduction

The goal of the 'Paleoecological Investigation of Recent Lake Acidification' (PIRLA) study is to examine whether selected lakes have been artifi-

cially acidified, and if so, when the acidification occurred (Charles & Whitehead 1986). Changes in lakewater acidity cannot be detected unless earlier conditions are known, but comparative data are almost never available. Therefore, paleoecological methods must be used to define both pre-disturbance conditions and rates of change of limnological characteristics.

Paleolimnologists assume that lake sediments accumulate in an ordered manner through time, and that any alteration of the order can be

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detected and corrected. Lake sediments contain a vertical sequence of physical, chemical, and biological fossils, the characteristics of which are functions of environmental conditions in and around the lake. The timing, and rate, of changes of sedimentary features are important aspects of paleolimnological studies. Such measurements of time require dating of levels in sediment cores.

Several methods for dating cores are commonly used. PIRLA relies mostly on measurement of sedimentary ^{210}Pb, a naturally occurring radionuclide, and application of one particular dating model for calculating dates and accumulation rates. ^{210}Pb-dating models are described mathematically in numerous papers, but actual calculation methods are never explicit. Estimates of dating uncertainty are seldom presented in published papers or reports. This paper describes and justifies the methods used by the PIRLA project to calculate ^{210}Pb dates and to estimate their uncertainties. The methods are applied to all the PIRLA cores, and examples of the results are presented.

Geochemistry of ^{210}Pb

^{210}Pb is a member of the ^{238}U decay series of radionuclides. A precursor of ^{210}Pb, ^{226}Ra, exists in most rocks, soils, and sediments. ^{226}Ra decays to ^{222}Rn, a gas which escapes from free surfaces into the atmosphere. ^{222}Rn has a half-life of 3.8 days, and decays through several short-lived radionuclides to ^{210}Pb. The average atmospheric residence time for ^{222}Rn is about 4.2 days, during which the gas is distributed widely (Turekian et al., 1977). ^{210}Pb atoms adsorb onto atmospheric particulates, which are deposited on land and water.

The sources of ^{210}Pb to lakewater are many and varied. The amount of the ^{210}Pb that falls onto land and is transported to lakewater varies with the biogeochemical and hydrological characteristics of the catchment. In general, adsorbed ^{210}Pb is bound very tightly to catchment soil particles, and little remobilization occurs (Smith & Siccama, 1981; Benninger et al., 1975). However,

some studies demonstrate that riverine input to lakes can be a major source of ^{210}Pb (Benoit & Hemond, 1987; Stiller & Imboden, 1987). A substantial fraction of lakewater ^{210}Pb can be derived from rapid flux of groundwater rich in dissolved ^{222}Rn (Norton et al., 1985). *In situ* production by ^{226}Ra and ^{222}Rn decay can produce nearly all the ^{210}Pb in large bodies of water with very long water residence times (Stiller & Kaufman, 1984). Regardless of the ultimate sources, if the total flux of ^{210}Pb to a lake and its sediments has been constant, then current dating models are valid.

Some investigations have questioned the use of ^{210}Pb dating for acidified lakes because either the flux of ^{210}Pb to lake sediment decreases or ^{210}Pb is leached more readily from sediments with increasing lakewater acidity (Dillon et al., 1987). However, an experimental study has demonstrated that stable lead is not released from sediments of two Adirondack lakes (Woods Lake and Lake Sagamore) until the pH of interstitial water dropped below 4.0 (Davis et al., 1982), and significant releases did not being until the pH was lower than 3.0 in one case and 2.0 in another. All PIRLA study lakes have pH values >4.0 in their water column. The constant-flux assumption has not been refuted, and it is still an underlying assumption in PIRLA dating procedures.

Once incorporated into the sediments, ^{210}Pb is buried by subsequent sedimentation. If no mixing occurs and sediment accumulation rate does not change, the distribution of ^{210}Pb activity vs. depth in a sediment core is described by an exponentially declining curve (Fig. 1). The curve of total activity does not converge to zero towards the bottom, but instead to a finite value that is called 'supported' ^{210}Pb. Supported ^{210}Pb in the sediments is derived from the *in situ* decay of ^{238}U daughter products. ^{210}Pb dating is based only upon 'unsupported' activity, or that derived from external sources. Measurement of supported activity in the presence of unsupported ^{210}Pb is problematic, and our approach is discussed in a later section of this report.

^{210}Pb behaves chemically much as stable Pb, and is probably subject to the same processes governing post-depositional mobility. All dating

Pb–210 ACTIVITY

$(\text{pCi or mBq or dpm g}^{-1})$

$$A_x = \int_x^\infty C_x \, P_x \, dx$$

$$(\text{for depth} = \text{cm})$$

$$A_x = \int_x^\infty C_x \, dx$$

$$(\text{for depth} = \text{g cm}^{-2})$$

$$t = k^{-1} \ln(A_x / A_o)$$

$$r = k \, \bar{A}^1 C$$

$$k = 0.03114 \text{ yr}^{-1}$$

Fig. 1. Idealized ^{210}Pb profile. X is the depth in the core in cm or g cm^{-2}; C is the activity of ^{210}Pb at a particular depth (in this example x_0 and x_1); ρ_x is the dry density of sediment (g cm^{-3}) at depth x (note that if depth is measured as cumulative mass – g cm^{-2} – then this term drops out); t is the age of the level x; k is the decay constant for ^{210}Pb (.03114 yr^{-1}); A_x and A_o are the total, integrated, unsupported ^{210}Pb at depth x and the surface, respectively; r is the accumulation rate of sediment at any selected depth with a characteristic A and C.

Theory of ^{210}Pb dating

Calculation of ^{210}Pb dates is dependent on the model that describes distribution of radioactivity in the sedimentary column. There are two popular models representing the end points of a continuum of sedimentary processes. The models can be modified to include various expressions of

models assume negligible post-depositional mobility (except for some models that include terms for mixing) and may be in error for that reason.

mixing. The Constant Initial Concentration, or Constant Activity Model (hereafter called the CIC model), assumes that ^{210}Pb activity in the uppermost layer of sediment is constant through time (Goldberg, 1963; Robbins, 1978). If the sedimentation rate changes, then the amount of ^{210}Pb swept from the water to the sediment changes proportionally. Increased bulk sedimentation rate results in increased ^{210}Pb sedimentation and constant activity in the uppermost layer of sediment. The Constant Rate of Supply, or Constant Flux (CRS model), assumes that the flux of ^{210}Pb to the sediment surface is constant through time (Goldberg, 1963; Appleby & Oldfield, 1978; Robbins, 1978). If the accumulation rate of bulk sediment changes, then sedimentary ^{210}Pb will be diluted or concentrated (Appleby & Oldfield, 1983; Binford & Brenner, 1986).

