Carbon-14 and the waste gas beta-gas radiation monitor

Degraded ability on 0EMF50L to accurately detect the quantity of noble gaseous activity contained within the waste gas tanks due to presence of Carbon-14

Sherlock Holmes states

- We must fall back upon the old axiom that when all other contingencies fail, whatever remains, however improbable, must be the truth.
  - "The Bruce-Partington Plans“

- Eliminate all other factors, and the one which remains must be the truth.
  -- *The Sign of the Four*

Waste Gas Monitor: 0EMF50L

- General Atomic Model RD32-06 dual range off-line beta-gas monitor.
- Installed in an “in-line” configuration for WGDT effluent discharges out to the unit 1 vent stack.
- Sensitivity:
  - Xe-133: 2.6E7 CPM/μCi/cc
  - Kr-85: 7.87E7 CPM/μCi/cc

At McGuire EMF stands for

- Electro-motive force?
- Effluent Monitoring Facilities?
- Expiring Many Failures?
- Expect Maintenance Frequently?
- Evaporating Monetary Funding?
- Exasperating Management Focus?
- Engineering Migraine Facilitation
- The sixth Electronic Monitoring System!

History of waste gas monitor

- December 1998 during waste gas decay tank (WGDT) release, waste gas monitor fails RP correlation check due to over-response at 2.6 times estimated response based on RP gamma spectroscopy analysis and is declared inoperable {12/03/1998}
- IAE checks detector calibration, and finds no problem
- Sample at monitor obtained during WGDT release, and compares favorably to initial sample analysis.
- Engineering tasked with analysis of the problem.

Possible Causes

- EMF equipment degradation
- EMI (noise)
- Gas migration underneath beta detector foil
- Excessive backpressure at sample chamber
- Pulse ringing (double counting)
- Waste gas tank stratification
- Calibration Methodology
- Sampling Method
- RP Count Room instrumentation
- Unaccounted activity in WGDT

Systematic elimination of any potential causes of problem

- Detector assembly replaced and calibrated (still overly responsive) {1/5/99}
- During WGDT release, testing coordinated to check for pulse distortion, sample chamber pressure and comparison to independent channel {4/29/99}
- All testing finds no problem with equipment

Analog to Digital Module

- Difference in digital module (RP-86A) to analog module (RP-30)
- Deviation appears to coincide with transition to digital module
- Kr-85 Primary gas standard ordered to test detector
- Testing to encompass both analog & digital modules

Primary Gas Cross-Check {7/19/99}

- Kr-85 from Analytics activity 1.32E-03μCi/cc
- New Detector tested: results 8.04E7CPM/(μCi/cc)
- RP-30A (analog) check against RP-86A (digital) module results for RP-30A
  - 8.08E7CPM/(μCi/cc) for scalar output
  - Analog meter indication @ 7.11E7CPM /(μCi/cc)
  - Output voltage response @ 7.45E7CPM/(μCi/cc)
- 0EMF50 actual detector & its readout module
  - results @ 7.88E7CPM/(μCi/cc)
- Elevated pressure test: sample chamber pressure elevated to 3 PSI and maintained 15 minutes
  - no appreciable increase observed

RD32 beta detector calibrated for Xe-133 & Kr-85 noble gases

- On site primary calibration documented in MCC 1346.05-00-0002 {1994}
  - Xe-133 sensitivity 2.60E7 CPM/(μCurie/cc)
  - Kr-85 sensitivity 7.87E7 CPM/(μCurie/cc)

- Original General Atomic Sensitivity {1975}
  - Xe-133 sensitivity 2.66E7 CPM/(μCurie/cc)
  - Kr-85 sensitivity 6.78E7 CPM/(μCurie/cc)
  - C-14 sensitivity 3.2E6 CPM/(μCurie/cc)

Problem Resolution

- Testing verified there was no equipment problem with 0EMF50
- RP increased correlation value for 0EMF50.
- 0EMF50 finally declared back operable on 9/9/1999 after 10 months logged inoperable due to correlation failure.

Engineering vs. Radiation Protection

- Engineering requested Corrective Action by RP: "RP to evaluate using an independent laboratory or other means to analyze waste gas sample for Beta activity to determine impact on correlation factor used in waste gas releases. It is possible that the Beta activity is a significant contributor to the correlation discrepancies because the process detector (EMF50) is only Beta sensitive."

- RP's Response: "At this time an independent sample to look for only beta emitting isotopes in the gaseous waste stream is not needed. RP is modifying their procedure to perform a correlation with each waste gas tank release inorder to trend the response against the new correlation factor."

Correlation Factor

- RP's Method to manage problem was to adjust EMF50's correlation value to Xe-133 based upon historical performance.
- 3.54E7 CPM/(μCi/cc) {6/18/97} [136%]
- 8.0E7 CPM/(μCi/cc) {8/31/99} [308%]
- 1.8E8 CPM/(μCi/cc) {12/19/00} [692%]
- 4.0E8 CPM/(μCi/cc) {8/16/01} [1538%]
- 2.08E9 CPM/(μCi/cc) {2/13/03} [8000%]

