ASSESSING INTERNAL CONTAMINATION LEVELS FOR FISSION PRODUCT INHALATION

Final Report

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By

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Purpose

The purpose of this project was to assess a portal monitor's ability to detect internal contamination due to the inhalation of fission products. With more than 100 power plants that contribute to the energy grid in the United States, it is reasonable to consider the possibility of a nuclear power plant accident. If such an accident were to occur, fission products would be released into the atmosphere and potentially affect the health of local citizens. The main public health concern would be internal contamination via fission product inhalation. In such an emergency, it would be necessary to triage a large number of people in a relatively quick manner to determine who would need further testing and who would not need followup, at least immediate followup. The methodology of how to deal with such an accident has not been fully explored. The portal monitor—TPM 903B—was chosen as the detection system for use in this study because of its availability in Georgia and its ability to quickly determine the total body count rate resulting from gamma-ray emissions from an internally contaminated individual.

Method of Analysis

To begin to assess the ability of the portal monitor to detect fission product internal contamination, a list of fission products needed for the study had to be developed. A pressurized light water reactor (PWR) was chosen to generate a list of fission products possibly released from a nuclear power plant accident. PWR's are the most common type of nuclear reactors in the United States; so this was a reasonable assumption. The text book *Nuclear Chemical Engineering* lists fission product concentrations in spent fuel. The fission products associated with a uranium-fueled PWR running at 1000MW_e with a three year fuel life was selected. Table 8.1 in this reference lists fission product concentrations at fuel discharge and various times post fuel removal. The activities of each fission product listed at normal discharge were used as the fission product activity for the release. The fission products contained in this list can be seen in Table 1.

Table 1. Fission Products present at fuel discharge. [Source]

Ag-110	Ce-144	Eu-156	Nb-95m	Rb-86	Sb-126m	Sr-89	Te-129
Ag-110m	Cs-134	H-3	Nd-147	Rh-103m	Se-79	Sr-90	Te-129m
Ag-111	Cs-135	I-129	Pd-107	Rh-106	Sm-151	Tb-160	Xe-131m
Ba-137m	Cs-136	I-131	Pm-147	Ru-103	Sn-117m	Tc-99	Xe-133
Ba-140	Cs-137	Kr-85	Pm-148	Ru-106	Sn-119m	Te-123m	Y-90
Cd-113m	Eu-152	La-140	Pm-148m	Sb-124	Sn-123	Te-125m	Y-91
Cd-115m	Eu-154	Nb-93m	Pr-143	Sb-125	Sn-125	Te-127	Zr-93
Ce-141	Eu-155	Nb-95	Pr-144	Sb-126	Sn-126	Te-127m	Zr-95

Next, it was important to determine the release fractions of each of these fission products from a PWR core during an accident scenario. The PWR release fractions found in the U.S. Nuclear Regulatory Commission's final report number 1465, "Accident Source Terms for Light-Water Nuclear Power Plants,"

were used [2]. The fission products from Table 1 were compared with the isotopes listed in Table 2. Table 2 lists the atmospheric release fractions of each group of fission products as a function of the type of accident. The ex-vessel accident scenario was chosen because fission products would actually be released into the atmosphere. The release fractions used were considered to be fractions of core inventory and representative of an ex-vessel release. The fission products from Table 1 that were not listed on Table 2 were eliminated by drawing the conclusion that they were not released into the atmosphere, and thus were not a concern for internal contamination. Table 3 shows the first list of fission products to assess.