Any given lake will fall somewhere between the two models (Robbins, 1978), depending on the total pool of ^{210}Pb in lakewater, partitioning of ^{210}Pb between particulate and dissolved forms, bulk sedimentation rate, and the rate at which water flows through the lake. If each of these factors is in steady state over the past 150–200 years, then the CRS and CIC models will both be valid.

The two models are expressed mathematically for calculating dates from ^{210}Pb activity (see Fig. 1 for graphical expression of each term). The CIC model is:

$$t_x = k^{-1} \ln(C_o \, C_x^{-1}), \tag{1}$$

where t is age in years, k is the decay constant for ^{210}Pb (0.03114 yr^{-1}), x is depth in core expressed as cumulative mass (g cm^{-2}), C_o is the specific unsupported activity at the sediment-water interface (pCi g^{-1}), C_x is the specific unsupported activity at depth x in the sedimentary section pCi g^{-1}). The CRS model is (Note that the depth term in the following equations is measured in cm. A better measurement would be in cumulative mass below the sediment-water interface – g cm^{-2}. If this measurement is used, as it was in the CIC model, then ρ_x is eliminated from each equation):

$$t_x = k^{-1} \ln(A_o A_x^{-1}), \tag{2}$$

where A_o = total, integrated, unsupported ^{210}Pb in the core (pCi cm^{-2}), A_x = integrated activity of unsupported ^{210}Pb below depth x, ($\int_x^\infty C_x\rho_x$ dx) (pCi cm^{-2}), ρ_x = dry mass per unit volume wet sediment (g cm^{-3}). Additionally, the bulk accumulation rate can be calculated explicitly by:

$$s_x = k\,A_x\,C_x^{-1} \tag{3}$$

The CIC model fails (because the assumption of constant activity is violated) if the distribution of ^{210}Pb activity with depth is non-monotonic. A mixing term can be included in both the CIC and CRS models (Oldfield & Appleby, 1984). Each of these models requires independent measurement of the depth of the zone of mixing and assumes instantaneous and complete mixing within the zone. A 1-cm error in the estimate of mixing depth can result in dates as much as 20–30% in error.

The PIRLA dates are calculated exclusively from the CRS model without a mixing term. This procedure was followed, despite some evidence of mixing in some cores, for several reasons given in the next paragraph. There are sites where the CRS model is not appropriate, for example in lakes with intermittent sediment slumping. In these cases the CIC model might be appropriate especially if the ^{210}Pb inventory is high and there are no non-monotonic aberrations in the ^{210}Pb profile. None of the PIRLA cores show any indication that the CRS model is not appropriate. Data required for calculation of dates using other models are available in the PIRLA data base management system (PIRLA/DBMS).

The first reason for using only one model is that studies in many temperate-zone lakes (Appleby & Oldfield, 1983) and in lakes in Florida (Binford & Brenner, 1986) indicate that ^{210}Pb activity is diluted by recent high sediment accumulation rates, which violates the major CIC assumption. Second, most of the CRS dates calculated for PIRLA cores agree with independent dating methods such as pollen horizons (Binford *et al.*, in prep.). Third, independent measurements of mixed-layer thickness are absent, dates from the non-mixing model are simpler to evaluate critically, and mixing has a significant effect on dates only when the thickness of the mixed zone exceeds about 15% of the depth of the unsupported ^{210}Pb profile (Oldfield & Appleby, 1984). Tinkering with different mixed-layer thicknesses is possible with the original data, and the interested reader is invited to do so. Any incorporation of mixed layers will result in a younger date at a given level. Thicker mixed layers result in younger dates for both CIC and CRS models.

Error analysis —Sources of error

All measurements of continuous variables are made with uncertainty, or error. Random errors, systematic errors, and blunders are some sources of error. This error analysis is concerned with some of the components of internal uncertainty, i.e. the variability controlled by the precision and accuracy of sedimentary measurements and the nature of the CRS model, and not that controlled by external forces such as application of incorrect models, inaccuracies of stratigraphic sampling, or glassware contamination.

Random errors arise from radioactive decay processes, natural variance of the ^{208}Po spike, measurement errors of dry mass per unit wet volume, estimation of supported ^{210}Pb, and variation of the efficiency of chemical extraction processes. Systematic errors occur if the ^{208}Po pike is improperly calibrated (although this error will not affect the accuracy of dates if the error is constant), and from the 'old-date error' of the uncorrected CRS model. Finally, blunders, or mistakes such as application of an inappropriate dating model, ^{208}Po spike addition, glassware contamination, incorrect labeling, etc. all cause increases in uncertainty. All the random errors except the last one and the 'old-date error' are considered in this analysis.

The amount of ^{210}Pb in a given mass of sediment is expressed as activity (either disintegrations per second – also known as Becquerels in SI units – or Curies) $\pm$ counting variability (one

standard deviation). Radioactive decay is a random process that follows a Poisson distribution. The true variance of a Poisson variable equals its true mean. Therefore the square root of a measurement provides an estimate of the standard deviation. Furthermore, the standard error of the mean (s.e.) for one observation from a known distribution is equal to the standard deviation.

^{210}Pb in sediment cores is measured by chemically extracting ^{210}Po, a daughter radionuclide, from a known mass of dry sediment and counting the alpha particles emitted over a given time, usually 25 000 to 75 000 seconds. A known amount of man-made ^{208}Po is added to the original sediment to provide an internal yield tracer. The original activity of ^{210}Po and, by assumption of secular equilibrium, ^{210}Pb is calculated by the proportion of ^{208}Po extracted by the process. Typical ^{210}Pb activities in the PIRLA project cores range from 0.1 pCi (3.7 mBq or 0.0037 dps) to 100 pCi, with attendant errors of from less than 10% to greater than 20%, depending on counting time, original activity, and mass of the sediment sample.

Supported ^{210}Pb can be estimated by several means, all of which are measured with finite precision. The PIRLA procedure is a semi-objective method that assumes the asymptotic activity below the bottommost horizon that contains measurable, unsupported ^{210}Pb is a constant through the upper section of the core (see Fig. 1). The mean and standard deviation of the bottom three determinations of ^{210}Pb activity are calculated. If the next higher (in the core) activity is less than the mean plus one standard deviation, the mean and s.d. are recalculated with the new level included. The procedure continues until the activity in the next higher level is greater than the mean plus one standard deviation. This level is then defined as the lowest interval with measurable, unsupported ^{210}Pb. The mean value for supported ^{210}Pb is then subtracted from total ^{210}Pb activity at each level. The standard deviation of supported ^{210}Pb is incorporated into the standard deviation of unsupported ^{210}Pb by the simple relation:

$$s_{unsupp} = (s^2_{supp} + s^2_{total})^{0.5} \qquad (4)$$

where s_{unsupp} is the standard deviation of unsupported ^{210}Pb, s^2_{supp} is the calculated variance of supported ^{210}Pb, s^2_{total} is the variance of total ^{210}Pb activity in an analyzed interval (counting error).

