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Jing-Jy Cheng and Charley Yu
Argonne National Laboratory

W. Alexander Williams and William Murphie
U.S. Department of Energy

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VALIDATION OF THE RESRAD-RECYCLE COMPUTER CODE*

Jing-Jy Cheng and Charley Yu
Argonne National Laboratory
9700 S. Cass Ave., Argonne, IL 60439

W. Alexander Williams and William Murphie
U.S. Department of Energy
19901 Germantown Road, Germantown, MD 20874-1290

ABSTRACT

The RESRAD-RECYCLE computer code was developed by Argonne National Laboratory under the sponsorship of the U.S. Department of Energy. It was designed to analyze potential radiation exposures resulting from the reuse and recycling of radioactively contaminated scrap metal and equipment. It was one of two codes selected in an international model validation study concerning recycling of radioactively contaminated metals. In the validation study, dose measurements at various stages of melting a spent nuclear fuel rack at Studsvik RadWaste AB, Sweden, were collected and compared with modeling results. The comparison shows that the RESRAD-RECYCLE results agree fairly well with the measurement data. Among the scenarios considered, dose results and measurement data agree within a factor of 6. Discrepancies may be explained by the geometrical limitation of the RESRAD-RECYCLE's external exposure model, the dynamic nature of the recycling activities, and inaccuracy in the input parameter values used in dose calculations.

INTRODUCTION

The RESRAD-RECYCLE computer code (1) was developed by Argonne National Laboratory (Argonne), under the sponsorship of the U.S. Department of Energy (DOE), to analyze potential radiation doses resulting from reuse and recycling of radioactively contaminated equipment and scrap metals. RESRAD-RECYCLE has a user-friendly interface and provides enough flexibility to simulate a variety of exposure conditions. The methodology used in RESRAD-RECYCLE is similar to that developed separately by the U.S. Nuclear Regulatory Commission (NRC) (2) and by the U.S. Environmental Protection Agency (EPA) (3). However, the NRC's and EPA's methodologies were implemented in spreadsheets and computer code that were not designed for general public use and that lack a user-friendly interface and the degree of flexibility incorporated into RESRAD-RECYCLE.

In 1999, an international effort was initiated by the Swedish Radiation Protection Institute, with the cooperation of the DOE and the Institut de Protection et de Sûreté Nucléaire (IPSN) of France, to compare dose results calculated with various computer programs with actual dose data measured for operation and handling activities involving radioactively contaminated scrap metal in a recycling facility. Other participants in the project included: (1) Studsvik RadWaste AB, Sweden, which has a facility that melts radioactively contaminated metal from the nuclear industry; (2) Akers AB, Sweden, a commercial manufacturer of steel and nonferrous metal rolls that accepts metal ingots from Studsvik; (3) Belgoprocess, Belgium, a company that is

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developing a process for milling very low-level radioactively contaminated concrete for recycling and was participating as an observer in this project; (4) Menon Consulting AB, Sweden, a consulting firm that managed and coordinated this validation project; and (5) Argonne, the developer of the RESRAD-RECYCLE code. For this validation project, two computer programs, RESRAD-RECYCLE and CERISE (4), were used to calculate radiation doses. Studsvik Radwaste AB collected the actual radiological dose data used for model comparison in this validation exercise.

Studsvik Radwaste AB, Sweden, has a facility that melts radiologically contaminated metal scrap. It has melted some 3,500 tons of carbon steel, stainless steel, brass, and aluminum since 1987. Slag and filtered dust generated from melting are conditioned and treated. Ingots are stored in the facility until radioactivity in them has decayed to a level below the limits set by the Swedish Radiation Protection Institute. The ingots are then released to commercial melting facilities, such as Akers AB, and mixed with uncontaminated scrap metal.

This paper describes Argonne's participation in this international validation project. It starts with discussion on the scrap metal selected for melting, which is then followed by discussions on the exposure scenarios, mass partitioning factors, radionuclide partitioning factors, exposure pathways, source geometry and exposure parameters, and dose conversion factors used in RESRAD-RECYCLE calculations. After that, the calculation results are presented and compared with the measured data. Finally, conclusions are drawn and possible explanations for the discrepancies are provided.