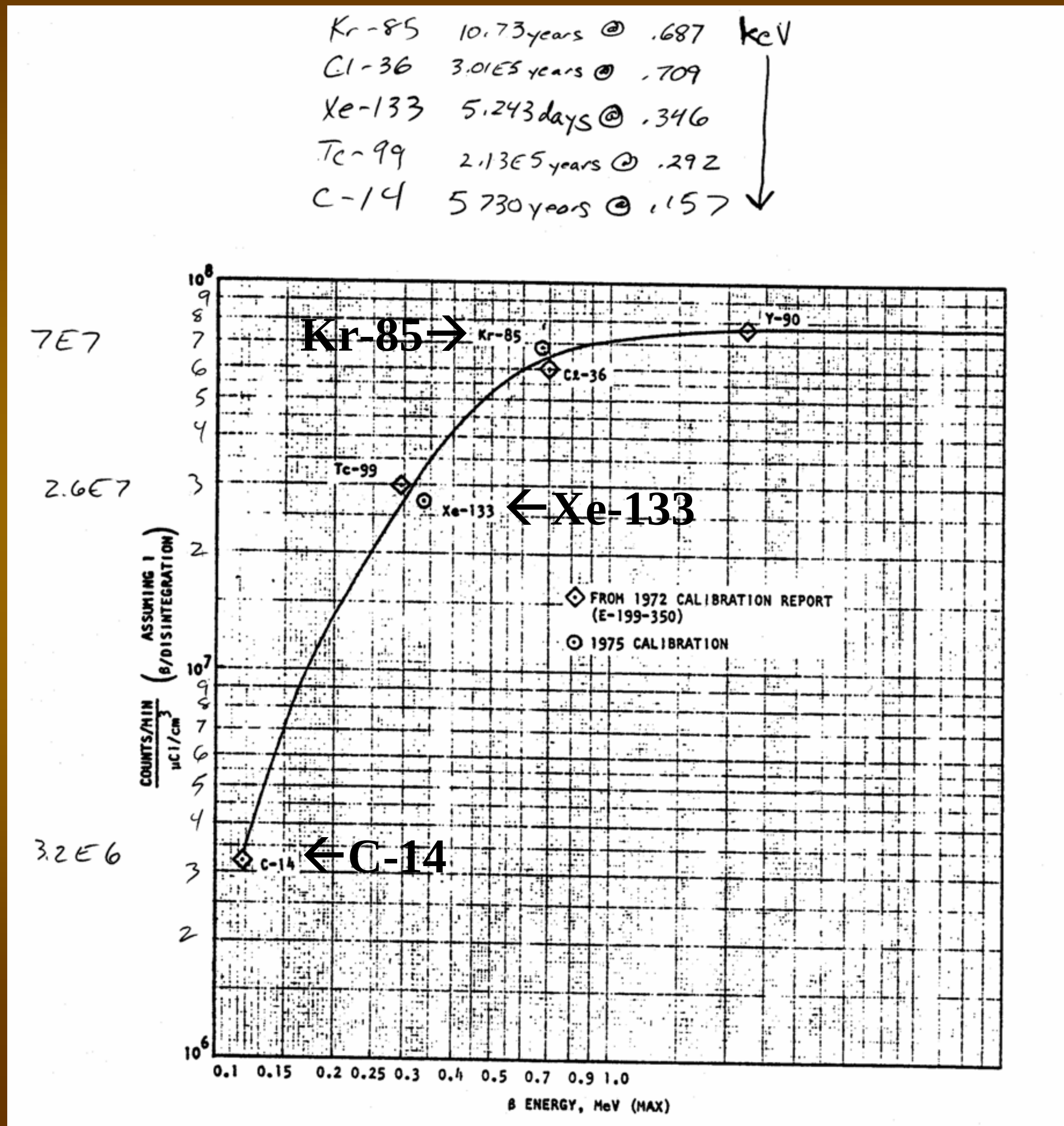
Waste Gas Release Anomalies in 2000

- Two WGDT releases 0EMF50L's response was twice expected range based upon 1999 correlation factor.
- Engineering requested insitu Kr-85 primary standard check on 0EMF50L.
  - (RP still claimed there exists some kind of problem with the instrumentation)
- Work rescheduled 3 times due to plant operational concerns with Waste Gas System out of service.
- On the third strike engineering gives up based upon RP recommendation: "Based on a consistent correlation result over the past few WG Tank releases I do not recommend checking the EMF with Kr-85. As long as the correlation results are consistent and we can accurately predict the EMF response during a release, I do not see a reason to perform the gas calibration."

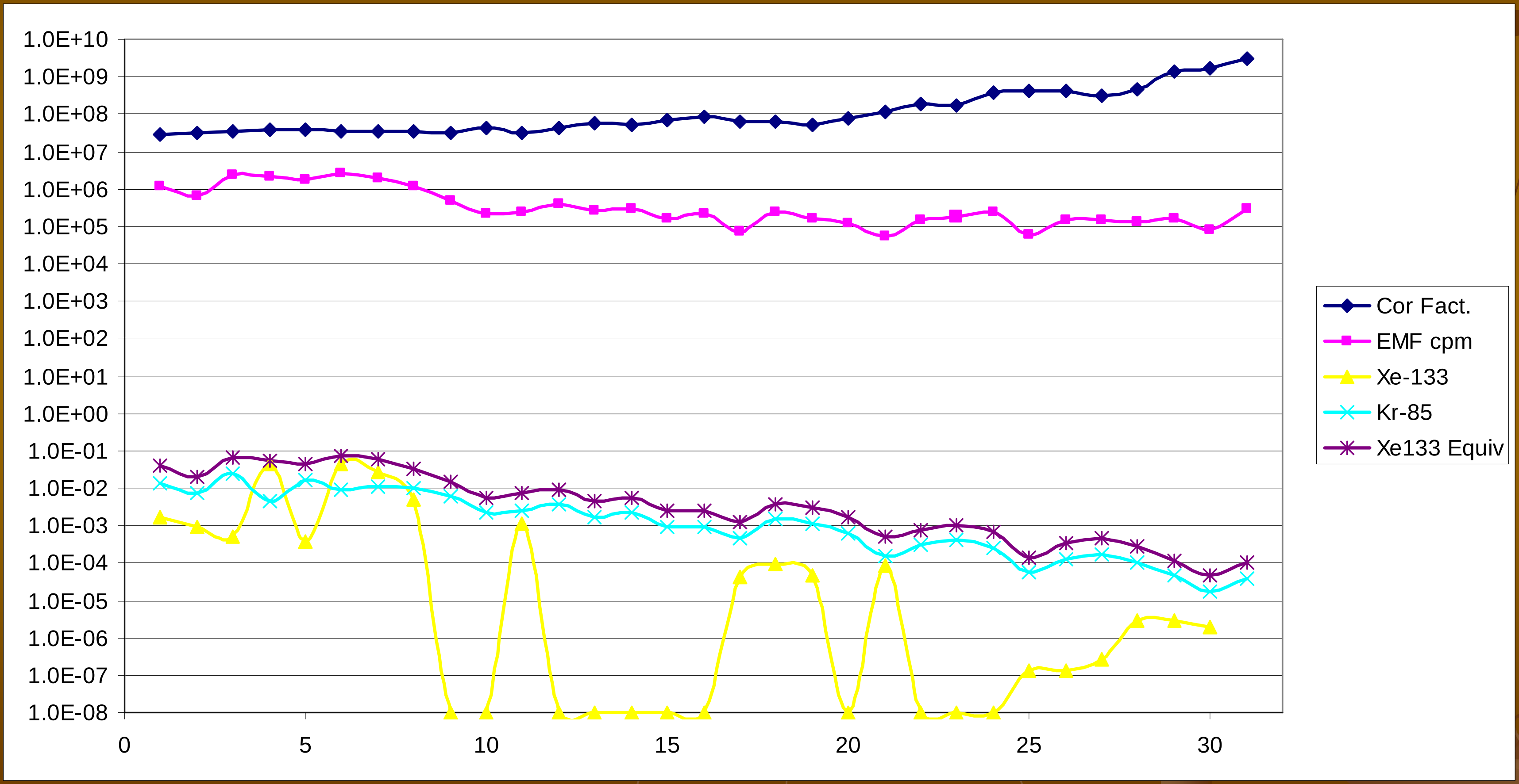
0EMF50L becomes more sensitive!

