10 CFR 61 RADIONUCLIDE CORRELATIONS FROM TMI-2

C.P. Deltete
Analytical Resources, Inc.
P.O. Box 6617
Wyomissing, PA 19610

P.J. Robinson
Electric Power Research Institute
3412 Hillview Avenue
Palo Alto, CA 94303

K.J. Hofstetter
GPU Nuclear
P.O. Box 480
Middletown, PA 17057

ABSTRACT

Since the implementation of 10 CFR 61 Part 61 in December 1983, a great deal of effort has been expended by individual utilities, as well as industry organizations such as the Electric Power Research Institute (EPRI), to develop 10 CFR 61 compliance programs. Much of this effort has been directed toward identifying and evaluating the use of scaling factors which could be used to correlate the presence of difficult to measure radionuclides (e.g., ^{99}Tc, ^{14}C, ^{63}Ni, ^{129}I, TRU, etc.), to more readily measured nuclides commonly found in power reactor waste streams. These efforts have generally concluded that insufficient, quantified analysis results are available to determine defensible scaling factors, largely because several of the isotopes of interest are present at such low concentrations as to make them below quantifiable limits of detection.

The 1979 accident at TMI-2, together with the resulting analysis and research conducted into the characteristics of radioactive waste from cleanup operations, has provided an opportunity to offer additional analytical data on 10 CFR 61 radionuclide correlations which may prove beneficial to the entire utility industry. In late 1986, EPRI's TMI-2 Technical Support Program conducted a detailed review and evaluation of all TMI-2 waste stream radionuclide analysis results since 1979 with the objective to compile a comprehensive data file of all such results which contain measurements of the difficult to measure radionuclides requiring identification by 10 CFR 61. An analysis of this data was then performed to determine appropriate correlations for use in the existing 10 CFR 61 compliance program at TMI-2.

INTRODUCTION

The objective of the investigation reported herein was to determine plant specific scaling factors at TMI-2 to infer the presence of difficult to measure radionuclides for which reporting under 10 CFR 61 is required. The specific radionuclides under consideration were ^{99}Tc, ^{14}C, ^{63}Ni, ^{129}I, and transuranics (TRU).

Initially, the investigation focused on liquid waste stream samples, since the most representative analyses results were available for liquid samples. Later efforts considered analyses results from solid samples. Only results applicable to liquid samples are reported in this paper. Furthermore, this paper only addresses non-transuranic nuclides which must be reported under the requirements of 10 CFR 61.

In all cases, a single ratio of the nuclide to be inferred, to a tracer nuclide, was desired. For consideration, the tracer nuclide required the following attributes, with respect to the nuclide to be inferred:

(1) is readily measured using analytical techniques at TMI-2;
(2) is prevalent in TMI-2 liquid waste streams; and
(3) has comparable transport and removal behavior.

METHODOLOGY

Data Collection

A review was made of all sample analyses on record since the March 1979 accident. Sample results were obtained and evaluated from both onsite and offsite analytical laboratory facilities. Despite the considerable number of liquid waste stream samples analyzed during the TMI-2 recovery program, very few were found to contain positive results for the isotopes under investigation. In fact, only 106 samples contained usable data, i.e., results which contained at least one ordered pair of inferred and scaling isotopes.

To evaluate the various liquid samples, the data was separated into categories representative of the source (Reactor Building Sump, Reactor Coolant System, or Auxiliary and Fuel Handling Building) and processing (Submerged Demineralizer System or EPICOR II) of major liquid streams. An identifier was used to label each sample with respect to the source of the water. The following identifiers were used:

Pt. #1	RB Sump water, influent to SDS
Pt. #2	RB Sump water, effluent from SDS
Pt. #5	RCS water, influent to SDS
Pt. #6	RCS water, effluent from SDS
Pt. #9	A&FHB water, influent to EPICOR II
Pt. #10	A&FHB water, effluent from EPICOR II

Additional liquid waste stream points were defined, however, insufficient sample results were available to perform a meaningful analysis of scaling factors.

An independent investigation was made for each of the radionuclides for which a scaling factor was desired. The available sample results in which both the desired nuclide and scaling nuclide were each identified, were combined into a data base, and decay corrected, for analysis. The accident date 3/28/79, was consistently used throughout the evaluation as a common date for referencing all sample results and nuclide ratios.