SCRAP METAL SELECTED

This validation project involved evaluating doses from the melting of a stainless steel nuclear fuel rack from the Swedish Ringhals Nuclear Power Plant. The rack was shipped to the Studsvik facility in a 20-foot-long container. The maximum dose rate measured on the outside of the container was 0.2 mSv/h. Radionuclide-specific measurements (made from outside the package) indicated an average radioactivity concentration of 109 Bq/g, mostly Co-60. Other radionuclides included Sb-125, Cs-134, Cs-137, and Eu-154.

SCENARIO DESCRIPTION

The fuel rack was melted in Studsvik and actual radiation dose rates were measured for each of the various operations involved in the process. Dust from the ventilation system and slag from the melting operation were also sampled and analyzed. To facilitate dose calculations, the geometry of the radiation source, exposure distance between the source and the worker, and the time span of each operation were developed on the basis of the real operations.

Eight exposure scenarios were developed to account for the various operations conducted during the melting process. These eight scenarios evaluated doses to the following work groups: (1) scrap truck drivers, (2) scrap unloaders, (3) scrap cutters/sorters, (4) furnace operators, (5) ingot handlers A, (6) ingot handlers B, (7) ingot fork driver, and (8) slag handler.

The truck drivers transported the fuel rack from Ringhals to Studsvik. The scrap unloaders unloaded the fuel rack from the transport vehicle. The scrap cutters/sorters disassembled the fuel rack and cut it into smaller pieces that could be fed to the furnace. The furnace operators loaded the fuel rack to the furnace and operated the furnace. After the ingot melt was poured into vertical molds, the ingot handlers A moved the molds away for cooling. After cooling, ingot handlers B removed the solid ingots from molds. The solid ingots were then put on wooden pallets in a

storage area by the ingot fork driver. During melting of the fuel rack, slag from the melt surface was removed by the slag handler with a special tool and was put in a metallic box in the same area for further processing.

MASS PARTITIONING FACTORS

The term “mass partitioning factor” refers to the fraction of throughput mass in the melting process that gets into the melting product. An ingot is the main product of the melting process. The mass partitioning of the slag is affected by the mass partitioning of the ingot. Dust and off-gas generated by the furnace are collected in the baghouse. Some of the baghouse contents may be released to the atmosphere through an emission stack.

Mass partitioning factors used in RESRAD-RECYCLE calculations were developed from the measured masses of the ingot product, the slag product, and filter dust (listed in Table I) and with application of the principle of mass conservation. The weight of the cutting swarf (2 kg) was neglected because (1) its mass was very small compared with the mass of the fuel rack (the initial throughput) (> 3,355 kg), and (2) the swarf is not a product of the melting process. Neglecting the cutting swarf had very little effect on the values of the partitioning factors, which were calculated as 98.35% for ingot, 1.64% for slag, and 0.01% for baghouse filter.

Table I. Measured Mass and Radionuclide Inventories

Inventories	Ingot	Cutting Swarf	Slag	Filter Dust	Total
Mass (kg)	3300	2	55	0.2	3357.2
Radionuclides (MBq)					
Co-60	518	0.34	8.7	0.02	527
Sb-125	12.2	0.01	0.08	0.0006	12.3
Cs-134	– ^a	–	0.09	–	0.09
Cs-137	–	–	36.3	0.0015	36.3
Eu-154	–	–	0.02	–	0.02

^a A dash (–) indicates activity was too low to be detected.

RADIONUCLIDE PARTITIONING FACTORS

During the melting process, radionuclides in the scrap metal could partition to one of the three products: ingot, slag, or dust particles. Radionuclides with low boiling points, typically concentrate in dust particles; those that oxidize easily tend to concentrate in slag. Distribution of radionuclides generally depends on chemical properties of the radionuclides, metallurgical composition of the scrap metal, presence of slag-forming substances added to the melt, melting temperature, and melting method (i.e., the type of furnace).

Radionuclide partitioning factors used for RESRAD-RECYCLE calculations were developed on the basis of the measured activity contents in ingot, slag, and dust filters (listed in Table I). Like the calculations for mass partitioning factors, the measured radionuclide contents in the cutting swarf were neglected and subtracted from the total contents. The total radionuclide contents, after the subtraction, were used to normalize the radionuclide contents in each of the melting products to obtain the radionuclide partitioning factors for that product. The calculated radionuclide partitioning factors are listed in Table II.

