Improving the control of systematic uncertainties in precision measurements of radionucleide half-life

S.Towers[a]

[a] Arizona State University, Tempe, AZ, USA; Purdue University, West Lafayette, IN, USA

Abstract

Many experiments designed to precisely determine the half-life of a radionucleide employ a long lived reference source to help determine the impact on the data of any systematic variation in the detector and associated electronics. The half-life of the radionucleide of interest is determined from the ratio of its decay rate data to the decay rate data from the reference source.

This correction procedure assumes that any underlying systematic affects the data and reference measurements in exactly the same way. In this paper we show that when some systematic effects affect the two differently, the ratio procedure can leave artifacts in the corrected data that can compromise an unbiased and precise assessment of the radionucleide half-life. We describe two methods that can help overcome this problem. We also describe several statistical tests that help determine which effects may underlie systematic variations in the data.

We discuss an illustrative example based on previously published ^{32}Si and ^{36}Cl data recorded by an experiment at Brookhaven National Laboratory. We correct the data for systematic variation related to climate variation and estimate the ^{32}Si half-life to be $T_{1/2} = 171.8 \pm 1.8$. The reduction in uncertainty in the ^{32}Si half-life, relative to the previous estimate based upon this data, is equivalent to that which would be achieved through increasing the size of the data set by almost 3.5 times.

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1. Introduction

Precision measurements of radionucleide half-lives are crucial to many different endeavours including, but not limited to, medical physics, dating of artifacts, cosmology, and our understanding of fundamental physics.

Precise and unbiased determination of the half-life of a radionucleide requires careful control of systematic effects both in the design of the detector apparatus and in the experimental method. One common method to control for systematic effects is to compare the decay rate observations of a radionucleide of interest to concurrent decay rate observations of a second "reference" radionucleide with a

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Email address: `smtowers@asu.edu` (S.Towers)

half-life that is precisely known from previous experiments (see, for instance, [1–7]). By "concurrent", we mean here either truly concurrent, or approximately concurrent within a short time scale relative to the time scale of systematic variations in the data.

To see how this works in practice, let us refer to the number of decays per unit time of the source of interest and the reference source as A and B, respectively:

$$\begin{aligned} A &= \lambda_A N_0^A e^{-\lambda_A t} \\ B &= \lambda_B N_0^B e^{-\lambda_B t}, \end{aligned} \tag{1}$$

where λ_A and λ_B are the decay rates of the two sources (and λ_B is assumed to be known with precision).

In the absence of systematic effects, we would obviously not need a reference source, and could use Equation 1 directly to fit the data for the source of interest to determine λ_A. In the presence of systematic effects common to both sources, f, we can fit the time series of A/B data for λ_A:

$$\begin{aligned} \frac{A}{B} &= \frac{\lambda_A N_0^A e^{-\lambda_A t}(1+f)}{\lambda_B N_0^A e^{-\lambda_B t}(1+f)} \\ &= \frac{\lambda_A N_0^A e^{-(\lambda_A-\lambda_B)t}}{\lambda_B N_0^B}. \end{aligned} \tag{2}$$

However, we describe in the following sections how use of just one reference source may not be sufficient to cancel systematic effects that differ somewhat between sources.[1] This leads to extra uncertainty and even potential bias in the estimate of λ_A. We discuss two methods that can be used to control for systematics that differ between sources. We also describe several statistical tests that can provide a powerful means to help determine if a suspected source of systematic variation truly is consistent with having a causative effect on the variations in the data.

We apply these methods in a case study of previously published ^{32}Si and ^{36}Cl data recorded by an experiment at Brookhaven National Laboratory.

