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Production of Radiocarbon by Neutron Radiation on Linen

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Abstract
Experiments were performed on modern linen to study the hypothesis that the 1988 radiocarbon dating of the Turin Shroud was in error by about 1300 years because the Shroud had received neutron radiation that produced contaminant radiocarbon by reactions with nitrogen indigenous to the linen. Measured spatial variations in linen nitrogen content caused radiocarbon age gradients as large as 261 yr-cm^{-1} for the neutron fluence needed to reduce its radiocarbon age by about 1300 years. Because of these variations and because measurements of radiocarbon and nitrogen contents were destructive, one to one comparisons could not be made between calculated and measured quantities of radiocarbon. Within this limitation, it was determined that the radiocarbon produced by neutron irradiation was not removed by high temperatures or by the chemical cleaning treatments used in the 1988 tests.

Keywords: Radiocarbon, neutron, radiation, nitrogen, linen.

1. INTRODUCTION

The 1988 tests determined that the Turin Shroud's radiocarbon date was approximately 1325 AD [1]. Radiocarbon dating is susceptible to error because of contamination, but the three laboratories used a number of standard chemical cleaning treatments to remove all usual types of contamination. Phillips [2] proposed a different type of contamination, nuclear radiation to produce unnatural radiocarbon that would artificially reduce the radiocarbon age of the Shroud. Hedges [3] stated that neutron reactions with trace nitrogen in the linen would be the principle mode for producing radiocarbon in the linen. Rinaudo [4], Moroni[5] and Barbesino [6] conducted experiments on a piece of 1st century Egyptian Lyma mummy linen that verified Hedges statement. Riani [7] conducted a robust statistical analysis of the 1988 data that rejected the null hypothesis that the ages measured by the three laboratories are homogeneous and suggest the presence of an important contamination in the 1988 Shroud samples. The effects of inhomogeneities had not been studied in the prior neutron experiments. This study supposed that neutron radiation can explain the erroneous radiocarbon date as well as the inhomogeneity and contamination of the Turin Shroud samples suggested by Riani [7],Walsh [8] and Van Haelst [9].

The three goals of these experiments were to (1) predict the concentration of radiocarbon produced using the measured nitrogen content of the linen and the measured neutron fluence,(2) measure the loss, if any, of the produced radiocarbon as a function of various heat and chemical treatments and (3) investigate inhomogeneities.

2. THEORY

Carbon has two stable isotopes, ^{12}C, which is 98.90% abundant and ^{13}C, which is 1.10% abundant. Radiocarbon, ^{14}C, is unstable and is only about 10^{-10}% abundant.^{14}C nuclei are continuously produced in the upper atmosphere by cosmic ray secondary neutron collisions with nitrogen to create ^{14}C and a proton; this reaction is denoted by $^{14}N(n,p)^{14}C$. This reaction is not constant, so the concentration of ^{14}C in the atmosphere varies over time. In 1950 a standard concentration of ^{14}C was defined as 100 percent Modern Carbon (pMC) when the ratio of the number of ^{14}C nuclei to the number of ^{12}C nuclei was equal to 10^{-12}[10].

When living plants take in carbon dioxide for their growth, they take in some ^{14}C and when the plant stops growing, this ^{14}C slowly emits electrons to decay back to ^{14}N with a half-life of 5730±40 years.

Thus, the percent Modern Carbon, *pMC(t)*, *t* years after it stops growing, is given by

$$pMC(t) = pMC_0\, e^{-\lambda t}, \qquad (1)$$

wherepMC_0 is the percent modern carbon in the plant when it stopped growing and λ is equal to1.2097×10^{-4} yr^{-1}, which corresponds to its 5730 year half-life.

Equation 1is solved below to yield the time, *t*, between the time the plant stopped growing and the time of measuring *pMC(t)*.

$$t = ln[pMC_0/ pMC(t)]/\lambda. \quad (2)$$

Since pMC_0 is not constant, t must be corrected to obtain the true time by using measured historical values of $pMC_0$from such things as tree rings.

The true age differs from the radiocarbon age by Δt if pMC_0 is incorrectly assumed to be equal to 100, but it is not. The value of Δt is given in Equation 3 below.

$$\Delta t = ln[pMC_0/100]/\lambda. \quad (3)$$

Figure 1 plots Equation 3 for different initial values of pMC_0 and shows that when pMC_0 is equal to 116.92 pMC, the radiocarbon age appears to be 1300 years younger than actual, as appeared to be the case in the 1988 radiocarbon dating of the Turin Shroud.