Table II. Radionuclide Partitioning Factors

Radionuclides	Ingot	Slag	Filter Dust
Co-60	9.83E-01	1.65E-02	1.00E-04
Sb-125	9.93E-01	6.50E-03	1.00E-04
Cs-134	0	1.00	0
Cs-137	0	1.00	4.00E-05
Eu-154	0	1.00	0

Radionuclide concentrations in the fuel rack were calculated from information on total mass and amount of radionuclides in the three melting products. Concentrations in the fuel rack were calculated as 157 Bq/g for Co-60, 3.66 Bq/g for Sb-125, 0.02682 Bq/g for Cs-134, 10.819 Bq/g for Cs-137, and 0.00596 Bq/g for Eu-154.

EXPOSURE PATHWAYS

The RESRAD-RECYCLE code evaluates three exposure pathways — external radiation, inhalation, and ingestion. To model external radiation exposure, the radiation source that the workers handled is simulated by a full or half cylinder with dimensions (radius and thickness) representing the source geometry. The RESRAD-RECYCLE code calculates an external dose conversion factor for each scenario on the basis of the dimensions of the cylindrical source, the exposure distance, and the density of the source material. Attenuation of external radiation can also be considered by specifying the material type, density, and thickness of shielding material located between the radiation source and the exposed workers.

The inhalation pathway considers radiation exposures resulting from inhalation of airborne dust particles. This pathway is evaluated for activities with the potential of causing suspension of source particles. An inhalation rate of 1.2 m³/h and a respirable fraction of 0.1 were assumed in the dose calculations. A dust loading factor, which is the concentration of airborne dust particles, is used to represent the air quality in the work place. Concentrations of radionuclides in the airborne dust particles are assumed to be the same as those in the source material, with one exception. For the scrap truck driver, scrap unloader, and scrap cutter/deliver, the source material is the scrap metal itself. For the ingot handlers and ingot fork driver, the source material is the ingot product. However, radiation exposures from the inhalation pathway was neglected because little dust loading occurred during the operations. For the slag handler, the source material is the slag product. For the furnace operator, dust particles are considered to originate from the melt

mixture inside the furnace. However, only volatile components of the mixture would become airborne, and a fraction of them would eventually be collected in the baghouse. Therefore, concentrations of radionuclides in the airborne dust particles are assumed to be the same as those calculated for baghouse dust particles.

For the ingestion pathway, it is assumed that the worker would incidentally ingest the dust particles that deposit on his hands or on the surface of surrounding materials with which his hands come in contact. The concentrations of radionuclides in the dust particles are assumed to be the same as those used for the inhalation pathway. In addition to incidental ingestion, inhalation could also contribute to the ingestion dose. Dust particles that are larger than the respirable size are assumed to enter the gastrointestinal tract after they are inhaled. Once these particles are absorbed into the blood stream, they would result in internal radiation exposure, and the resulting radiation doses are attributed to the ingestion pathway. Because dust loading was neglected for the ingot handlers and ingot driver scenarios, no ingestion exposure was assumed for these scenarios as well. For the other scenarios, an ingestion rate of 0.00625 g/h was assumed for the dose calculations.

SOURCE GEOMETRIES AND EXPOSURE PARAMETERS

Table III lists the source geometry and exposure parameters used by RESRAD-RECYCLE for dose calculations. These parameters were developed together by the participants of this project to reflect the actual working condition. Along with the scenario names, the dosimeter codes used in dose measurements are also listed.

For the scrap truck driver scenario, the external radiation was considered to be attenuated by the truck cab, which had a density of 7.86 g/cm³ and a thickness of 0.3 cm. During the handling of ingot melt, ingot handlers A were shielded from radiation by the molds, which had a density of 7.86 g/cm³ and a thickness of 8 cm. The slag handler was shielded by the slag container, which had a density of 7.86 g/cm³ and a thickness of 1.2 cm. The ingot fork driver took five ingots to storage at a time; therefore, dimensions of the radiation source were developed to consider potential radiation exposure from the five ingots.

The density of the fuel rack was estimated as 0.126 g/cm³, which is the ratio between the fuel rack mass to the fuel rack volume. This value is far from the theoretical value for a steel material and could result in some degree of inaccuracy in dose calculations.

DOSE CONVERSION FACTORS

The external radiation model in RESRAD-RECYCLE is based on the dose conversion factors given in the EPA's Federal Guidance Report (FGR) No. 12 (5) and the point kernel method. This model calculates an external dose conversion factor for each radionuclide and each scenario on the basis of a 1 Bq/g concentration in the source material. The dose conversion factor for a particular radionuclide is then multiplied by the source concentration and exposure duration to obtain the radiation dose for the external exposure pathway. Table IV lists the external dose conversion factors.

