# STUDY OF THE PARAMETERS AFFECTING THE CORRELATION OF BACKGROUND VERSUS COSMIC RADIATION IN CO<sub>2</sub> COUNTERS: RELIABILITY OF DATING RESULTS

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ABSTRACT. Systematic treatment of the data recorded by our guard counters and corrections introduced for meteorological factors has allowed observations on solar events clearly manifested in the readings. Examples are the solar flares of March 1989 and especially of June 1991, which caused a *ca*. 10% decrease in the cosmic radiation flux reaching the counters. A sinusoidal variation in the cosmic-ray flux with a period of one year is also clearly manifested in the data. The observation that the background in the <sup>14</sup>C measurements depends on the intensity of the cosmic radiation has led to the use of monthly correlations for the determination of the best background value to be used in the age calculations. This reduces the error significantly. However, various factors such as random statistical fluctuations of the background measurements may affect the slope of the correlations and consequently the calculated age of the samples. Long-term observations of the relation between background values and coincidence counts have led to constraints in the slope of the correlation. A simple extension of the fitting procedure is explored, which maintains the physically meaningful range of the slopes, but is flexible to adjust for the seasonally varying contributions to the variations of the cosmic-ray flux.

### INTRODUCTION

 $CO_2$  gas proportional counters, in conjunction with the production of very pure  $CO_2$  samples, have been shown to produce reliable, high-precision radiocarbon dating results that can be used for calibration purposes (Münnich 1957; Grootes 1977; Kromer and Münnich 1992; Stuiver and Reimer 1993). The background value of each counter must be understood and measured accurately. The stability of the background in such counters has been demonstrated recently (Kromer and Münnich 1992). The primary factor affecting the background variation is the cosmic-ray intensity reaching the counters (Loosli *et al.* 1980; Theodórsson 1996: 73–96, 216–220). Münnich (1957) suggested the use of monthly correlations with a varying slope for the calculation of the best background value for each sample. Calculation of the background from such correlation, rather than the use of a mean background value, considerably reduces the error in the background and consequently the error in the age (Schlitzer *et al.* 1985). The monthly correlations are important for refreshing the data and taking into account the readjustment of the high voltage settings, which is required approximately every year.

The slope of the correlation of background *versus* coincidence counts is governed basically by pressure and temperature change of the atmosphere (Håkansson 1980). However, due to statistical fluctuation of the background, a limited number of background measurements may not reproduce the expected slope well, causing artificial changes in calculated ages. This paper is therefore an attempt to better understand the factors affecting the slope of the correlation and especially the changes in the cosmic radiation intensity by using the guard counters that provide a very good system, as it appears, for monitoring the incoming radiation. The paper also suggests a preliminary tentative procedure for safeguarding the system against rapid statistical fluctuations and improving, to a certain degree, the reliability of the age results without unnecessarily increasing or decreasing the errors.

#### SAMPLE PREPARATION PROCEDURE

We prepare samples used by the Laboratory of Archaeometry of the National Center for Scientific Research "Demokritos" in Athens in the following manner. All organic samples are subjected to a chemical pretreatment to extract the compound containing the set of carbon atoms related to the event sought to be dated (Hedges 1992). They are then combusted by a de Vries-type continuous combustion system (de Vries and Barendsen 1953; Münnich 1957) and converted to CO<sub>2</sub>. All other oxides are removed by reaction with KMnO<sub>4</sub> and the CO<sub>2</sub> is precipitated to carbonate in a CaCl<sub>2</sub>/NH<sub>4</sub> solution. The sample is then reconverted to CO<sub>2</sub> by HCl. In the final purification step, the impurities of the gas are removed by passing the sample through a column filled with activated charcoal at 0°C (Kromer and Münnich 1992). The final step in the sample preparation procedure is the volume calibration, in which the mass of every sample is adjusted to exactly 8.47 g of CO<sub>2</sub>.

