Evidence for Correlations Between Nuclear Decay Rates and Earth-Sun Distance

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Unexplained periodic fluctuations in the decay rates of $^{32}$Si and $^{226}$Ra have been reported by groups at Brookhaven National Laboratory ($^{48}$Si), and at the Physikalisch-Technische Bundesanstalt in Germany ($^{226}$Ra). We show from an analysis of the raw data in these experiments that the observed fluctuations are strongly correlated in time, not only with each other, but also with the distance between the Earth and the Sun. Some implications of these results are also discussed, including the suggestion that discrepancies in published half-life determinations for these and other nuclides may be attributable in part to differences in solar activity during the course of the various experiments, or to seasonal variations in fundamental constants.

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Following the discovery of radioactivity by Becquerel in 1896, an intense effort was mounted to ascertain whether the decay rates of nuclides could be affected by external influences including temperature, pressure, chemical composition, concentration, and magnetic fields. By 1930, Rutherford, Chadwick, and Ellis concluded that “The rate of transformation of an element has been found to be a constant under all conditions.” (For decays resulting from K-capture, or for beta-decays in strong ambient electromagnetic fields, the situation is slightly more complicated, since these decays are influenced by the electron wave functions which can be affected by external pressure or fields.) For $^{32}$Si and $^{226}$Ra, which decay by beta- and alpha-emission, respectively, fluctuations in the counting rates (in the absence of strong external electromagnetic fields) should thus be uncorrelated with any external time-dependent signal, as well as with each other. In what follows we show that neither of these expectations is realized in data we have analyzed for $^{32}$Si and $^{226}$Ra, thus suggesting that these decays are in fact being modulated by an external influence.

Between 1982 and 1986, Alburger, et al. measured the half-life of $^{32}$Si at Brookhaven National Laboratory (BNL) via a direct measurement of the counting rate as a function of time. If $\dot{N}(t)$ denotes the number of surviving atoms starting from an initial population $N_0$ at $t = 0$, then the familiar exponential decay law, $N(t) = N_0 e^{-\lambda t}$, leads to $\dot{N} \equiv dN/dt = -\lambda N_0 e^{-\lambda t}$ where $\lambda = \ln(2)/T_{1/2}$. A plot of $\ln(\dot{N}(t))$ as a function of time is then a straight line whose slope is $\lambda$, which then gives the half-life $T_{1/2}$. At the time this experiment was initiated, the $^{32}$Si half-life was believed to be in the range of $60 \lesssim T_{1/2} \lesssim 700$ yr, and hence a multiyear counting experiment was needed to obtain a measurable slope. As in other counting experiments, the counting rate for $^{32}$Si was continually monitored in the same detector against a long-lived comparison standard, which in the BNL experiment was $^{36}$Cl ($T_{1/2}$=301,000 yr). Since the fractional change in the $^{36}$Cl counting rate over the four year duration of the experiment was only $O(10^{-5})$, which was considerably smaller than the overall uncertainty of the final result, $T_{1/2}(^{32}$Si$)=172(4)$ yr, the $^{36}$Cl decay rate was assumed to be constant. Any time dependence for $^{36}$Cl beyond the expected statistical fluctuations was then presumed to arise from various systematic effects, such as drift in the electronics. By computing the ratio $^{32}$Si/$^{36}$Cl $\equiv N(^{32}$Si$)/N(^{36}$Cl$)$, these apparatus-dependent systematic effects should have largely cancelled, and hence this ratio was used to obtain the half-life of $^{32}$Si. On the other hand, barring an accidental cancellation, time-dependent contributions to the $^{32}$Si and $^{36}$Cl decay rates themselves would not cancel in the ratio $^{32}$Si/$^{36}$Cl.

