

# **A Model to Determine the Radiological Implications of Non-fixed Radioactive Contamination on the Surfaces of Packages and Conveyances**

J. S. Hughes<sup>1</sup>, S. M. Warner Jones<sup>1\*</sup>, M. T. Lizot<sup>2</sup>, M-L. Perrin<sup>2</sup>, S. Thierfeld<sup>3</sup>, E. Schroedl<sup>4</sup>, G. Schwarz<sup>4</sup>, R. Rawl<sup>5</sup>, M. Munakata<sup>6</sup>, M. Hirose<sup>7</sup>

- 1. NRPB, Chilton, Didcot, Oxfordshire, UK.
- 2. IRSN, BP 17, Fontenay-aux-Roses, F-92262 France.
- 3. Brenk Systemplanung GmbH, Heider-Hof-Weg 23, 52080 Aachen, Germany.
- 4. GRS, Schwertnergasse 1, 50667 Cologne, Germany
- 5. ORNL, PO Box 2008, Oak Ridge, TN 37831-6472, USA.
- 6. JAERI, Tokai-mura, Naka-gun, Ibaraki-ken, 319-1195, Japan
- 7. NFT Co. Ltd., 1-1-3, Shiba Daimon, Minato-ku, Tokyo 105-0012, Japan
- \* Present address: Amersham plc, Little Chalfont, Buckinghamshire, UK

### **1. INTRODUCTION**

The surfaces of packages and conveyances used to transport radioactive materials can sometimes become contaminated with radioactive material. This usually occurs as a result of the transfer of radioactive material from areas in which these packages and conveyances are handled. This contamination may subsequently be transferred to transport equipment, workers and to areas accessible to the public. This can represent a significant radiation safety issue that requires careful management.

The current regulatory limits for non-fixed contamination on packages and conveyances have been in use for over 40 years, and are based on a simple exposure model. However, the bases on which these limits were derived have been subject to changes, as a result of successive revisions of international recommendations. In recognition of this need for a review and analysis of the current contamination limits an IAEA Co-ordinated Research Project (CRP) on the "Radiological Aspects of Package and Conveyance Non-Fixed Contamination" was initiated to review the scientific basis for the current regulatory limits for surface contamination. The CRP was also to develop guidance material for evaluating the radiological significance of surface contamination to workers and the public in light of state-of-the-art research, technical developments and current transport practices. The specific objectives of the work undertaken within this multi-national CRP were, in accordance with the terms of reference:

- To ensure that appropriate models exist for all package types including consideration of the aspects pertinent for assessing and revising a surface contamination model for transport,
- To collect where possible contamination, operational and dosimetric data to ensure modelling consistency,
- To use models for assessing the limitations and optimisation of radiation doses incurred in transport operations, and
- To consider preventive methods for package and conveyance contamination.

The CRP was to include a review of the dose pathways for non-fixed surface contamination on packages and conveyances under routine conditions of transport and to quantify the resulting internal and external radiation dose to transport workers and the public (per unit of surface contamination on packages). This assessment was to take into account the different categories of radioactive material packages used in contemporary practices and related transport operations. Six countries and one international organization agreed to undertake research to support the project. These were: France, Germany, Japan, Sweden, United Kingdom, United States of America, and the World Nuclear Transport Institute (WNTI).

# **2. NON-FIXED SURFACE CONTAMINATION IN TRANSPORT**

Surface contamination on packages used to carry radioactive materials tends to be associated with certain types of packages and transport operations. New packages are assembled in clean environments and are very unlikely to have contamination on external surfaces. In the nuclear industry, some waste packages, and especially irradiated nuclear fuel flasks are sometimes found to be contaminated, as a result of being loaded and unloaded in contaminated environments, e.g. fuel storage ponds. These flasks are decontaminated before being dispatched but the phenomenon of "sweating" or "weeping" often results in the reappearance of non-fixed contamination on the flask surface during or following transport.

Limits on non-fixed and fixed contamination on the external surfaces of transport packages, and equipment, have been included in the IAEA Transport Regulations since their inception [1]. The non-fixed contamination limits, which are based on a radiological model developed by Fairbairn [2], are specified in the Regulations as 4 Bq cm<sup>-2</sup> for beta and gamma emitters and low toxicity alpha emitters, and 0.4 Bq cm<sup>-2</sup> for all other alpha emitters. These limits are applicable when averaged over any area of 300  $\text{cm}^2$  of any part of the surface.

# **3. THE FAIRBAIRN MODEL**

The Fairbairn model limits its consideration to inhalation of airborne contamination and transfer of contamination to the hands under a specified set of exposure scenarios. The permissible levels of contamination are constrained so as not to result in an airborne concentration greater than the maximum permissible concentration in air (MPCa) specified in the 1959 recommendations of the International Commission on Radiological Protection (ICRP) [3]. These levels should also constrain the dose to contaminated hands to what was considered to be good-practice in the 1960s. These constraints were applied to the alpha and beta-emitting radionuclides then considered the most hazardous, <sup>239</sup>Pu and <sup>90</sup>Sr respectively, giving the limits noted above.