2. Control of systematic effects

As just discussed, a reference source can be used to correct decay rate data for systematic effects that are common to both the reference and the radionucleide of interest. However, things become more complicated if there are systematic effects that are not common to both sources. In this case, we have

$$\frac{A}{B} = \frac{\lambda_A N_0^A e^{-\lambda_A t}(1+f+g_A)}{\lambda_B N_0^B e^{-\lambda_B t}(1+f+g_B)}, \tag{3}$$

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[1] An example of such an effect would perhaps be differing attenuation of the decay products of the sources in the air or detector material due to the differing energy spectra of the decay products. The time variations of such effects could conceivably be related to temporal variations in the environmental conditions in the vicinity of the detector.

where f is the common systematic effect, and g_A and g_B are the systematic effects that differ between sources. If we assume that f, g_A, and g_B are all much less than 1, we obtain

$$\frac{A}{B} \sim \frac{\lambda_A N_0^A e^{-\lambda_A t}(1+f+g_A)(1-f-g_B)}{\lambda_B N_0^B e^{-\lambda_B t}}, \tag{4}$$

which yields, when we expand and assume that second order terms can be neglected,

$$\frac{A}{B} \sim \frac{\lambda_A N_0^A e^{-\lambda_A t}(1+g_A-g_B)}{\lambda_B N_0^B e^{-\lambda_B t}}. \tag{5}$$

We now see that if $g_A \neq g_B$, the ratio A/B will have extra variation due to systematic effects that have not been cancelled out. This extra variation, if it remains uncorrected, will not only inflate the uncertainty on the estimate of λ_A, but also potentially bias the estimate of λ_A if $\Delta g = g_A - g_B$ has trend in time.

2.1. Parameterization when cause of variation is unknown

When significant differing variation is observed between the decay rate observations of an interest and reference source, but the cause of the variation is unknown, the use of two reference sources can be employed in a subsequent experiment to control for the variation.

We begin with the assumption that the sources of systematic variation g_A and g_B have the same underlying functional form, except scaled by a constant such that $g_B = b g_A$ (here we assume that $b \neq 0$). We obtain

$$\frac{A}{B} = \frac{\lambda_A N_0^A e^{-\lambda_A t}\left(1+g_A(1-b)\right)}{\lambda_B N_0^B e^{-\lambda_B t}}. \tag{6}$$

A second reference source with well-known decay rate λ_C can help us to parameterize the temporal variation of g_A, via

$$\frac{C}{B} \sim \frac{\lambda_C N_0^C e^{-\lambda_C t}(1+c g_A - b g_A)}{\lambda_B N_0^B e^{-\lambda_B t}}. \tag{7}$$

Solving for g_A yields

$$g_A = \frac{1 - \frac{C}{B}\frac{\lambda_B N_0^B e^{-\lambda_B t}}{\lambda_C N_0^C e^{-\lambda_C t}}}{b-c}. \tag{8}$$

This now becomes a parameterization of the functional form of g_A, down to two unknown constants, N_0^B/N_0^C and $c-b$.

Substituting this formulation for g_A into Equation 6 now allows the use of decay rate data from the two reference sources, B and C, to control for systematic effects. The decay rate, λ_A, can be obtained from a fit to Equation 6 that also estimates the two extra constants N_0^B/N_0^C and $d = (1-b)/(b-c)$.

2.2. Parameterization when the cause of variation is known or suspected

If some variable is suspected to systematically affect the decay rate observations from the two sources differently, and we have measurements of that variable concurrent to the decay rate data, it is a simple matter to parameterize $\Delta g = g_A - g_B$ in terms of that variable and include the parameterization in Equation 5. Potential causes of variation could include environmental variables, like temperature or humidity [2, 17].[2]

This is of course similar to any data correction procedure, but here we describe how to test a suspected cause of systematic variation to determine if it is truly consistent with potentially being causative. However, before we move on, it should be noted that even if a variable suspected to have causative effect passes the statistical tests outlined here, causation still has not been proven. However, if the statistical tests are *not* passed, the variable can be ruled out as having significant causative effect on variations in the data.

For instance, let's assume that Δg is suspected to have an approximately linear relationship with the air pressure, P, and we have a time series of measurements of P concurrent with the decay rate data measurements (ie; we hypothesize $\Delta g = q + sP$, where q and s are parameters to be determined). With this parameterization we would thus regress the data on the RHS of Equation 5 to estimate λ_A, N_0^A/N_0^B, and q and s to determine if s is significantly different from zero. A useful cross check of true dependence of Δg on P is to then take the first time derivative of Equation 5 and fit it to the first derivative of the time series; the fit should return a value of s that is statistically consistent with the first estimate.[3] This first derivative method is also useful as a low frequency noise filter when performing spectral analyses of the time series, and has long been used for this purpose[12, 13].