The number of sample results for each nuclide under investigation in the data base does not represent the total number of positive results for that nuclide, since the nuclide to be inferred may have been measured in a sample in which the scaling nuclide was not identified. In these cases, the sample was not included in the data base.

Data Analysis

A prerequisite for selecting a specific nuclide as the preferred scaling isotope was the establishment of a definitive relationship, or correlation, between the scaling and inferred nuclides. The existence of such a relationship was evaluated by performing a linear regression analysis of the logarithm of the decay corrected sample results. As with any linear regression analysis, the calculated correlation coefficient quantifies the "goodness of fit" of the data, with a value of 1.0 indicating a perfect linear fit of the data.

The linear correlation for each potential nuclide combination was determined using the equation $Y=mX+b$, with the understanding that the desired nuclide ratio, Y/X, is not constant over the range of X nuclide values if the Y-intercept, b, is other than zero. For the case of each of the nuclide combinations evaluated, the most appropriate linear regression yielded a non-zero Y-intercept. In order to obtain a single nuclide ratio, Y/X, a determination was made of the sample results most representative of current and future waste streams, and then these results were used to calculate an average nuclide ratio. This subjective determination considered such factors as sample origin, sample chronology, spread of sample data, continued availability of comparable liquid wastes, effects of prior processing, etc. In determining the most appropriate ratio of sample results, preference was given to more recent data rather than older historical data.

Upon selection of the final sample result ratios to be used for each radionuclide correlation, the maximum, minimum, average, and standard deviation of the linear ratios was determined. In addition, for the logarithm of the ratios, the log mean average and log mean dispersion were calculated. Consideration of these arithmetic values, with the desirability to limit the maximum-to-minimum ratio to less than 10, resulted in the final nuclide ratio to be used as a scaling factor. This is consistent with the NRC's 10 CFR 61 Branch Technical Position requirement of a decade agreement.

A propagation of error analysis was also performed for each of the radionuclide ratios determined to be most suitable for continued 10 CFR 61 use at TMI-2. The method considered both counting errors (as reported by the analytical laboratory, in absolute terms or as a percentage of the measured value) and equipment calibration errors (considered to be 4% of the sample concentration). These two fractional errors for each sample result were combined using a square root-of-the-sum-of-the-squares (SRSS) method, and the fractional error in the ratio Y/X was determined by combining the individual fractional errors in quadrature sum using the method presented in "Radiation Detection and Measurement," by Glenn F. Knoll. The resulting one sigma uncertainties are presented together with the scaling ratio.

Data Limitations

Several limitations to the data obtained at TMI-2 had to be considered while evaluating the radionuclide correlations. These limitations were primarily due to the unique nature of the TMI-2 accident, as well as the radionuclide transport and removal mechanisms observed during cleanup operations. Each of the more significant limitations in the data are summarized below.

- Production ceased as of 3/28/79 for both fission and activation products. The total inventory was fixed as of this date, so that only the release and transport from the core, and throughout the plant, affected the actual absolute activities measured, and the relative correlations between individual isotopes.
- Because of the extreme selectivity of the Submerged Demineralizer System (SDS), zeolites for removal of only certain isotopes, (i.e., ^{137}Cs, ^{90}Sr) these were not considered as prime candidates for scaling nuclides.
- Most of the sample results available at TMI-2 reflect historical waste streams which do not currently exist at TMI-2. The only liquid stream which remains representative of water typically processed is the RCS. Therefore, preference was given to RCS sample results over the RB sump or other streams.
- At the time of the 3/28/79 accident, the TMI-2 core had a very low power history (approximately 97 EFPD), which resulted in a lower inventory of fission products available for release from the core than if the plant had been in operation longer. Consequently, the concentrations of nuclides observed throughout TMI-2 liquid waste streams, in both absolute terms and relative to other nuclides, are less than could otherwise be measured in another plant, should it experience a comparable core damage transient.

- o With the limited operational history at TMI-2, there was little opportunity for the activation of corrosion and wear products, thereby resulting in low concentrations of isotopes such as ^{60}Co and ^{54}Mn.

RADIONUCLIDE ANALYSIS

Ni 63 Observations

Several radionuclides (^{55}Fe, ^{60}Co, ^{125}Sb, and ^{137}Cs), were considered to infer the presence of ^{63}Ni in the liquid waste streams under consideration. After reviewing all of the available data, ^{55}Fe, although exhibiting a linear correlation to ^{63}Ni, was rejected as the preferred scaling factor since it has been measured infrequently at TMI-2 due to its decay properties. This nuclide will, however, be considered as an alternative for inferring ^{63}Ni.

