Radiation Therapy Implications of Anomalous Variations of the Nuclear Decay Law

James S. Welsh

April 2012

Advisory Committee on the Medical Uses of Isotopes

Special thanks to:

- George J. Buse
- Ephraim Fischbach
- Nicholas Flores
- J. Thomas Gruenwald
- Daniel Javorsek II
- Jere H. Jenkins
- Robert H. Lee
- Daniel W. Mundy
- Peter A. Sturrock

Radioactive decay is expected to follow a mathematically precise exponential function

- From an educational website on radioactivity: “No operation or process of any kind (i.e., chemical or physical) has ever been shown to change the rate at which a radionuclide decays.”

RADIATIONS

FROM

RADIOACTIVE SUBSTANCES

by

SIR ERNEST RUTHERFORD, O.M., D.Sc., Ph.D., LL.D., F.R.S.

NOBEL LAUREATE

Cavendish Professor of Experimental Physics in the University of Cambridge

JAMES CHADWICK, Ph.D., F.R.S.

Fellow of Gonville and Caius College, Cambridge

and

C. D. ELLIS, Ph.D., F.R.S.

Fellow of Trinity College, Cambridge

Re-issue of the edition of 1930

CAMBRIDGE

AT THE UNIVERSITY PRESS

1951

Generally true but well-known exceptions do exist

- Electron Capture (e.g. ^{7}Be, ^{109}In and ^{110}Sn)
- Isomeric Transitions
  - ^{99m}Tc was the first metastable isotope to demonstrate observable half-life changes due to chemical environment

Another possible exception?

- Recent studies of the measured decays of some radioisotopes have suggested anomalous variations in decay rate
- These observations raise the question of whether such variations could have clinical relevance:
  – Temporary brachytherapy (both LDR and HDR)
  – Teletherapy
  – Gamma Knife radiosurgery treatments

- “While looking into the rate of radioactive decay of several isotopes as a possible source of random numbers……researchers pored through published data on specific isotopes and found disagreement in the measured decay rates – odd for supposedly physical constants…”
- “Checking collected data they came across something even more surprising: long-term observation of the decay rate of Si-32, Cl-36 and Ra-226 seemed to show a small seasonal variation. The decay rate was ever so slightly faster in winter than in summer.”
  - “Apparently radioactivity is stronger in winter than in summer.”

- “In February 2011 Earth witnessed a large coronal mass ejection... For most people this just meant a brighter Aurora Borealis.”
- “However, for radioactive elements across the planet the flare may have caused a decrease in radioactive decay…”

- “Purdue scientists noticed the decay rate of a medical isotope dropped during the solar flare, and what's more, the decline started before the flare did.”
- “…could be useful for protecting satellites and astronauts -- if there is a correlation between decay rates and solar activity, changed decay rates could provide early warning of an impending solar storm.”

- “…in classrooms around the world, students are taught that the rate of decay of a specific radioactive material is a constant. This concept is relied upon, for example, when anthropologists use carbon-14 to date ancient artifacts and when doctors determine the proper dose of radioactivity to treat a cancer patient…”

Upon more detailed reading

- While examining data (prospectively collected from 2Dec 06 – 3Jan 07 at Purdue University; datasets acquired at Brookhaven National Laboratory, Physikalisch-Technische Bundesanstalt and Children's Nutrition Research Center; and published data) researchers found disagreement in measured decay rates

Nuclear Instruments and Methods in Physics Research A286 (1990) 576–583
North-Holland

THE HALF-LIFE OF ^{137}Cs – A CRITICAL REVIEW

M.J. WOODS

Division of Radiation Science and Acoustics, National Physical Laboratory, Teddington, Middlesex TW11 0LW, UK

During a recent coordinated research programme organised by the International Atomic Energy Agency, the half-lives of 36 radionuclides were evaluated to assess their suitability as standards for detector calibrations. That exercise highlighted several radionuclides, including ^{137}Cs, for which there were significant discrepancies between the published data. Half-life values for ^{137}Cs have been examined in more detail in an attempt to discover some of the causes of the observed differences and suggest more specific recommendations for metrologists and evaluators.

M.J. Woods, S.E.M. Lucas / Nucl. Instr. and Meth. in Phys. Res. A 369 (1996) 534–538 537

Table 2
Published half-life values

Author(s)	Year	Ref.	Half-life*	Std. dev	Used in evaluation
Wiles and Tomlinson	1955	[4]	10117	±146	No
Anikina	1958	[5]	10702	±584	Yes
Flynn et al. (a)	1965	[6]	10227	±146	Yes
Flynn et al. (b)	1965	[6]	10410	±329	Yes
Anspach et al.	1965	[7]	10527	±51	Yes
NBS	1977	[1]	10636	±88	No
Lagoutine et al.	1978	[8]	10282	±13	No
Ramthun	1983	[9]	10589	±92	No
Debertin	1985	[1]	10848	±110	No
Kochin et al.	1989	[10]	10665	±37	No
Schötzig et al.	1991	[11]	10513	±14	Yes
Martin et al.	1994	[12]	10561	±14	Yes
This work	1995		10495	±4	Yes

* Where published values have been expressed in years, they have been converted to days using a factor of 365.25.

Earth and Planetary Science Letters, 78 (1986) 168–176
Elsevier Science Publishers B.V., Amsterdam – Printed in The Netherlands

[6]

Half-life of ^{32}Si

D.E. Alburger, G. Harbottle and E.F. Norton

Brookhaven National Laboratory, Upton, NY 11973 (U.S.A.)

Received August 15, 1985; revised version received March 3, 1986

Beta rays from a ^{32}Si-^{32}P source, produced in 1968-69 via the $^{30}Si(t,p)^{32}Si$ reaction using a Van de Graaff beam at $E_t = 3.4$ MeV, were counted with an end-window gas-flow proportional counter system including an automatic precision sample changer. Comparison counts were taken on the β rays from a ^{36}Cl source. Measurements beginning February, 1982 were made at approximately 4-week intervals, each consisting of a total of 40 hours of counting on each sample. The decay rate was determined from the $^{32}Si/^{36}Cl$ ratio of counts. Small periodic annual deviations of the data points from an exponential decay curve were observed, but are of uncertain origin and had no significant effect on the result. Based on the analysis of 53 points taken in 48 months, the value $T_{1/2} = 172(4)$ yr is adopted for the half-life of ^{32}Si. This result is substantially greater than two previously reported measurements of 108(18) yr and 101(18) yr but is lower than values based on geophysical evidence.