This method assumes not only that supported ^{210}Pb is a constant through a sedimentary section, but also that the variance of supported ^{210}Pb is also a constant. The assumption is probably valid in PIRLA lakes, which were selected in part because their drainage basins were minimally disturbed. If a lake's drainage basin is not disturbed by cultural activities or subject to catastrophic events, the erosion rate over 100–150 years will be approximately constant.

Dry density ((ρ) in the notation of Appleby & Oldfield, 1978) of the sediment is an important term in CRS model calculations. ^{210}Pb activity is expressed per gram dry mass, and must be converted to activity per unit volume. Dry density of sediment cores from Florida lakes was measured by volumetric sampling. Dry density of all other PIRLA cores was calculated as a function of water, organic, and inorganic content:

$$\rho_x = \frac{D(2.5I_x + 1.6C_x)}{D + (1 - D)(2.5I_x + 1.6C_x)} \qquad (5)$$

where ρ is dry density (g_{dry} cm$^{-3}_{wet}$), x is depth in the core (cm or g cm^{-2}), D is proportion dry weight of unit wet volume, I is the inorganic proportion of dry material (density = 2.5 g cm^{-3}), C is the organic proportion of dry material (density = 1.6 g cm^{-3}).

Both methods have difficulties. Both require accurate measurement of water content, which changes constantly by evaporation during the measurements. Accurate volumetric sampling of wet sediments is very difficult, but good precision can be attained by experienced technicians. On the other hand, determinations of dry mass are made with almost no error. Calculated dry density can be in error because the densities used in the equation are estimates themselves, and vary with the nature of the organic and inorganic material.

Comparison of measured and calculated ρ of core sediments from Florida PIRLA lakes dem-

onstrates that calculation results in about a 5% overestimate relative to measurement (Fig. 2). However, neither method can be considered better than the other. The slope of the major axis (type II regression) is 1.09 ± 0.015 (95% c.i.). There is no difference between measured and calculated densities (paired t-test; $t = -0.97$, $p = 0.3335$, 111 degrees of freedom), nor between variances of measured and calculated densities (F test; $F = 1.205$, $p = 0.15$). That the two methods have such similar results is especially interesting because the organic and inorganic density estimates were empirically determined from sediments of lakes in New England (Stephen Norton pers. comm.). Eroded soil is often a major contributor to lake sediments (Mackereth, 1966), and the compositions of Florida soils and New England soils are quite different.

Although dry density is a variable, it is assumed to be measured without error for calculations of ^{210}Pb dates. Variance estimates of dry density for sediments with different properties are not available.

The only systematic error considered by this study is the possibility of an improperly calibrated ^{208}Po spike. Inter-laboratory comparisons early in the project indicated that no bias occurred in one of the two sites for ^{210}Pb dating (Table 1). This result does not prove that no bias exists, but

it seems unlikely for two independent laboratories to have the same systematic error. Although not discussed specifically, other sources of systematic

MEASURED VS. CALCULATED DRY DENSITY
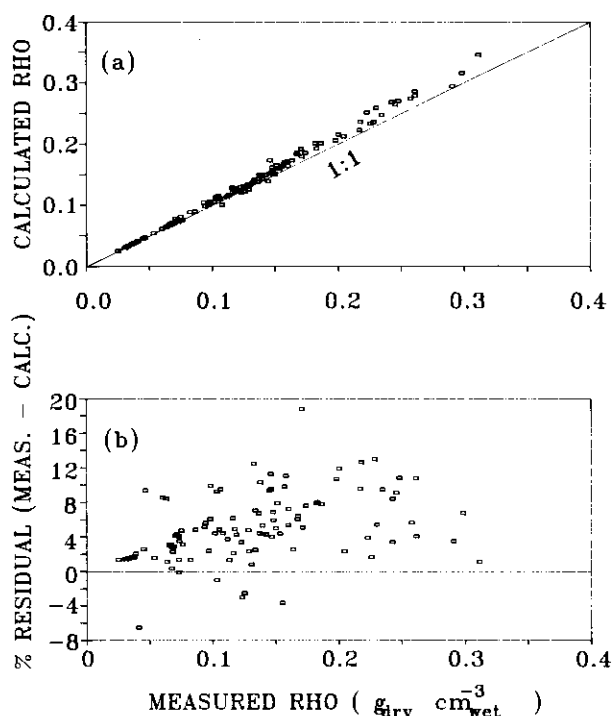
Fig. 2. Comparison of measured and calculated dry density for sediment in 12 cores from the Florida PIRLA lakes.

Table 1. Results of inter-laboratory comparison of ^{210}Pb measurements. The Florida lab used a ^{208}Po spike of 6.52 pCi, the Maine lab used 3.26 pCi.

Lab	Lake	Depth	Count #	Mass (g)	Count Time	^{208}Po Counts	% Yield	^{210}Pb Counts	^{210}Pb Activity
Florida	Fish River	0–8 cm	1	0.7367	50 000 sec	1193	65.9	846	6.47 ± 0.31
Florida	Fish River	0–8 cm	2	0.6225	50 000	925	51.1	610	6.91 ± 0.28
Florida	Fish River	0–8 cm	3	0.6397	50 000	1216	67.2	818	6.85 ± 0.24
Maine	Fish River	0–8 cm	1	0.500	40 000	753	100	793	6.87 ± 0.49
Maine	Fish River	0–8 cm	2	0.501	40 000	774	100	844	7.09 ± 0.50
Maine	Fish River	0–8 cm	3	0.501	40 000	658	90.1	687	6.79 ± 0.52
Florida	Fourth	45–55 cm	1	0.7973	50 000	950	52.5	24	0.21 ± 0.04
Florida	Fourth	45–55 cm	2	0.5417	50 000	1105	61.1	45	0.49 ± 0.07
Florida	Fourth	45–55 cm	3	0.6989	50 000	1259	69.6	45	0.33 ± 0.05
Maine	Fourth	45–55 cm	1	1.500	40 000	674	93.1	62	0.20 ± 0.03
Maine	Fourth	45–55 cm	2	1.500	40 000	681	94.1	66	0.21 ± 0.03
Maine	Fourth	45–55 cm	3	1.500	40 000	797	100	65	0.18 ± 0.03
Maine	Fourth	45–55 cm	4	1.500	40 000	643	88.8	62	0.21 ± 0.03

error are also discounted by the inter-laboratory comparisons.