Table 2. PWR release fractions [Source]

Nuclide	Ex-Vessel release
I, Br	0.25
Cs, Rb	0.35
Te, Sb, Se	0.25
Ba, Sr	0.1
Ru, Rh, Pd, Mo, Tc, Co	0.0025
La, Zr, Nd, Eu, Nb, Pm, Pr, Sm, Y, Cm, Am	0.005
Ce, Pu, Np	0.005

Releases are Fractions of Core Inventory. Source: NUREG-1465

Table 3. List of released fission products

Ba-140	Eu-155	Pm-147	Sb-126m	Te-127
Ba-137m	Eu-156	Pm-148m	Sb-126	Te-129m
Ce-141	I-129	Pm-148	Se-79	Te-129
Ce-144	I-131	Pr-143	Sr-89	Y-90
Cs-134	La-140	Pr-144	Sr-90	Y-91
Cs-135	Nb-93m	Rb-86	Sm-151	Zr-93
Cs-136	Nb-95m	Rh-103m	Tc-99	Zr-95
Cs-137	Nb-95	Rh-106	Te-123m	
Eu-152	Nd-147	Sb-124	Te-125m	
Eu-154	Pd-107	Sb-125	Te-127m	

Because of the large number of fission products to be investigated and the knowledge that a select few would actually drive the internal dose, a toxicity index was developed to help eliminate those isotopes that would not be significant contributors to the dose. The toxicity index used in this research was

defined by Equation 1. Any fission products with a toxicity index of less than 1.0 x 10^{-14} were assumed to not be major dose contributors and were thus eliminated from the list of isotopes of concern.

$$T.I. = \mathrm{Re}\,leaseFraction * Activity * DoseCoefficient \qquad (1)$$

The computed toxicity indexes for all of the fission products can be seen in Table 4. The remaining fission products after toxicity elimination can be seen in Table 5.

Table 4. Fission Products and Adult Toxicity Indexes

Isotope	Toxicity Index	Isotope	Toxicity Index
Ba-140	1.40613E-12	Rb-86	1.55269E-15
Ce-141	2.56342E-13	Rh-103m	7.97755E-17
Ce-144	2.84892E-12	Rh-106	----
Cs-134	5.50953E-12	Ru-103	8.86394E-14
Cs-135	6.69703E-19	Ru-106	8.69307E-13
Cs-136	2.4819E-13	Sb-124	6.32223E-15
Cs-137/Ba-137m	1.685E-12	Sb-125	1.01241E-13
Eu-152	2.54918E-17	Sb-126m	1.03653E-18
Eu-154	1.8018E-14	Sb-126	3.8624E-16
Eu-155	2.5054E-15	Se-79	2.49899E-18
Eu-156	3.72784E-14	Sm-151	2.4278E-16
I-129	3.23587E-18	Sr-89	6.97724E-13
I-131	1.54763E-11	Sr-90	1.80269E-12
La-140	8.00781E-14	Tc-99	1.38833E-18
Nb-93m	1.26551E-20	Te-123m	5.9093E-18
Nb-95m	1.19354E-15	Te-125m	2.56289E-14
Nb-95	1.20464E-13	Te-127m	2.76598E-13
Nd-147	6.83485E-14	Te-127	2.2676E-14
Pd-107	6.67466E-23	Te-129m	9.16297E-13
Pm-147	2.42459E-14	Te-129	3.02282E-14
Pm-148m	1.07542E-14	Y-90	5.8737E-15
Pm-148	2.12236E-14	Y-91	4.0395E-13
Pr-143	1.39687E-13	Zr-93	9.16653E-19
Pr-144	9.7717E-16	Zr-95	3.18675E-13

Table 5. Fission products exceeding the toxicity index cut-off

Isotope	Toxicity Index	Isotope	Toxicity Index
Ba-140	1.40613E-12	Pr-143	1.39687E-13
Ce-141	2.56342E-13	Rh-106	----
Ce-144	2.84892E-12	Ru-103	8.86394E-14
Cs-134	5.50953E-12	Ru-106	8.69307E-13
Cs-136	2.4819E-13	Sb-125	1.01241E-13
Cs-137/Ba-137m	1.685E-12	Sr-89	6.97724E-13
Eu-154	1.8018E-14	Sr-90	1.80269E-12
Eu-156	3.72784E-14	Te-125m	2.56289E-14
I-131	1.54763E-11	Te-127m	2.76598E-13
La-140	8.00781E-14	Te-127	2.2676E-14
Nb-95	1.20464E-13	Te-129m	9.16297E-13
Nd-147	6.83485E-14	Te-129	3.02282E-14
Pm-147	2.42459E-14	Y-91	4.0395E-13
Pm-148m	1.07542E-14	Zr-95	3.18675E-13
Pm-148	2.12236E-14	----	----

Next, it was important to separate the list of fission products into groups based on the type of radiation that they emit. Group 1, as seen in Table 6, consisted of any fission product that released gamma rays. The remaining fission products were placed in Group 2, as seen in Table 7. This separation based on gamma-emission was performed because the portal monitor is only sensitive to gamma radiation. All other types of radiation emitted are not detected by the portal monitor. Fission products in Group 2 will be considered separately.