- 2002: correlation is at 4E8 that is 15 times nominal Xe-133 sensitivity for an RD32 beta-gas monitor.
- Two WGDT still much higher than expected
- Engineering bull-dogs issue by coordinating Kr85 primary gas standard insitu check to prove beyond any reasonable doubt that there still exists NO equipment problems. {7/18/02}
- Test performed on 8/27/02
  - Kr-85 activity @ 1.15E-03 μCi/cc
  - 0EMF50L sensitivity @ 7.88E7 CPM/(μCi/cc)

Calibration Curve of RD32 beta detector



Historical Response of 0EMF50

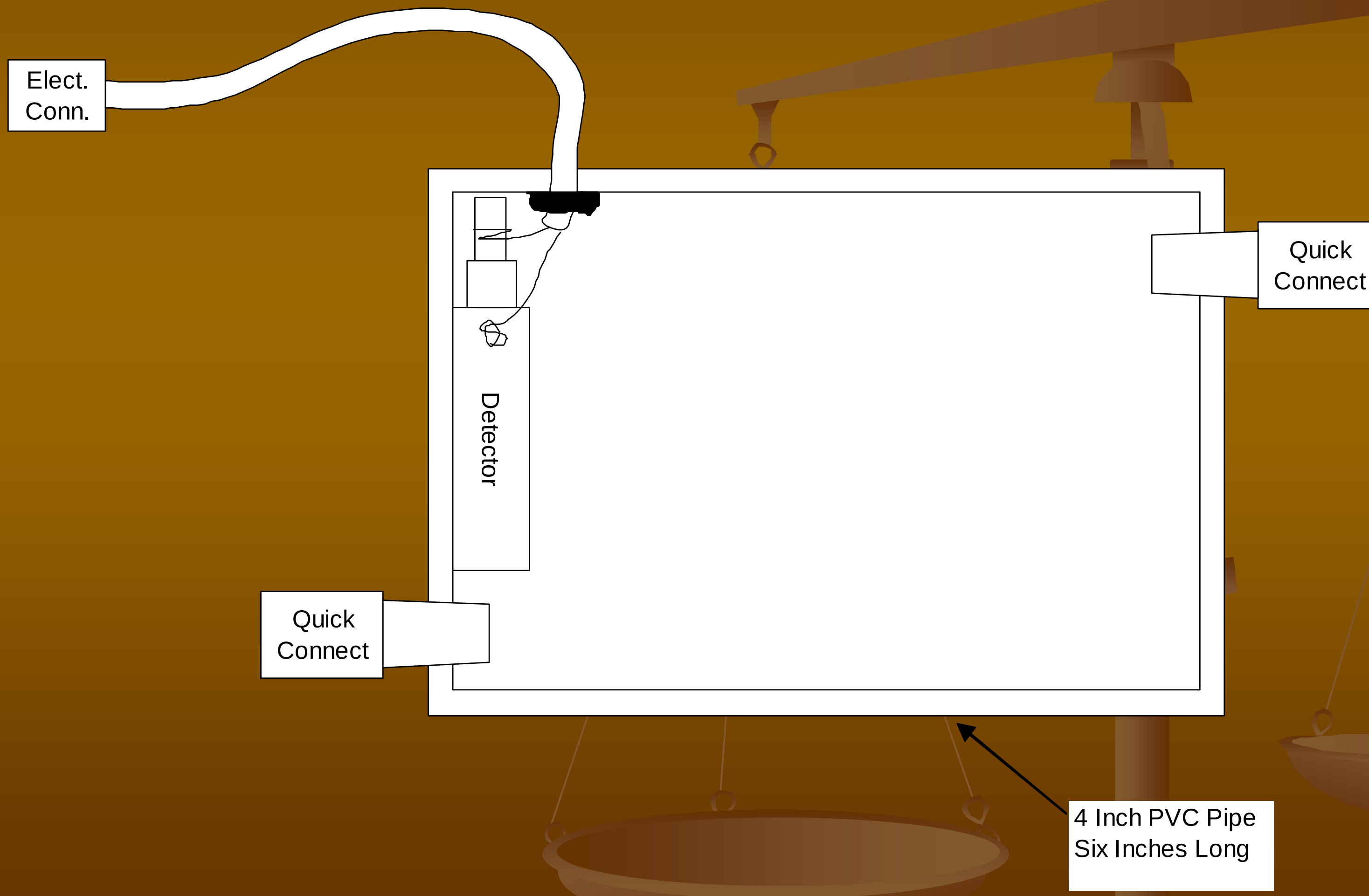


Actual Performance

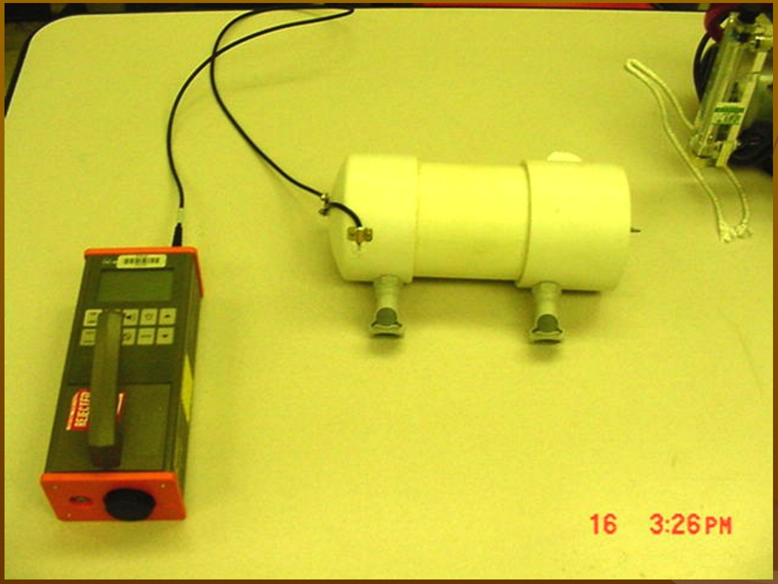
Date	WGDT	Countroom Sample Analysis Isotope			Estimated	
		Xe-133	Kr-85	Xe-Eq	Corr	Noble Gas
05/21/03	D	3.81E-05	1.14E-03	2.96E-03	6.16E+06	9.07E+04
6/2/03	F	9.93E-06	4.57E-04	1.18E-03	2.45E+06	3.62E+04
6/19/03	E	1.83E-05	3.96E-04	1.03E-03	2.14E+06	3.16E+04

Date	WGDT	EMF50 response			Estimated	
		Actual			C-14	
		CPM	RD32 Error	Corr error	Count	μCi/cc
05/21/03	D	2.45E+05	170.15%	-96.02%	1.54E+05	4.82E-02
6/2/03	F	1.80E+05	396.82%	-92.67%	1.44E+05	4.49E-02
6/19/03	E	1.41E+05	345.53%	-93.42%	1.09E+05	3.42E-02