Beyond linear regression analyses, there are statistical methods, such as correlation analysis, that can instead be used to determine whether or not a variable is consistent with potentially having a causative effect on the data. It has to be kept in mind that correlation between two quantities X and Y is necessary to prove causation, but correlation itself does not prove causation if the existence of a third and countervailing causative variable has not been ruled

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[2] In recent times environmental data are readily available online for any urban center in the world close to an airport[9, 10]. Even though the data are for exterior climate conditions, it should be noted that air pressure is the same indoors and out, unless a building is pressure controlled. Specific humidity (the mass of water contained in a kg of dry air, and a function of air pressure, temperature, and relative humidity) is also the same indoors and out unless the building is humidified or de-humidified (which can occur if an air-conditioning unit operates in the summer). In addition, the average indoor air temperature is correlated to outdoor air temperature due to temperature threshold behavior in the duty cycle of the building HVAC thermostats [11].

[3] Note that if the time between subsequent measurements of A, A_{i-1} and A_i, is relatively small, the first derivative of the time series is approximated by

$$A_i' = (A_i - A_{i-1})/(t_i - t_{i-1}). \tag{9}$$

out. One way to examine the possible countervailing effect of third variable, Z, is to calculate what are known as *partial correlation coefficients.* The partial correlation coefficient of X and Y, controlling for Z is

$$r_{XY|Z} = \frac{r_{XY} - r_{XZ}r_{YZ}}{\sqrt{1-r_{XZ}^2}\sqrt{1-r_{YZ}^2}}, \tag{10}$$

where r_{XY}, r_{XZ}, and r_{YZ} are the correlations between X and Y, X and Z, and Y and Z, respectively. If $r_{XY|Z}$ is consistent with zero then X and Y are consistent with being independent [14]. Partial correlation analyses are equivalent to regression analyses where the variable X is regressed on Y and Z. We include a discussion of partial correlation methods here because of their applicability to analyses that use correlation methods alone to conclude evidence of causality (e.g. reference [19]).

Time series with seasonal variation can present particular analysis challenges because they can appear to be significantly correlated even when the underlying cause of seasonality in the two series is completely unrelated. For example, assume we have two unrelated time series

$$\begin{aligned} X &= \mu_X(1+\epsilon_X\cos 2\pi\omega(t-\phi_X)), \qquad \text{and} \\ Y &= \mu_Y(1+\epsilon_Y\cos 2\pi\omega(t-\phi_Y)) \end{aligned} \tag{11}$$

with same period, $1/\omega$, but different phases ϕ_X and ϕ_Y, and different relative amplitudes of annual variation, ϵ_X and ϵ_Y. The expected correlation between the two time series is [20]

$$r = \cos 2\pi\omega(\phi_X - \phi_Y), \tag{12}$$

which is maximally positive when the series are in phase, maximally negative when they are π out of phase, and zero when they are $\pm\pi/2$ out of phase. Thus, if the two time series are close to being in phase, if researchers assume that the null hypothesis is that $r = 0$, an erroneous conclusion might be drawn that significant correlation due to potential causality. While we use the example of first order harmonics here, note that any periodic time series can be expressed as an infinite sum of sines and cosines; the first order harmonic is the first order approximation to a periodic time series.

Because of the risk of assuming that two correlated but unrelated seasonal time series is evidence of a cause/effect relationship, it is a worthwhile cross-check to examine what are known as “seasonal differences” of the times series, where for each data point we take the difference between the data for that day (or weekly or monthly average) and the data taken one year prior (note that this can only be done if the time series spans a period of several years). If data has annual periodicity, examination of potential systematic variations that have a time scale much less than one year can be facilitated by first removing the variation in the data due to the annual periodicity via the seasonal differencing method. Seasonal differencing methods have long been used for this purpose in time series analyses in various fields, including biology and econometrics[15, 16].