## DESCRIPTION AND MANAGEMENT OF THE COUNTING SYSTEM

The laboratory possesses two similar gas proportional counters, named counter 2 and counter 3, manufactured at the Institute of Environmental Physics, Heidelberg University, as part of a joint research project funded by Volkswagen Stiftung. Counter 3 lies over counter 2. They are 81.2 cm long, 8-cmdiameter (4 L) electrolytic copper cylindrical tubes (Schoch et al. 1980), closed at the end with 5-mmthick flat quartz plates. The anode is 50-µm-diameter tungsten wire. Each counter is inside a 120-cmlong, double-walled, propane flow anticoincidence guard counter with 9 anode wires. Between the inner and guard counters are 2-cm-thick cylindrical lead blocks that prevent most of the radiation (mainly y rays) generated in the walls of the guard counter from entering the inner counter. The whole system is shielded on all sides against cosmic rays with 10-cm-thick lead blocks. It is located in the basement of a two-story building (2 ceilings, ca. 40 cm concrete in total), 3 m below ground level (Dep et al. 1994). Air conditioning keeps the room temperature constant at 22°C. The data acquisition unit is a personal computer, which records the counter parameters every 2 h. The working gas pressure of the sample is ca. 1.2 atmospheric. Typically, the samples are measured for two or three days. Standard and background measurements are performed once or twice a month. We use the laboratory substandard "Wilhelm" (Karlen et al. 1966), prepared at the Radiocarbon Laboratory of the Institute of Environmental Physics of the University of Heidelberg. The activity of the standard is 10.196 times the activity of NIST HOxI. The background count rate is on the order of 4 cpm. A counting time for the background of ca. 4000 min ensures the collection of >10,000 counts. Thus, the statistical error of each background measurement is <0.8% (0.03-0.04 cpm).

Every month, we produce a new background *versus* cosmic radiation (coincidence counts) correlation (Fig. 1) containing the data of the ten most recent background measurements. The background value, which is subtracted from the anti-coincidence counts of each sample, is calculated by linear regression of the experimental points of this correlation (Münnich 1957; Kromer and Münnich 1992):

$$Y = B \times X + A \tag{1}$$

where Y is the background count rate and X is the coincidence count rate recorded in each background measurement. B is the slope of the line and A is the constant value, *i.e.*, the intercept at zero cosmic radiation. The errors of A and B are calculated according to standard techniques (Bevington 1969; Gomer 1989) and used in the calculation of the confidence interval of the age error. Approximately every year, the high voltage settings of the counters are increased to ensure optimum operation of the counters always in the middle of the plateau (Knoll 1979). A new background *versus* cosmic radiation correlation begins after every change in the operating high voltage. The background error calculated from the monthly background correlations is ca. 0.25%, instead of ca. 1%, which is the standard deviation of the mean background value derived from background measurements performed in the period of a year. The correlation therefore provides a significant improvement in the precision of the dating.



Fig. 1. A typical 10-point correlation of background versus coincidence counts (cosmic radiation). R = correlation coefficient, N = number of experimental points.

# **RESULTS AND OBSERVATIONS**

## **Cosmic-Ray Flux Fluctuation**

The guard counters provide a very convenient method for monitoring incoming radiation. Since the main component of this radiation is due to the cosmic-ray flux reaching our counter installation, their recordings have been used for observing its fluctuations.

Figure 2 presents the cosmic radiation intensity recorded in guard counter 2 of the Athens laboratory from September 1986 to April 1997, as well as that recorded in guard counter 12 at the Heidelberg laboratory from January 1990 to April 1997. The data show that the cosmic radiation intensity is subject to amplitude modulation with a period of one year, with troughs in August and peaks in February. These are sometimes distorted by strong isolated depressions that may be due to solar flares and magnetic winds that produce a shielding effect from the cosmic radiation reaching the Earth (Tandon and Bhatia 1965; Xiaoping *et al.* 1985; Murali *et al.* 1991). Two such events, clearly shown in Figure 2a, occurring in March 1989 and June 1991, were identified as solar flares in the literature (Morton and Mathews 1993; Walker and Wong 1993; M.O.P. 1991). Of special interest is the event in June 1991, which produced a decrease of 9.5% in cosmic radiation. It lasted for about two months

and was evidently associated with a strong solar flare that had intensified the shielding effect of the magnetic field of the Earth. This event was also recorded in Heidelberg (Fig. 2b) and in the Southern Hemisphere (Pretoria, South Africa; J. C. Vogel, personal communication).



Fig. 2. Variation of the daily cosmic radiation recorded by the guard counter 2 in the Athens laboratory (curve a) from September 1986 to April 1997 compared to the equivalent variation recorded by guard counter 12 in the Heidelberg laboratory (curve b) from January 1990 to December 1995

The yearly periodicity is probably due to temperature variations throughout the atmosphere, which affect the altitude of the main muon production level (Theodórsson 1996). It could also be due to the co-rotation anisotropy between the sun and the interplanetary magnetic field, or to the Earth's orbital motion effect, although there are strong objections against the last two reasons (Thambyahpillai 1975). The annual periodicity is apparently overlapped by a long-term periodicity that follows the 11-yr solar cycle. It is interesting to note, in support of the last observation, that the beginning of the cosmic-ray intensity recordings in the Athens laboratory coincides with the number 22 11-yr solar cycle that started in 1986 (N.O.A.A. 1993, 1994).