The transport and removal characteristics of ^{125}Sb are different than ^{63}Ni, and there is no chemical reason for the two nuclides to correlate. Therefore this nuclide was also rejected. In the case of ^{137}Cs, there was no linear correlation with ^{63}Ni and, therefore, no basis to develop a scaling factor. In addition, ^{137}Cs is selectively removed by the SDS system, making it a poor choice as a scaling nuclide.

The evaluation of ^{60}Co to infer the presence of ^{63}Ni resulted in two important reasons to select this nuclide as the preferred scaling nuclide for ^{63}Ni. First, a positive relationship existed between ^{63}Ni and ^{60}Co in the data base samples. Secondly, although ^{60}Co is not particularly abundant in TMI-2 liquid waste streams, this nuclide is easy to measure with installed equipment.

A total of 24 samples in which both ^{63}Ni and ^{60}Co had positive (i.e., non-LLD) values were available. Of these samples, 18 (75%) were considered useable for a detailed analysis based upon an assessment of the representativeness of each sample. The other samples were rejected based upon a qualitative assessment of the time, conditions, and source of each sample.

Figure 1 presents a logarithmic plot of the available isotopic data, in units of uCi/ml. The results of the "best fit" linear regression analysis are also displayed on this graph.

From the available data, ^{60}Co was determined to be the most technically accurate and defensible scaling factor to infer the presence of ^{63}Ni in TMI-2 liquid waste streams. Using a total of 18 data points, an average linear ratio ($^{63}Ni/^{60}Co$) of 1.65+0.07 was determined for the reference date of 3/28/79.

Tc 99 Observations

The radionuclides, ^{60}Co, ^{106}Ru, ^{125}Sb, and ^{137}Cs, were considered to infer the presence of ^{99}Tc in the liquid waste streams under consideration. After reviewing the available data, ^{106}Ru was quickly rejected since there were too few measurable data points to develop any conclusions about the relationship between ^{99}Tc and ^{106}Ru.

Likewise, there was insufficient ^{125}Sb data to infer a relationship with ^{99}Tc. In the case of ^{137}Cs, there existed a strong linear correlation with ^{99}Tc, however, measured decontamination factors across the SDS system differed by several orders of magnitude for these two nuclides. Consequently, ^{137}Cs was considered a poor choice to infer ^{99}Tc in the liquid streams at TMI-2.

The evaluation of ^{60}Co to infer the presence of ^{99}Tc resulted in two important reasons to select this nuclide as the preferred scaling nuclide for ^{99}Tc. First, a very high linear relationship existed between ^{99}Tc and ^{60}Co in the data base samples. Secondly, although ^{60}Co is not particularly abundant in TMI-2 liquid waste streams, this nuclide is easy to measure with installed equipment.

A total of 15 samples in which both ^{99}Tc and ^{60}Co had positive (i.e., non-LLD) values were available for inclusion in the data base. Of these samples, 8 (53%) were considered useable for a detailed analysis based upon an assessment of the representativeness each sample. Of the samples excluded, 6 were anlayzed by an off-site laboratory. Each of these samples had reported ^{99}Tc levels an order of magnitude lower than comparable samples analyzed by the TMI-2 laboratory, and were rejected from further evaluation since the results were not conservative in relation to onsite results. The other samples were rejected based upon a qualitative assessment of the time, conditions, and source of each sample.

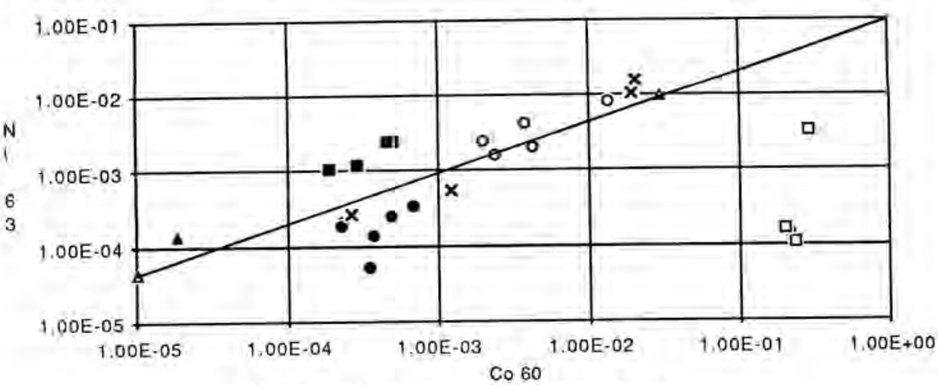

Fig. 1. Ni 63 vs. Co 60 Correlations.