RP's response to rule out any unaccounted beta activity within WGDT



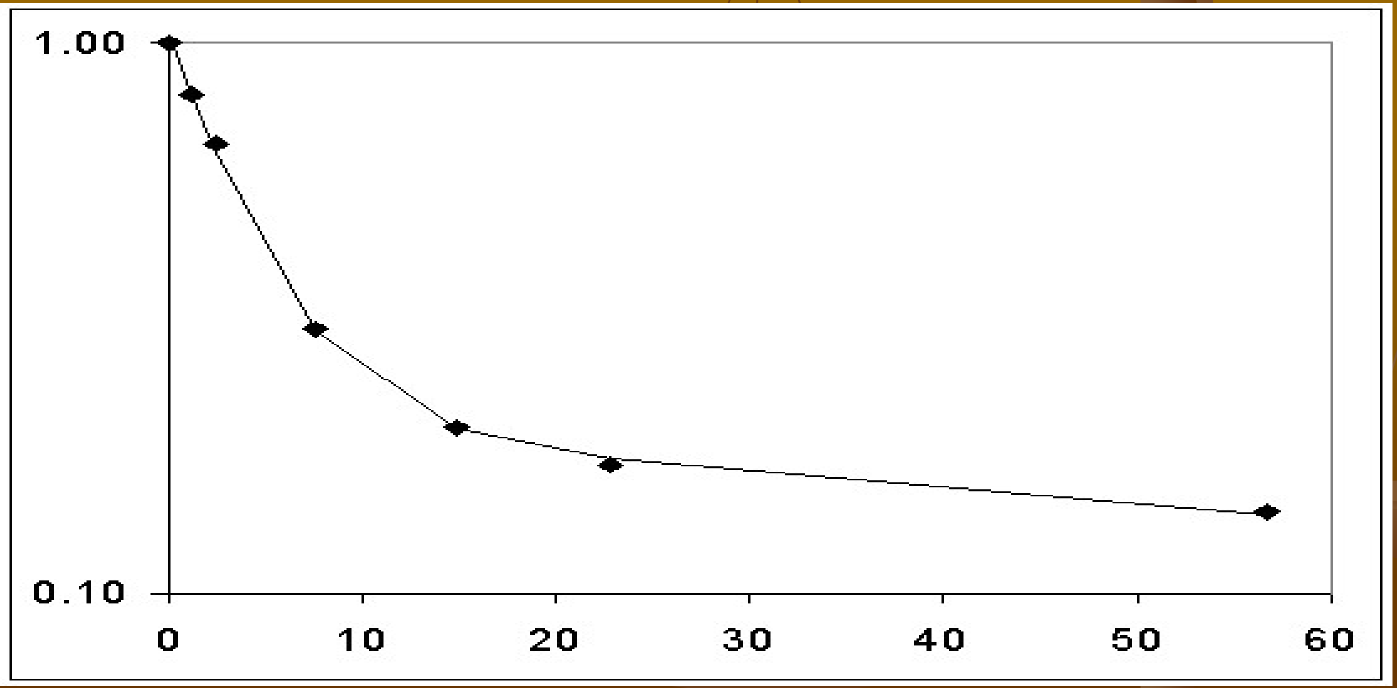
RP’s redundant beta detector



- High Tech
  - (frisker in a pipe)
- Testing with spare RD32 detector, RP pipe detector, sample pump and marinelli installed in series in the RP lab area using actual WGDT samples.
- Results was RP pipe detector over-ranged when exposed to WGDT sample.
- When sample was diluted until RP pipe detector was on-scale, gamma analysis indicated minimal to no activity in sample.
- RP finally agrees there does appears to be a “mystery beta”

Determining the "mystery" beta

- 83% with a HVL of 2.5 mg/cm^2
- 11% with a HVL of 35 mg/cm^2
- 10% with an infinite HVL
- 2.5 mg/cm^2 HVL fits nicely with C-14. The 35 mg/cm^2 HVL is consistent with Kr-85, which emits a gamma and was therefore identified in the sample. The infinite HVL component was due to the gas in the space between the detector window and the attenuator surface (plus a trace of gamma from the Kr-85). I had estimated this gap to be about 0.25 cm. The observed 10% infinite half life component implies a gap of about 0.36 cm. These results are consistent with a mixture of Kr-85 & C-14 in which the activity of C-14 is about 10 times greater than the Kr-85.



Validation of C-14

- Sample from a WGDT was analyzed on liquid scintillation counter.
- Upon correctly calibrating counter for C-14, WGDT sample analysis verifies beta spectrum consistent with Carbon-14.
- RP compared the liquid scintillation spectrum of a sample of known C-14 against the spectrum of the impinger samples of the waste gas samples. The spectra were nearly identical in shape. John finds that the waste gas sample is consistent with C-14 and thinks that our initial determination that the endpoint energy was significantly below that of C-14 was due to quenching since the LS is currently calibrated only for tritium, which has a much lower energy. All evidence now indicates that the pure beta emitter in the waste gas is C-14.
- (Na-Na-Na-Na-Na!!! I told you so!!!)

Characteristics of Beta Particles

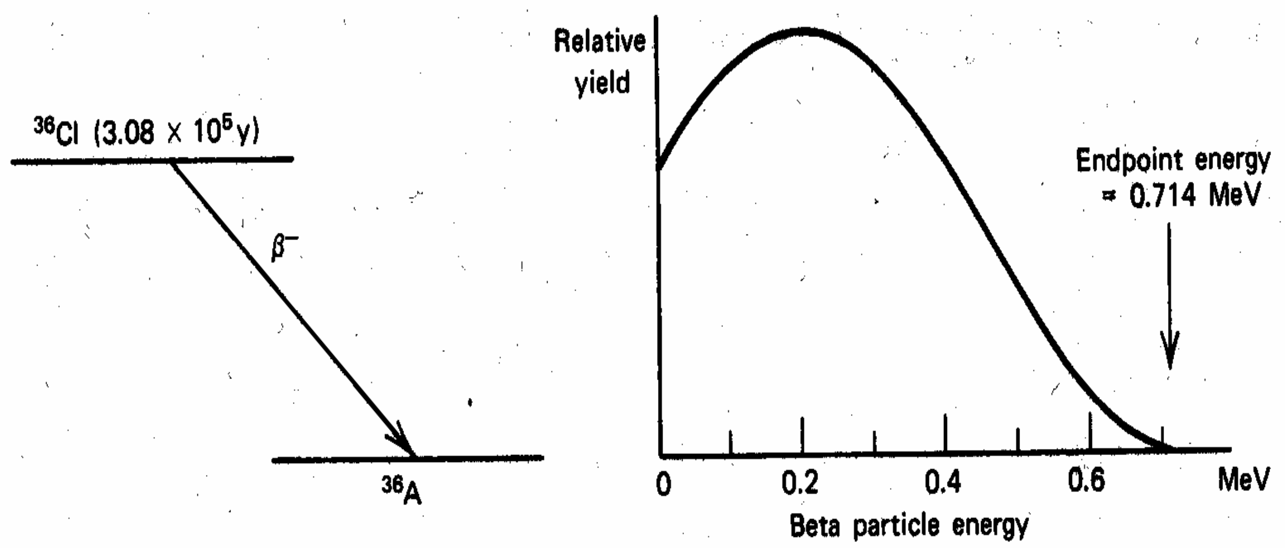
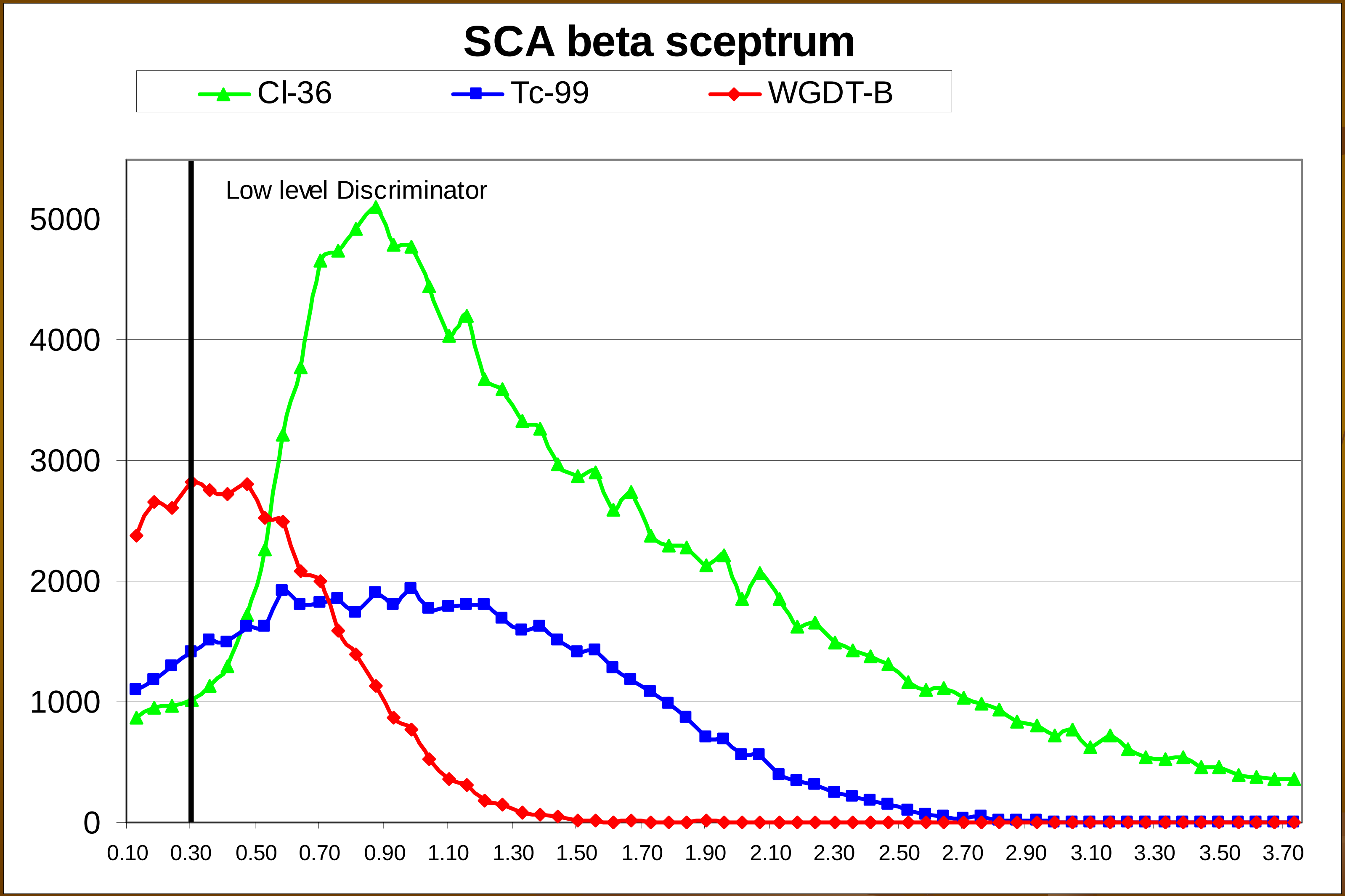