Table 6. Group 1 Fission Products

Group 1 Fission Products	
Ba-140	Nd-147
Ce-141	Nb-95
Ce-144	Pm-148
Cs-134	Pm-148m
Cs-136	Ru-103
Ba-137m/Cs-137	Sb-125
Eu-154	Te-127
Eu-156	Te-129
I-131	Te-129m
La-140	Zr-95

Table 7. Group 2 Fission Products

Group 2 Fission Products
Sr-89
Sr-90
Y-90*
Y-91
Ru-106
Pr-143
Pm-147

*Y-90 is only used in the Child Phantom

The fission products in Group 1 were assessed using gamma energy and intensity. Based on the specifications of the portal monitor, an energy and intensity cut off were determined. The energy cut off was determined to be 40keV. Thermoscientific, the maker of the TPM-903B portal monitor, reports that the cut-off energy for the detector is 60keV. In previous validation of the detector, the energy cut off of 60keV was validated. In order to be as conservative as possible, the energy cut-off of 40keV was utilized. The cut off for the gamma ray intensity was determined to 0.05%. The gamma rays that did not meet the specifications were eliminated and not evaluated in this research. Te-125m was eliminated from the list of fission products because it did not emit gamma rays that were above both of the cut offs. Te-125m does not emit any other type of radiation, so it was not placed in Group 2 and so was eliminated completely from the list of fission products of concern.

Earlier work conducted by Randahl Palmer at Georgia Tech was utilized in the Monte Carlo N-Particle Transport Code (MCNP) detector model. Palmer previously validated the MCNP detector model of the TPM-903B by using point source measurements and developed scaling factors to determine the difference between the experimental and calculated count rates.

Two MIRD phantoms were chosen for the anatomical modeling —an adipose male and child phantoms. These MIRD phantoms had been previously modified by Dr. Hertel and his graduate students to include adipose tissue, esophageal tissue, and intestinal walls. These phantoms were placed in the MCNP model of the detector oriented perpendicularly to the detector so that their anterior and posterior are facing the detector legs. The isotopes used in this research, compared to the isotopes used by Palmer, required the use of additional organs in the adipose male phantom due to different biokinetics. The additional source organs modeled in the adipose male phantom were the thyroid, the pancreas, and the spleen.

For each fission product, the gamma energies and intensities corresponding to this fission product were used to populate the source term input.

A tally of the number of source particles detected by the MCNP modeled portal monitor was performed for each organ. The sum of the counts per second per Bq of activity was computed. This sum was computed for each of the source organs modeled. To determine the internal distribution of each inhaled isotope, the Dose and Risk Calculation software (DCAL), developed by the U.S. Environmental

Protection Agency, was used. The adult age of 25 years (9125 days) was used for both the Adipose Male and Child phantom models. This was done so that the counts computed would be as conservative as possible. An equivalent dose was computed along with the compartment and source region activity. The method of intake used was inhalation. The lung absorption type—fast, medium, or slow—was determined for each isotope individually. Also, an environmental exposure input was used as opposed to an occupational exposure. The Activity Median Aerodynamic Diameter, AMAD, was assumed to be the default value of 1 micro-meter. DCAL takes these input values to determine the distribution of a particular isotope within the internal organs as a function of time. The output values are categorized by source region, i.e. lungs, thyroid, bladder, etc. These output values are reported as a fraction of unit activity.