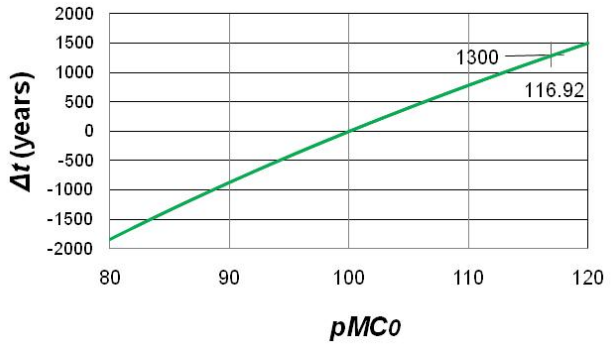

Figure 1. Radiocarbon age appears Δt years younger if the initialpercent Modern Carbon,pMC_0, is greater than 100 and no correction is made.

Because linen contains trace amounts of nitrogen, if linen receives neutron radiation, additional ^{14}C will be produced in the linen. This will increase its pMC_0 valueby an unknown amount and causes an error in the true age unless the neutron fluence and nitrogen content are known to make the proper correction using Equation 3.The increase, ΔpMC, in pMC_0caused by the $^{14}N(n,p)^{14}C$ neutron reaction is given by the following equation.

$$\Delta pMC = 10^{12} n_0 \sigma [M_C/f_C]\ [f_N/M_N] \times 100\ pMC, \quad (4)$$

Where n_0 is the neutron fluence in n·cm^{-2},
σ is the 1.81×10^{-26}m^2 cross section for thermal neutrons,
f_C is the weight fraction of ^{12}C(for the linen used, this was 0.4322, obtained from total carbon weight fraction by multiplying it by the 0.9890 natural abundance of ^{12}C),
$f_N$is the weight fraction of ^{14}N(obtained from total nitrogen weight fraction by multiplying bythe 0.9963 natural abundance of ^{14}N),
M_C is the isotopicweight of^{12}C (12.000 g) and
$M_N$is the isotopic weight of ^{14}N (14.003 g).

3. EXPERIMENTAL

Linen Used: A sheet of unbleached modern plain-woven flax linen[11], which measured 0.99 m in the warp direction by 1.56 m in the weft direction, was used in this study. It was thoroughly washed in a mild detergent and rinsed repeatedly in distilled water before using. The areal density of the linen was 25 mg·cm^{-2}, the mass density of the fibers determined by weighing in air and water was 1.45 g·cm^{-3}and the thread density was 13 threads/cm in the warp and 15 threads/cm in the weft directions. Its measured total carbon weight fraction was 0.4370±0.0002. Three pieces were taken from this sheet at two major locations for neutron irradiation at three different times, as shown in Figure 2. Two ≈0.5 g samples were taken from region 2; one was irradiated in carbon dioxide with 1.07×10^{14} n·cm^{-2} and the other in air with 1.04×10^{14}n·cm^{-2}. Approximately 12 g of Region 5 was irradiated with 1.07×10^{14}n·cm^{-2} and ≈0.5 g of this piece, denoted as Region 4, was used for radiocarbon measurements. Samples measured for radiocarbon content weighed between 7 and 20 mg.

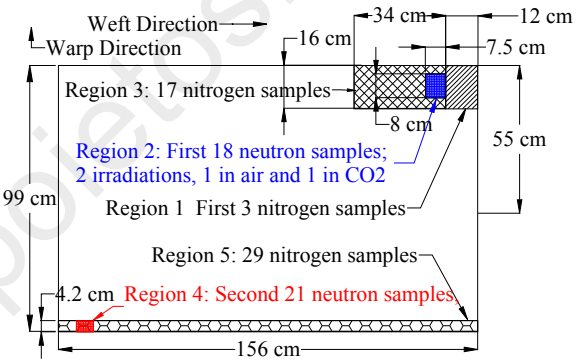

Figure 2. Unbleached modern flax linen used in experiments, showing locations of samples.

Nitrogen Concentration Measurements [12]**:**The first measurements of nitrogen weight fractions were made using three samples each weighing about 1.5 g taken from Region 1. The result was 720±32 ppm, so this value was used in calculations for the 18 neutron-irradiated samples in the adjacent Region 2. Subsequent nitrogen measurements were made on 10 mg samples to better conform to the sizes of the radiocarbon samples and so that spatial variations in nitrogen content could be determined; the accuracy of these measurements was5%.Seventeen measurements in Region 3surrounding Region 2 yielded a nitrogen content of 614±50 ppm. Region 4 contained another 21 neutron–irradiated samples; surrounding this region, 29 samples in Region 5 were measured for nitrogen content and yielded a weight fraction of 566±62 ppm, so this value was used for calculations for Region 4 samples.