Figure 1-1 The decay scheme of ^{36}Cl and the resulting beta particle energy distribution.

- Each specific beta decay is characterized by a fixed decay energy or Q-value. The beta particle appears with an energy that varies from decay to decay and can range from zero to the "beta endpoint energy," which is numerically equal to the Q-value.

Radiation Detection and Measurement, Glenn F. Knoll Second Addition Copyright 1989.

Beta spectrums observed by 0EMF50



Carbon-14

- Pure beta emitter (undetectable via gamma spectral analysis using existing count-room laboratory analysis)
- Long-lived (5730 years half-life)
- Production Method: Air absorption within reactor coolant will produce C-14 via a neutron/proton reaction with nitrogen.

$$ {}_{0}n^{1} + {}_{7}N^{14} \rightarrow {}_{1}p^{1} + {}_{6}C^{14} $$

$$ {}_{6}C^{14} + 4H \rightarrow CH_4 $$

- Methane extracted from primary via WG System
- Tail end of Carbon-14 beta spectrum detectable by existing RD32 calibration methodology.

Finding source of C-14

This calculation determines the amount of C-14 that would be produced in the McGuire RCS from nitrogen in the form of ammonia as a result of the N-14(n,p)C-14 reaction.

$A = \lambda N \sigma \Phi t$

$\lambda = 3.835 \times 10^{-12}\ sec^{-1}$

$\Phi = 5 \times 10^{13}\ n/cm^2 \bullet sec$ (at 100% power)

$\sigma = 1.82 \times 10^{-24}\ cm^2$

t = days X 86,400 sec/day

$N = ppb\ NH_3 \times (4.91 \times 10^{20})$

Results:

The conversion for DPS to Curies is 3.7×10^{10} dps/Curie. Combining the above gives the C-14 generation at 100 % power:

C-14 (Ci) = 4.00×10^{-7} (ppb NH3) (days)

Insufficient quantity to account for observed response in WGDT.

Looking elsewhere

This calculation determines the amount of C-14 that would be produced in the McGuire Reactor Coolant System from oxygen contained in the water coolant molecules as a result of the O-17 (n,α) C-14 reaction.

$$A = \lambda N \sigma \Phi t$$

$$N = \left(702\,ft^3\right)\left(\frac{28317cm^3}{ft^3}\right)\left(\frac{0.710g}{cm^3}\right)\left(\frac{1moleWater}{18.015gWater}\right)\left(\frac{0.00038mole^{17}O}{moleWater}\right)\left(\frac{6.022X10^{23}\,atoms}{mole}\right)$$

N = 1.793 X 10^{26} λ = = 3.835 X 10^{-12} sec^{-1}

σ = 0.167 X 10^{-24} cm^2 Φ = 4 X 10^{13} n/cm^2•sec (100%)

t= days X 86,400 sec/day

Combining the above gives the C-14 activity generation at 100 % power per day as 0.0107 Ci or on an Effective Full Power Year basis as 3.91 Ci ^{14}C per EFPY.

Primary source of Carbon-14

- Production Method
  - Water will produce C-14 via a neutron/alpha reaction with O-17.

$$_{0}n^{1} + {}_{8}O^{17} \rightarrow {}_{2}\alpha^{4} + {}_{6}C^{14}$$

$$_{6}C^{14} + 4H \rightarrow CH_{4}$$

- Methane extracted from primary via Waste Gas System
- Tail end of Carbon-14 beta spectrum detectable by existing RD32 calibration methodology.

Design Function of 0EMF50

- Protect the public from excessive exposure to noble gaseous activity.
  - To exceed the ODCM limits, a concentration of noble gas exceeding full range of 0EMF50 is required.
- Assurance that laboratory analysis agrees with gaseous activity discharges to the environment.
  - Only gamma emitting isotopes are required for accounting in effluent releases. (C-14 does not require accounting)
  - Ability of 0EMF50 to accurately detect the quantity of Xe-133 or Kr-85 is considered degraded due to the masking affect from Carbon-14.

Options to address Nonconforming - degraded condition of 0EMF50

- Option 1: Do nothing
- Option 2: Sample for Carbon-14
- Option 3: Alternative Calibration
- Option 4: New Gamma monitor
- Option 5: Delete the waste gas monitor

Option 1: Do nothing

- Continue with existing correlation ratio to Xe-133, elevating alarm setpoint, and additional sampling at 0EMF50 during GWR.
- Cost: minimal, extra labor for sampling and analysis during GWR
- Drawback
  - Only effective if ratio of C-14 to noble gas remains stable. As noble gas inventory changes from fuel leaks, ratio of C-14 to noble gases within waste gas tanks changes.
  - Does not address the nonconforming/degraded condition with 0EMF50

Option 2: Quantify amount of Carbon-14

- Requires sampling and laboratory analysis to quantify amount of C-14 activity in waste gas decay tanks
- Cost: Unknown, no qualified sampling method exists for C-14. Sampling method depends on primary form of carbon molecules in waste gas tanks, but the exact form is unknown (methane, CO2, carbon monoxide).
- Drawback: C-14 does not require reporting. McGuire would be deviating from industry requirements if we begin accounting for C-14.