The internal dose conversion factors used by RESRAD-RECYCLE were obtained from the EPA's Federal Guidance Report No. 11 (6). For some radionuclides, more than one value is listed in the EPA report to account for different chemical forms. In that case, the most conservative value was used in the calculations to obtain conservative dose results. The RESRAD-RECYCLE inhalation and ingestion dose conversion factors are listed in Table V.

Table III. Source Geometry and Exposure Parameters

Worker Scenario	Dose Codes	Source Geometry	Mass (t)	Density (g/cm ³)	Thickness (cm)	Radius (cm)	Distance (cm)	Time (h)	Source Material ^a for the External Pathway	Source Material ^a for the Internal Pathways	Dust Loading (g/m ³)	Number of Workers
Scrap truck driver ^b	610	1 full cylinder	3.3	0.126	400	145	150	0.15	Scrap	Scrap	1 x 10 ⁻⁴	2
Scrap unloader	611	1 full cylinder	3.3	0.126	400	145	50	3.7	Scrap	Scrap	1 x 10 ⁻⁴	2
Scrap cutter/sorter	612	1 full cylinder	3.3	0.126	400	145	30	9.95	Scrap	Scrap	1 x 10 ⁻³	2
Furnace operator	613	1 full cylinder	3.3	7.86	100	40	145, 90°	6.3	Scrap	Baghouse filter	1 x 10 ⁻³	2
Ingot handling A (shielded) ^d	614 A	1 full cylinder	3.2	7.86	100	40	100	0.7	Ingot	none	0	1
Ingot handling B (unshielded)	614 B	1 full cylinder	3.2	7.86	100	40	50	1.5	Ingot	none	0	1
Ingot fork driver	615	1 full cylinder	3.2	7.86	250	23	200	0.2	Ingot	none	0	1
Slag Handler ^e	617	1 full cylinder	0.055	1.5	20	25	50	0.2	Slag	Slag	1 x 10 ⁻³	1

^a Radionuclide concentrations in the specified materials were used in the dose calculations for the various worker scenarios.

^b External radiation was attenuated by a shielding material with a density of 7.86 g/cm³ and a thickness of 0.3 cm.

^c Off-center distance.

^d External radiation was attenuated by a shielding material with a density of 7.86 g/cm³ and a thickness of 8 cm.

^e External radiation was attenuated by a shielding material with a density of 7.86 g/cm³ and a thickness of 1.2 cm.

Table IV. External Dose Conversion Factors [(Sv/h)/(Bq/g)]

Radionuclides	Scrap Truck Driver	Scrap Loader	Scrap Cutter/Sorter	Furnace Operator
Co-60	5.60E-08	1.42E-07	1.79E-07	1.11E-08
Sb-125	9.51E-09	2.48E-08	3.14E-08	1.65E-09
Cs-134	3.58E-08	9.19E-08	1.16E-07	6.53E-09
Cs-137	1.30E-08	3.33E-08	4.22E-08	2.35E-09
Eu-154	2.61E-08	6.80E-08	8.60E-08	5.11E-09
Radionuclides	Ingot Handler A	Ingot Handler B	Ingot Fork Driver	Slag Handler
Co-60	1.06E-09	9.98E-08	3.09E-09	2.22E-08
Sb-125	3.56E-11	1.51E-08	4.62E-10	3.34E-09
Cs-134	2.58E-10	5.94E-08	1.82E-09	1.33E-08
Cs-137	8.15E-11	2.14E-08	6.56E-10	4.75E-09
Eu-154	4.06E-10	4.62E-08	1.43E-09	1.01E-08

Table V. Internal Dose Conversion Factors (Sv/Bq)

Radionuclides	Inhalation	Ingestion
Co-60	5.91E-08	7.28E-09
Sb-125	3.30E-09	7.59E-10
Cs-134	1.25E-08	1.98E-08
Cs-137	8.63E-09	1.35E-08
Eu-154	7.73E-08	2.58E-09

RESULTS AND COMPARISONS

Tables VI-XIII list calculation and measurement dose results for the eight exposure scenarios. Measured external radiation exposures, excluding background levels, are available for four operational activities and are compared with the calculation results.