Figure 3 plots the results of all the nitrogen measurements. The normalized distributions of nitrogen weight fractions for samples plotted in Figure 3 are shown in Figure 4. In the following, the measured radiocarbon that was produced by neutron irradiation was compared with the calculated amount using Equation 4. For this comparison to be valid, it is necessary to measure both the weight fraction of nitrogen, f_N, and the radiocarbon content, pMC, in the same

sample. Unfortunately, both measurements are destructive and the nitrogen content varies significantly from place to place in the linen, so precise one-to-one comparisons were not possible. Fortunately, the Region 2 samples that were studied appeared to be fairly homogeneous because the measured results were consistent with a nitrogen weight fraction of 720 ppm.

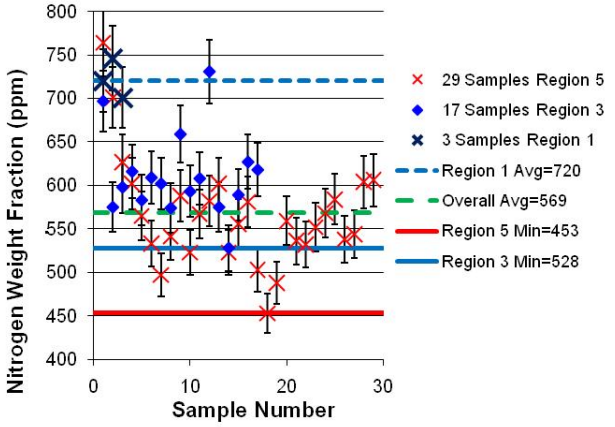

Figure 3. Measured total nitrogen contents for all samples.

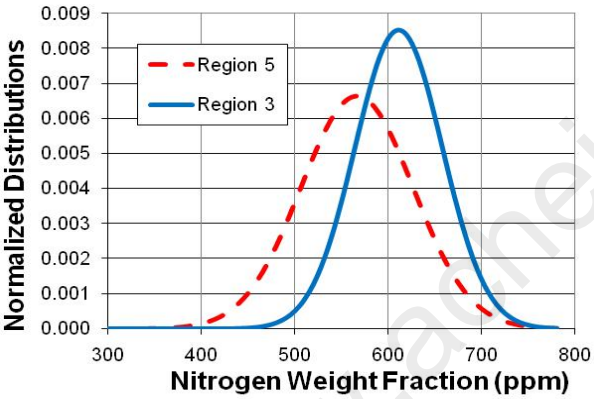

Figure 4. Normalized distributions of nitrogen weight fractions.

Radiocarbon Measurements [13, 14]**:**All radiocarbon measurements had errors of about ±0.75 pMC; none were larger than 1.1 pMC. The percent Modern Carbon of a virgin linen sample from Region 2 measured 107 pMC; this is larger than 100 pMC because of the remnants of radiocarbon produced in the atmospheric nuclear tests in the 1950's. A linen piece from Region 2 was neutron irradiated in carbon dioxide gas with a neutron fluence of 1.07×10^{14}n·cm^{-2}. Equation 4, using the average720 ppm nitrogen content, predicted an increase of 27.55±1.38 pMC which when added to the 107 pMC yields 134.55±1.38 pMC. Errors of ±5% are included because nitrogen measurements had this accuracy. The measured value was 134.89±0.76pMC. This good agreement demonstrates the validity of Equation 4.

This good agreement is in contrast to the result obtained for linen irradiated with a fluence of 1.04×10^{14}n·cm^{-2} in a2.7cm^{3}polypropylene vial containing four small1 mm diameter holes to the outside air. The calculated value was 133.77±1.34 pMC, but the measured value was 224.6±0.9 pMC. Subtracting the baseline value 107 pMC from each of these indicates that the increase in radiocarbon when irradiated in air was 117.6 instead of the predicted 26.77 pMC, a factor of 4.39 times more than predicted. Calculations described below in Effects of Chemical Pretreatments show that collisions of neutrons with nitrogen in the air that surrounds the sample create radiocarbon that diffuse into the sample to cause this increase in radiocarbon. The radiocarbon produced in the air most likely combines with oxygen in the air and diffuses into the linen as carbon dioxide, $^{14}C^{16}O_2$.

Experiments were conducted to determine ifthe radiocarbon that was produced in the surrounding air would diffuse out at room temperature. After waiting 116 days, the measured radiocarbon content had dropped only 12.2 pMC to 212.4±1.1 pMC, which was too slow to obtain an accurate rate at which the radiocarbon was diffusing from the linen. To speed the diffusion process as well as obtain temperature dependent diffusion data, elevated temperature diffusion experiments were conducted. Five samples were heated for 75 minutes at different temperatures,130, 175, 200, 225, and 245 ˚C. Figure 5 shows these samples after heating along with a non-heated sample.