Each source organ MCNP tally was multiplied by the DCAL output for that particular source organ. The blood DCAL was distributed as instructed by ICRP publication 72. All of the combined products of the MCNP tally and DCAL time distribution were summed for each time to determine the total body counts per second per becquerel as a function of time post inhalation. Because eventually, the fission product counts per second will be summed to determine the total counts per second associated with an inhalation of all of the released fission products, it was important to weight this count rate by the fraction of release and core activity. The fraction of release was normalized for each isotope of concern. The core activity of each isotope was normalized as well for each isotope of concern. The product of the normalized release fraction and normalized core activity was determined to be the relative abundance of the fission product in the inhalation. This relative abundance was multiplied by all of the total counts per second per becquerel for each time post inhalation. This product would give the number of counts per second for each individual fission product. The counts per second per becquerel were determined by dividing the expected counts per second per becquerel by the previously determined scaling factor. This scaling factor was taken to be the average of the scaling factors computed by Palmer. Then, using the corresponding dose coefficient in rem/Bq and the conversion between rem and Sv for 250mSv, the total body counts per second per 250mSv was determined for various times post fission product inhalation. Because the dose coefficient takes into account all types of radiation, it was determined that all radiation emitted by fission products in Group 1 was accounted for.

After determining the total body counts per second per 250mSv for each gamma-emitting fission product, it was important to sum the expected counts per second per 250mSv for each fission product. The sum was completed for various times post fission product inhalation. The sum of each individual fission product's CPS per 250mSv as registered by the detector is equivalent to the number of counts per second per 250mSv that will be registered by the detector for an inhalation of all of the gamma-emitting fission products.

Now it was important to consider the fission product dose contributions for those fission products in Group 2. The radiation emitted by these fission products will not be registered by the detector; however, since it will be contributing to the dose, it should be taken into account when determining the counts per second per 250mSv that corresponds to the trigger level. First, all fission products in Group 2 were analyzed to determine the kinds of radiation emitted. All members of Group 2 are beta emitters. In order to account for the beta radiation, a ratio of the dose coefficients—weighted based on relative

isotope abundance—was computed. This ratio was of the weighted dose coefficients for fission products in Group 1 divided by the sum of the weighted dose coefficients for fission products in Group 1 and the weighted dose coefficients for fission products in Group 2. This ratio was then multiplied by the total body CPS per 250mSv determined previously for the Group 1 fission products. The ratio is shown in Equation 2.

$$Ratio = \frac{\sum DC_i^1 * R_i^1 * A_i^1}{\sum DC_i^1 * R_i^1 * A_i^1 + \sum DC_i^2 * R_i^2 * A_i^2} \quad (2)$$

In Equation 2, DC represents the dose coefficients, R represents the corresponding normalized release fraction, and A represents the normalized activity of the fission product, with the superscripts describing the group of the fission product. The final result was a trigger level, in counts per second, that corresponded to a committed effected dose of 250mSv for all contributing types of radiation.

Assumptions

Several assumptions were made in the completion of this research. The validity of these assumptions relies on the underlying principle that the results of this research will only be used as a first cut screening tool. No conclusions can be drawn on the precise amount of dose absorbed by an individual based on the detector reading. The results of this research are meant to simply identify individuals who need further, more accurate methods of testing to determine the individual's dose.

The first assumption made was that the reactor responsible for emitting the fission products is a uranium-fueled Pressurized Water Reactor (PWR), operating at 1000MW_e with a fuel life of three years. While there is no way to be certain specifically what kind of reactor could potentially release fission products, assuming that it is a PWR is a valid assumption because the majority of reactors operating in the United States are PWR's. Additionally, not all reactors are fueled with Uranium.

The second assumption made was that the fission product concentrations released from the reactor were equivalent to their relative concentrations at the end of the fuel cycle and behaved in the manner predicted by the release fractions found in NUREG document 1465. Additionally, the assumption was made that the reactor release of fission products could be characterized as an ex-vessel release which means the fission products are escaping into the open atmosphere. Also, it was assumed that all fission products released into the atmosphere were inhaled at once in the relative concentrations predicted by their core activity and ex-vessel release fraction.