These changes were also studied in relation to some meteorological parameters such as surface atmospheric pressure, relative humidity and temperature, obtained from the Bulletin of the National Observatory of Athens (N.O.A. 1991, 1992, 1993, 1994, 1995, 1996). Figure 3 shows the variation of the cosmic-ray flux as recorded by guard counter 2 and the abovementioned meteorological parameters *versus* time, taken in 2-h intervals in December 1993. Only the atmospheric pressure (Fig. 3c) shows a clear anti-correlation with cosmic radiation (Fig. 3d). The dependence of the cosmic-ray intensity on the changes of the atmospheric pressure arises partly as a mass absorption effect and partly from the finite lifetime of the muon (Thambyahpillai 1975) and it is more clearly observed during the cold months when the variations in pressure are much stronger (Håkansson 1980). Figure 4 shows a typical



Fig. 3. Variation of the temperature (a), relative humidity (b), atmospheric pressure (c) and cosmic-ray flux (d), as recorded by guard counter 2 taken in 2-hr intervals in December 1993

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correlation of the cosmic radiation with the atmospheric pressure during a cold month in Athens (December 1993). The slope of this correlation is 0.19% per mmHg. Equivalent correlation in Heidelberg shows similar behavior in the same period, resulting in a slope of 0.23% per mmHg. The spread of the pressure values during the cold months is on the order of 30 mmHg, instead of 10 mmHg during the hot months. Therefore, the winter correlations are more representative of the overall behavior of the cosmic radiation with pressure. The error in the measurements is of the same order as the size of the points on the graph. Most of the points deviate >2 $\sigma$  from the best-fit line, and this may be explained by the fact that the intensity of the cosmic-ray flux reaching the surface of the Earth is not only a pressure-dependent phenomenon (Dutt and Thambyahpillai 1965; Håkansson 1980; Theodórsson 1996). This is reflected in our data by the fact that the cosmic radiation versus atmospheric pressure of the years 1992–1996 shows two distinct parallel straight lines with approximately the same slopes, one from May to September (hot months) and the other from October to April (cold months).



Fig. 4. Variation of the cosmic-ray flux as recorded in 2-hr intervals by guard counter 2 vs. atmospheric pressure in December 1993. The equation is a linear regression fit. R = correlation coefficient, N = number of experimental points.

For 1991, when the strong solar event occurred, there are three distinct correlations with increasing slopes from June to November 1991, stabilizing by December 1991 (Fig. 5). The final slope in 1991 approaches the same value as that of the years 1992–1996, indicating that the relationship between cosmic radiation reaching the Earth and atmospheric pressure is basically always the same. To exclude the possibility that the annual periodicity, shown in Figure 2, is due to an atmospheric pressure effect, the recordings of guard counter 2 were normalized to the same pressure. The atmospheric pressure value of 752.7 mmHg, which is the mean value from June 1991 to December 1996,



Fig. 5. Variation of the cosmic radiation recorded by the guard counter 2 versus the atmospheric pressure taken in 2-hr intervals from June to December 1991. The equations are produced from the corresponding linear regression fit of each group of points. R = correlation coefficient, N = number of experimental points.

was chosen for normalizing all the guard recordings. The normalized values were calculated from the equation (Facorellis 1996)

$$G_{n} = G_{m} - (752.7 - P_{m}) * |S|$$
(2)

where  $G_n$  is the guard normalized value,  $G_m$  is the measured guard value,  $P_m$  is the corresponding atmospheric pressure and S = -1.9 cpm × mmHg<sup>-1</sup> is the slope of the linear regression fits of the cosmic radiation *versus* atmospheric pressure. The value for the slope S was taken as the weighted mean of the stabilized part of 1991 (November, December) and the years 1992–1996. In Figure 6, the normalized guard values are plotted *versus* time for the years 1991–1996. It is obvious that the annual periodicity in the cosmic-ray intensity is still evident, suggesting that it is not a pressuredependent phenomenon. The June 1991 strong solar event is responsible for the exceptionally low values in the initial part of the graph.