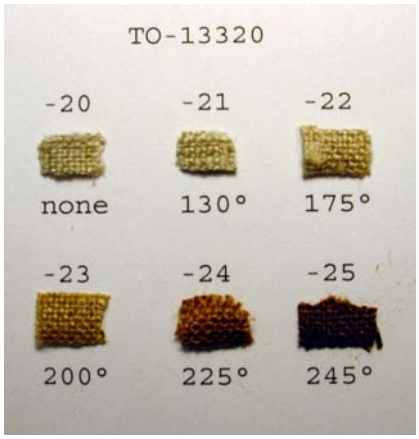

Figure 5. Appearance of six linen samples after heating for 75 minutes at indicated temperature.

The radiocarbon that remained after the heat treatments was measured and is shown in Figure 6; the extra points are for repeat measurements. The line at 133.77 pMC is the predicted value from Equation 4. If no points lie below this line, it indicates that only the radiocarbon produced from the nitrogen in the air diffused out of the sample and that the radiocarbon produced from the nitrogen within the linen did not leave the samples during heating at temperatures up to 245 ˚C for 75 minutes.

The diffusion rates for the heated samples were computed using the unheated sample as the reference. Two room

temperature diffusion rates were determined from room temperature measurements at times of 114, 294 and 410 days after irradiation. The diffusion rates are plotted in an Arrhenius format in Figure 7. It is important to note that in computing the diffusion rates, the diffusion was not assumed to be linear because it is actually exponential.

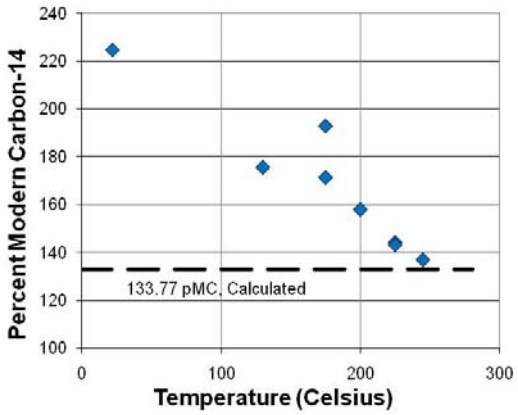

Figure 6. Measured radiocarbon remaining after 75 minute heat treatments between 130 and 245 ˚C.

Thus, more than two points are required to compute the exponential diffusion rate; the third point was the level to which the radiocarbon declined. The predicted value was 133.77±1.34 pMC; other values were also tried and the value of 133.4 pMC gave the lowest R^2 value in the Arrhenius plot. This further verifies the validity of Equation 4. It also verifies that only the radiocarbon produced from the nitrogen in the air diffused out of the sample and that the radiocarbon produced from the nitrogen in the linen remained in the sample at temperatures up to 245 ˚C for 75 minutes.

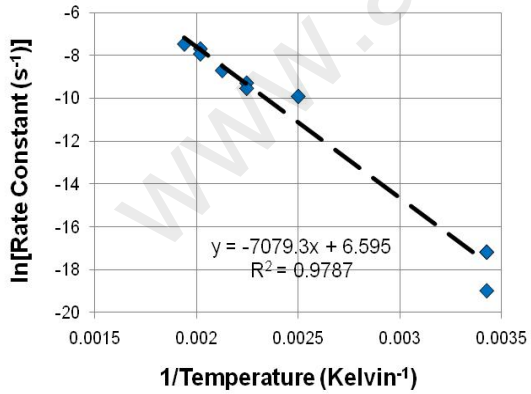

Figure 7. Least squares fit to Arrhenius plot of diffusion rates.

The Arrhenius plot provides the rate constants k(T) for the diffusion process for all temperatures, T, that lie on the straight line fit. The result is given in Equation 5.

$$k(T) = e^{6.595} \times e^{-7079.3/T} = 731.4\, e^{-7079.3/T} \qquad (5)$$

From Equation 5, the rate constant at room temperature (293 K) for diffusion of the radiocarbon produced from the nitrogen in the air is 2.34×10^{-8} sec^{-1}, or 0.74 yr^{-1}. Thus, after 6 years the fraction of this radiocarbon remaining in the linen will be $e^{-0.74\times6}$= 0.012, or 1.2%. This is very slow for diffusion of gases in polymers. The quantity 7079.3 in the exponent of Equation 5 corresponds to an activation energy of 58.9 kJ/mole, which is not an uncommon value for diffusion of carbon dioxide or other gases through amorphous polymers. However, the pre-exponential factor of 731.4 is very small for diffusion in fibrous polymers. The reason is probably because flax fibers are highly crystalline and diffusion is virtually zero through crystalline material. Thus, the diffusion most likely occurs along the labyrinth of amorphous material between the crystals that hold them together. This torturous path greatly slows the diffusion and could explain the very small pre-exponential factor.