Since these limits were derived there have been a number of changes in radiation protection philosophy and dosimetry, mainly as a result of the recommendations of the International Commission on Radiological Protection. These include changes in the dose coefficients for inhalation of radionuclides and a change in the specification of the annual dose limit for workers. Also, during the period since the contamination limits were derived much experience has been gained in their use, and in contemporary transport operations. These developments have created the conditions in which a review of these limits is required.

Some inherent limitations of the Fairbairn model have also been recognised. For example, the limited range of radionuclides and exposure pathways considered, the high occupancy times assumed, uncertainties in the resuspension mechanism, out-dated dose coefficients and dose criteria, and the fact that the possible exposure of members of the public were not considered.

# **4. THE DEVELOPMENT OF THE BASIC MODEL**

It was agreed early in the CRP study that a single model should be developed to provide most of the dose calculations. This model is called the Basic Model. Participants from France, Germany, United Kingdom and WNTI met three times to co-ordinate the development of the Basic Model and input from other participants in the CRP was considered during this time. Information on worker operations, including handling times and distances from packages was provided by many of the organisations involved in the study. Contributions were made from other relevant studies, such as work on resuspension mechanisms, previous assessments of exposures from contamination in transport, and work on the prevention of contamination and decontamination techniques.

The approach taken was to develop a model with model parameters that do not necessarily represent a particular transport situation or geographical environment. Rather, the essential elements of the transport phases are abstracted to form a general model for those operations. Such a model should provide a practical means for the assessment of radiation exposures of workers and the public that is unlikely to be exceeded under actual transport conditions.

The methodology, assumptions and other parameters used in the Basic Model were considered in detail and final agreement on these was achieved at the third CRP meeting in Berlin in November 2002. Calculations were performed independently by the Basic Model group participants to verify the accuracy of the spreadsheet calculations being used. Forty-nine radionuclides were modelled during the development and testing of the Basic Model. When completed, the Basic Model was then used to calculate values for all radionuclides listed in the IAEA Transport Regulations[4].

# **5. THE BASIC MODEL**

In order to assess the exposures of workers and members of the public to radioactive contamination on packages and conveyances, it is necessary to consider the various stages of transport and how individuals may become exposed. These include the design of the package, loading, preparation for transport, transport/ storage and final receipt and checking by the consignee. The transport operations in the Basic Model begin with the final inspection stage immediately before the transport shipment. The model is intended to cover all the main types of transport operations. It was considered that focusing on different package types, each representing the main types of transport, was an efficient way of achieving this approach. The Basic Model was developed around the transport

procedures in a way that follows the steps taken during a typical shipment. The exposure of workers and the public was then considered at each phase of the transport process, for each of the package types.

Four categories of packages are considered, covering the main types of shipments:

- Small manually handled packages (SM), such as those used for industrial and medical materials;
- Small remotely handled packages (SR), such as waste or  $UO<sub>2</sub>$  drums moved using fork-lift trucks;
- Large remotely handled packages (LR), such as standard ISO freight containers; and
- Irradiated nuclear fuel flasks (FF).

The transport process was separated into a series of five steps, each sub-divided into individual operations, and these are shown in Table 1. Groups of workers (A, B, C etc.) carrying out these operations were defined, whose duties are as follows, and are shown in Table 1:

- A carries out final checks, e.g. a health physics worker.
- B a loading operator, e.g. a fork-lift truck driver.
- C the driver of the conveyance.
- F a general worker at a transfer site.
- G a loading operator e.g. a crane operator.
- H a health physics worker at transfer site.
- T a health physics worker at consignee's site.
- U an unloading worker at consignee's site.

Some of these operations may be performed by several workers. Times are given for each operation either per package (pa), conveyance (co) or task (ta). Within the Basic Model, annual times are calculated from the individual task time and number of operations/ packages handled per annum. Annual times taken for individual tasks were summed for each of the worker groups. In a few cases, the total working time is in excess of the maximum annual transport working time, which was assumed to be 1500 h. This total is for transport operations, the remainder of the annual working time is assumed to be taken up with non-transport activities. In these instances, the doses were scaled from the calculated total time to the total working time of 1500 h. The 'excess time' would be distributed among other workers in that group. It is assumed that the driver of the conveyance (Worker C) spends 600 h per annum driving the conveyance in the movement phase. This was chosen as a representative fraction, about onethird, of the annual working time. For all workers a maximum working year was assumed to be 250 days.