#### **Stability and Variation of Background**

The above observations provide the necessary information for a better understanding of the background variations. Figures 7a and 7b present the fluctuation of the background (anticoincidence counts) and the cosmic radiation (coincidence counts), respectively, in sample counter 2 for a period of *ca*. 6 yr (October 1990 to April 1996). Figure 7c presents the corresponding counts of guard counter 2 for the same period. One can see that the background, coincidence and guard counts exhibit exactly the same behavior, indicating a direct relation between background and cosmic-ray



Fig. 6. Variation of the cosmic radiation intensity normalized to the mean atmospheric pressure value (752.7 mmHg) taken in 2-hr intervals recorded by the guard counter 2 from June 1991 to December 1996

flux. The overall fluctuation of the background is 0.47 cpm, which is quite small considering the long time period studied. This time spanned includes several successions of hot and cold months, the strong solar flare of June 1991, as well as the gradual shift of the intensity of the cosmic radiation to higher values as the current 11-yr solar cycle reaches its completion.

The direct relationship of the background value in both counters with the intensity of the cosmic radiation should therefore give monthly background correlations with constant slope. However, this is far from reality. Actually, the slope of the correlations with 10 background measurements fluctuates, producing deviations from the mean value that frequently reach 50%, and more rarely, approach or even exceed 100%. It is also important to note that the slope can even take, in some rare cases, negative values, which have no physical meaning.

The long-term observations and correlations, however, better reflect the true dependence of background on the cosmic radiation. By plotting all the points between consecutive changes of the counter high voltage (measurements performed during *ca.* 1 year), overall continuous correlations are produced. In the last 7 years, seven such long-term correlations were produced for both counters (Table 1). Although they still show some considerable changes in the slope B and the constant A, the weighted mean of these parameters is the same, for both counters within 1 $\sigma$  (counter 2: B = 0.0064  $\pm$  0.0007 and A = 0.99  $\pm$  0.34, counter 3: B = 0.0068  $\pm$  0.0008 and A = 1.01  $\pm$  0.42). This fact suggests that the dependence of the background on the intensity of the cosmic radiation follows a particular rate of increase ( $\approx$ 0.0066 cpm for every coincidence count). That is, irrespective of the abso-



Fig. 7. (a) Variation of the background in counter 2 from October 1990 to April 1996. The statistical error of each background measurement is 0.03–0.04 cpm. (b) Variation of the coincidence counts (cosmic radiation) as recorded by the sample counter 2 during the above background measurements. (c) Variation of the cosmic-ray flux as recorded by the guard counter 2 during the above background measurements.

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lute values of background, which may vary between summer and winter or by other atmospheric changes, the slope is practically unchanged. The similar construction of the two counters results in similar slopes. These results clearly point to the conclusion that the abrupt variations of the slope of the short-term correlations are due to random statistical fluctuations of the background measurements and are not justified as real changes of quality or behavior of the cosmic radiation.

The above observations and treatment of the data have revealed some sources of inaccuracy that need attention, as they may affect the calculated ages to a lesser or greater extent. We have performed some calculations in an attempt to estimate the total variation in the age of a sample that may occur by using monthly background correlations of 10 points. We use three different 10-point slopes (0.0110, 0.0050 and 0.0014) obtained in three consecutive monthly background correlations (October, November and December 1991) of counter 3. Obviously, if the coincidence count value of a particular sample falls near the middle of the range of coincidence counts observed in the counter, the effect of different slopes on the background value calculated for this sample would be negligible and thus the effect on the age also negligible. However, if the coincidence counts happen to be near the highest or the lowest side of the range, then statistical differences may occur in the calculated background value and therefore in the age, for a sample measured during a period of high slope (October 1991) or of low slope (December 1991). For example, assuming the slope of November 1991 in counter 3 to be the closest one to the "true" slope, then we calculate background deviations from the "true" value of 0.07 and -0.03 cpm for a sample with high coincidence counts measured in October 1991 (high slope) and December 1991 (low slope), respectively. Consequently, this leads to age shifts to younger ages by 28–55 yr, depending on the age of the sample, if it is measured in the preceding period of high slope, and to older ages by 12-24 yr if it happened to be measured during the following period of low slope.

These apparent shifts in the age of a sample calculated at extreme cases, and due to the instability of the slope of the monthly background correlations, may obviously be more important in measurements of samples that need to be dated with very high precision, *e.g.*, tree-ring samples for calibration purposes. It can also be more serious with routine samples and especially after changes in the counter high voltage settings in cases where one needs a quick but reliable age.