Analysis

For each fission product in Group 1, the gamma ray intensities and energies were exported from the program RadToolBox. The energy cut off for the TPM-903B is approximately 60keV. It was determined

to include only gamma energies that were greater than 40keV when populating the source definition for each fission product. The intensity cut off was reasonably determined to be 0.05%.

The source definitions for each fission product in Group 1 were written into the adipose male phantom MCNP code. A list of the organs of concern was made encompassing all organs affected by all of the fission products. This list was used to modify the adiopose male phantom file to include each organ of interest. This way the only difference between each input file for each fission product was the energy and intensity spectrum, which is specific to the isotope of interest. A master Child phantom input file was constructed in the same manner. A unit source was placed in each organ of interest and a pulse-height tally was performed on each organ of interest. All 20 input files were run and tallies for each source organ were computed. These tallies were then used in conjunction with the Dose and Risk Calculation software output for each coinciding fission product to determine which source organs were of interest specific to each isotope. The distributions of the fission products in the organs of interest for each isotope were then multiplied by the tallies computed in MCNP. For each time post inhalation, the total body count sum was computed by summing over all of the organs of interest. This would give the number of counts per becquerel of fission product if all of the fission products were released in the same concentrations.

The weighting factor for each fission product was computed as the product of the normalized release fraction and the normalized activity. The total body count sum was multiplied by the weighting factor. The scaling factor relating the MCNP model of the detector to the actual detector for the TPM-903B was previously determined for a variety of radioisotopes. The average of the scaling factors was computed to be 0.958. This scaling factor was used for each fission product to determine the actual detector response per bequerel of activity. RadToolBox provided the dose coefficients for each fission product. The dose coefficients for each fission product were used to determine the counts per second (CPS) per 250mSv over a period of 30 days.

Once the total body counts per second per 250mSv was computed for each fission product individually, the total body CPS per 250mSv were summed to determined the total body CPS per 250mSv from the gamma-emitting fission product contribution.

Because not all of the fission products are gamma-emitting, it is important to determine a way to take into account the other kinds of radiation. The fission products in Group 2 are all beta-emitting fission products. The dose coefficients for each Group 2 fission product were determined using RadToolBox. The ratio computed per Equation 2 was 0.881 for the adipose male. The ratio computed per Equation 2 was 0.893 for the child. These ratios were then multiplied by the total total body CPS per 250mSv from the gamma-emitting fission product contribution resulting in the total body CPS per 250mSv representing all forms of contributing radiation.

Results

The trigger levels corresponding to a committed effective dose of 250mSv over times post inhalation are displayed in Tables 8 and 9 for the Adipose Male and Child phantoms, respectively.

Table 8. Trigger levels for the Adipose Male

Days following exposure		Total Body Count
		cps per 250 mSv
	0.25	6.94E+05
	0.5	3.88E+05
	1	1.48E+05
	2	7.42E+04
	3	6.69E+04
	4	6.38E+04
	5	6.11E+04
	6	5.86E+04
	7	5.61E+04
	8	5.36E+04
	9	5.13E+04
	10	4.90E+04
	20	3.00E+04
	30	1.90E+04

Table 9. Trigger Levels for the Child

Days following exposure		Total Body Count
		cps per 250 mSv
	0.25	3.93E+05
	0.5	2.29E+05
	1	1.01E+05
	2	6.02E+04
	3	5.54E+04
	4	5.30E+04
	5	5.09E+04
	6	4.90E+04
	7	4.72E+04
	8	4.54E+04
	9	4.37E+04
	10	4.21E+04
	20	2.93E+04
	30	2.18E+04

Graphical representations of these trigger levels can be seen in Figures 1 and 2 for the adipose male and child, respectively.