Table VI. Calculated and Measured Doses (Sv) for Scrap Truck Driver Scenario (dose code 610)

Radionuclides	External Radiation	Inhalation	Ingestion
Co-60	1.23E-06	1.57E-11	1.74E-11
Sb-125	4.62E-09	1.92E-14	3.98E-14
Cs-134	1.22E-10	5.12E-16	7.31E-15
Cs-137	2.08E-08	1.66E-13	2.34E-12
Eu-154	2.24E-11	7.97E-16	2.40E-16
Total (Individual)	1.26E-06	1.59E-11	1.97E-11
Total (Collective)	2.52E-06	3.16E-11	3.94E-11
Measured Dose	< 1.00E-06	NA ^a	NA
Calculation / Measurement	– ^b	–	–

^a NA = not available.

^b A dash (–) indicates that no actual measured dose was available for calculation of the ratio.

Table VII. Calculated and Measured Doses (Sv) for Scrap Unloader Scenario (dose code 611)

Radionuclides	External Radiation	Inhalation	Ingestion
Co-60	7.72E-05	3.86E-10	4.28E-10
Sb-125	2.97E-07	4.74E-13	9.82E-13
Cs-134	7.74E-09	1.26E-14	1.80E-13
Cs-137	1.32E-06	4.10E-12	5.77E-11
Eu-154	1.44E-09	1.97E-14	5.91E-15
Total (Individual)	7.88E-05	3.91E-10	4.87E-10
Total (Collective)	1.56E-04	7.82E-10	9.74E-10
Measured Dose	3.80E-05	NA	NA
Calculation / Measurement	4.11	–	–

^a NA = not available.

^b A dash (–) indicates that no actual measured dose was available for calculation of the ratio.

Table VIII. Calculated and Measured Doses (Sv) for Scrap Cutter/Sorter Scenario (dose code 612)

Radionuclides	External Radiation	Inhalation	Ingestion
Co-60	2.62E-04	1.04E-08	7.81E-08
Sb-125	1.01E-06	1.28E-11	1.79E-10
Cs-134	2.64E-08	3.40E-13	3.29E-11
Cs-137	4.49E-06	1.10E-10	1.05E-08
Eu-154	4.90E-09	5.29E-13	1.08E-12
Total (Individual)	2.68E-04	1.05E-08	8.89E-08
Total (Collective)	5.36E-04	2.10E-08	1.78E-07
Measured Dose	1.07E-04	NA ^a	NA
Calculation / Measurement	5.01	– ^b	–

^a NA = not available.

^b A dash (–) indicates that no actual measured dose was available for calculation of the ratio.

Table IX. Calculated and Measured Doses (Sv) for Furnace Operator Scenario (dose code 613)

Radionuclides	External Radiation	Inhalation	Ingestion
Co-60	1.97E-06	6.57E-09	4.95E-08
Sb-125	2.96E-09	8.08E-12	1.13E-10
Cs-134	1.13E-10	0	0
Cs-137	1.78E-08	2.79E-11	2.67E-09
Eu-154	3.17E-11	0	0
Total (Individual)	1.99E-06	6.61E-09	5.22E-08
Total (Collective)	3.98E-06	1.32E-08	1.04E-07
Measured Dose	2.20E-05	NA ^a	NA
Calculation / Measurement	0.18	– ^b	–

^a NA = not available.

^b A dash (–) indicates that no actual measured dose was available for calculation of the ratio.

Table X. Calculated and Measured Doses (Sv) for Ingot Handling Scenario (dose code 614A)

Radionuclides	External Radiation	Inhalation	Ingestion
Co-60	1.09E-07	0	0
Sb-125	8.14E-11	0	0
Cs-134	0	0	0
Cs-137	0	0	0
Eu-154	0	0	0
Total (Individual)	1.10E-07	0	0
Total (Collective)	1.10E-07	0	0
Measured Dose	< 1.00E-06	NA ^a	NA
Calculation / Measurement	– ^b	–	–

^a NA = not available.

^b A dash (–) indicates that no actual measured dose was available for calculation of the ratio.