Phys. Med. Biol., 1990, Vol. 35, No 8, 1079–1088. Printed in the UK

The effective half-life of a broad beam ^{238}PuBe total body neutron irradiator

Kenneth J Ellis

USDA/ARS Children's Nutrition Research Center, Department of Pediatrics, Baylor College of Medicine, Houston, TX 77030, USA

Received 12 January 1990, in final form 2 April 1990

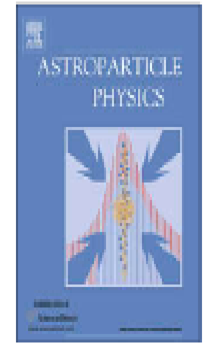
Abstract. A broad-beam ^{238}PuBe neutron irradiator has been previously developed for exclusive use in *in vivo* neutron activation analysis in humans. The initial calibrations of the facility provided only a fixed value for the thermal neutron flux. Adjustment of this flux value for decay of the neutron source was later introduced and was based on the physical half-life of ^{238}Pu. The current findings would suggest, however, that a more appropriate value for the effective half-life for the total body irradiator is 141.7 ± 2.5 y. In addition, variations in the induced counts for a Mn standard on a yearly basis indicate that seasonal differences of approximately 0.5% can be present between the winter and summer months.

Astroparticle Physics 32 (2009) 42–46

Contents lists available at ScienceDirect

Astroparticle Physics

journal homepage: www.elsevier.com/locate/astropart



Evidence of correlations between nuclear decay rates and Earth–Sun distance

Jere H. Jenkins [a], Ephraim Fischbach [a,*], John B. Buncher [a], John T. Gruenwald [a], Dennis E. Krause [a,b], Joshua J. Mattes [a]

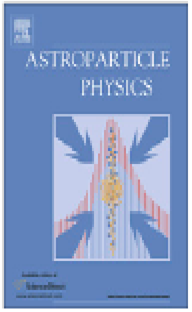
[a] *Physics Department, Purdue University, 525 Northwestern Avenue, West Lafayette, Indiana 47907, USA*
[b] *Physics Department, Wabash College, Crawfordsville, Indiana 47933, USA*

Astroparticle Physics 34 (2010) 121–127

Contents lists available at ScienceDirect

Astroparticle Physics

journal homepage: www.elsevier.com/locate/astropart



Power spectrum analysis of BNL decay rate data

P.A. Sturrock [a,*], J.B. Buncher [b], E. Fischbach [b], J.T. Gruenwald [b], D. Javorsek II [c], J.H. Jenkins [b], R.H. Lee [d], J.J. Mattes [b], J.R. Newport [b]

[a] *Center for Space Science and Astrophysics, Stanford University, Stanford, CA 94305-4060, USA*
[b] *Department of Physics, Purdue University, West Lafayette, IN 47907, USA*
[c] *416th Flight Test Squadron, 412th Test Wing, Edwards AFB, CA 93524, USA*
[d] *Department of Physics, United States Air Force Academy, CO 80920, USA*

Pergamon

Appl. Radiat. Isot. Vol. 49, No. 9–11, pp. 1397–1401, 1998

Printed in Great Britain
0969-8043/98 $19.00 + 0.00

PII: S0969-8043(97)10082-3

Half-life Measurements of Europium Radionuclides and the Long-term Stability of Detectors

HELMUT SIEGERT, HEINRICH SCHRADER* and ULRICH SCHÖTZIG

Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, D-38116 Braunschweig, Germany

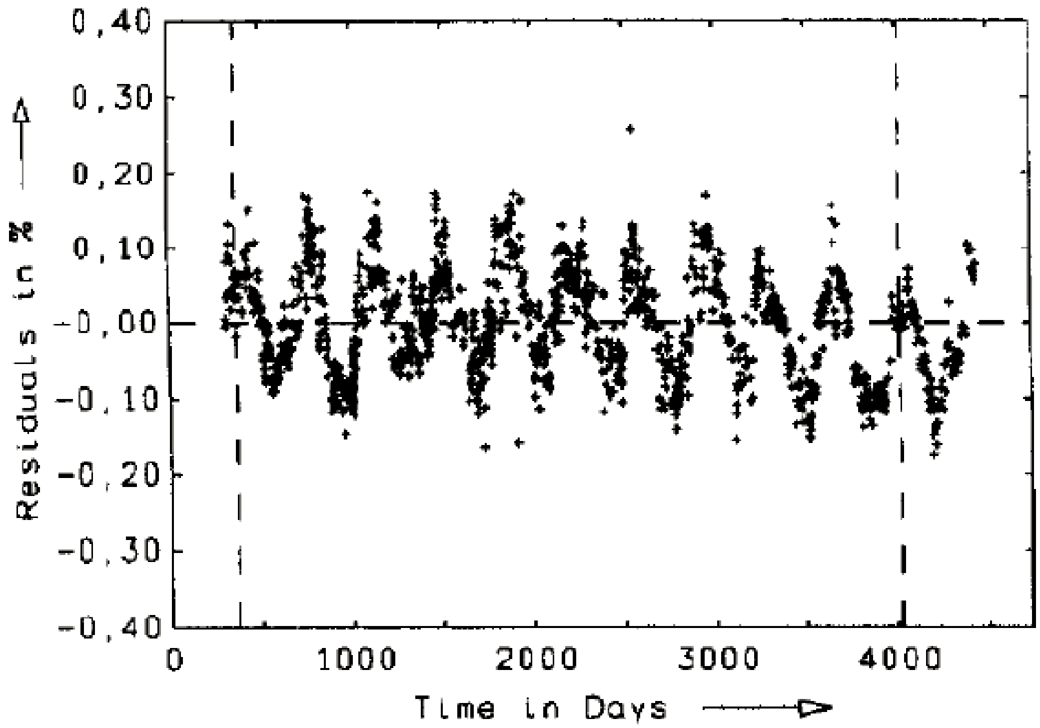


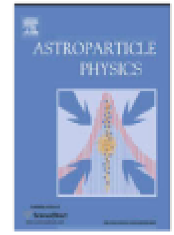
Fig. 1. Residuals of the ionization chamber measurement data of ^{226}Ra as a function of time from a fit with an exponential decay function. A datum point is an average value and contains about 30 individual measurements of current taken over about 3 days and corrected for background. The vertical dotted lines are positioned at 1st January at an interval of 10 years.