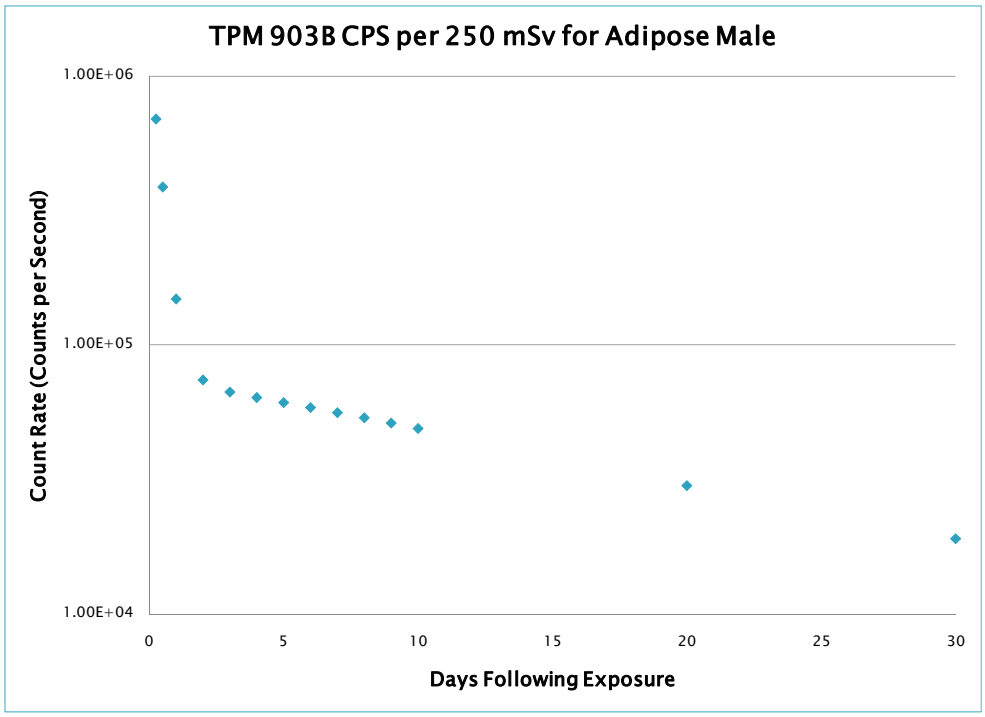

Figure 1. Adipose male trigger levels as a function of time post inhalation

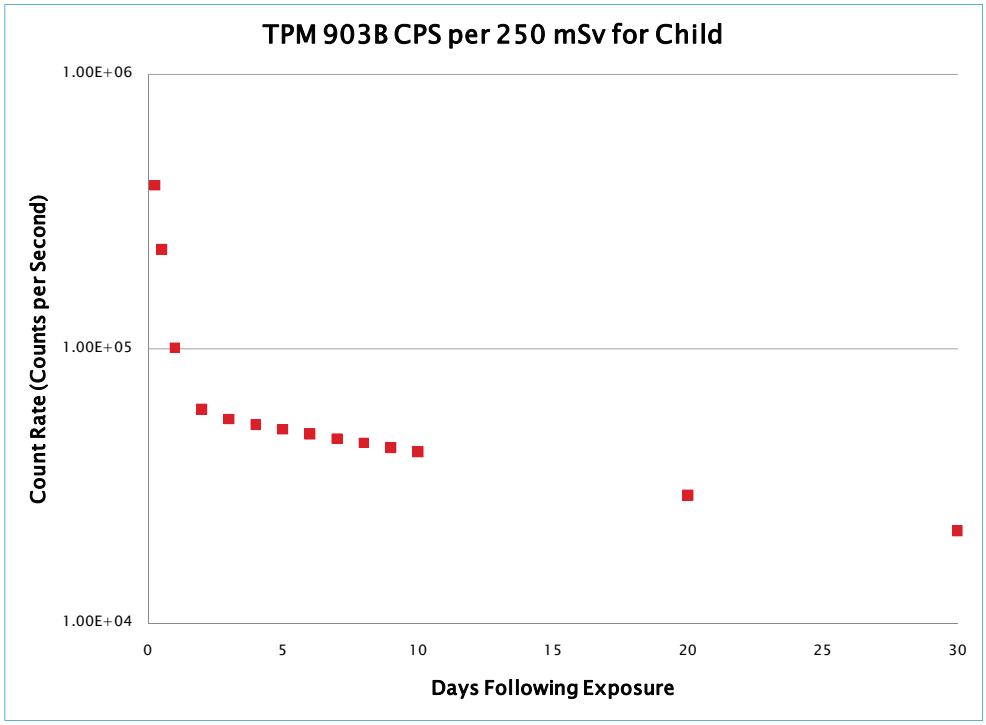

Figure 2. Child trigger levels as a function of time post inhalation

Detailed procedure sheets can be found in Appendix A.