Table XI. Calculated and Measured Doses (Sv) for Ingot Handling Scenario (dose code 614B)

Radionuclides	External Radiation	Inhalation	Ingestion
Co-60	2.20E-05	0	0
Sb-125	7.42E-08	0	0
Cs-134	0	0	0
Cs-137	0	0	0
Eu-154	0	0	0
Total (Individual)	2.21E-05	0	0
Total (Collective)	2.21E-05	0	0
Measured Dose	NA ^d	NA	NA
Calculation / Measurement	– ^b	–	–

^a NA = not available.

^b A dash (–) indicates that no actual measured dose was available for calculation of the ratio.

Table XII. Calculated and Measured Doses (Sv) for Ingot Fork Driver Scenario (dose code 615)

Radionuclides	External Radiation	Inhalation	Ingestion
Co-60	9.10E-08	0	0
Sb-125	3.02E-10	0	0
Cs-134	0	0	0
Cs-137	0	0	0
Eu-154	0	0	0
Total (Individual)	9.13E-08	0	0
Total (Collective)	9.13E-08	0	0
Measured Dose	NA ^a	NA	NA
Calculation / Measurement	– ^b	–	–

^a NA = not available.

^b A dash (–) indicates that no actual measured dose was available for calculation of the ratio.

Table XIII. Calculated and Measured Doses (Sv) for Slag Handling Scenario (dose code 617)

Radionuclides	External Radiation	Inhalation	Ingestion
Co-60	6.57E-07	2.10E-10	1.58E-09
Sb-125	8.58E-10	1.02E-13	1.43E-12
Cs-134	3.69E-09	4.17E-13	4.03E-11
Cs-137	6.20E-07	1.35E-10	1.29E-08
Eu-154	7.07E-10	6.48E-13	1.32E-12
Total (Individual)	1.28E-06	3.46E-10	1.45E-08
Total (Collective)	1.28E-06	3.46E-10	1.45E-08
Measured Dose	1.80E-06	NA ^a	NA
Calculation / Measurement	0.71	– ^b	–

^a NA = not available.

^b A dash (–) indicates that no actual measured dose was available for calculation of the ratio.

Among the three exposure pathways analyzed, radiation exposure from the external radiation pathway was far more significant than radiation exposure from the two internal radiation pathways (inhalation and ingestion). Radiation exposures incurred by the scrap unloaders and scrap cutters/sorters were greater than those incurred by the other workers because of the closer exposure distances and longer exposure times experienced by the scrap unloaders and scrap cutters/sorters.

In general, RESRAD-RECYCLE results agree fairly well with the measurement data. The ratios between the calculated results and the measurement data are 4.11, 5.01, 0.18, and 0.71 for the scrap unloader, scrap cutter/sorter, furnace operator, and slag worker, respectively. For the furnace operator scenario, potential shielding from the furnace wall was considered in dose calculations. In reality, for part of the operation, the operators were exposed to radiation without shielding. As a result, radiation dose calculated by RESRAD-RECYCLE (about 4.0E-6 Sv) was less than the actual exposure (2.2E-5 Sv) measured by dosimeters. Because of the modeling requirement for a deterministic analysis, fixed values have to be specified for the exposure distances. This condition is not consistent with the dynamic nature of worker activities; during the activities the worker changes his position constantly instead of fixing himself at a specific position. The limitation of the RESRAD-RECYCLE external radiation model also contributes to the discrepancies between the calculation results and the measurement data. To facilitate external dose calculations, RESRAD-RECYCLE requires that the radiation source be represented by either a half or a full cylinder with appropriate radius and thickness. Most often, radiation sources do not conform to this geometry and a certain degree of discrepancy in the dose results is inevitable.

CONCLUSIONS

The RESRAD-RECYCLE code was selected to participate in an international validation project to compare modeled dose calculation results with actual measurement data for radiation exposures during recycling of radioactively contaminated scrap metal. In the project, RESRAD-RECYCLE was partially validated by the measurement data taken from the Studsvik recycling facility in Sweden.

Agreement between the RESRAD-RECYCLE results and the measurement data is fairly well. Among the scenarios considered, dose results and measurement data agreed within a factor of 6. Discrepancies result from (1) the limitation of the RESRAD-RECYCLE external radiation model, which considers only cylindrical geometry for radiation sources, (2) the limitation of deterministic analyses that can not fully consider the dynamic nature of worker activities (e.g., fixed exposure distances were used in dose calculations), and (3) the inaccuracy of the exposure parameters developed to represent the actual exposure conditions (e.g., the assumption of a fully shielded operation versus an actual operation that is shielded only part of the time.)

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