Astroparticle Physics 31 (2009) 407–411

Contents lists available at ScienceDirect

Astroparticle Physics

journal homepage: www.elsevier.com/locate/astropart



Perturbation of nuclear decay rates during the solar flare of 2006 December 13

Jere H. Jenkins, Ephraim Fischbach *

Physics Department, Purdue University, West Lafayette, IN 47907, USA

A R T I C L E I N F O

Article history:
Received 2 April 2009
Received in revised form 20 April 2009
Accepted 25 April 2009
Available online 7 May 2009

Keywords:
Flares
Particle emission: Sun – nuclear reactions
Neutrinos: physical processes

A B S T R A C T

Recently, correlations have been reported between fluctuations in nuclear decay rates and Earth–Sun distance, which suggest that nuclear decay rates may be affected by solar activity. In this paper, we report the detection of a significant decrease in the decay of ^{54}Mn during the solar flare of 2006 December 13, whose X-rays were first recorded at 02:37 UT (21:37 EST on 2006 December 12). Our detector was a 1 μCi sample of ^{54}Mn, whose decay rate exhibited a dip coincident in time with spikes in both the X-ray and subsequent charged particle fluxes recorded by the Geostationary Operational Environmental Satellites (GOES). A secondary peak in the X-ray and proton fluxes on December 17 at 12:40 EST was also accompanied by a coincident dip in the ^{54}Mn decay rate. These observations support the claim by Jenkins et al. that nuclear decay rates may vary with Earth–Sun distance.

- “…swings seemed to be in synch with the Earth’s elliptical orbit, with the decay rates oscillating as the Earth came closer to the sun and then moving away…”
- “…took another look at the decay data… found a recurring pattern of 33 days. It was a bit of a surprise, given that most solar observations show a pattern of about 28 days – the rotation rate of the surface of the sun…. But the core of the sun – where nuclear reactions produce neutrinos – apparently spins more slowly than the surface

Neutrinos to blame?

- Radioactive decay itself produces neutrinos, so depending on the shape of the sample, the flux of neutrinos varies
  - A sphere should have a greater internal flux of neutrinos than a thin foil sample, for example
- Could half-life of a radionuclide depend on its shape?

Nuclear Instruments and Methods in Physics Research A 622 (2010) 93–96

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Study of the dependence of ^{198}Au half-life on source geometry

R.M. Lindstrom [a], E. Fischbach [b], J.B. Buncher [b], G.L. Greene [c,d], J.H. Jenkins [e,b,*], D.E. Krause [f,b], J.J. Mattes [b], A. Yue [c]

[a] Analytical Chemistry Division, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA
[b] Physics Department, Purdue University, 525 Northwestern Avenue, West Lafayette, IN 47907, USA
[c] Physics Department, University of Tennessee, Knoxville, TN 37996, USA
[d] Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA
[e] School of Nuclear Engineering, Purdue University, 400 Central Dr, West Lafayette, IN 47907, USA
[f] Department of Physics, Wabash College, Crawfordsville, IN 47933, USA

Nuclear Instruments and Methods in Physics Research A 659 (2011) 269–271

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Absence of a self-induced decay effect in ^{198}Au

Richard M. Lindstrom [a], Ephraim Fischbach [b,*], John B. Buncher [c], Jere H. Jenkins [d], Andrew Yue [e]

[a] Analytical Chemistry Division, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA
[b] Department of Physics, Purdue University, West Lafayette, IN 47907, USA
[c] Department of Physics, Wittenberg University, Springfield, OH 45501, USA
[d] School of Nuclear Engineering, Purdue University, 400 Central Dr., West Lafayette, IN 47907, USA
[e] Department of Physics, University of Tennessee, Knoxville, TN 37996, USA

Neutrinos not to blame?

- Researchers from NIST and Purdue tested this decay anomaly by comparing two shapes (spheres and thin foils) of Au-198 with identical mass and activity
- The maximum neutrino flux produced by decay in the sphere was several times greater than the flux of solar neutrinos at the surface of the Earth
  - So an effect should be observed if this was the mechanism
- If neutrinos are truly affecting the decay rate, the atoms in the spheres should decay slower than in the foil
- No significant difference in decay rate between the shapes was detected

Neutrinos not to blame?

- But is there a possible problem with this experiment?
- The neutrinos allegedly causing the observed changes are not those produced by the sample itself (electron antineutrinos in the case of Au-198)
- But rather are supposedly solar neutrinos (which due to neutrino oscillations might have a different effect)

An obvious challenge

- Were the observed variations in decay rate caused by changes in the responses of the experimental apparatus between summer and winter rather than the isotopes themselves?

Nuclear Instruments and Methods in Physics Research A 620 (2010) 332–342

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Analysis of environmental influences in nuclear half-life measurements exhibiting time-dependent decay rates

Jere H. Jenkins [a,b,*], Daniel W. Mundy [c], Ephraim Fischbach [b]

[a] School of Nuclear Engineering, Purdue University, West Lafayette, IN 47907, USA
[b] Physics Department, Purdue University, West Lafayette, IN 47907, USA
[c] Department of Radiation Oncology Physics, Mayo Clinic, Rochester, MN 55905, USA

ARTICLE INFO

Article history:
Received 29 December 2009
Received in revised form
12 March 2010
Accepted 18 March 2010
Available online 27 March 2010

Keywords:
Ionization chambers
Proportional counters
Beta decay
Gamma decay
Energy loss

ABSTRACT

In a recent series of papers evidence has been presented for correlations between solar activity and nuclear decay rates. This includes an apparent correlation between Earth–Sun distance and data taken at Brookhaven National Laboratory (BNL), and at the Physikalisch-Technische Bundesanstalt (PTB). Although these correlations could arise from a direct interaction between the decaying nuclei and some particles or fields emanating from the Sun, they could also represent an "environmental" effect arising from a seasonal variation of the sensitivities of the BNL and PTB detectors due to changes in temperature, relative humidity, background radiation, etc. In this paper, we present a detailed analysis of the responses of the detectors actually used in the BNL and PTB experiments, and show that sensitivities to seasonal variations in the respective detectors are likely too small to produce the observed fluctuations.