## SAFEGUARDING THE SYSTEM AND IMPROVING RELIABILITY

Therefore, it would be desirable to safeguard one's system for situations of this kind and improve the reliability of the resulting ages. We now turn to the procedure to correct for a varying background component due to statistical fluctuations that are not related to the cosmic-ray flux as demonstrated above. The procedure must be flexible to adjust for changes in counter parameters such as high-voltage or discriminator thresholds, yet reflect the dependence on the physical meteorological condition of the atmosphere. Accordingly, we use a sliding correlation of background versus coincidence counts, calculated over the most recent 10 measurements. Applied without constraints, the correlation may lead to unacceptably large statistical fluctuations of up to 0.1 cpm for extreme cases as observed in fall 1991. On the other hand, keeping the slope numerically constant would not allow the adjustment of background parameters to accommodate for the seasonal variations discussed above.

We have investigated a simple procedure to control the fluctuation of the slope. By extending the straight line to zero cosmic radiation, we can introduce a constraint at the intercept with the y-axis. This means introducing a "fixed" point to be used together with the experimental points for stabilizing the slope. A "fixed" positive background point with a certain degree of freedom at zero cosmic radiation would prevent the slope from turning to unjustifiably high or low values. At the same time,

it would give the necessary freedom for changes, if the absolute background values change, due to seasonal variations or genuine cosmic events. The proposed values for such a "fixed" point could be  $1 \pm 0.5$  cpm for both counters. This value represents the weighted mean of the constant A and its maximum standard deviation after 7 years of overall continuous background correlations including *ca.* 140 background measurements in each counter (Table 1).

It is important to note that the use of a slope constraint in the form of a "fixed" point for zero cosmic radiation does not include the assumption of a strictly linear relation of background *versus* coincidence count rate over the full range of cosmic-ray flux variation, which needs to be checked in the future. On the other hand, the use of such a constraint in the background correlation should fulfill two further criteria: 1) The absolute age should not deviate from the "real" one, and 2) the age error must not be artificially decreased or increased.

Counter 2	Points	Time range	Slope B	σΒ	Constant A	σΑ	
	35	10/1990-11/1991	0.0044	0.0011	1.79	0.49	
	17	12/1991-8/1992	0.0106	0.0034	-1.13	1.59	
	16	9/1992-3/1993	0.0099	0.0016	-0.18	0.75	
	14	3/1993-9/1993	0.0092	0.0045	-0.41	2.12	
	21	10/1993–9/1994	0.0062	0.0019	0.94	0.86	
	30	11/19946/1996	0.0064	0.0024	0.89	1.16	
	10	9/1996-3/1997	0.0016	0.0100	3.00	4.81	
Weighted Mean			0.0064	0.0007	0.99	0.34	
Counter 3	Points Time range		Slone D	۳D	Constant A	- 1	
	1 Onits	Thile Talige	Slope D	OB	Constant A	0A	
	18	10/1990-4/1991	0.0060	0.0019	1.34	0.99	
	18 34	10/1990–4/1991 5/1991–8/1992	0.0060 0.0055	0.0019 0.0014	1.34 1.60	0.99 0.70	
	18 34 14	10/1990–4/1991 5/1991–8/1992 9/1992–3/1993	0.0060 0.0055 0.0088	0.0019 0.0014 0.0021	1.34 1.60 0.12	0.99 0.70 1.10	
	18 34 14 16	10/1990-4/1991 5/1991-8/1992 9/1992-3/1993 3/1993-9/1993	0.0060 0.0055 0.0088 0.0047	0.0019 0.0014 0.0021 0.0024	1.34 1.60 0.12 2.14	0.99 0.70 1.10 1.26	
	18 34 14 16 22	10/1990–4/1991 5/1991–8/1992 9/1992–3/1993 3/1993–9/1993 10/1993–9/1994	0.0060 0.0055 0.0088 0.0047 0.0085	0.0019 0.0014 0.0021 0.0024 0.0026	1.34 1.60 0.12 2.14 -0.09	0.99 0.70 1.10 1.26 1.35	
	18 34 14 16 22 28	10/1990-4/1991 5/1991-8/1992 9/1992-3/1993 3/1993-9/1993 10/1993-9/1994 11/1994-6/1996	0.0060 0.0055 0.0088 0.0047 0.0085 0.0097	0.0019 0.0014 0.0021 0.0024 0.0026 0.0030	1.34 1.60 0.12 2.14 -0.09 -0.50	0.99 0.70 1.10 1.26 1.35 1.59	
	18 34 14 16 22 28 10	10/1990-4/1991 5/1991-8/1992 9/1992-3/1993 3/1993-9/1993 10/1993-9/1994 11/1994-6/1996 9/1996-3/1997	0.0060 0.0055 0.0088 0.0047 0.0085 0.0097 0.0121	0.0019 0.0014 0.0021 0.0024 0.0026 0.0030 0.0059	1.34 1.60 0.12 2.14 -0.09 -0.50 -1.93	0.99 0.70 1.10 1.26 1.35 1.59 3.13	