Option 3a: Increase the LLD to attenuate C14 beta spectrum via new primary calibration

- Cost: Estimated at $20K or higher. A primary calibration is performed with NIST traceable levels of Xe-133 and Kr-85 radioactive gases.
- Benefits: Assures 0EMF50 remains sensitive to required radioactive noble gases while eliminating Carbon-14's affect and provides traceability for all subsequent calibrations at new discrimination level.
- Drawbacks:
  - Requires use of RD32 beta gas monitor with capability to vent radioactive gas standards out the unit vent stack.
  - Complicated test that may require several shifts. Because of short half-life of Xe-133 (5 days) testing must be coordinated for procurement of gas standards and scheduling work.

Option 3b: Perform double calibration

- Trace-ability back to NIST is required for compliance with SLC TR 16.11-7.
- Perform first calibration at existing discrimination level (NIST traceability)
- Perform second calibration at elevated discrimination level (attenuate C-14).
- Estimate noble gas response by ratio of observed calibration source responses.

- Drawback
  - Exact response to radioactive noble gases is not documented, but only estimated.
  - Requires addition maintenance resources for all subsequent calibrations.

Option 4: Install a gamma sensitive monitor as 0EMF50 over the existing beta sensitive monitor

- Cost: $300,000 (similar to EMF33)
- Benefits:
  - Monitors only SLC required gamma emitting isotopes
  - Minimizes affect from Carbon-14
- Drawback:
  - Cost not supported by payback analysis
  - Year or more delay before implementing

Option 5: Delete 0EMF50

- 0EMF50 performs a redundant function with the unit vent stack noble gas monitor 1EMF36L.
- Benefits:
  - Eliminates resource dedication to support monitor
  - No longer nonconforming/degraded status
- Drawback: Significant reduction in sensitivity between 0EMF50 & 1EMF36 due to dilution affect of unit vent stack (over 1000/1)

Management's Choice

- Option 3b (alternative calibration)
- Recalibrate 0EMF50L at higher discrimination level to attenuate response from Carbon-14
- Minor Modification developed.
- OE15424: 03-032445 issued to INPO for Carbon-14 in Waste Gas System Waste Stream.

McGuire's ODCM Requirements

- Waste Gas Storage Tanks analysis for principal gamma emitters
- MNS in addition samples for Tritium & particulates for each tank
- Carbon-14 does not require analysis or reporting
- USNRC,NRR was contacted, and confirmed that there are no NRC regulatory requirements to measure or report C-14 in our effluents. In general NUREG-1301, RG 1.21, and our SLCs require reporting "principal gamma emitters", H-3, Sr-89, Sr-90, and gross alpha in gaseous effluents.

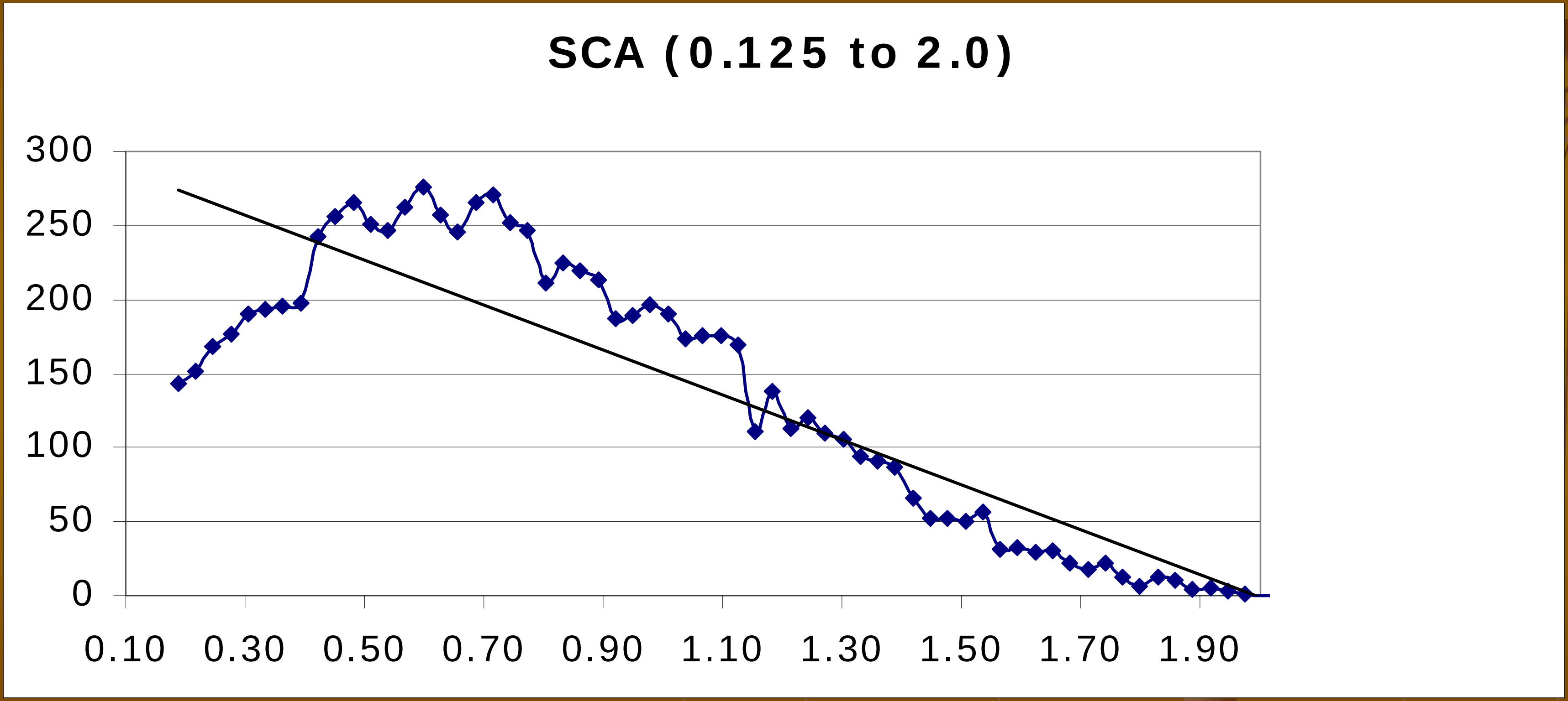
Alternative Calibration

- Alternative Calibration processed as corrective modification
- Carbon-14 source obtained.
- Calibration performed within one shift
- Cheep!
  - Carbon-14 source: $635.00
  - Maintenance 20 Man hours
  - Engineering 20 hours to write cal procedure