The unmodified CRS model generates a systematic error during calculation of dates. A_x is found by numerically integrating an exponential curve, which mathematically never converges on zero. The numerical calculation, though, disregards a small fraction of the integrated area at the bottom of the curve. If the small area is not estimated and added to A, then dates earlier than about 80–100 years ago begin to diverge greatly from the 'true' date (see also Robbins, 1978; 337–355 for an analytical discussion), because the small area becomes a larger proportion of A_x. CRS dates presented in this report are calculated by adding the small fraction of A as determined by a numerical technique developed by Peter Appleby (pers. comm.) and modified slightly by Dan Engstrom (pers. comm.). The method is described in a later section.

Error analysis – Estimating error

Several methods for estimating dating uncertainty are available. The simplest and most appropriate carry error estimates for input data through the model calculations and assign an aggregate error to the output. The two methods used for this report are First-Order-Analysis and Monte Carlo Simulation Analysis.

First-Order Analysis, also called general error propagation (Meyer 1975), approximates the variance of a dependent variable by applying mathematical rules to the combination of variances of the independent variables (Reckhow & Chapra, 1983). The fundamental rule is derived by using only the first two terms of the Taylor series expansion (linearization) to approximate the variance of a function:

$$s^2(f(x)) = s^2(x)\,(\partial f(x)/\partial x)^2 x \qquad (6)$$

for any one variable, and:

$$s_y^2 = \sum_i (\partial f/\partial x_i)^2 s^2 x_i +$$
$$+ 2 \sum_i \sum_j (\partial f/\partial x_i) s_{x_i} s_{x_j} p_{x_i \cdot x_j} \qquad (7)$$

for two independent variables where $p_{x_i \cdot x_j}$ is the correlation coefficient between x_i and x_j. This equation is general and can be extended to more than two variables. The full derivation can be found in Meyer (1975).

If the independent variables are uncorrelated, then the covariance term is 0, and (7) reduces to (8):

$$s_y^2 = (\partial f/\partial x_i)^2 s_{x_i}^2 \qquad (8)$$

The $\partial f/\partial x_i$ term expresses a 'sensitivity factor' (Reckhow & Chapra, 1983), which can also be interpreted in part as the effect of non-linear and non-monotonic characteristics of the function.

Computationally, the relation reduces to a few simple rules if one assumes that the variables are independent and normally distributed, and the function is linear in the neighborhood of the measurement value:

$$s_{A+B}^2 = s_A^2 + s_B^2 \qquad (9)$$

$$s_{A-B}^2 = s_A^2 + s_B^2 \qquad (10)$$

$$s_{AB}^2 = AB\,[(s_A/A)^2 + (s_B/B)^2] \qquad (11)$$

$$s_{A/B}^2 = A/B\,[(s_A/A)^2 + (s_B/B)^2] \qquad (12)$$

Other formulas are used for exponential and other more complex functions. Each step of the CRS model calculations is an arithmetic operation covered by the rules. Despite the obvious relationship between ^{210}Pb activities of various levels, the variables used in each calculation step are assumed to be uncorrelated.

The equation for calculating First-Order error of the age at a particular level in a core is:

$$s_t^2 = k^{-2}A_o^{-2} \cdot s_{Ao}^2 + k^{-2}A_x^{-2} \cdot s_{Ax}^2 \qquad (13)$$

where s_t^2 is the variance of the age (calculated from (2)), s_{Ao}^2 is the variance of total, integrated ^{210}Pb, and s_{Ax}^2 is the variance of integrated ^{210}Pb at level x. The variances of integrated ^{210}Pb are derived from calculations made throughout the entire procedure and described for each intermediate variable in a later section. This formula is also applicable to CIC dates (C replaces A). The concentrations are usually reasonably independent.

However, the dependence between A and A_o increases further up the profile for the CRS model. Thus eq. 13 gives a conservative (i.e. larger) estimate of the error than alternative formulations using more independent variables.

First-order analysis may not be applicable to many problems for several reasons (Reckhow & Chapra, 1983). First, if the model is non-linear, the approximation by the first two terms of the Taylor expansion is less accurate than for linear models. Second, the accuracy of a non-linear error estimate is a function of the magnitude of the error of the independent variable. As the error in x becomes larger, non-linearity magnifies the error of y. Finally, if the standard deviation of x is not a good measure of variation (for example x's with skewed distributions), the error estimate will be faulty. First-order analysis must be tested by an alternative means.

Monte Carlo simulation is the second method used in the PIRLA study for estimating errors in dates and is used as a test of the first-order analysis. The method is named for the role of random chance (as with the gambling tables in Monte Carlo) in the outcome of calculations. Independent variables are assigned a particular probability density function (p.d.f.) that describes the chance of randomly obtaining a particular value for the variable. The p.d.f. can be any distribution (uniform if there is equal probability of any value within a range, normal or Gaussian for normally distributed variables, etc.) that can be described by one or a few parameters (range, standard deviation and mean, respectively). A single value for each of the independent variables is sampled at random from the distribution, then the dependent variable is calculated. The sampling and calculation are repeated many times, and the result is a set of synthetically derived, dependent variables that have a range, a mean, and a variance.

Monte Carlo methods are computer-intensive and cumbersome for simple problems, but very effective for complex models with several variables and non-linear characteristics. Obviously, it helps if the p.d.f. for each independent variable is known. Estimates and approximations of the p.d.f. can be made, and discussions of which statistics should be used are available in Reckhow & Chapra (1983), Mosteller & Tukey (1977), Meyer (1975) and other standard references on data analysis.

CRS-model calculations in the PIRLA study

This section describes the calculations of CRS-model dates, not including error calculations or correction of the old-date error. Table 2 is a spreadsheet-format example, done with invented

Table 2. Spread-sheet representation of CRS-model, ^{210}Pb dating calculations. This table does not include the 'old-date error' correction.