The BNL data for the ratio $^{32}$Si/$^{36}$Cl revealed an unexpected annual variation of $^{32}$Si/$^{36}$Cl which could not be accounted for by the known effects of temperature, humidity, or pressure variations in their detector. We obtained the raw data from the BNL experiment in conjunction with an independent effort to apply a new randomness test to nuclear decays, and the BNL data are shown in Fig. 1. When comparing the results from experiments on different nuclides, it is convenient to study the function $U(t) \equiv \dot{N}(t)/\dot{N}(0)$ $\exp(+\lambda t)$ rather than $\dot{N}(t)$ itself, since $U(t)$ should be time-independent for all nuclides. For $^{32}$Si, we used $\lambda = 4.0299 \times 10^{-3}$yr$^{-1}$ from Ref. 6. Figure 1 exhibits $U(t)$ for the $^{32}$Si/$^{36}$Cl BNL data, along with a plot of $1/R^2$, where $R$ is the distance between the Earth and the Sun. An annual modulation of the $^{32}$Si/$^{36}$Cl ratio is clearly evident, as was first reported in Ref. 6. The Pearson correlation coefficient, $r$, between the raw BNL data and $1/R^2$ is $r=0.52$ for $N=239$ data points, which translates to a formal probability of $6 \times 10^{-18}$ that this correlation would arise from
two data sets which were uncorrelated. As shown in Figure 2, the correlation coefficient increases to $r=0.65$ for $N=235$ data points when a 5 point rolling average is applied. There is also a suggestion in Figs. 1 and 2 of a phase shift between $1/R^2$ and the BNL data, which we discuss in greater detail below.

![FIG. 1: Plot of $U(t)$ for the raw BNL $^{32}\text{Si}/^{36}\text{Cl}$ ratio along with $1/R^2$ where $R$ is the Earth-Sun distance in units of $1/(\text{a.u.})^2$. $U(t)$ is obtained by multiplying each data point by $\exp(\lambda t)$ where $\lambda = \ln(2)/T_{1/2}$ and $T_{1/2}=172$ yr for $^{32}\text{Si}$. The left axis gives the scale for the normalized $U(t)$, and the right axis denotes the values of $1/R^2$ with $1/(\text{a.u.})^2$ obtained from the U.S. Naval Observatory (USNO). The fractional change in $^{32}\text{Si}$ counting rates between perihelion and aphelion is approximately $3 \times 10^{-3}$. As noted in the text, the correlation coefficient between the BNL data and $1/R^2$ is $r=0.52$ for $N=239$ points. The formal probability that the indicated correlation could have arisen from uncorrelated data sets is $6 \times 10^{-18}$.](image)

The strong correlation between the BNL decay data and the annual modulation of the Earth-Sun distance suggests that the $^{32}\text{Si}/^{36}\text{Cl}$ ratio may be responding to some influence originating from the Sun. If this is indeed the case, then the effects of this influence would be expected to be present in other decays as well. Although there are hundreds of potentially useful nuclides whose half-lives have been measured, the data from many of the experiments we examined were generally not useful, most often because data were not acquired continuously over sufficiently long time periods. However, we were able to obtain the raw data from an experiment carried out at the Physikalisch-Technische Bundesanstalt (PTB) in Germany [9,10], measuring the half-life for $^{185}\text{Eu}$, in which $^{226}\text{Ra}$ was the long-lived comparison standard. This experiment, which extended over 15 years, overlapped in time with the BNL experiment for approximately 2 years, and exhibited annual fluctuations in the $^{226}\text{Ra}$ data similar to those seen at BNL. Figure 3 exhibits the PTB data as a 5 point rolling average, and it is evident from the figure that the PTB data closely track the annual variation of $1/R^2$. The Pearson correlation coefficient $r$ for the data in Fig. 3 is $r=0.66$ for $N=1968$ data points, corresponding to a formal probability of $2 \times 10^{-246}$ that this correlation could arise from two data sets which were uncorrelated. As in the case of the BNL data, there is also a suggestion of a phase shift between $1/R^2$ and the PTB data (see below), although this phase shift appears to be smaller than for the BNL data.