Figure 2 presents a logarithmic plot of the available isotopic data in units of uCi/ml. The results of the "best fit" linear regression analysis are also displayed on this graph.

^{129}I. First, neither nuclide appeared to be removed by the SDS system, and secondly, the two nuclides have comparable chemical characteristics. Therefore, from the available data, ^{125}Sb was determined to be the best candidate evaluated for inferring the presence of ^{129}I.

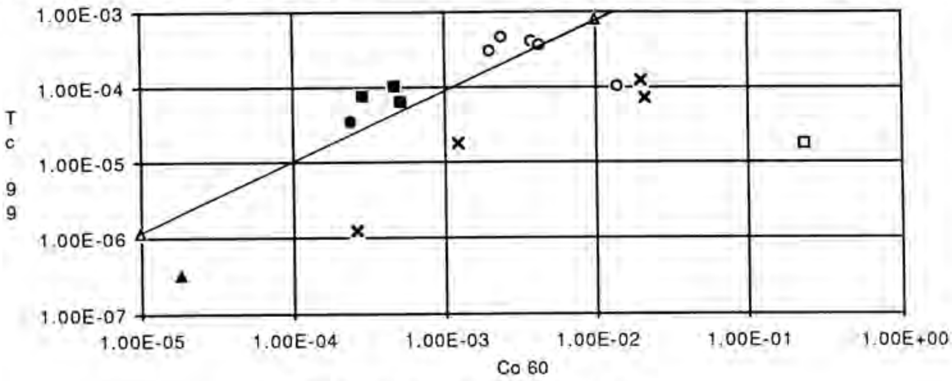

Fig. 2. Tc 99 vs. Co 60 Correlations.

From the available data, ^{60}Co was determined to be the most technically accurate and defensible scaling factor to infer the presence of ^{99}Tc in TMI-2 liquid waste streams. Using a total of 8 data points, an average linear ratio ($^{99}Tc/^{60}Co$) of 0.11+0.01 was determined for the reference date of 3/28/79.

I 129 Observations

Several different radionuclides were considered to infer the presence of ^{129}I in the liquid waste streams under consideration. The nuclides investigated were ^{60}Co, ^{106}Ru, ^{125}Sb, and ^{137}Cs. After reviewing all of the available data, ^{60}Co was rejected as the preferred scaling factor since it appeared to be independent of ^{129}I, and no definite relationship could be established.

Likewise, there was insufficient ^{106}Ru data to infer a relationship with ^{129}I. In the case of ^{137}Cs, there existed considerable sample data in which both nuclides were measured, however, there was no observable relationship between these two nuclides.

A total of 17 samples in which both ^{129}I and ^{125}Sb had positive (i.e., non-LLD) values were available for inclusion in the data base. Of these samples, 10 (59%) were considered useable for a detailed analysis based upon an assessment of the representativeness of each sample. Of the samples excluded, 6 samples originated from reactor building sump (RBS) water and were generally outside the range of the remaining RCS samples by an order of magnitude. Since nearly all RBS water has been processed through SDS, the necessity of a scaling factor responsive to this source of liquid waste stream is greatly reduced. Consequently, these samples were rejected from further evaluation in favor of sample results from the RCS, which remains the primary liquid waste stream to be processed.

Figure 3 presents a logarithmic plot of the available isotopic data in units of uCi/ml. The results of the "best fit" linear regression analysis are also displayed on this graph.