Row	1 INTTOP	2 INTBOT	3 TOTACT	4 UNSUPACT	5 RHO	6 PERCORG	7 RHOXCEE	8 INTEGSEG	9 INTEGINT	10 SUMBOT	11 SUMTOP	12 TTOP	13 TBOT	14 SEDRATE	15 ORGSEDRT
1	0.00	0.50	50.50	50.00	0.10	0.45	5.00	2.50	2.14	11.88	14.38	0.0	6.1	0.008	0.004
2	1.00	1.50	30.88	30.38	0.12	0.44	3.65	1.82	1.49	7.94	9.37	12.4	18.9	0.009	0.004
3	2.00	2.50	18.96	18.46	0.13	0.44	2.40	1.20	1.01	5.22	6.42	25.4	32.0	0.010	0.004
4	3.00	3.50	11.72	11.22	0.15	0.44	1.68	0.84	0.70	3.37	4.21	38.6	45.6	0.011	0.005
5	4.00	4.50	7.31	6.81	0.17	0.43	1.16	0.58	0.47	2.09	2.67	52.7	60.1	0.012	0.005
6	5.00	5.50	4.64	4.14	0.18	0.43	0.75	0.37	0.29	1.25	1.62	67.7	75.3	0.012	0.005
7	6.00	6.50	3.02	2.52	0.18	0.43	0.45	0.23	0.18	0.73	0.96	82.9	90.3	0.012	0.005
8	7.00	7.50	2.03	1.53	0.18	0.43	0.28	0.14	0.11	0.41	0.55	97.7	104.8	0.013	0.006
9	8.00	8.50	1.43	0.93	0.20	0.43	0.19	0.09	0.07	0.21	0.30	112.1	117.7	0.013	0.006
10	9.00	9.50	1.06	0.56	0.20	0.43	0.11	0.06	0.04	0.08	0.13	127.2	134.5	0.014	0.006
11	10.00	10.50	0.84	0.44	0.20	0.42	0.08	0.03	0.003	0.003	0.03	141.2	147.7	0.015	0.006
12	11.00	11.50	0.60	0.00	0.20	0.42	0.00	0.00	0.00	0.00	0.00	–	–	–	–
13	12.00	12.50	0.40	0.00	0.20	0.42	0.00	0.00	0.00	0.00	0.00	–	–	–	–
14	13.00	13.50	0.60	0.00	0.20	0.42	0.00	0.00	0.00	0.00	0.00	–	–	–	–
15	14.00	14.50	0.40	0.00	0.20	0.42	0.00	0.00	0.00	0.00	0.00	–	–	–	–

data, that is useful for defining each variable and how it is calculated. The spreadsheet format is convenient for demonstrating the calculations because the entire spreadsheet is expressed in matrix, or array, notation. A matrix or array is a group of numbers (called elements) arranged in rows and columns. Each element is referred to by its location in the array. Some elements are the result of calculations that depend on other elements. The elements are then described by equations, and not by variables.

For example, consider a matrix, called X, of the form:

Columns

	1	2	3
Rows	7	8	15
	12	16	18

Element 2,3 (in row 2, column 3) is 15. Element 2,3 is the sum of elements 2,1 and 2,2. The equation is expressed as $X_{(2,3)} = X_{(2,1)} + X_{(2,2)}$. Note that the elements in column 3 are the sums of columns 1 and 2 in each row. The general notation is then $X_{(r,3)} = X_{(r,1)} + X_{(r,2)}$.

The calculation matrix for the CRS-model dating is called $CAL_{(r,c)}$. The following description gives the equation, in array notation, of each column in the spreadsheet. Each row of the spreadsheet represents an analyzed interval in the core. The variables, or columns, in order of measurement or calculation are:

INTTOP –
the top of the analyzed interval in cm: element $CAL_{(r,1)}$.

INTBOT –
the bottom of the analyzed interval in cm: $CAL_{(r,2)}$.

TOTACT –
The total ^{210}Pb activity in pCi g^{-1}: $CAL_{(r,3)}$.

UNSUPACT –
Unsupported ^{210}Pb activity in pCi g^{-1}: $CAL_{(r,4)}$.

RHO –
Dry mass cm^{-3} wet sediment: $CAL_{(r,5)}$.

PERCORG –
Proportion organic matter in g g^{-1}: $CAL_{(r,6)}$.

RHOXCEE –
RHO times UNSUPACT in pCi cm^{-3}: $CAL_{(r,7)} = CAL_{(r,5)} \times CAL_{(r,4)}$. This is the first step of calculations, and is the amount of ^{210}Pb in 1 cm^3 of wet sediment.

INTEGSEG –
^{210}Pb activity integrated over the thickness of the analyzed interval, or RHOXCEE times the thickness of the analyzed interval (INTBOT – INTTOP) in pCi cm^{-2}: $CAL_{(r,8)} = CAL_{(r,7)} \times (CAL_{(r,2)} - CAL_{(r,1)})$. If the interval is 1 cm

16	17	18	19	20	21	22	23	24	25	26	27	28
CUMMASST	CUMMASSB	SDACT	SDRHO	SDUNSUP	SDRHOCEE	SDINTSEG	SDINTINT	SDSUMBOT	SDSUMTOP	SDTTOP	SDTBOT	SDSEDRT
0.000	0.05	4.00	–	4.00	0.40	0.200	0.05	0.233	0.307	0.962	0.923	0.005
0.105	0.17	3.00	–	3.00	0.36	0.180	0.05	0.132	0.223	0.995	0.860	0.006
0.228	0.29	1.50	–	1.501	0.20	0.098	0.03	0.072	0.121	0.904	0.806	0.005
0.363	0.44	0.65	–	0.652	0.10	0.049	0.01	0.045	0.067	0.841	0.798	0.005
0.518	0.60	0.30	–	0.306	0.05	0.026	0.01	0.035	0.043	0.843	0.845	0.004
0.690	0.78	0.20	–	0.207	0.04	0.019	0.01	0.028	0.034	0.920	0.950	0.005
0.870	0.96	0.18	–	0.188	0.03	0.017	0.005	0.021	0.027	1.060	1.056	0.006
1.050	1.14	0.10	–	0.114	0.02	0.010	0.004	0.018	0.021	1.206	1.285	0.006
1.235	1.33	0.09	–	0.105	0.02	0.011	0.004	0.014	0.018	1.518	1.561	0.007
1.435	1.54	0.08	–	0.097	0.02	0.010	0.003	0.010	0.014	1.878	1.746	0.009
1.635	1.74	0.07	–	0.089	0.02	0.009	0.003	0.003	0.009	2.067	1.053	0.011
1.835	1.94	0.06	–	0.081	0.02	0.008	–	–	–	–	–	–
2.035	2.14	0.05	–	0.074	0.01	0.007	–	–	–	–	–	–
2.235	2.34	0.06	–	0.081	0.02	0.008	–	–	–	–	–	–
2.435	2.54	0.05	–	0.074	0.01	0.007	–	–	–	–	–	–

thick, then INTBOT – INTTOP is equal 1 and INTEGSEG = RHOXCEE.