Since the BNL and PTB data each exhibit strong correlations with the annual variation of $1/R^2$, it is not surprising that these data correlate with each other. Figure 4 exhibits this correlation along with the annual variation of $1/R^2$. The Pearson correlation coefficient for the BNL and PTB data is $r=0.88$ for $N=35$ points, which corresponds to a formal probability of $4 \times 10^{-12}$ that this correlation could have arisen from two uncorrelated data sets. Moreover, the difference in latitude between BNL and PTB, as well as the difference in their climates, argues against an explanation of this correlation in terms of seasonal variations of climatic conditions such as temperature, pressure, and humidity etc., which could have influenced the respective detection systems. As an example, radon concentrations are known to fluctuate seasonally, as has been noted in Ref. [10], and it was suggested that the decay of $^{222}\text{Rn}$ could lead to a seasonally dependent charge distribution on the experimental apparatus. However, this effect is extremely small given the low counting rates that typically arise from radon background [11], and in any case, the PTB data shown in Fig. 5 were corrected for background.

![FIG. 2: Plot of the 5 point rolling average of $U(t)$ for the BNL $^{32}\text{Si}$ data shown in Figure 1. Each data point represents the average of 5-points centered on the original datum, which serves to smooth short term fluctuations in the $^{32}\text{Si}/^{36}\text{Cl}$ ratio arising from influences other than a possible annual $1/R^2$ variation. As noted in the text, the correlation coefficient between the BNL data and $1/R^2$ is $r=0.65$ for $N=235$ points. The formal probability that the indicated correlation could have arisen from uncorrelated data sets is $1 \times 10^{-20}$.](image)
The preceding considerations, along with the correlations evident in Fig. 4, suggest that the time-dependence of the $^{32}\text{Si}/^{36}\text{Cl}$ ratio and the $^{226}\text{Ra}$ decay rate are being modulated by an annually varying flux or field originating from the Sun, although they do not specify what this flux or field might be. The fact that the two decay processes are very different (alpha decay for $^{226}\text{Ra}$ and beta decay for $^{32}\text{Si}$) would seem to preclude a common mechanism for both. However, recent work by Barrow and Shaw [12, 13] provides an example of a type of theory in which the Sun could affect both the alpha- and beta-decay rates of terrestrial nuclei. In their theory, the Sun produces a scalar field $\phi$ which would modulate the terrestrial value of the electromagnetic fine structure constant $\alpha_{EM}$. This could, among other effects, lead to a seasonal variation in alpha and beta decay rates, both of which are sensitive to $\alpha_{EM}$ [14]. We note from Fig. 3 that the fractional difference between the $^{226}\text{Ra}$ counting rates at perihelion and aphelion is $\approx 3 \times 10^{-3}$, and this would require that the coupling constant $k_\alpha$ of $\phi$ to $\alpha_{EM}$ should be $k_\alpha \approx 3 \times 10^6$. However, this is substantially larger than the value $k_\alpha = (-5.4 \pm 5.1) \times 10^{-8}$ inferred from a recent trapped ion experiment [12, 15]. Although the specific model of Refs. [12, 13, 14] would not account for the $^{32}\text{Si}$ and $^{226}\text{Ra}$ data quantitatively, variants of this model might work. This includes models in which separate scalar fields $\phi_1$ and $\phi_2$ couple, respectively, to $\alpha_{EM}$ and to the electron-proton mass ratio $m_e/m_p$.

Another interesting possibility is that terrestrial radioactive nuclei are interacting in a novel way with the neutrino flux $\Phi_\nu$ emitted from the interior of the Sun. This flux also varies with $1/R^2$, and the resulting seasonal modulation of $\Phi_\nu$ has been observed by Super-Kamiokande [16, 17]. This possibility is supported by the data we report in Ref. [18] in which we present evidence for the possible detection of a change in the decay rate of $^{54}\text{Mn}$ during the solar flare of 13 December 2006. As noted in Ref. [18], the coincidence in time between the change in the $^{54}\text{Mn}$ counting rate and the solar flare, along with other observations, is consistent with a mechanism based on a change in $\Phi_\nu$ during the solar flare.

We note that irrespective of the origin of the solar flare data, or of the correlations evident in Figs. 14, the existence of these effects may explain discrepancies in various half-life determinations reported in the literature. Examples are $^{32}\text{Si}$, $^{44}\text{Ti}$ and $^{137}\text{Cs}$, among many others [6, 19, 20, 21]. If nuclides such as $^{32}\text{Si}$, $^{36}\text{Cl}$, and $^{226}\text{Ra}$ respond to changes in the solar neutrino flux due to the time-dependence of $1/R^2$, then they can also respond to changes in intrinsic solar activity which are known to occur over time scales both longer and shorter than one year. Thus, depending on when half-life measurements were made, and on the specific techniques employed, it is possible that some of the half-life discrepancies reported in the literature could be reconciled if appropriate data on solar activity become available.