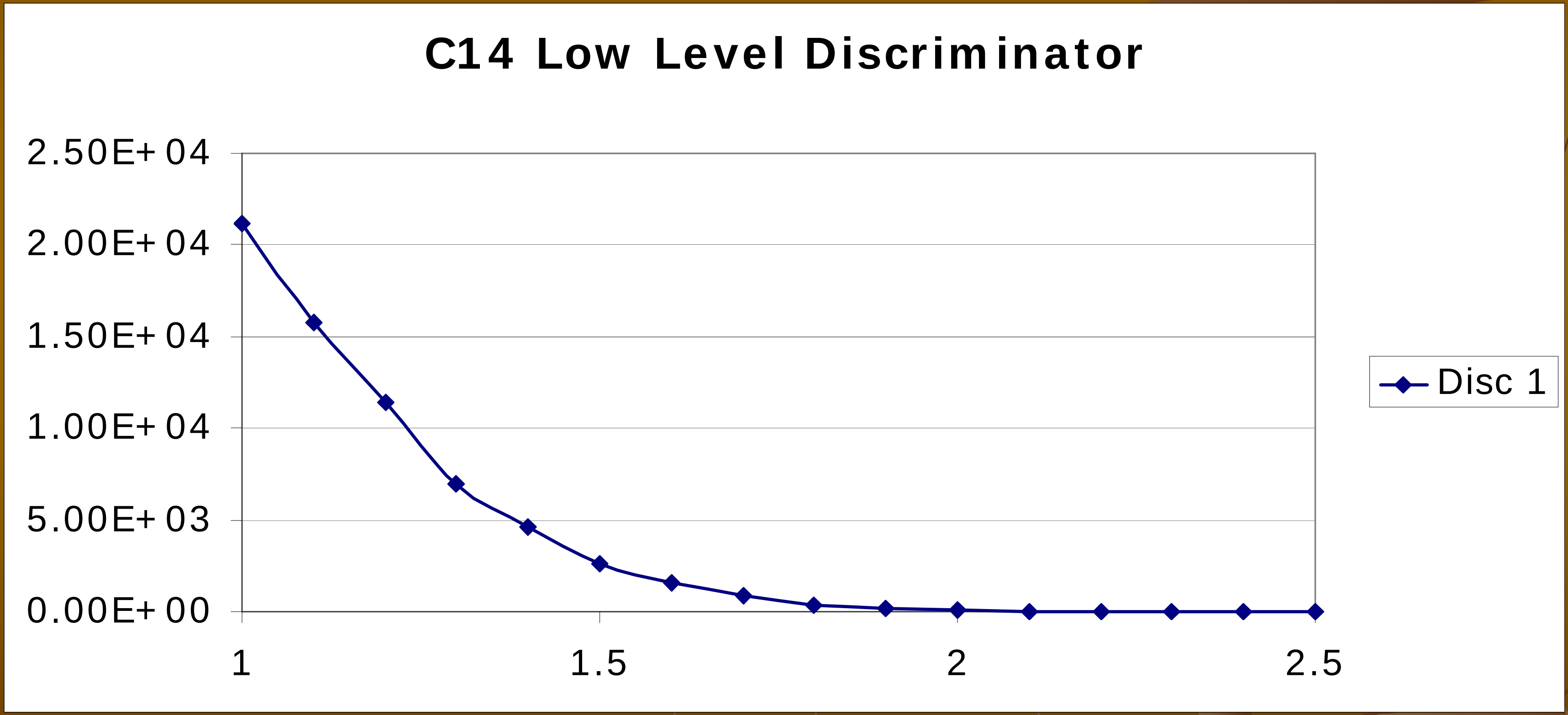
Calibration Method

- 1^{st} Cal traceable to primary calibration
  - Three differing activities of Cl-36
  - Check against Tc-99 & C-14
  - Adjust HV & LLD to obtain required countrates
- 2^{nd} Cal to attenuate Carbon-14
  - Install C-14 source
  - Increase LLD above beta end-point
  - Recount Mid level Cl-36 & Tc-99
  - Ratio response for 2^{nd} / 1^{st}
  - Tc-99 response estimates Xe-133 sensitivity reduction
  - Cl-36 response estimated Kr-85 sensitivity reduction

Analyzer Trace



Discriminator Noise Test



Alternative Calibration

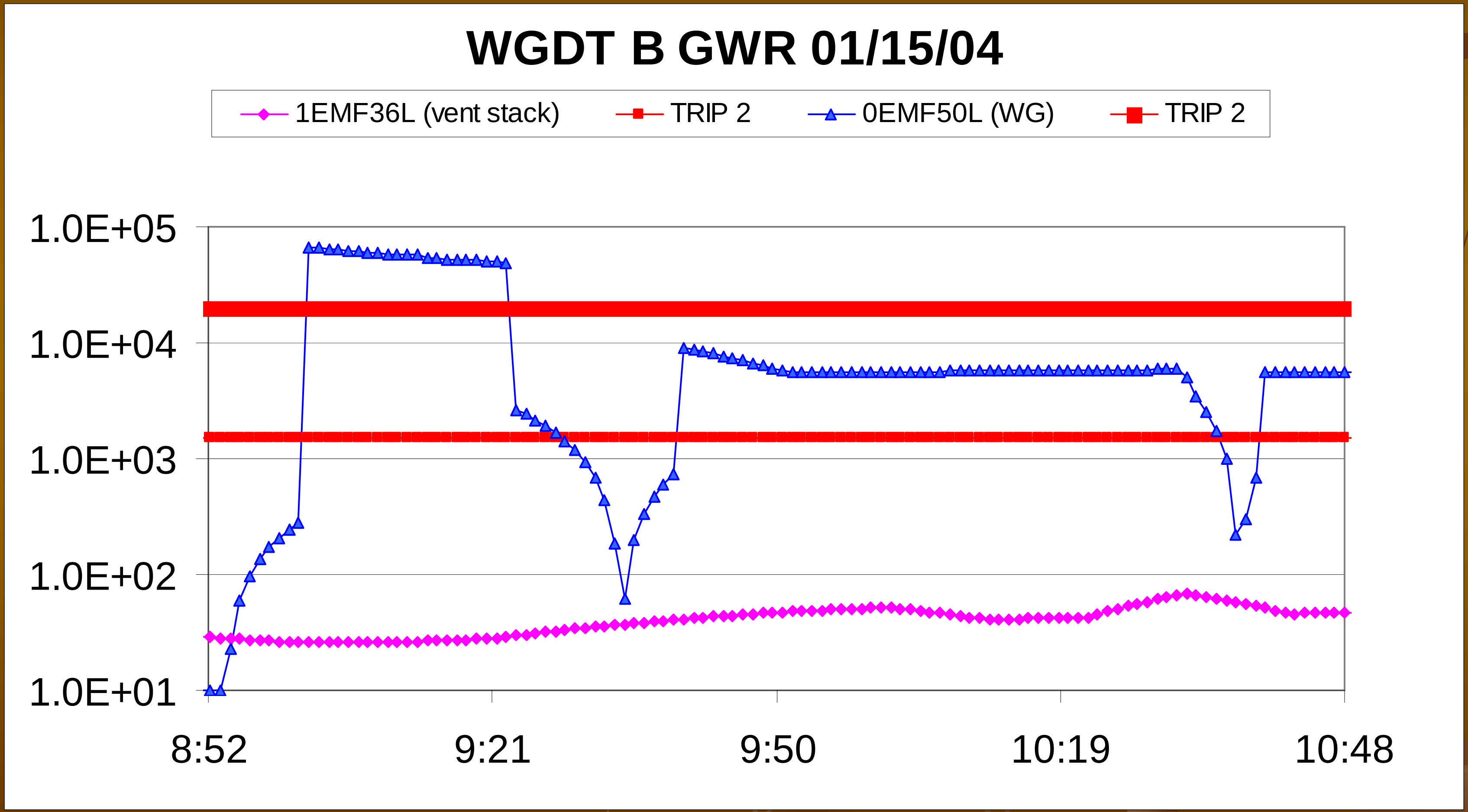
- LLD increased from 0.5 VDC to 2.1 VDC
- Cl-36 transfer source attenuated approximately 60%
- Tc-99 transfer source attenuated approximately 80%
- Xe-133 sensitivity estimated @ 5.78E6 CPM/μCi/cc
- Kr-85 sensitivity estimated @ 2.91E7 CPM/μCi/cc