TABLE 1.Description of Chemical Treatments Used

Type of Pretreatment	Chemical Pretreatment
Arizona A	0.5% HCl for 2 hours at room temperature (RT) 0.25% NaOH for 2 hours at RT Dilute acid wash
Arizona B	Commercial detergent (1.5% SDS) for 1 hour at RT Wash with distilled water 0.1% HCl for 1 hour at RT Commercial detergent (1.5% Triton X-100) for 1 hour at RT Soxhlet extraction with ethanol for 1 hour Wash with water for 1 hour at 70°
Oxford A	Petroleum ether for 1 hour at 40° 1M HCl for 2 hours at 80° 1M NaOH for 2 hours at 80° Dilute acid wash
Oxford B	Oxford A followed by 2.5% NaOCl (pH=3) for 30 min at RT
Zurich A	Ultrasonic wash in distilled water for 1 hour at RT
Zurich B	Zurich A followed by 0.5% HCl for2 hours at RT 0.25% NaOH for 2 hours at RT Dilute acid wash
Zurich C	Zurich A followed by 5% HCl for2 hours at 80° 2.5 % NaOH for 2 hours at RT Dilute acid wash

Effects of Chemical Pretreatments: Neutron-irradiated samples from both Regions 2 and 4 were subjected to chemical pretreatments like those used in the 1988 radiocarbon dating of the Turin Shroud. Table 1 describes the chemical treatments that were used to duplicate those used by Oxford, Zurich and Arizona. Figure 8 shows the results of the chemical treatments for samples in Regions 2

and 4. Not all pretreatments were performed on Region 2 samples because they were all consumed in the thermal treatments used to perform the Arrhenius analysis.

The consistently larger amount of radiocarbon in Region 2 samples than in the Region 4 samples requires explanation. It is a result of three things. (1) The Region 2 samples had a nitrogen content of about 720 ppm and Region 4 samples had about 566 ppm. (2) The Region 2 samples had more air surrounding them during radiation than did Region 4 samples and since radiocarbon is produced in the air during irradiation, more radiocarbon diffused into the Region 2 sample. (3) A longer time elapsed between irradiation and radiocarbon measurement for the Region 4 sample, allowing a greater amount of radiocarbon from the air to diffuse out again.

In the following these three things are incorporated into a simple correction which assumes all the radiocarbon produced in the air enters the linen. Since the nitrogen in the air and the nitrogen in the linen sample both receive the same neutron fluence, the radiocarbon produced in the air is equal to the radiocarbon produced in the linen times the ratio of the mass of nitrogen in the air surrounding the linen and the mass of nitrogen in the linen. This correction is simple, but it involves many steps. The correction is best described using Table 2because the table lists the Items used in the steps and also presents intermediate results that can be used for comparing the differences in the experiments conducted on Region 2 and Region 4 samples. Table 2 is for samples that received no chemical or thermal treatments because these treatments remove radiocarbon produced in the air, reducing the differences.

The Items 1 through 6 in Table 2 require no explanation. The discrepancies between calculated and measured values are shown as Item 7. The calculated radiocarbon for Region 2 is 90.83pMC smaller than measured, but the calculated radiocarbon for Region 4 is only 5.94 pMC smaller than measured. Items 8 through 16 are needed to compute the relative amounts of nitrogen in the air and the nitrogen in the sample. Looking at Item 16, the Region 2 sample was surrounded by 5.83 times more nitrogen than was contained in the sample, while this ratio is only 1.44 for the Region 4 sample. Thus, Item 17 shows the amount of radiocarbon produced in the air is much larger for Region 2 than it is for Region 4.

TABLE 2. Corrections to Account for Additional Radiocarbon Produced in Air Surrounding Sample

Item	Description	Unit	Symbol	Region 2	Region 4	Comment
1	Nitrogen content	ppm	f_N	720	566	Average in adjacent regions
2	Neutron fluence	$n \cdot cm^{-2}$	n_0	1.04×10^{14}	1.07×10^{14}	Measured
3	Radiocarbon increase	pMC	ΔpMC	26.77	21.66	Eq 4, using Items 1 and 2
4	Baseline radiocarbon	pMC	pMC_0	107	107	Measured
5	Calcuated total radiocarbon	pMC	pMC_c	133.77	128.66	Item 3 + Item 4
6	Measured radiocarbon	pMC	pMC_m	224.6	134.6	Measured
7	Uncorrected discrepancy	pMC	D_0	90.83	5.94	Item 6 - Item 5
8	Sample mass	g	m_s	0.51	12	Measured
9	Fiber mass density	$g \cdot cm^{-3}$	ρ_f	1.45	1.45	Measured
10	Fiber volume	cm^3	V_f	0.35	8.3	Item 8 ÷ Item 9
11	Container volume	cm^3	V_c	2.7	19	Measured
12	Air volume	cm^3	V_a	2.35	10.7	Item 11 - Item 10
13	Nitrogen content of air at STP	$g \cdot cm^{-3}$	ρ_{N_air}	9.11×10^{-4}	9.11×10^{-4}	Calculated from published data
14	Mass of nitrogen in air	mg	m_{N_air}	2.14	9.8	Item 12 × Item 13
15	Mass of nitrogen in sample	mg	m_{N_sample}	0.367	6.8	Item 1 × Item 8
16	Ratio of nitrogen masses (air/sample)		R	5.83	1.44	Item 14 ÷ Item 15
17	Radiocarbon produced in air	pMC	ΔpMC_{air}	156.01	31.09	Item 3 × Item 16
18	1st corrected radiocarbon	pMC	pMC_{cor1}	289.78	159.75	Item 5 + Item 17
19	1st corrected discrepancy	pMC	D_1	-65.18	-25.15	Item 6 - Item 18
20	Time from radiation to measurement	day	δt	114	395	Measured
21	Remaining radiocarbon from air	pMC	$\Delta pMC_{air}(\delta t)$	123.82	13.96	Eq 5, using Items 17 and 20
22	2nd corrected radiocarbon	pMC	pMC_{cor2}	257.59	142.62	Item 5 + Item 21
23	2nd corrected discrepancy	pMC	D_2	-32.99	-8.02	Item 6 - Item 22