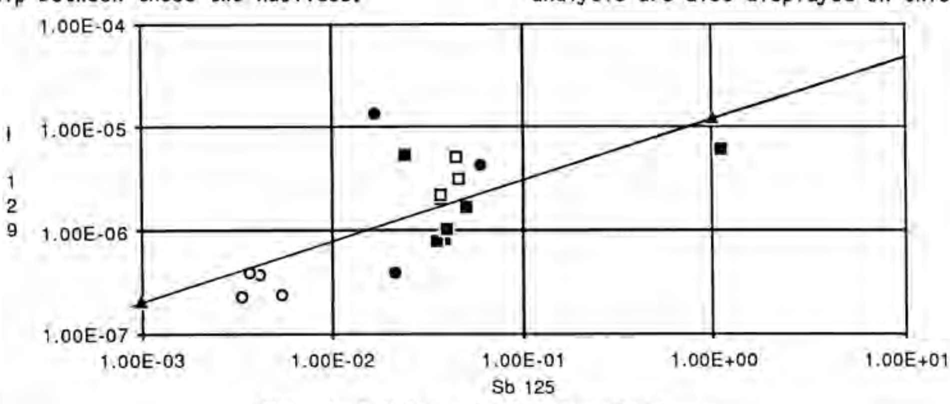

Fig. 3. I 129 vs. Sb 125 Correlations.

The evaluation of ^{125}Sb to infer the presence of ^{129}I, although yielding only a weak linear relationship between the two nuclides, resulted in two reasons to select this nuclide as the preferred scaling nuclide for

From the available data, ^{125}Sb was determined to be the most technically accurate and defensible scaling factor to infer the presence of ^{129}I in TMI-2 liquid waste streams. Using a total of 10 data points, an average linear ratio ($^{129}I/^{125}Sb$) of 6.9E-05+5E-06 was determined for the reference date of 3/28/79.

C 14 Observations

A total of 11 samples had positive (i.e., non-LLD) ^{14}C values, and were available for inclusion in the data base. None of these samples were found to be correlated with any of the other nuclides commonly measured in TMI-2 liquid waste streams.

Consequently, rather than developing a linear ratio to scale ^{14}C and another nuclide, an upper limit of observed ^{14}C values in liquids was determined which could be used to conservatively estimate the ^{14}C present in a given volume of liquid. One of the 11 samples was slightly greater (by a factor of less than 3) than the defined maximum and was the only sample not specifically used to define the maximum.

Figure 4 presents a logarithmic plot of the available ^{14}C data.

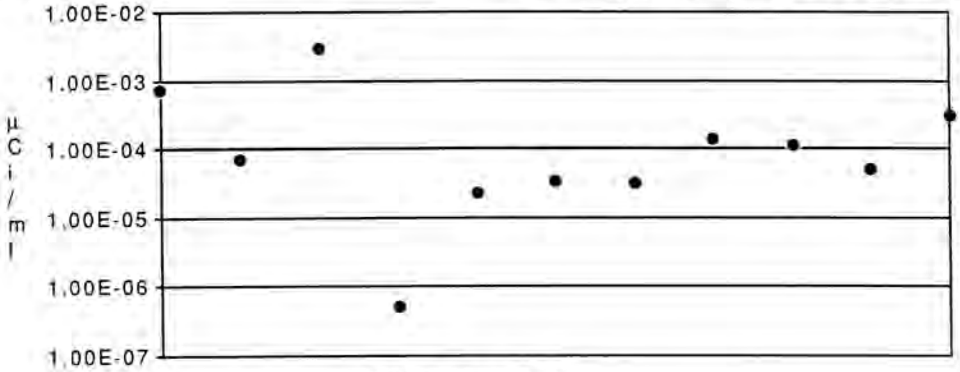

Fig. 4. C 14 Data.

Based on nearly all of the available samples, 1.0E-03 was determined to be a conservative upper bound for ^{14}C measured in liquid waste streams. Consequently, for practical application, this value was recommended for use as the concentration of ^{14}C present in liquids (or residual moisture) for the determination of the total activity of ^{14}C in a waste package.

CONCLUSIONS

Updated radionuclide correlations to infer the presence of difficult-to-measure 10 CFR 61 isotopes have been determined for liquid waste streams at TMI-2. The correlations were thoroughly evaluated using sample data which date back to 1979, and include offsite, as well as onsite, analytical laboratory results. These correlations are expected to have several benefits. First, the updated correlations are based upon redionuclides which continue to be readily measured in TMI-2 liquid waste streams. Consequently, routine measurement results should continue to be available to support the inference of the difficult-to-measure nuclides. Secondly, these new scaling factors are now available to reflect the changing liquid waste streams at TMI-2. Water from the Reactor Coolant System (RCS) is now the primary liquid waste processed, and the current knowledge of radionuclide correlations is advantageous to understand the transport and deposition of these nuclides on processing media shipped for disposal.