Response

- "…we present a detailed analysis of the responses of the detectors actually used.…and show that sensitivities to seasonal variations in the respective detectors are likely too small to produce the observed fluctuations.
- The observed variations in decay rates were highly unlikely to have come from temperature or humidity or some other environmental influence on the detection systems
- Also, there does not seem to be any environmental influence with a 33-day or a half-year period, further calling this possibility into question

Further criticism

- Ra-226 decays by alpha emission… but an annual variation in decay rate was observed
    - So does this type of variability work for alpha as well as beta decay?
- But Cooper showed the RTG (radioisotope thermoelectric generator) power output of NASA’s Cassini satellite was uninfluenced by distance from the sun
    - The RTG used Pu-238, an alpha emitter
    - Cassini’s orbit went as close to the sun as Venus and as far as Saturn

A possible explanation?

- While both Pu-238 and Ra-226 are alpha emitters, the Ra-226 that showed variations was in equilibrium
  - When secular equilibrium is reached (in about 200 years), 42% of the photon emissions are due to beta-decaying daughters of Ra-226
  - An ionization chamber cannot discriminate whether the decay is from the daughters which decay via beta emission or the alpha-emitting parent

A possible explanation?

- In other words, while Ra-226 decays by alpha decay, the daughters (which contribute significantly to what is measured) undergo beta decay and these may exhibit an annual cycle
- In contrast, the Pu-238 in the RTG was not yet in secular equilibrium and therefore non-alpha emitting daughters were not able to contribute to a meaningful degree and variation was not observed

Another challenge

- Norman et al calculated ratios between rates of two kinds of decay, to cancel out any changes in the equipment between summer and winter
- Assumed that if there were an annual variation, it would affect different decay processes differently, so the ratios would change
  - Looked at the ratios between Am-241/Sn-121m, Ba-133/Ag-108m and Na-22/Ti-44
- Found that the ratios did not change much annually

Response

- Am-241 decays primarily by alpha decay, but it remains possible that its decay products decay by beta decay and are subject to an annual influence
- A more important point is that different radionuclides are inherently behave differently
  - And even with beta decay, may not be influenced by the sun as much as others.
  - (Recall that not all electron capture decaying nuclides are equally susceptible to chemical effects e.g. K-40 < Be-7)

Summary

- Anomalous periodic variations have been characterized by both
  - Oscillations with a strong annual periodicity and
  - Short-duration deviations from expected decay rates
- Short-duration deviations result in apparent decay rate changes persisting hours to days
- Annual periodicity has been observed in 14 separate nuclides (including ^{60}Co, ^{90}Sr/^{90}Y, ^{137}Cs and ^{226}Ra)
- Annual oscillation amplitude varies by nuclide and is typically less than 0.5%

Summary

- Short-duration deviations have only been observed in four nuclides thus far (54Mn, 60Co, 90Sr/90Y and 226Ra)
- Preliminary analysis of the short-duration deviations suggest changes in the apparent half-life persisting for up to two days
- It remains unknown whether short-duration decay rate variations exist in other key medical isotopes
    - (e.g. ^{106}Ru/^{106}Rh, ^{103}Pd, ^{125}I, ^{131}Cs, ^{192}Ir)
- It also remains unknown whether short-duration variability, if it does exist in these isotopes, results in clinically relevant dosimetric changes

Summary

- Some of this raw data was acquired during calibration sequences, precision measurements or in establishing references
  - procedures familiar to medical physicists
- Additional investigations could include:
  - Analysis of archived data on these isotopes and
  - Careful examination of existing calibration data from active clinics that have been sampled at frequencies sufficient to detect these rate variations

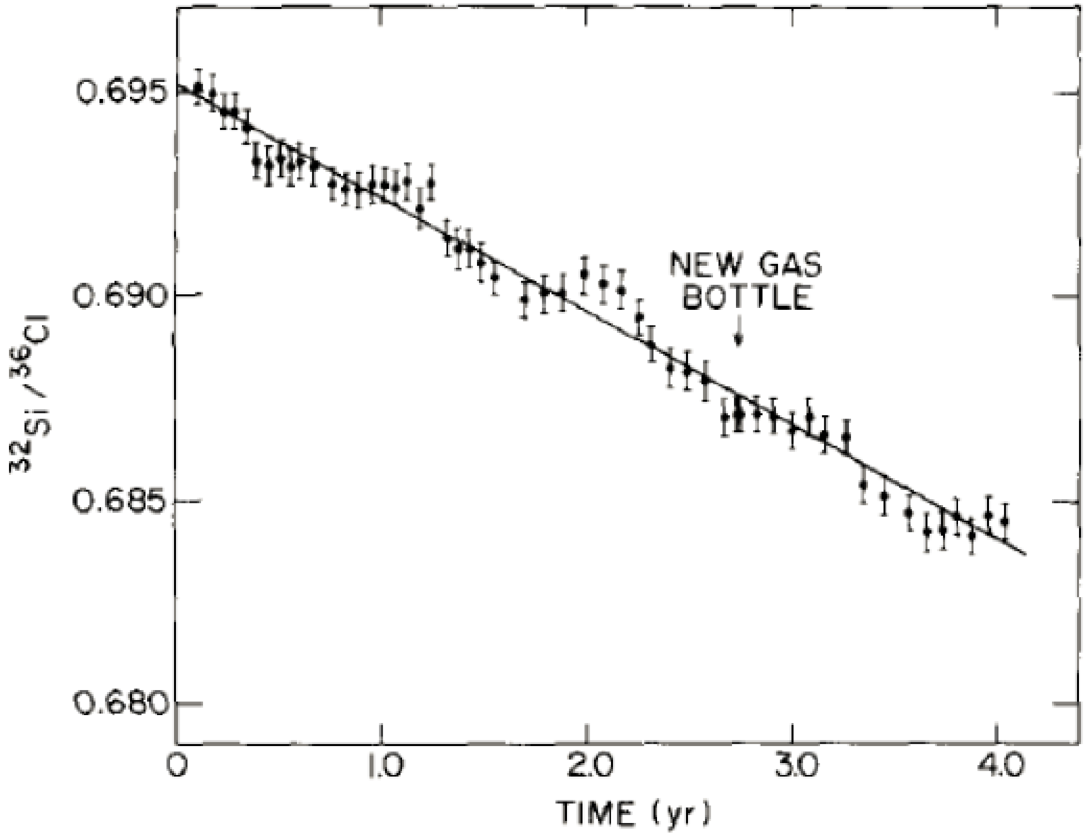


Fig. 2. Ratio of $^{32}Si/^{36}Cl$ counts measured for 53 points over a period of 48 months. Points are averages of 4 runs, each with 10 hr on each sample. Error bars are (arbitrarily) three times the statistical uncertainties and the solid line is an exponential computer fit, although the ordinate is linear for convenience in plotting. The results of the fit are $T_{1/2} = 171.6$ yr with an uncertainty of 3.3 yr and a standard deviation of 3.2 yr.