Adding Item 17 to the original calculated radiocarbon, Item 5, produces the 1st corrected result that is now significantly larger than measured, as seen in Items 18 and 19.

Item 20 shows the large time delays between neutron irradiation and radiocarbon measurement. During this time delay, some of the radiocarbon produced in the air that entered the sample slowly diffused out again. Item 21 was obtained using Equation 5 to determine how much radiocarbon, Item 17, would remain after the time delay, Item 20. Adding this amount to the original calculated radiocarbon, Item 5, produces Items 21 and 22 that are significantly improved over Items 18 and 19, but still larger than measured.

A possible reason for the 2nd corrected calculated radiocarbon to be larger than the measured radiocarbon is that not all the radiocarbon produced in the air entered the sample as was originally assumed in this analysis. Neglected in this assumption is the possible loss of radiocarbon out of the containers used during irradiation. The Region 2 sample was in a polypropylene container containing four small 1 mm diameter holes open to the outside the air, which would allow the escape of radiocarbon, perhaps accounting for the -32.99 pMC discrepancy. The Region 2 sample, which has a discrepancy of only -8.02 pMC was irradiated in a tightly wrapped plastic film that was placed inside another container with the remaining volume filled with plastic foam, so it was less likely to lose as much radiocarbon through the container walls as was the Region 2 sample.

It must be pointed out that the nitrogen contents (Item 1) in Table 2 are not the nitrogen contents of the actual samples that were used, but are the averages of the nitrogen contents of samples in adjacent regions. Thus, exact agreements between calculated and measured values are not expected and good agreements may be fortuitous. Nevertheless, the corrections presented in Table 2explain why Region 2 samples had larger radiocarbon contents than Region 4 samples.

The lower bounds marked in Figure 8 are explained as follows. The smallest nitrogen concentration measured in Region 3, which surrounds Region 2, was 528 ppm. From this and the neutron fluence of that this linen received, Equation 4predicts that the pMC for Region 2 will not fall below 126.6±0.98pMC. If it falls below, then some radiocarbon that was produced within the linen is removed by the chemical treatment. Likewise, the smallest nitrogen concentration measured in Region 5, surrounding Region 4 and which received 1.07×10^{14}n·cm^{-2}, was 453 ppm; this results in a lower bound of 124.3±0.87 pMC for Region 4. Only one sample falls below its lower bound and that is the Region 4 sample that received the Zurich C treatment, which actually was not as harsh as the Oxford B treatment.

Effects of a Thermal Treatment followed by Chemical Pretreatments: Using some samples from the irradiated linen that was used in the chemical pretreatment experiments, similar experiments were conducted, but the samples were heated for 75 minutes at 175 ˚C prior to the chemical treatments. The temperature of 175 ˚C was chosen because Figure 4 shows that this heat treatment produces a small but noticeable color change, more than any color change that was observed on the samples used in the 1988 tests.

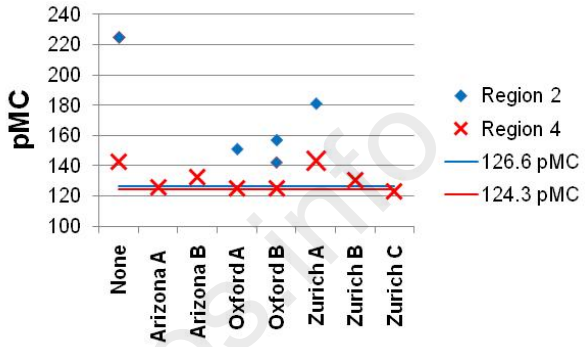

Figure 8. Percent Modern Carbon remaining in samples from Regions 2 and 4 after chemical treatments.