 TABLE 1. Linear Fitting Equations of Long-Term Variation of Background Count Rate versus

 Coincidence Count Rate of Counters 2 and 3 within the Same High-Voltage Operation

## Effect of the Constraint on Absolute Age

To check the effect of using a fixed point on the actual absolute age, one needs samples with accurately known absolute ages. Such samples can be dendrochronologically dated tree rings or historical samples of known age. The comparison with known-age samples may not be a very conclusive task since it involves using a calibration curve, which induces further errors and possible age shifts. However, this attempt was thought necessary for the completion of this work and for excluding the possibility of large discrepancies.

For this purpose wood samples were used, kindly offered by Dr. Patrick Gassmann, director of the Laboratory of Dendrochronology of the Cantonal Archaeological Museum of Neuchâtel in Switzerland. Two of the samples originated from two Swiss lake settlements, one from the Late Bronze age, in Cortaillod-Est (Gassmann 1984) and the other from the Neolithic period, in Hauterive-Champréveyres (Benkert and Egger 1986). Both samples were prepared and dated. They had 242 and 30 tree rings, respectively. From the first sample (DEM-387) 30 consecutive rings were selected with a true age  $1031 \pm 15$  yr BC. For the second sample (DEM-388), 22 rings were used with true age 3819  $\pm 11$  yr BC. Table 2 presents the calibrated ages of the samples (Pearson and Stuiver 1993; Stuiver and Pearson 1993) as they result from the different background correlations, together with their true ages. The calibration was performed with the Radiocarbon Calibration Program Rev. 3.0.3c of the Quaternary Isotope Laboratory of the University of Washington (Stuiver and Reimer 1993). As the true age of the samples is known, age ranges derived from the calibration program other than ones that included the true age were rejected irrespective of the calculated probability. From Table 2 it can be seen that both the correlations with a fixed point and without (free) produce practically the same <sup>14</sup>C age result for both samples.

TABLE 2. Ages and errors of two tree-ring samples obtained with free background correlations and the proposed fixed point and associated errors (see text). The true age is derived dendro-chronologically.

Correlation	DEM-387 age (BP)	Cal age (BC)	1σ probability	DEM-388 age (BP)	Cal age (BC)	1σ probability
Free	2859 ± 26	1039–979	75%	5054 ± 23	3815-3797	20%
Fixed point	2861 ± 27	1044–979	76%	5060 ± 23	3818-3800	21%
True age		1031 ± 15			3819 ± 11	

# Effect of the Constraint on the Age Error

To investigate the effect of the correlations with a fixed point on the age *error* compared to the *error* of the free correlations, a large number of samples were tested. These samples were measured at least once in both counters at different times. Their ages and associated errors were calculated using correlations including the fixed point  $1 \pm 0.5$  cpm for both counters. They were then compared to the corresponding age errors calculated from the correlation without any fixed point (free). The age errors in most cases are the same or different by only 1–2 yr. The good agreement of the results demonstrates that the introduction of a constraint in the form of a "fixed" point with the value of 1 and variance  $\pm 0.5$  does not decrease or increase the error in the age, even when the sample is measured more than once in different counters.

In summary, the advantage of using a fixed point in the background correlation is that it prevents the slope of the linear regression fit from deviating significantly from the mean value of the background correlations. This is particularly valuable in the case of abrupt changes due to random statistical fluctuations. In these cases, the use of a fixed point allows the accurate calculation of the age and associated realistic error of the samples. The suggested constraint is quite simple as one can easily introduce a "fixed" point or delete it according to the conditions without any change in the software or the calculation routines.

# CONCLUSION

This work has shown that the guard counter proved to be a very convenient system for monitoring the intensity of the cosmic radiation received by the Earth, allowing interesting observations on secular variations and solar events, such as those during March 1989 and June 1991.