INTEGINT –

Interpolated ^{210}Pb activity integrated over the thickness of the non-analyzed interval. The interpolation is done by the log rule of integration: $CAL_{(r,9)}$ = $((CAL_{(r+1,7)} - CAL_{(r,7)})/\ln(CAL_{(r+1,7)}/CAL_{(r,7)})) \times (CAL_{(r+1,1)} - CAL_{(r,2)})$. This value will be zero in cores with analyses done on contiguous intervals, which is the recommended procedure for use with CRS model, but which was not done for PIRLA cores.

SUMBOT –

Total ^{210}Pb activity integrated from the bottom of the core to the bottom of the interval in pCi cm^{-2}: $CAL_{(r,10)}$ = $CAL_{(r+1,11)}$ + $CAL_{(r,9)}$.

SUMTOP –

Total ^{210}Pb activity integrated from the bottom of the core to the top of the interval in pCi cm^{-2}: $CAL_{(r,11)}$ = $CAL_{(r,10)}$ + $CAL_{(r,8)}$.

TTOP –

Time in years before the core was taken at the top of an analyzed interval: $CAL_{(r,12)}$ = $(\ln(CAL_{(1,11)}/CAL_{(r,11)}))/k$. Note that this equation is equivalent to (2).

TBOT –

Time in years before the core was taken at the bottom of an analyzed interval: $CAL_{(r,13)}$ = $(\ln(CAL_{(1,11)}/CAL_{(r,10)}))/k$. This equation is also equivalent to (2).

SEDRATE –

Accumulation rate of bulk sediment in g cm^{-2} yr^{-1}: $CAL_{(r,14)}$ = $k ((CAL_{(r,11)} + CAL_{(r,10)})/2)/CAL_{(r,4)}$. This equation is equivalent to (3).

ORGSEDRT –

Accumulation rate of organic sediment in g cm^{-2} yr^{-1}: $CAL_{(r,15)}$ = $CAL_{(r,14)} \times CAL_{(r,6)}$.

CUMMASST –

Cumulative mass from the top of the core to the top of the analyzed interval in g cm^{-2}: $CAL_{(r,16)}$.

CUMMASSB –

Cumulative mass from the top of the core to the

bottom of the analyzed interval in g cm^{-2}: $CAL_{(r,17)}$.

SDACT –

Standard deviation of total ^{210}Pb activity. This variable is the measured counting error: $CAL_{(r,18)}$.

SDRHO –

Standard deviation of ρ: $(CAL_{(r,5)})$. This variable is assumed to be zero in the PIRLA cores.

SDUNSUP –

Standard deviation of unsupported ^{210}Pb activity. This variable includes the standard deviation of supported ^{210}Pb activity and the counting error $(CAL_{(r,18)})$ combined by the rule (10) mentioned above: $CAL_{(r,20)}$ = $(CAL^2_{(r,18)} + s^2_{\text{supported}})^{0.5}$.

SDRHOCEE –

Standard deviation of the calculation variable $CAL_{(r,7)}$: $CAL_{(r,21)}$ = $(CAL^2_{(r,19)} \times CAL^2_{(r,4)} + CAL^2_{(r,20)} \times CAL^2_{(r,5)})^{0.5}$.

SDINTSEG –

Standard deviation of the calculation variable $CAL_{(r,8)}$: $CAL_{(r,22)}$ = $(CAL^2_{(r,21)} \times (CAL_{(r,2)} - CAL_{(r,1)})^2)^{0.5}$.

SDINTINT –

Standard deviation of the calculation variable $CAL_{(r,9)}$: $CAL_{(r,23)}$ = $((.25 \times (CAL^2_{(r+1,22)}) \times (CAL_{(r+1,1)} - CAL_{(r,2)})^2)^{0.5}$.

SDSUMBOT –

Standard deviation of the calculation variable $CAL_{(r,10)}$: $CAL_{(r,24)}$ = $(CAL^2_{(r,23)} + CAL^2_{(r+1,25)})^{0.5}$.

SDSUMTOP –

Standard deviation of the calculation variable $CAL_{(r,11)}$: $CAL_{(r,25)}$ = $(CAL^2_{(r,22)} + CAL^2_{(r,24)})^{0.5}$.

SDTTOP –

Standard deviation of the age at the top of the analyzed interval, $CAL_{(r,12)}$: $CAL_{(r,26)}$ = $((k^{-2}CAL^{-2}_{(1,11)} \times CAL^2_{(1,25)}) + (k^{-2}CAL^{-2}_{(r,11)} \times CAL^2_{(r,25)}))^{0.5}$.

SDTBOT –

Standard deviation of the age at the bottom of the analyzed interval, $CAL_{(r,13)}$: $CAL_{(r,27)}$ = $((k^{-2}CAL^{-2}_{(1,11)} \times CAL^2_{(1,25)}) + (k^{-2}CAL^{-2}_{(r,10)} \times CAL^2_{(r,24)}))^{0.5}$.

SDSEDRT –

Standard deviation of the accumulation rate of bulk sediment, $CAL_{(r,14)}$: $CAL_{(r,28)} = (k^2 \times CAL_{(r,11)}/CAL_{(r,4)} \times (((CAL_{(r,25)}/CAL_{(r,11)})^2 + (CAL_{(r,20)}/CAL_{(r,4)})^2_j))^{0.5}$.

SDORGRT –

Standard deviation of the accumulation rate of organic sediment, $CAL_{(r,15)}$. This variable is not calculated because of the unknown variation of organic percentage determinations.

Correction of the old-date error of the CRS model

The distribution of unsupported ^{210}Pb activity in perfect sediment cores is described by an exponential curve, which mathematically never reaches zero. CRS calculations, though, require the assignment of a level at which that activity goes to zero. A tiny amount of total, integrated activity is therefore omitted from A_x and A_o. The amount is negligible for recent dates, but at older ages, as ^{210}Pb approaches zero unsupported activity, the neglected fraction represents an increasing proportion of A_x. The result is an increasing divergence of the CRS-model date from the true date. This section describes the correction applied to data from the PIRLA cores.

The method basically adds a small amount of integrated ^{210}Pb to A_x at each level. The problem is to decide how much to add, and where in the core to begin the addition. To do this, the sediment accumulation rate is first assumed to be constant at levels between 80% and 100% of cumulative mass, where 100% of the cumulative mass is found at the assigned level of zero unsupported [210]Pb. If the accumulation rate has been constant, then the ^{210}Pb activity will be a perfectly exponential curve in this interval. The graph of the logarithm of ^{210}Pb activity is then a straight line. The straight line is drawn mathematically from the depth of 80% cumulative mass to the depth of 99% cumulative mass, then extrapolated to 100% cumulative mass. The area of the curve between 99% and 100% cumulative mass is then the amount of A_x to add to each level, and the intercept at 100% cumulative mass is the spot at which to begin the additions. The calculations are iterated three times, and the final dates are calculated from the corrected values for A_x.