In the case of radionucleide decay data, if a seasonal variable like temperature is suspected to cause variations in the data, it should be true that a an anomalously hot or cold day at a season of the year (when compared to the temperature typically observed at that location at that season) is associated with anomalies in the decay rates (when compared to the decay rates typically observed in that season). The seasonal differencing method cancels the effect of any annual harmonic modulation, leaving just the day-to-day variations that allow us to determine whether or not temperature anomalies really do appear to be correlated to variations in the data.

In the next section we will discuss the application of regression, first derivative, partial correlation, and seasonal differencing analyses as a means to disentangle various potential underlying causes of variation in radionucleide decay data recorded at Brookhaven National Laboratory.

3. Case study: Brookhaven ^{32}Si and ^{36}Cl data

Beginning in 1982 and spanning a period of over four years, an experiment at Brookhaven National Laboratory (BNL) collected decay data from a ^{32}Si source, and concurrently collected decay data from a ^{36}Cl reference source [2]. Based upon this data, the analysis estimated the ^{32}Si half-life to be $T_{1/2} = 172(4)$ years. The BNL data, normalized by decay rate, are shown in Figure 1. As seen in the Figure, the normalized decay rates are highly correlated, indicating significant common systematic variation.

As also seen in Figure 1, the ratio of the normalized decay rates shows significant annual variation, indicating that there is some source (perhaps more than one) of systematic variation that differs between the two radionucleides. Semkow (2009) hypothesizes that environmental variables like temperature may be the cause [17], and indeed the Brookhaven experimenters themselves noted dependence of their data on temperature [2]. In contrast, Jenkins *et al* ([18, 19]) posit that environmental variables cannot account for a significant fraction of the observed variation, and the variation is instead primarily due to novel physics associated with interaction of the decaying nuclei with particles or fields from the Sun [18–20]. Under this latter scenario, the variation in the decay rates would be proportional to $1/R^2$, where R is the Earth-Sun distance. Thus under the hypothesis of Jenkins *et al*, the decay rates would vary harmonically with a period of one year, with an annual maximum around January 4th of each year. The hypothesis of Jenkins *et al* is that the variation related to novel physics affects ^{32}Si decays differently than those of ^{36}Cl and thus only becomes apparent after systematic variations common to both sources have been accounted for.

In the analysis of the Brookhaven data we give full consideration to both the hypothesis of Jenkins *et al* and that of Semkow. We employ the statistical tests described in the previous section to determine which effects do not appear to be consistent with being related to the variations in the data. In order to assess any potential dependence of the data on climate, online daily climate data are obtained for the Brookhaven locale over the period of experiment [9, 10].[4]

We first employ a partial correlation analysis to help distinguish between variables that may underly variation in the data, and those that appear correlated to the data for spurious reasons not related to cause and effect. The correlations between the normalized ^{32}Si/^{36}Cl data and temperature, air pressure, and $1/R^2$ are -0.62, -0.16, and 0.53, respectively, all of which are significant to $p < 0.01$. The partial correlations between the data and temperature and pressure, controlling for $1/R^2$, are both significant to $p < 0.01$. In contrast, the partial correlation of the data and $1/R^2$, controlling for temperature and air pressure, is 0.09, which is not significant ($p = 0.17$). We thus find the variations data are not consistent with being related to variations in $1/R^2$.

To examine the use of linear regression methods for this same purpose, we begin by linearly regressing the normalized ^{32}Si/^{36}Cl data on $1/R^2$, and the daily average temperature, specific humidity, relative humidity, and air pressure. Only temperature and air pressure yield regression coefficients significantly different from zero (both with $p < 0.001$), and account for 60% of the variation in the data (temperature alone accounts for over 55% of the variation, but air pressure does have a slight but significant contribution to the fit). The phase of the data is 38 ± 5 days after January 1$^{\mathrm{st}}$, and the phase of the temperature and air pressure regression is 38 ± 2 days. We note the phase of the data is statistically inconsistent with the phase of $1/R^2$. In addition, the regression coefficient associated with $1/R^2$ is not significant when included in the regression fit that also includes temperature and air pressure ($p = 0.10$). In Figure 1 we show the regression with $1/R^2$, and the regression with temperature and air pressure, overlaid on the ^{32}Si/^{36}Cl data.