The effect of atmospheric pressure, relative humidity and temperature on the intensity of the cosmic-ray flux reaching the surface of the Earth was investigated and found to be rather minute. Based on our daily measurements, the cosmic-ray flux fluctuation from September 1986 to April 1997 exhibits a short-term annual periodicity and a long-term one that follows the 11-yr solar cycle. The use of monthly correlations of background *versus* cosmic radiation is an important procedure to improve accuracy in  $CO_2$  counters. However, our study showed that extreme statistical variations may cause significant changes in the slope of the background correlation. This can induce smaller or larger shifts in the calculated age of samples when measured in a certain counter.

The long-term observation of the background and the intensity of the cosmic-ray flux during the last 7 years showed that this Bkg/CR relation is constant with a specific value of slope whose mean is equal in both counters. Based on that, a refinement to the background correlation procedure proposed here may stabilize the slope and assure the reliability of results in all circumstances. This can be achieved very simply by introducing a "fixed" point with a certain variance in the monthly background correlation, which in the case of the counters of the Athens laboratory would be  $1 \pm 0.5$  cpm. The effect of such a constraint on the age and errors of the results was checked by dendrochronologically dating tree-ring samples and the error treatment of a large number of ordinary samples. The coherence of the results proved that the use of a fixed point in the background correlation may provide a very valuable safeguard to the system.

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## REFERENCES

- Benkert, A. and Egger, H. 1986 Dendrochronologie d'un site du Bronze Final Hauterive-Champréveyres (Suisse). Société Préhistorique Française 10: 486– 502.
- Bevington, P. R. 1969 Data Reduction and Error Analysis for the Physical Sciences. New York, McGraw-Hill Book Company: 92-118.
- de Vries, H. and Barendsen, G. W. 1953 Radiocarbon dating by a proportional counter filled with carbon dioxide. *Physica* 19: 987–1003.
- Dep, L., Elmore, D., Fabryka-Martin, J., Masarik, J. and Reedy, R. C. 1994 Production rate systematics on insitu cosmogenic nuclides in terrestrial rocks: Monte Carlo approach of investigating <sup>35</sup>Cl(n,g) <sup>36</sup>Cl. Nuclear Instruments and Methods in Physics Research B92: 321-325.
- Dutt, J. C. and Thambyahpillai, T. 1965 Atmospheric effects on the cosmic-ray intensity at a depth of 60 m.w.e. underground at London. *Journal of Atmospheric and Terrestrial Physics* 27: 349–358.
- Facorellis, Y. (ms.) 1996 Study of the conditions and parameters for high precision dating with <sup>14</sup>C. Ph.D. Thesis, University of Patras, Greece.
- Gassmann, P. 1984 Dendrochronologie: 100,000 cernes sur Cortaillod-Est. Archéologie Suisse 7(2): 62–68.

- Gomer, T. (ms.) 1989 Elektronik und Datenverarbeitung einer Vielfachzahlrohranlage. Thesis, Institut für Umweltphysik an der Universität Heidelberg, Germany: 24-34.
- Grootes, P. M. (ms.) 1977 Thermal diffusion, isotope enrichment and radiocarbon dating beyond 50,000 years BP. Ph.D. Thesis, Groningen, The Netherlands.
- Håkansson, S. 1980 Temperature-dependent seasonal variation of the background in counters used for radiocarbon dating. *In* Stuiver, M. and Kra, R. S., eds., Proceedings of the 10th International <sup>14</sup>C Conference. *Radiocarbon* 22(2): 448–454.
- Hedges, R. E. M. 1992 Sample treatment strategies in <sup>14</sup>C dating. In Taylor, R. E., Long, A. and Kra, R. S., eds., Radiocarbon After Four Decades: An Interdisciplinary Perspective. New York, Springer-Verlag: 165– 183.
- Karlen, I., Olsson I. U., Kallberg P. and Kilicci, S. 1966 Absolute determination of the activity of two <sup>14</sup>C dating standards. Arkiv Geofysik 6: 465–471.
- Knoll, G. F. 1979 Radiation Detection and Measurement. New York, John Wiley and Sons: 182–185.
- Kromer, B. and Münnich, K. O. 1992 CO<sub>2</sub> gas proportional counting in radiocarbon dating: Review and perspective. *In* Taylor, R. E., Long, A. and Kra, R. S.,

eds., Radiocarbon After Four Decades: An Interdisciplinary Perspective. New York, Springer-Verlag: 184–197.