Conclusion

The TPM-903B portal monitor was determined to be a good response tool in triaging members of the public exposed to an ex-vessel release of fission products. The counts per second for these trigger levels are far above the lower limit of detection for post inhalation times of up to 30 days. There were only 2 phantom types used in this research. It might be worthwhile to investigate fission product inhalation using a wider variety of phantom types.

References

[1] Soffer, L., et al. Accident Source Terms for Light-Water Nuclear Power Plants. Final Report. Washington: U.S. Nuclear Regulatory Commission, 1995.

[2] Computerized Radiological Risk Investigation System for Assessing Doses and Health Risks from Atmospheric Releases of Radionuclides. Oak Ridge, Tennessee: Oak Ridge National Laboratory.

[3] Palmer, R.; Hertel, N. Simulation and Testing of Radiation Detection Instruments as Monitors of Internal Contamination Levels. Atlanta, Ga: Georiga Institute of Technology, 2010.

Appendix A

TPM-903B (Adipose Male with Inhaled Fission Products)
Basic Operation

- Attach aluminum feet to bottom of the PVC pipes.
- String cables through top PVC pipe and place on top of the two sides.
- Connect the cables to the bottom side of the display unit.
- Connect the portal monitor to AC power or D-cell batteries and turn on.
- The background will automatically be acquired once the portal monitor is turned on. Record the background value.
- Calibrate the portal monitor, following the instructions listed in the manual.
- Set the background count parameter to 20 seconds and turn off the occupation alarm by setting the nsigma parameter to n=99.
- Have the victims form a line at least 15 feet from the portal monitor.
- Have each victim stand sideways inside the center of the portal monitor, facing the display unit.
- Once victim enters the portal monitor manually set the mode to background mode by pushing the # button. After the victim has been in the portal monitor for approximately one minute write down the count rate.
- After a count rate has been obtained, subtract the background count from the number on the display and compare the result to the proper trigger level.

Trigger Levels if Inhaled

Time (days)	Fission Products (cps)
0.25	6.94E+05
0.5	3.88E+05
1	1.48E+05
2	7.42E+04
3	6.69E+04
4	6.38E+04
5	6.11E+04
6	5.86E+04
7	5.61E+04
10	4.90E+04
20	3.00E+04
30	1.90E+04

TPM-903B (Child with Inhaled Fission Products)
Basic Operation

- Attach aluminum feet to bottom of the PVC pipes.
- String cables through top PVC pipe and place on top of the two sides.
- Connect the cables to the bottom side of the display unit.
- Connect the portal monitor to AC power or D-cell batteries and turn on.
- The background will automatically be acquired once the portal monitor is turned on. Record the background value.
- Calibrate the portal monitor, following the instructions listed in the manual.
- Set the background count parameter to 20 seconds and turn off the occupation alarm by setting the nsigma parameter to n=99.
- Have the victims form a line at least 15 feet from the portal monitor.
- Have each victim stand sideways inside the center of the portal monitor, facing the display unit.
- Once victim enters the portal monitor manually set the mode to background mode by pushing the # button. After the victim has been in the portal monitor for approximately one minute write down the count rate.
- After a count rate has been obtained, subtract the background count from the number on the display and compare the result to the proper trigger level.

Trigger Levels if Inhaled

Time (days)	Fission Products (cps)
0.25	3.93E+05
0.5	2.29E+05
1	1.01E+05
2	6.02E+04
3	5.54E+04
4	5.30E+04
5	5.09E+04
6	4.90E+04
7	4.72E+04
10	4.21E+04
20	2.93E+04
30	2.18E+04