To further check the potential relationship of climate variations to the variations in the normalized data, we regress the first derivatives of the time series. We again find that only temperature and air pressure yield regression coefficients significantly different from zero, and the regression coefficients are statistically consistent with those obtained from the first fit. The first derivative of the data regressed on the first derivative of $1/R^2$ does not yield a significant regression coefficient, indicating that $1/R^2$ is inconsistent with having a causative effect on the variations in the data.

We perform one further cross-check to confirm that the variations in the Brookhaven data are indeed consistent with potentially being related to climate by taking the seasonal differences of both the normalized data and the climate time series (thus cancelling out any possible dependence on $1/R^2$ in the seasonally differenced data). Regressing the seasonal differences of the data on the seasonal differences of the climate variables again reveals that only temperature and air pressure play a significant role, accounting for 50% of the variation in the seasonally differenced data. The regression coefficients from the fit to the seasonally differenced data are statistically consistent with those obtained from

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[4] We point out here that these climate data, while readily accessible online now, were not easily accessible to the researchers who originally conducted the Brookhaven experiment.

the regression fit to the original time series.

Finally, we fit an exponential curve, with corrections for temperature and air pressure variations, to the Brookhaven ^{32}Si/^{36}Cl data, as seen in Figure 1. The fit of an exponential without correction for systematic climate-related variation yields an estimate for the ^{32}Si half-life of $T_{1/2} = 171.6 \pm 3.3$, in agreement with the estimate in Reference [2]. In contrast, correction for systematic climate-related variation yields an estimated half-life $T_{1/2} = 171.8 \pm 1.8$, with uncertainty 1.87 times smaller than the half-life estimate obtained without correction for the systematic variation (an uncertainty reduction equivalent to increasing the data set by a factor of almost 3.5).

4. Discussion and Summary

The unbiased and precise determination of radionucleide half-lives requires good understanding of systematic variations. We have shown that when data from one reference radionucleide are used to control for systematic effects, residual variations can still remain in the data if there are sources of systematic variation that are not common to both radionucleides. This residual variation inflates the uncertainty on the radionucleide half-life, and can also bias the half-life estimate if the residual variations have temporal trend. We have shown how a second reference source can be used to correct for this variation if the cause of the variation is unknown.

If the cause of systematic variation is known or suspected, and a time series of measurements of that causative variable have been made coincident with the data, we have shown that parameterization of the systematic variation in terms of the causative variable can lead to significant reduction in the uncertainty of the estimate of the radionucleide half-life. We have discussed statistical methods that can be used to assess whether or not a variable is consistent with potentially being truly causative of systematic variations in the data. These methods include regression, regression on the first derivatives, partial correlation analysis, and seasonal differencing analysis.

Using previously published Brookhaven ^{32}Si and ^{36}Cl data as an illustrative example, we have shown that the ^{32}Si and ^{36}Cl data appear to have systematic variations that differ that between the radionucleides. Both climate variation and novel physics have been proposed as causes of the differing systematics [2, 17–20]. Using regression, first derivative regression, partial correlation, and seasonal differencing analyses, we have determined that the systematic variations in the Brookhave data are statistically inconsistent with being caused by novel physics, but are consistent with being potentially related to variations in temperature and air pressure. Data dependence on temperature was previously noted in Reference [2], but the apparent potential relationship to air pressure is somewhat surprising given that Reference [2] mentions that the Brookhaven detector was enclosed in a pressure regulated chamber. However, Reference [2] also mentions that the regulation would at times fail when the air pressure was low, which perhaps explains the slight but significant correlations we observe between variations in the data and variations in air pressure. In addition, pressure dependence can potentially arise if pressure regulation is relative to outside air pressure.