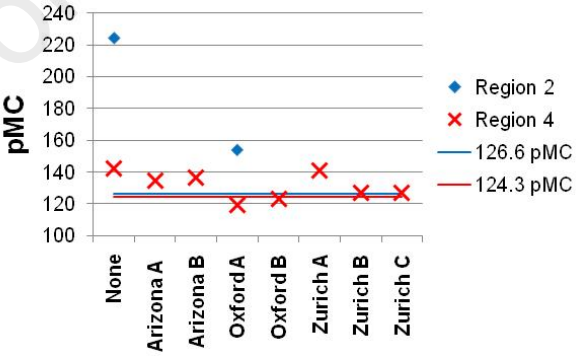

Figure 9. Percent Modern Carbon remaining in samples from Regions 2 and 4 after both thermal and chemical treatments.

Figure 9 shows the results of these experiments. Region 4 samples Oxford A and B fall slightly below the lower bound for this region, but this is attributed to a large variation in nitrogen content. As an example of the inconsistencies caused by variations in nitrogen contents, notice that Oxford A is lower than Oxford B even though Oxford B is a much harsher treatment. Comparing Figures 8 and 9, one might expect that samples both heat and chemically treated would have lower radiocarbon contents than those only chemically treated, but that is not true for all chemical treatments. This inconsistency is most likely caused by the variations in nitrogen content that occur throughout the linen. Within this error, it appears that no radiocarbon created within the flax fibers was removed by heat and chemical treatments.

Gradients of Nitrogen Inhomogeneity: While measurements of nitrogen concentrations at random

locations in the linen confirmed that the nitrogen content is inhomogeneously distributed, it did not provide information on the spatial gradients of nitrogen variation and how this might affect the radiocarbon dating of closely-spaced samples. Therefore, closely-spaced samples were cut from 1 cm by 5 cm linen pieces obtained from Region 3 that surrounds Region 2, and Region 5 that surrounds Region 4, and their nitrogen contents were measured. Figure 10 shows how the samples were cut. The perimeters of the 1 cm×1 cm squares, which weighed 10 mg, were measured for nitrogen content. The centers, which weighed 15 mg, were retained if additional measurements were required.

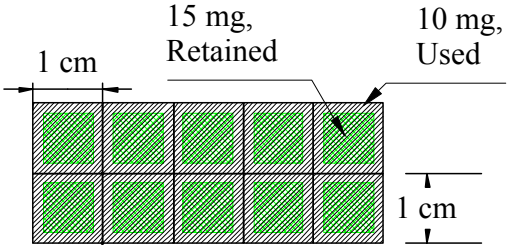

Figure 10. Squares of linen were cut for measuring gradients of nitrogen concentrations in Regions 3 and 5.

The results are plotted in Figures 11 through 14. Figure 11 shows the nitrogen weight fractions for samples in region 3 and Figure 12 shows how these samples would radiocarbon date in 1988 if irradiated in 33 AD with a neutron fluence of 8.3×10^{13}n·cm^{-2}. Equations 3 and 4 were used to determine this neutron fluence to make the linen appear 1300 years younger if the nitrogen weight fraction was the average of all samples measured in this linen, namely, 569 ppm. The time period of 1300 years corresponds to the suspected age error in the 1988 radiocarbon dating of the Turin Shroud. Figures 13 and 14 are like Figures 11 and 12 except they are for samples taken from Region 5.

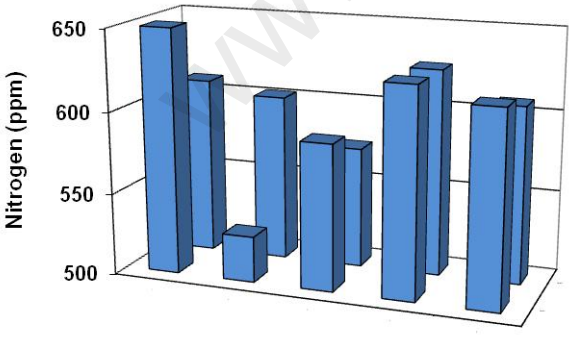

Figure 11. Measured nitrogen weight fractions in linen samples from Region 3. The bar locations correspond to the 1 cm spacing of the samples shown in Figure 10.

Figures 11 and 12 for Region 3 show nitrogen concentrations ranging between 528 and 653 ppm and the corresponding radiocarbon dates ranging between 1241 and 1502 AD instead of 33 AD. The gradients are as large as 125 ppm/cm and 261 years/cm.

Figures 13 and 14 for Region 5 show nitrogen concentrations ranging between 532 and 606 ppm and the corresponding radiocarbon dates ranging between 1241 and 1405 AD. The gradients are as large as 60 ppm/cm and 126 years/cm.