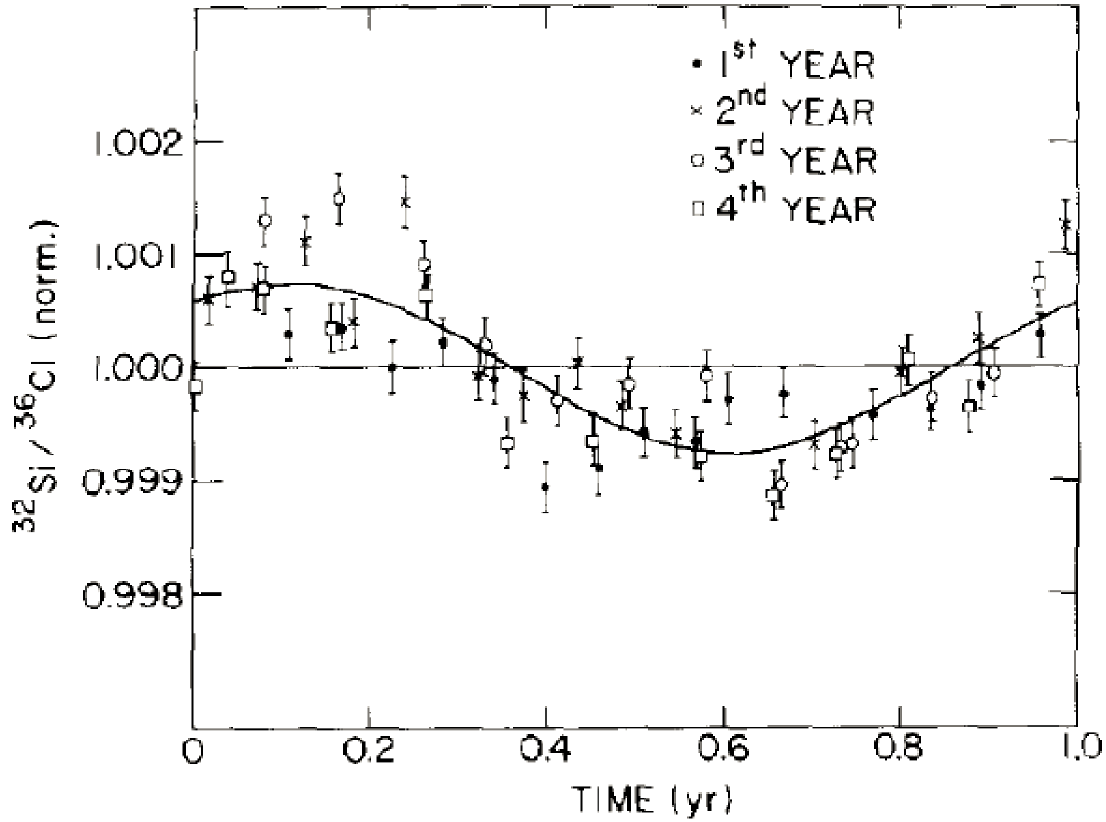
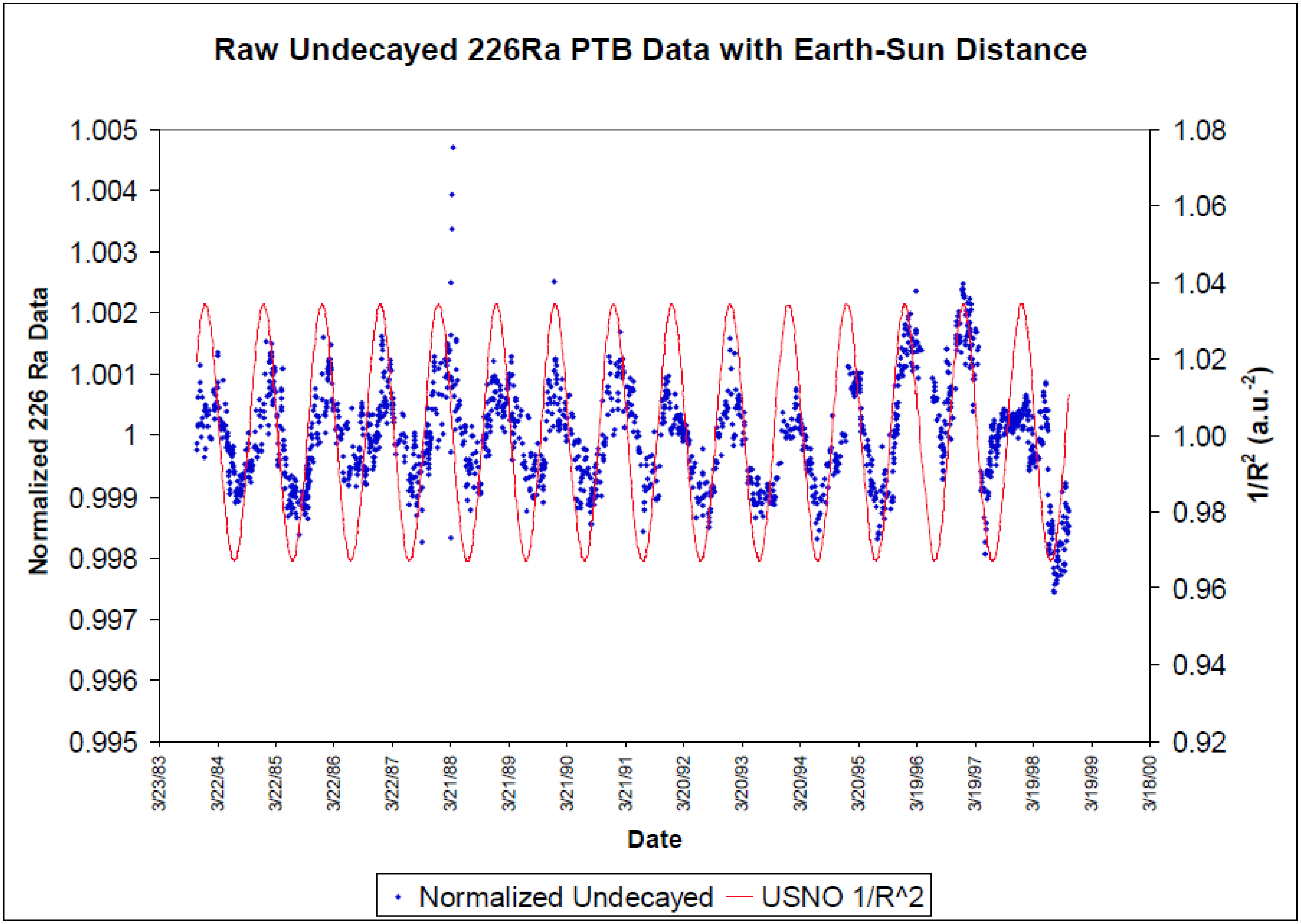