Extrapolating CRS-model dates

Dates determined by any method can be extrapolated beyond the data. In this case, extrapolated dates cannot be considered ^{210}Pb dates, but are rough guesses based on dubious assumptions. Extrapolated dates may be useful for comparison with other stratigraphic markers, and therefore have been calculated for the non-Florida PIRLA cores. These extrapolated dates are to be discussed only with full recognition of the assumptions and limits of ^{210}Pb dating in general and the CRS model in particular.

CRS-model dates are calculated from the distribution of activity of unsupported ^{210}Pb in sediment cores. If a sample is taken from a level in a core deep enough to contain unmeasurable unsupported ^{210}Pb, then CRS-model dates cannot be calculated. However, there are a number of reasons to attempt extrapolation of dates below the level of unsupported [210]Pb. Horizon markers such as changes in pollen or diatom assemblages, ash layers, or other stratigraphic features that can be linked to specific historical events may occur deep in a core. Matching the markers with an independent measurement of time strengthens the paleolimnological analysis.

Methods to extrapolate ^{210}Pb dates do exist, but any extrapolation is a poor approximation of a true date, and any correspondence with horizon markers may be pure coincidence. The most appropriate method assumes that a single, long-term equilibrium rate of sediment accumulation can be derived from ^{210}Pb analysis in the upper part of a core. The rate is then extended to sedimentary levels below unsupported ^{210}Pb to calculate older dates. Any change in accumulation rate renders the extrapolation invalid.

Equilibrium accumulation rates for PIRLA cores (except for those from Florida) were chosen by examining profiles of accumulation rates. The value depends on the expert judgement of the person assigning the equilibrium accumulation rate. If the rate was relatively constant over the depth of a core, then the mean rate and its standard deviation were calculated by including all the levels. If the rate increased near the top of the

core, several deep levels of relatively constant accumulation rate, which is assumed to be the pre-disturbance rate, were used to calculate the mean and standard deviation. If the rate varied with no apparent trends, then the entire core was again used to calculate the mean and standard deviation. Obviously, the standard deviation of the third situation is large. The resulting extrapolated dates consequently have large errors, which can be greater than the age itself.

For example, suppose a core with five analyzed levels has accumulation rates of:

Depth (cm)	Accumulation rate $(\mathrm{g\ cm^{-2}\ yr^{-1}})$
0–2	0.0335
2–4	0.0259
4–6	0.0145
6–8	0.0154
8–10	0.0150
10–12	beyond [210]Pb

The obvious equilibrium accumulation rates is 0.0150 with a standard deviation of 0.00045 (the mean and s.d. of 0.0145, 0.0154, and 0.0150). The cumulative mass of the sediment at each level is also known from the measurement or calculation of dry densities (see the earlier section on measured vs. calculated dry densities). An extrapolated date is calculated by the equation:

$$t_e = t_b + ((m_x - m_b)/S_e) \qquad (14)$$

where t_e is the extrapolated age at depth x in years, t_b is the age at the bottom of the bottommost level with unsupported ^{210}Pb, m_x is the cumulative mass at depth x in $\mathrm{g\ cm^{-2}}$, m_b is the cumulative mass at bottom of bottommost level with unsupported ^{210}Pb, S_e is the equilibrium rate of sediment accumulation. The standard deviation of t_e is calculated by deriving the equation for the variance in the same manner as discussed in an earlier section, and is:

$$s.d.t_e = (s.d.^2 t_b + t_e\,(s.d.S_e/S_e)^{0.5})^2)^{0.5} \qquad (15)$$

where $s.d.t_e$ is the standard deviation of t_e, $s.d.t_b$ is the standard deviation of t_b, and $s.d.S_e$ is the standard deviation of S_e.

Extrapolated dates should be used only with great caution, and they should not be called ^{210}Pb dates because they are not. They are at best educated guesswork, and are possibly justified if there is evidence of little or not disturbance in the drainage basin prior to the basal ^{210}Pb date. [210]Pb dates extrapolated into the future have the same degree of validity.

Results and discussion

Age of intervals vs. error

Selected results of error analysis data are presented in this section to illustrate relationships between dates, uncertainties, and other factors. The full set of calculated errors is available in Binford (1986) and from the PIRLA data base.

Of all the 51 cores, Core 1 from Lake Lou in Florida had the greatest error at the bottom interval with measurable unsupported [210]Pb (29%, 155 ± 45 years s.d.). The smallest error at the bottom of the datable section was from Core 1, Lake Barnes in the Adirondacks (1.5%, 128 ± 2.02 years). Extreme errors (maximum and minimum) for each region are shown in Table 3.

The error (95% confidence interval) of a ^{210}Pb date calculated both by first-order propagation and Monte Carlo simulation through the CRS model increases non-linearly with the date (Fig. 3). This relationship is expected because of the logarithmic nature of ^{210}Pb distribution in a sediment column and the random nature of radioactive decay. As measured ^{210}Pb decreases lower in the sediment column, measurement and other errors increase in proportion. Note that only the 95% C.I. (85 and 89 years) from Lake Lou was greater than 40 years. No Monte Carlo 95% intervals were greater than 50 years. At ages of about 140 years, most 95% C.I.'s cluster around 15–25 years, and 95% Monte Carlo intervals are usually 5–20 years, with a few near 40–50.

Table 3. Regional extremes for standard deviations of dates in lowermost sedimentary sections with measurement ^{210}Pb.

Region	Lake	Core	Date ± Max. s.d.	Date ± Min. s.d.
Adirondacks	Arnold	2	148 ± 24 (16.4%)	
	Barnes	1		128 ± 4 (3.1%)
New England	Mud	1	137 ± 21 (15.4%)	
	Little Long	1		154 ± 6 (4.0%)
Florida	Lou	1	155 ± 45 (29.3%)	
	Mirrow	4		148 ± 6 (4.1%)
Northern Great	Otto Mielke	1	154 ± 31 (20.3%)	
Lakes	Brown	14		130 ± 4 (3.4%)

CRS DATE VS. 95% C.I.