Alternative Calibration Results

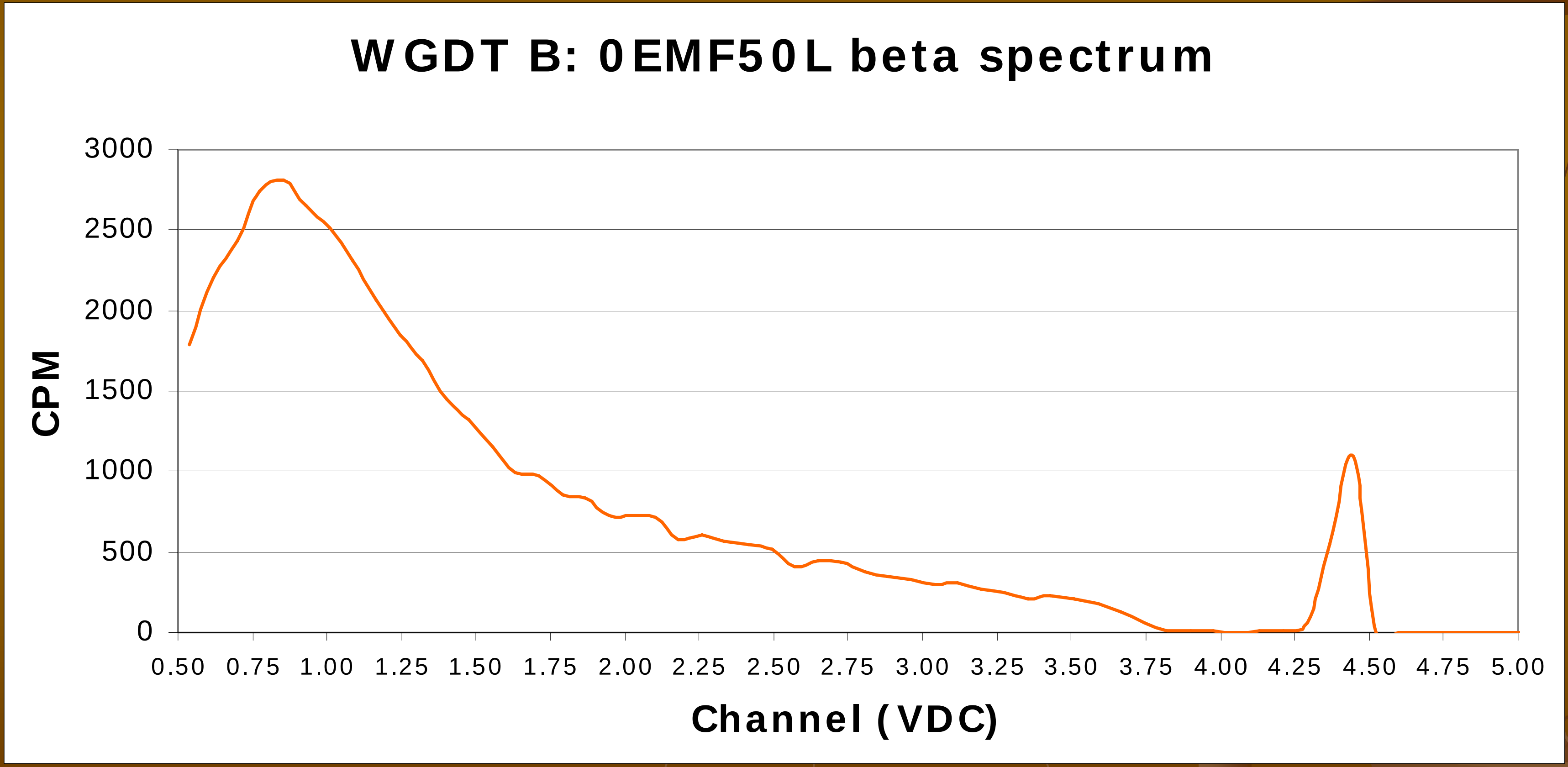
Date	WGDT	Countroom Sample Analysis: Isotope Xe-133	Kr-85	Xe-Eq	EMF50 response: Bkgd	Estimated Correlation	Estimated Noble Gas	Actual CPM
05/21/03	D	3.81E-05	1.14E-03	2.96E-03	3.00E+01	6.16E+06	9.07E+04	2.45E+05
6/2/03	F	9.93E-06	4.57E-04	1.18E-03	3.00E+01	2.45E+06	3.62E+04	1.80E+05
6/19/03	E	1.83E-05	3.96E-04	1.03E-03	3.00E+01	2.14E+06	3.16E+04	1.41E+05
11/6/03	C	1.51E-05	1.16E-02	5.82E-02	1.00E+01	3.36E+05	9.13E+05	3.03E+05
1/15/04	B	6.18E-07	2.30E-04	1.16E-03	1.00E+01	6.70E+03	1.81E+04	5.81E+03
2/8/05	B	0.00E+00	3.66E-03	1.84E-02	1.00E+01	1.07E+05	2.88E+05	9.88E+04

Date	WGDT	Estimated C-14 Count	Estimated C-14 μCi/cc	RD32 Corr Error	Correlation Error	C-14 alternative calibration Kr-85	C-14 alternative calibration Xe-133	XeEq Corr Kr85
05/21/03	D	1.54E+05	4.82E-02	170.15%	-96.02%	NA	NA	3.03
6/2/03	F	1.44E+05	4.49E-02	396.82%	-92.67%	NA	NA	3.03
6/19/03	E	1.09E+05	3.42E-02	345.53%	-93.42%	NA	NA	3.03
11/6/03	C	NA	NA	-66.77%	-9.86%	2.91E+07	5.78E+06	5.03
1/15/04	B	NA	NA	-67.96%	-13.31%	2.91E+07	5.78E+06	5.03
2/8/05	B	NA	NA	-65.70%	-7.24%	2.91E+07	5.78E+06	5.03

Additional Insurance



Waste Gas Beta Spectrum



Estimating Carbon-14 Response

Actual	RD32 =	438340	97650	=	C14 Corr
Estimated	RD32 =	288042	106506	=	C14 Corr
Error	RD32 =	52.18%	-8.32%	=	C14 Corr