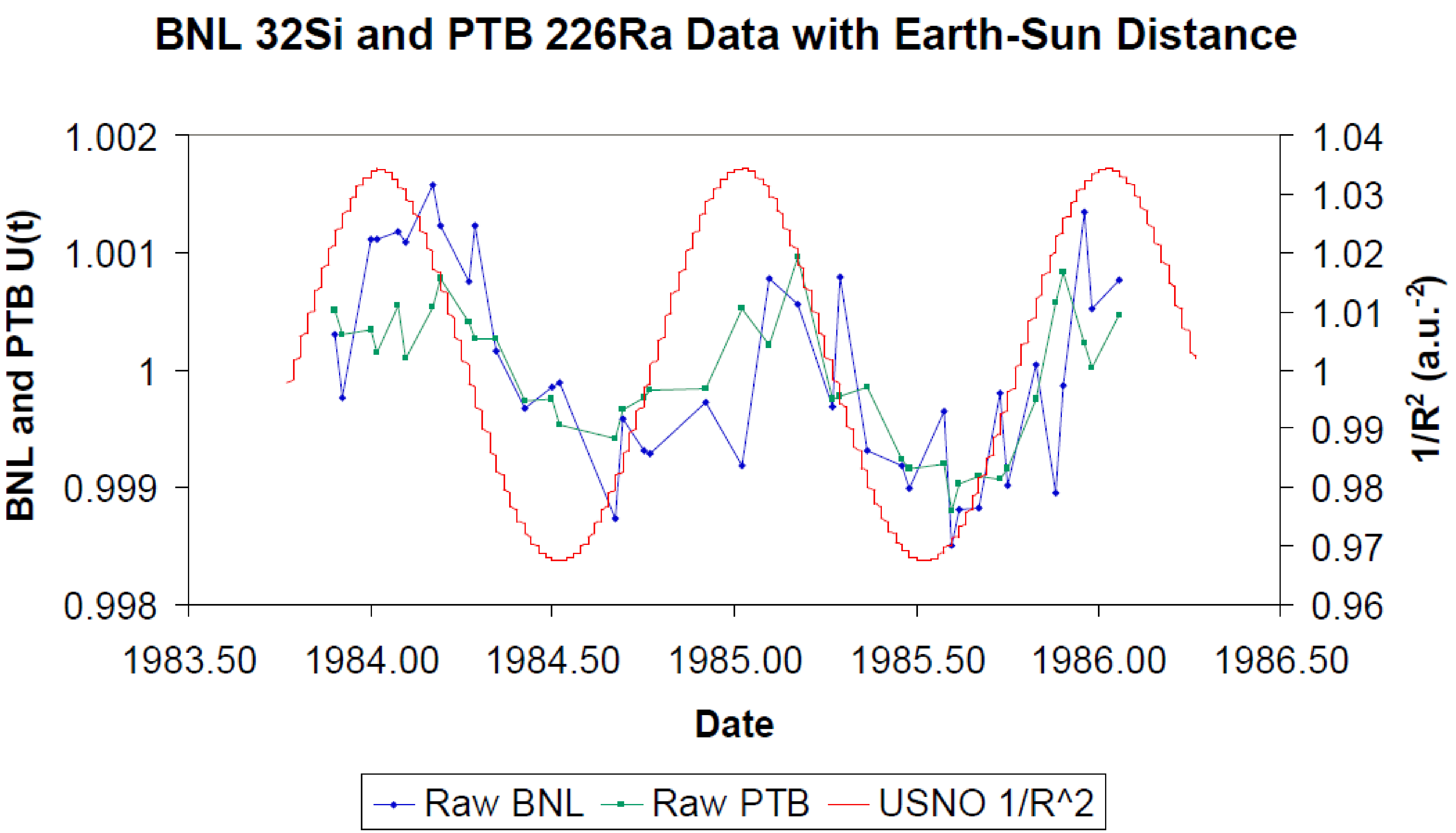