Log 95% C.I. = 0.0078 * CRS Date + 0.1646
$R^2 = 0.82$
$p < 0.01$

95% C.I. (yr)

CRS DATE (yr)

M.C. MEDIAN DATE VS. 95% INTERVAL

Log 95% Int. = 0.0137 * M.C. Date − 0.4325
$R^2 = 0.82$
$p < 0.01$

95% INTERVAL

MONTE CARLO MEDIAN DATE (yr)
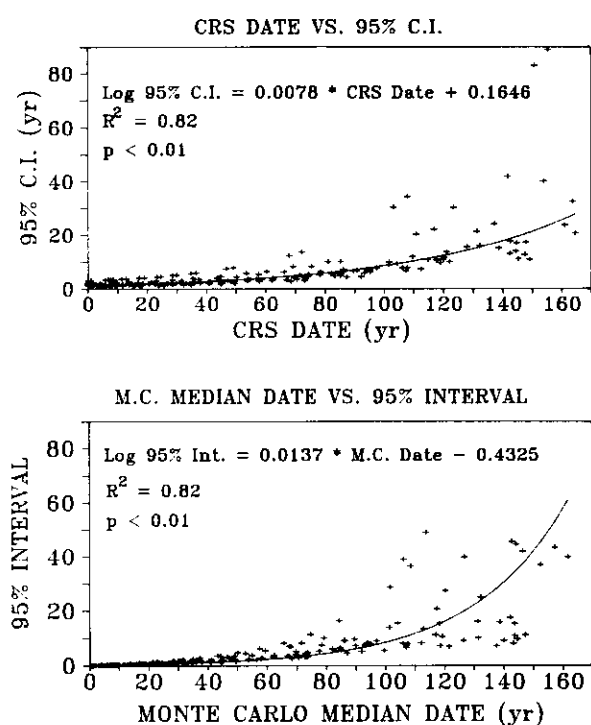
Fig. 3. Variation of 95% confidence interval with CRS date and 95% occurrence interval for Monte Carlo medians for Florida lake cores. 95% confidence intervals are calculated as 1.96 × the standard deviation (see text for explanation). Monte Carlo intervals include the range of 950 of the 1000 output dates at each level.

Comparison of first-order error analysis and Monte Carlo simulations

Monte Carlo simulations were performed on 12 cores from north Florida lakes; Each simulation calculated 1000 different ^{210}Pb profiles and dates

for a core. Simulated ^{210}Pb activity at each level was designated by sampling from a normal distribution with a mean equal the measured activity and standard deviation equal the measured counting error. The RANNOR function of the Statistical Analysis System (SAS Institute 1985: page 267) provided the pseudo-random, normally distributed variate. Simulated ^{210}Pb activities were then used to calculate CRS-model, ^{210}Pb dates. Each level within a core then has 1000 dates with a particular distribution determined by counting error and the CRS equation. The median date and range within which 95% of all calculated dates fall are compared with the 95% confidence interval of the First-Order error analysis.

Standard CRS dates are slightly older (mean difference 0.6%) than Monte Carlo median dates (Fig. 4). This result is unexpected, and indicates a slight bias in either the standard CRS model, or in the use of a normal distribution to sample ^{210}Pb activity for Monte Carlo analysis.

Although the First-Order 95% confidence intervals and the Monte Carlo 95% ranges are slightly different, the general agreement is good until the C.I. exceeds 15 years (Fig. 5). The average age of a 15-yr C.I. is about 130 CRS-model years (see Fig. 3: regression line). This disconformity suggests that errors calculated for dates older than 130 years are unreliable. Consequently, the actual dates are also uncertain, and should be used with care and skepticism.

This comparison suggests that First-Order error analysis is sufficient for all PIRLA cores at dates less than 130 years. Older dates must be
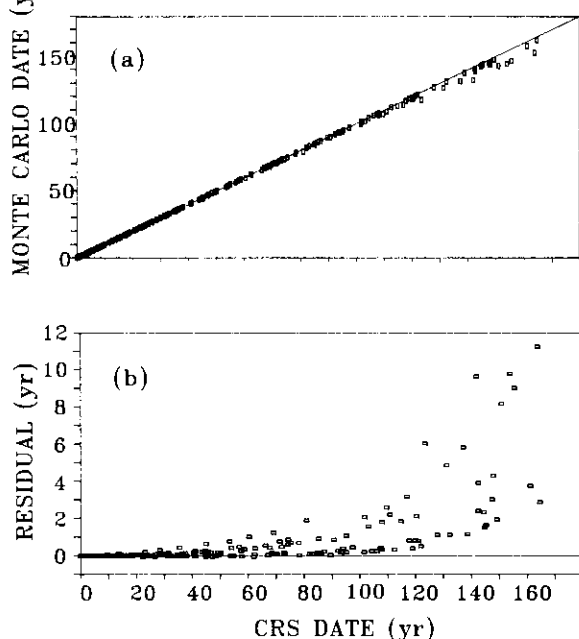
MONTE CARLO VS. FIRST-ORDER ANALYSIS

Fig. 4. Comparison of simple CRS dates and median dates from Monte Carlo simulation.
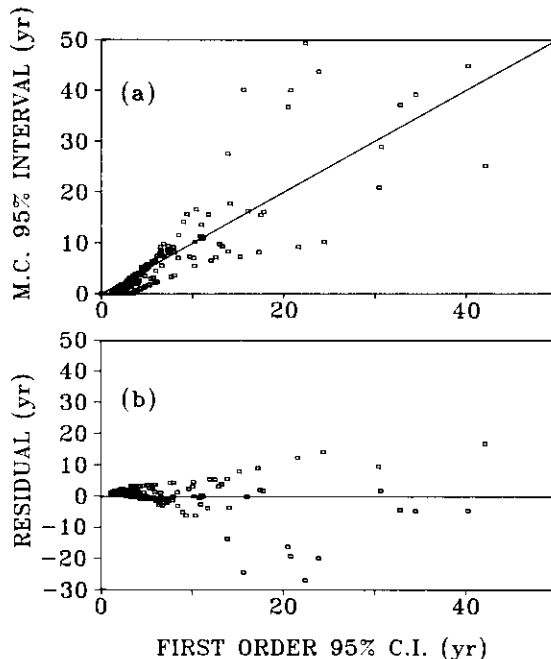
MONTE CARLO VS. FIRST-ORDER ERRORS

Fig. 5. Comparison of First-Order confidence intervals and Monte Carlo simulation 95% occurrence intervals. Note that two data points (CRS error = 83 and 89 years, M.C. error = 42 and 46 years) are not shown.

treated as approximations, and checked carefully for correspondence with other markers that indicate dates. First-Order methods are computationally simpler and less expensive, and therefore more efficient for analyzing many cores. This analysis does not necessarily extend to either CRS models with mixing terms or to CIC models. CIC errors are calculated by a completely different method (see Robbins, 1978). First-Order error analysis of mixing-CRS models would probably be valid, but the procedure has not yet been evaluated.

Acknowledgements

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