- Loosli, H. H., Heimann, M. and Oeschger, H. 1980 Lowlevel gas proportional counting in an underground laboratory. *In* Stuiver, M. and Kra, R. S., eds., Proceedings of the 10th International <sup>14</sup>C Conference. *Radiocarbon* 22(2): 461–469.
- Magnetic Observatory of Pendeli 1991 Magnetic Bulletin. Athens, Greece, Institute of Geology and Mineral Exploration.
- Morton, Y. T. and Mathews, J. D. 1993 Effects of the 13-14 March 1989 geomagnetic storm on the *E*-region tidal ion layer structure at Arecibo during AIDA. *Journal of Atmospheric and Terrestrial Physics* 55(3): 467-485.
- Münnich, K. O. 1957 Messung naturlichen Radiokohlenstoffs mit einem CO<sub>2</sub> proportional Zahlrohr. Einige Anwendungen der Methode., Inaugural-dissertation zur Erlangung der Doktorwurde hohen Naturwissenschaftlich-mathematischen Fakultat der Ruprecht-Karl-Universitat zu Heidelberg, Germany.
- Murali, A. K., Prabhakaran Nayar, S. R. and Somayajulu, V. V. 1991 Role of periodic fluctuations in solar windmagnetosphere coupling. *Journal of Atmospheric and Terrestrial Physics* 53(10): 881–887.
- National Observatory of Athens 1991 Climatological Bulletin, Institute of Meteorology and Physics of the Atmospheric Environment, Athens.
- \_\_\_\_\_1992 Climatological Bulletin, Institute of Meteorology and Physics of the Atmospheric Environment, Athens.
- 1993 Climatological Bulletin, Institute of Meteorology and Physics of the Atmospheric Environment, Athens.
- \_\_\_\_1994 Climatological Bulletin, Institute of Meteorology and Physics of the Atmospheric Environment, Athens.
- \_\_\_\_1995 Climatological Bulletin, Institute of Meteorology and Physics of the Atmospheric Environment, Athens.
- 1996 Climatological Bulletin, Institute of Meteorology and Physics of the Atmospheric Environment, Athens.
- National Oceanic and Atmospheric Administration 1993

Solar-Geophysical Data Prompt Reports I, No. 586: 24.

\_\_\_\_1994 Solar-Geophysical Data Comprehensive Reports II, No. 599: 15.

- Pearson, G. W. and Stuiver, M. 1993 High-precision bidecadal calibration of the radiocarbon time scale, 500-2500 BC. In Stuiver, M., Long, A. and Kra, R. S., eds., Calibration 1993. Radiocarbon 35(1): 25-33.
- Schlitzer, R., Roether, W., Weidmann, U., Kalt, P. and Loosli, H. H. 1985 A meridional <sup>14</sup>C and <sup>39</sup>Ar section in northeast Atlantic deep water. *Journal of Geophysical Research* 90(C4): 6945–6952.
- Schoch, H., Bruns, M., Münnich, K. O. and Münnich, M. 1980 A multi-counter system for high-precision <sup>14</sup>C measurements. *In* Stuiver, M. and Kra, R. S., eds., Proceedings of the 10th International <sup>14</sup>C Conference. *Radiocarbon* 22(2): 442–447.
- Stuiver, M. and Pearson, G. W. 1993 High-precision bidecadal calibration of the radiocarbon time scale, AD 1950–500 BC and 2500–6000 BC. In Stuiver, M., Long, A. and Kra, R. S., eds., Calibration 1993. Radiocarbon 35(1): 1–23.
- Stuiver, M. and Reimer, P. J. 1993 Extended <sup>14</sup>C data base and revised CALIB 3.0 <sup>14</sup>C age calibration program. *In Stuiver, M., Long, A. and Kra, R. S., eds., Calibra*tion 1993. *Radiocarbon* 35(1): 215-230.
- Tandon, J. N. and Bhatia, V. B. 1965 Post-storm cosmicray increases and the radiation belt. *Journal of Atmo*spheric and Terrestrial Physics 27: 635–640.
- Thambyahpillai, T. 1975 Sidereal daily variations in cosmic-ray intensity and their relationships to solar modulation and galactic anisotropies. NATO ASI Series, Series C, Mathematical and Physical Sciences 14: 37– 59.
- Theodórsson, P. 1996 Measurement of Weak Radioactivity. London, World Scientific: 333 p.
- Walker, G. O. and Wong, Y. W. 1993 Ionospheric effects observed throughout east Asia of the large magnetic storm of 13–15 March 1989. Journal of Atmospheric and Terrestrial Physics 55(5): 995–1008.
- Xiaoping, Z., Yunfang, L., Donqi, Y. and Qihua, G. 1985 The response of geomagnetic spectral composition to solar wind during storm time. *Journal of Atmospheric* and Terrestrial Physics 47(8–10): 1017–1022.