These gradients in radiocarbon dates calculated from the measured nitrogen concentrations in Regions 3 and 5 are comparable with the gradients of about 41 yr/cm observed for the 1988 radiocarbon results.

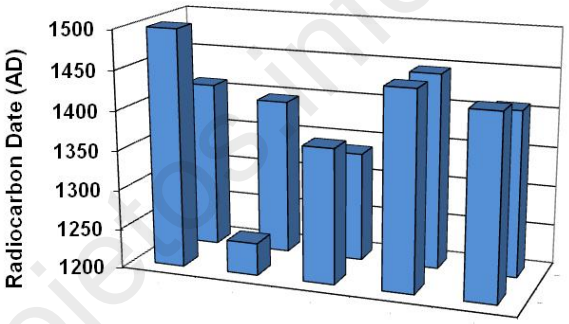

Figure 12. Calculated radiocarbon dates of the samples from Region 3 shown in Figure 11; see text.

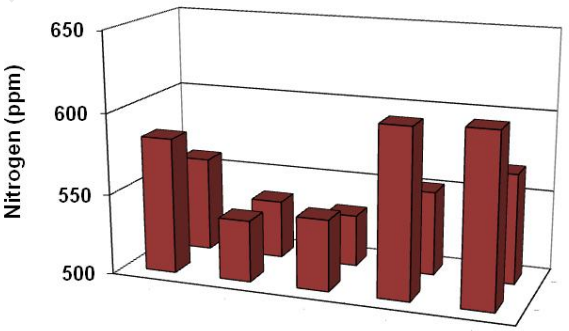

Figure 13. Measured nitrogen weight fractions in linen samples from Region 5. The bar locations correspond to the 1 cm spacing of the samples shown in Figure 10.

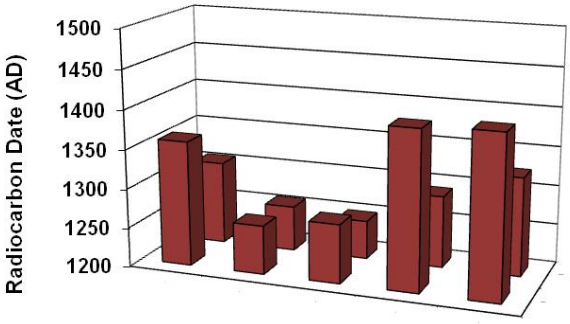

Figure 14. Calculated radiocarbon dates of the samples from Region 5 shown in Figure 13; see text.

4. CONCLUSION

Neutron irradiation of flax linen increases the radiocarbon content by two distinct modes. The first mode is by nuclear reactions with nitrogen indigenous to the flax. Within the uncertainties caused by variations in nitrogen content of the linen, the amount of radiocarbon produced by this mode can be calculated from the neutron fluence and the nitrogen content of the linen. Within these uncertainties, this radiocarbon was not removed by high temperatures or by the chemical cleaning treatments used in the 1988 tests. Thus, this radiocarbon acts as a contamination that is indistinguishable from the natural radiocarbon in the linen and it will act to reduce the radiocarbon age of the linen.

The second mode of generating radiocarbon is by nuclear reactions with the nitrogen in the air that surrounds the linen and then enters the linen. These experiments have shown that the amount of radiocarbon produced by this mode and which enters the linen can be estimated if the linen is of known volume, is in a closed container of known volume, and it remains there for a few days. Experiments were not conducted to determine how much radiocarbon produced by this mode enters the linen if the linen is not in a closed container or if it is removed from the container immediately after irradiation. The most important finding of this research concerning radiocarbon produced in the air surrounding the linen is that it is not permanently bound to the linen and 99% of it diffuses out in about six years at room temperature. Furthermore, virtually all of this radiocarbon can be removed in minutes by exposure to elevated temperatures or by the chemical cleaning treatments used in the 1988 tests. Thus, if the Turin Shroud received neutron irradiation, its carbon-14 content would increase, causing its radiocarbon age to be younger despite experiencing thermal heating greater than it received in the 1532 Chambery fire and by the pretreatment cleaning procedures employed in the 1988 tests. In addition, significant spatial variations of nitrogen were measured in the modern linen used in these experiments. Calculations showed that these variations would cause radiocarbon age gradients as large as 261 yr/cm if it received the neutron fluence needed to reduce its age by 1300 years. If the Turin Shroud had received neutron irradiation and the Shroud's natural variations of nitrogen content were similar to these found in the linen used in this study, these radiocarbon age gradients could explain Riani's [7] conclusions that the Shroud's radiocarbon dates were inhomogeneous.

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13. Neutron irradiations were performed at the University of Missouri Research Reactor in Columbia MO.

14. Radiocarbon measurements, sample heat treatments and chemical treatments were performed at IsoTrace Radiocarbon Laboratory, Accelerator Mass Spectrometry Facility at the University of Toronto.