Fig. 4. Points from Fig. 2 corrected for decay using $T_{1/2} = 172$ yr, normalized to 1.000 for the average of all points, and plotted with four 12-month groups superposed. Error bars are statistical uncertainties and $T = 0$ is January 1. A clear annual effect is evident and an arbitrary sine function fit gives an amplitude of 3.4 standard deviations, a maximum on February 9, and a minimum on August 6.

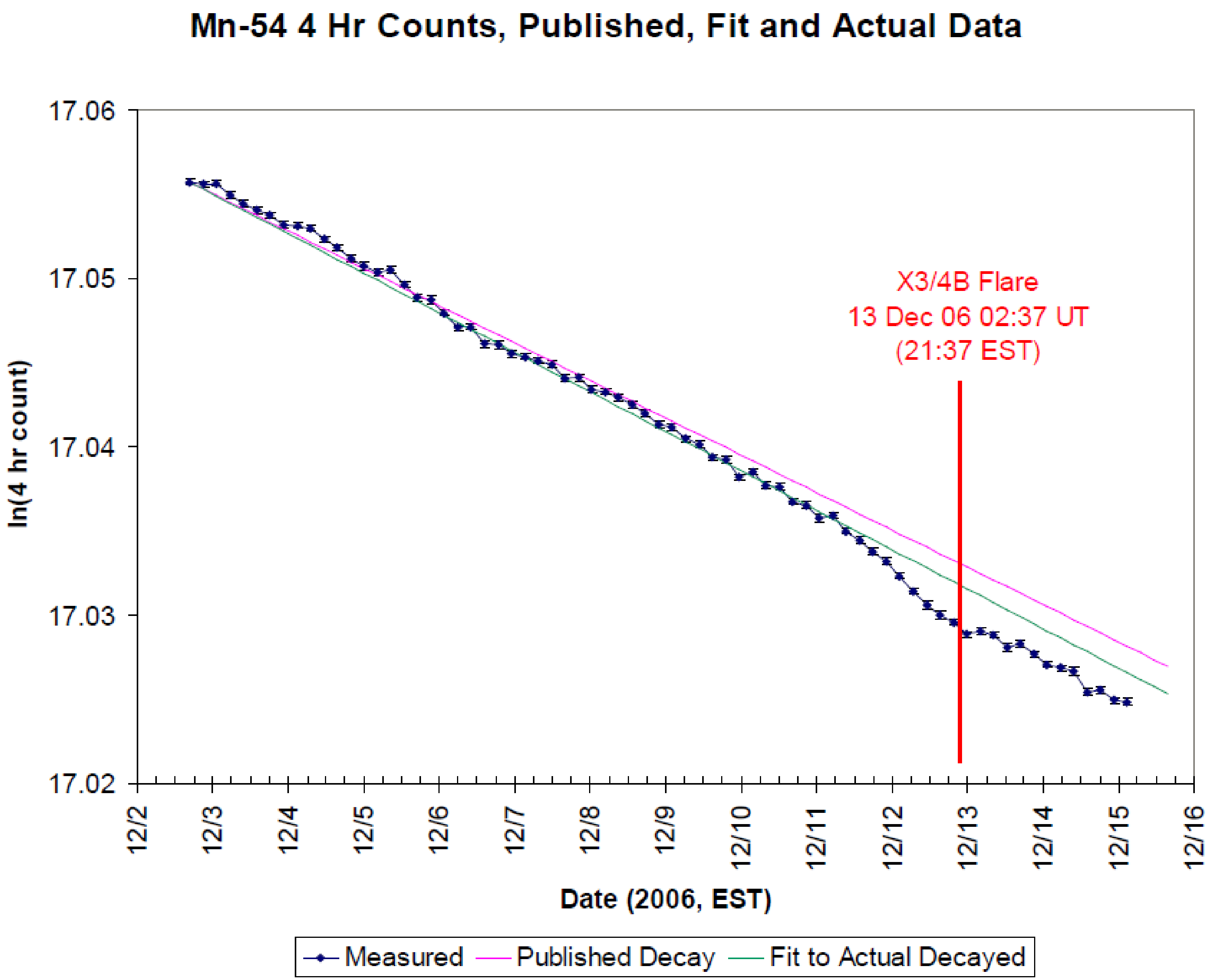


Pearson Correlation Coefficient *r*=0.62, *N*=1974, Prob=5.13x10^{-210}

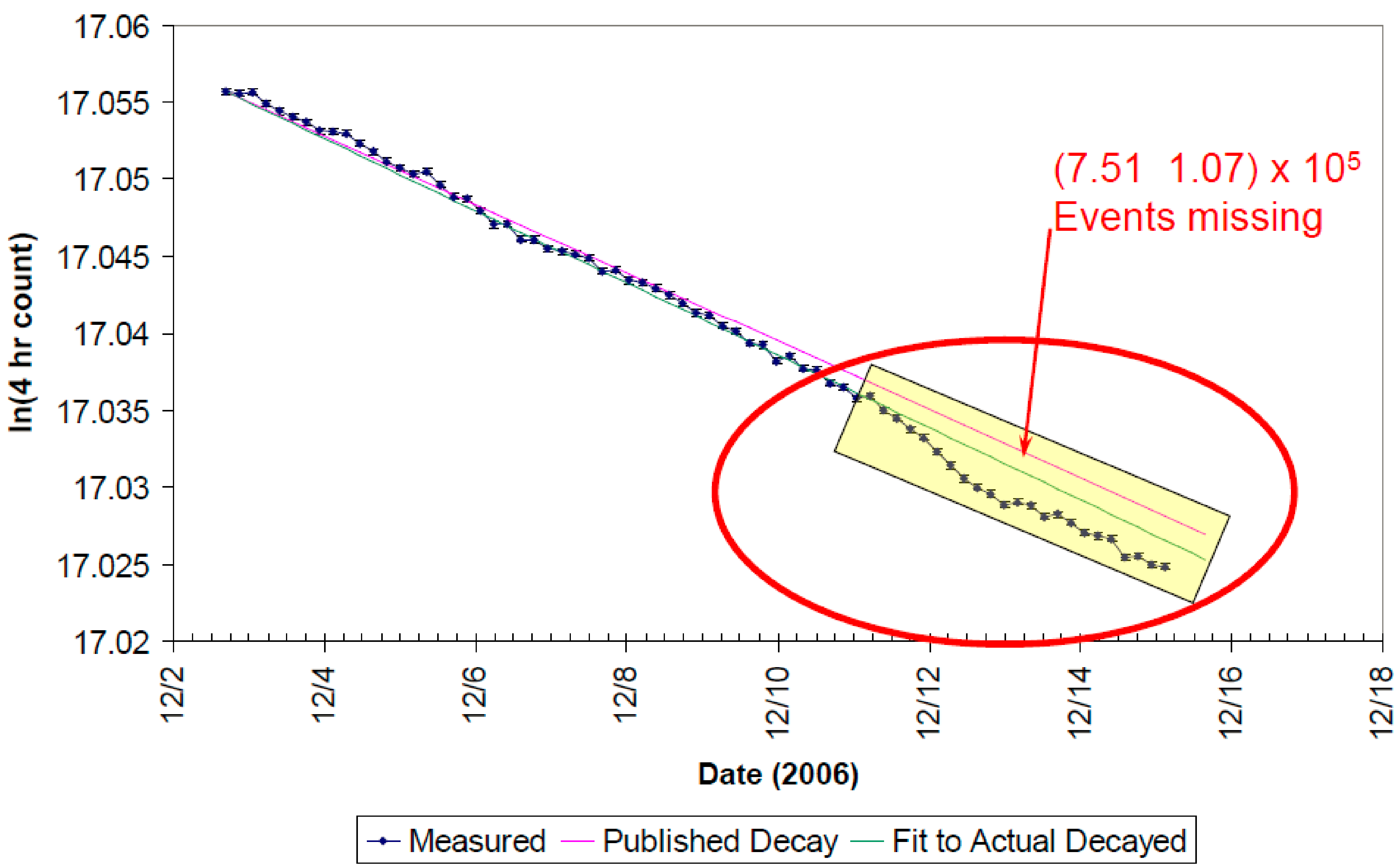
Data from Siegert, et al., Appl. Radiat. Isot. **49**, 1397 (1998) Fig. 1