Returning to Fig. 4 we briefly explore the suggestion noted above of a possible phase shift of $1/R^2$ relative to both the BNL and PTB data. Although this may be an experimental artifact arising from binning effects, etc., such a phase shift could also arise from other smaller contributions to periodic variations in neutrino flux. Possibilities for such contributions were explored in Ref. [17].
where a search was made for short time variations in the observed flux at Super-Kamiokande arising from either the 7.25° inclination of the solar axis relative to the ecliptic, or from fluctuations in the temperature of the solar core. A modulation of the neutrino flux arising from a coupling between a neutrino magnetic moment and a latitudinally inhomogeneous solar magnetic field [22] could also account for a possible phase shift. Although there is no compelling evidence at present for such short time variations at Super-Kamiokande [16, 17], the statistical power of the BNL, PTB, and similar data sets may prove to be a useful tool in the search for such effects. Yet another possible explanation for the apparent phase shift could be a seasonally-varying velocity-dependent effect similar to that observed by the DAMA/LIBRA collaboration [23].

In summary, we have presented evidence for a correlation between changes in nuclear decay rates and the Earth-Sun distance. While the mechanism responsible for this phenomenon is unknown, theories involving variations in fundamental constants could give rise to such effects. These results are also consistent with the correlation between nuclear decay rates and solar activity suggested by Jenkins and Fischbach [18] if the latter effect is interpreted as possibly arising from a change in the solar neutrino flux. These conclusions can be tested in a number of ways. In addition to repeating long-term decay measurements on Earth, measurements on radioactive samples carried aboard spacecraft to other planets would be very useful since the sample-Sun distance would then vary over a much wider range. The neutrino flux hypothesis might also be tested using samples placed in the neutrino flux produced by nuclear reactors.

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[1] H. Becquerel, Comptes Rendus 122, 501 (1896).
[2] S. E. Rutherford, J. Chadwick, and C. Ellis, Radioactivity (Cambridge University Press, 1930).
[3] L. Fassio-Canuto, Phys. Rev. 187, 2141 (1969).
[4] H.-P. Hahn, H.-J. Born, and J. Kim, Radiochimica Acta 23, 23 (1976).
[5] T. Ohtsuki, H. Yuki, M. Muto, J. Kasagi, and K. Ohno, Phys. Rev. Lett. 93, 112501 (2004).
[6] D. E. Alburger, G. Harbottle, and E. F. Norton, Earth and Planet. Sci. Lett. 78, 168 (1986).
[7] S.-J. Tu and E. Fischbach, Phys. Rev. E 67 (2003).
[8] S.-J. Tu and E. Fischbach, Int. J. of Mod. Phys. C 16, 281 (2005).
[9] H. Schrader, private communication.
[10] F. Wissman, Rad. Prot. Dos. 118, 3 (2006).
[11] J. D. Barrow and D. J. Shaw, arXiv:0806.4317v1 [hep-ph] (2008).
[12] D. J. Shaw, arXiv:gr-qc/0702090v1 (2007).
[13] J. Uzan, Rev. Mod. Phys. 75, 403 (2003).
[14] T. Rosenband et al., Science 319, 1808 (2008).
[15] J. Yoo et al., Phys. Rev. D 73, 112001 (2006).
[16] J. Jenkins and E. Fischbach, arXiv:astro-ph/0808.3156 (2008).
[17] D. E. Alburger and G. Harbottle, Phys. Rev. C 41, 2321 (1990).
[18] I. Ahmad et al., Phys. Rev. Lett. 80, 2550 (1998).
[19] M. Woods, Nucl. Inst. Meth. Phys. Res. A286, 576 (1990).
[20] P. A. Sturrock, G. Walther, and M. S. Wheatland, Ap.J. 507, 978 (1998).
[21] R. Bernabei et al., Eur. Phys. Jour. C (In press) (2008).