Using an exponential curve with corrections for temperature and air pressure variation we fit the Brookhaven ^{32}Si/^{36}Cl data for the ^{32}Si half life. We obtain the ^{32}Si half-life estimate $T_{1/2} = 171.8 \pm 1.8$. This has uncertainty 1.87 times smaller than the original half-life estimate in Reference [2] (which was obtained with an exponential fit with no correction for systematic variation), $T_{1/2} = 171.6 \pm 3.3$. The reduction in uncertainty when climate-related systematic variations are accounted for is equivalent to increasing the size of the data set by a factor of almost 3.5.

We hope that the methods outlined in this paper will be helpful to researchers who wish to understand and control the systematic variations in their radionucleide decay data.

References

[1] da Silva, MAL, *et al* (2004) *Use of the reference source method to determine the half-lives of radionuclides of importance in nuclear medicine*, Applied radiation and isotopes, 60(2): 301-305.

[2] Alburger, DE, Harbottle, G, Norton, EF (1986) *Half-life of* ^{32}*Si* Earth Plan. Sci. Lett. 78: 168-176

[3] Woods, MJ and Collins, SM (2004) *Half-life data-a critical review of TECDOC-619 update*, Applied radiation and isotopes 60(2) : 257-262

[4] Gostely, JJ (1992) *A determination of the half-life of 137Cs*, International journal of radiation applications and instrumentation. Part A. Applied radiation and isotopes, 43(7): 949-951.

[5] Allison, JW (1960) *Half-life of iridium 192*, British Journal of Applied Physics, 11(7) :302.

[6] Van Ammel, R and Pomme, S and Sibbens, G (2004) *Experimental verification of the half-life of¡ sup¿ 65¡/sup¿ Zn*, Applied radiation and isotopes, 60(2): 337-339.

[7] Schrader, H (1989) *Measurement of the half-lives of 18F, 56Co, 125I, 195Au and 201Tl*, International Journal of Radiation Applications and Instrumentation. Part A. Applied Radiation and Isotopes, 40(5): 381-383.

[8] Schrader, H (2004) *Half-life measurements with ionization chambers – A study of systematic effects and results.* Applied Radiation and Isotopes, 60: 317-323

[9] The National Climatic Data Center (NCDC) of the National Oceanic and Atmospheric Administration (NOAA), `http://www.ncdc.noaa.gov`. (accessed Sep 19, 2012)

This website gives access to hourly climate data for many locations worldwide. For full details on how to access NCDC climate data from this website, please contact the author (S.Towers).

[10] Daily averages of airport climate data from `http://www.wunderground.com/history/airport/AAA/YYYY/MM/1/MonthlyHistory.html?format=1` where `AAA` is the IATA airport code for the airport, `YYYY` is the year, and `MM` is the month. (accessed Sep 19, 2012)

[11] De Carli, M, *et al* (2007) *People's clothing behaviour according to external weather and indoor environment* Building and Environment, 42(12): 3965-3973.

[12] Morrey, JR (1968) *On determining spectral peak position from composite spectra with a digital computer.* Anal. Chem. 40:905-914.

[13] Horlick, G (1972) *Digital data handling of spectra utilizing Fourier transformations.* Anal. Chem., 1972, 44 (6): 943-947.

[14] Kunihiro, B, Shibata, R, and Sibuya, M (2004) *Partial correlation and conditional correlation as measures of conditional independence.* Australian and New Zealand Journal of Statistics 46 (4): 657-664.

[15] Hamilton, JD (1994) *Time Series Analysis.* Princeton University Press, ISBN 0-691-04289-6

[16] Tsay, RS (2005) *Analysis of Financial Time Series.* John Wiley & Sons, ISBN 0-471-690740

[17] Semkow, TM, *et al* (2009) *Oscillations in radioactive exponential decay*, Physics Letters B, 675(5): 415-419.

[18] Jenkins, JH, Mundy DW, and Fischbach, E (2010) *Analysis of environmental influences in nuclear half-life measurements exhibiting time-dependent decay rates* Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 620(2):332-342.