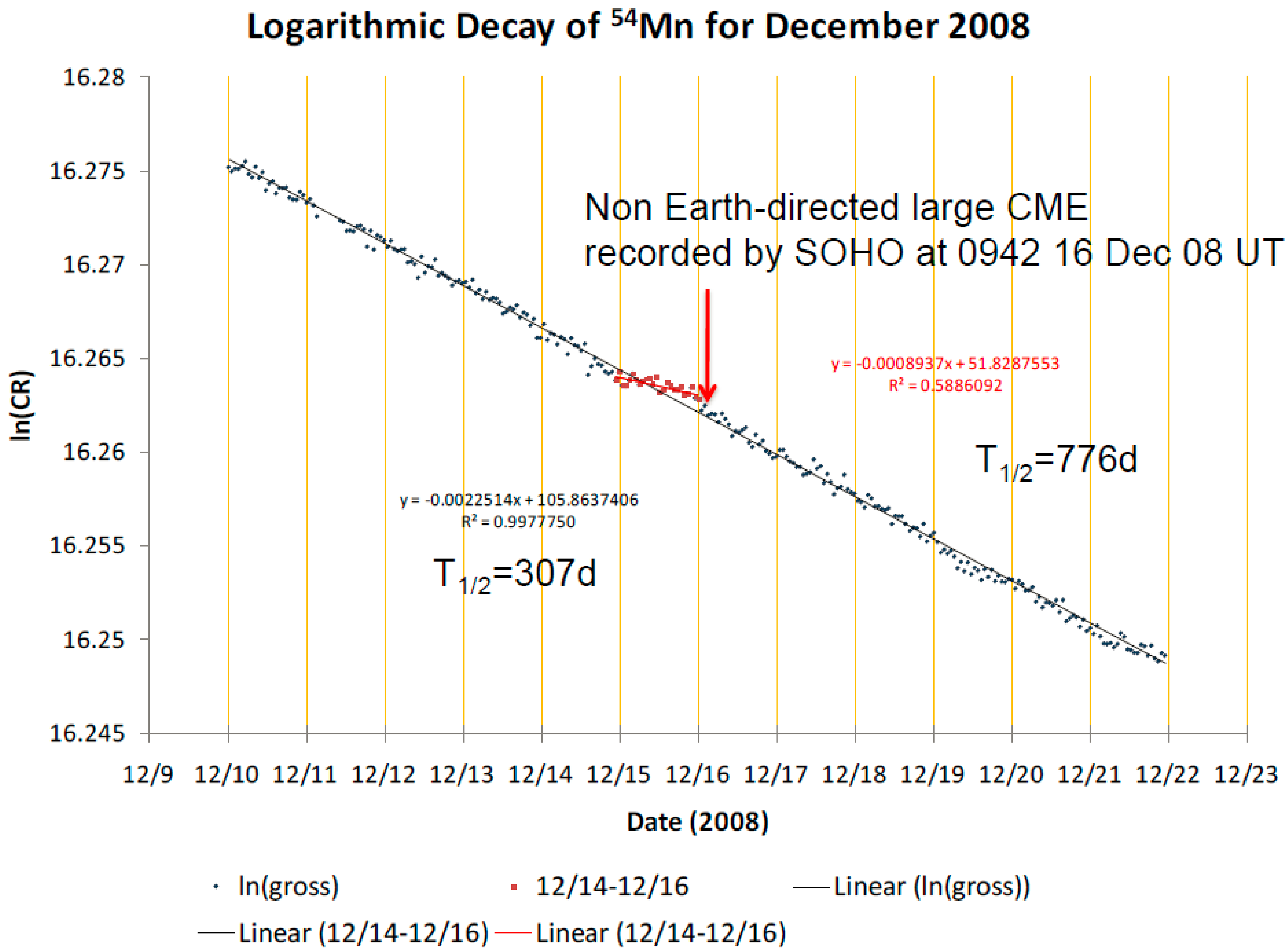


Pearson Correlation Coefficient r**=0.66,** N**=39, Prob=5.8x10^{-6}**


Mn-54 4 Hr Counts, Published, Fit and Actual Data
ln(4 hr count)
17.06
17.05
17.04
17.03
17.02
X3/4B Flare
13 Dec 06 02:37 UT
(21:37 EST)
12/2
12/3
12/4
12/5
12/6
12/7
12/8
12/9
12/10
12/11
12/12
12/13
12/14
12/15
12/16
Date (2006, EST)
Measured
Published Decay
Fit to Actual Decayed

Physics 167 Mn-54 4 Hr Counts, Published, Fit and Actual Data





Logarithmic Decay of 54Mn for December 2008
16.28
16.275
16.27
16.265
16.26
16.255
16.25
16.245
ln(CR)
Non Earth-directed large CME
recorded by SOHO at 0942 16 Dec 08 UT
y = -0.0008937x + 51.8287553
R² = 0.5886092
T1/2=776d
y = -0.0022514x + 105.8637406
R² = 0.9977750
T1/2=307d
12/9 12/10 12/11 12/12 12/13 12/14 12/15 12/16 12/17 12/18 12/19 12/20 12/21 12/22 12/23
Date (2008)
ln(gross)
12/14-12/16
Linear (ln(gross))
Linear (12/14-12/16)
Linear (12/14-12/16)

Purdue Experiments