[19] Jenkins, JH, *et al* (2009) *Evidence of correlations between nuclear decay rates and Earth–Sun distance*, Astroparticle Physics, 32(1): 42-46.

[20] Javorsek, D *et al* (2010) *Power Spectrum Analyses of Nuclear Decay Rates.* Astroparticle Physics, 34(3): 173-178.

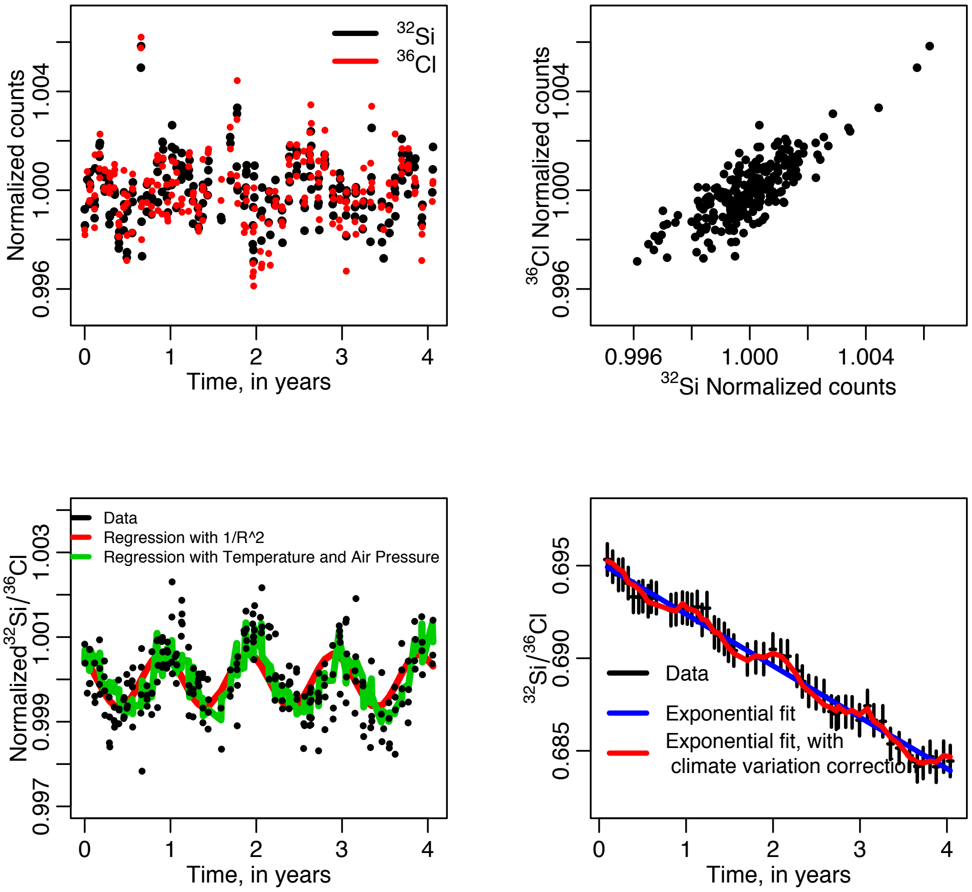

Figure 1: The first plot shows ^{32}Si and ^{36}Cl decay data recorded over a four year period at the Brookhaven National Laboratory. The data are normalized by estimated half-life. The second plot shows the normalized ^{32}Si data vs the ^{36}Cl data; the data are highly correlated, indicating that they share significant common systematic variation. Taking the ratio of the normalized ^{32}Si/^{36}Cl data reveals annual variation, indicating that the decay data of the two radionucleides also have differing sources of systematic variation, and/or sources of systematic variation with differing amplitude (third plot). Overlaid is the regression fit with $1/R^2$ (note that $1/R^2$ is out of phase with the data), and the regression fit with temperature and air pressure (which is in phase with the data). In the fourth plot we show the ^{32}Si/^{36}Cl data. Overlaid are the best-fit regression with an exponential, and the best-fit regression with an exponential with correction for systematic variation related to variations in temperature and air pressure.