The following exposure pathways are used to assess doses to transport workers and members of the public from operations involving the shipment of contaminated packages. The pathways of importance to workers are skin contamination, inadvertent ingestion following skin contamination, inhalation of resuspended activity and external exposure to contamination on the package and on the ground. Members of the public should not come into direct contact with packages, therefore only inhalation and external exposure from the contaminated packages are considered. Critical groups of members of the public are assumed to be in the vicinity of packages being handled during phases 3 to 5 of the transport steps shown in Table 1. The annual doses of the groups present at each step are assessed separately, and the limiting step is the one resulting in the highest dose, as indicated in Table 2.

Inhalation is an important pathway and resuspension is a key element of the Basic Model. The Fairbairn model [2] uses a conservative choice of value for the resuspension factor. However, resuspension factors may vary over several orders of magnitude, depending on the surface, material and environmental conditions. For the Basic Model a range of working conditions are considered and therefore it is more appropriate to use the resuspension rate. This has the units of  $h^{-1}$  (derived from Bq  $h^{-1}$  per Bq), and describes the percentage of surface activity that is resuspended into the surrounding air. It mainly depends on the properties of the surface, the interaction between matter on the surface, and the air speed. It is independent of the volume of air into which resuspension occurs. A value of 10 $^4$  h<sup>-1</sup> was chosen for the resuspension rate, based partly on some limited experimental work in France and Japan, which give resuspension rate values in a range from 10<sup>-3</sup> h<sup>-1</sup> to 10<sup>-7</sup> h<sup>-1</sup> [5].





# **6. EXPOSURE PATHWAYS**

The annual effective dose per unit surface contamination from the following exposure pathways were calculated and summed for each worker group and each public critical group.

#### *Skin dose*

It is assumed that a worker handles the contaminated packages and a fraction (20%) of the activity level on the surface (Bq cm<sup>-2</sup>) is transferred to the hands, over a total area of 400 cm<sup>2</sup>. It is further assumed that 20% of the level on the hands is transferred to an area of the face (100 cm<sup>2</sup>). Exposure times for the hands and face are assumed to be 4 h and 8 h per day respectively. The annual equivalent doses, per unit activity on the package, to each tissue are obtained by multiplying the annual exposure time by the fraction of activity transferred, and by skin dose coefficients of beta and gamma dose rates per unit contamination level (Sv h<sup>-1</sup> per Bq cm<sup>-2</sup>). The contribution to the annual effective dose is obtained by multiplying the equivalent dose by the tissue weighting factor for skin (0.01) and the ratio of the exposed area to the total UV-exposed area of skin (3000 cm<sup>2</sup>).

### *Ingestion*

When the hands are contaminated it is assumed that there is a possibility of transfer to the mouth and subsequent ingestion of activity. It is assumed that 1% of the activity on the hands is ingested daily. The annual effective dose is then calculated by multiplying the annual activity ingested by the dose coefficient for ingestion.

#### *Inhalation*

The rate of activity resuspended from the package surface is calculated by multiplying the activity per unit area (Bq cm<sup>-2</sup>), by the resuspension rate (10<sup>-4</sup> h<sup>-1</sup>). The indoor air concentration is then obtained by dividing this rate by the room volume and the air exchange rate. Outdoors the air concentration is obtained be dividing the resuspended activity by the wind velocity and the cross sectional area of the volume into which the activity disperses,  $Q(m^2)$ . The value of Q is taken to be ten times the cross sectional area of the package, or load, at distances up to 3 m. Further multiplying factors are applied at greater distances. The annual effective dose from inhalation received by a worker or member of the public is then obtained from the product of the air concentration, breathing rate, exposure time and dose coefficient for each radionuclide.

### *External exposure*

Gamma dose rates from non-fixed surface contamination, at a level of 1 Bq  $cm<sup>-2</sup>$ , on the surface of each package category were calculated for each radionuclide, using the 'Microshield' code [6]. Dose rates were calculated at a number of distances from the package surface, and at 1 m above the ground where packages are regularly handled. Ground contamination, due to the accumulation of material washed off packages, is assumed to occur at 10% of that on the package surface.

### *Other pathways*

A number of minor exposure pathways were investigated to assess the magnitude of annual exposure to workers and public. These included cloud-shine, deposition of resuspended activity onto the ground, deposition of resuspended activity onto skin and clothing, and public exposure from an extended area of accumulated contamination, such as a railway line. Public exposures from the transfer of contamination into the environment, including the biosphere, were also assessed. All of these pathways were found to give annual doses very much less than the main pathways and were therefore not considered further.

### **7. RESULTS OF THE DOSE CALCULATIONS**

It was agreed that the Basic Model should provide individual doses for at least 49 selected radionuclides. A selection of these results is given in Table 2 in terms of normalised effective dose (mSv  $y^{-1}$  per Bq cm<sup>-2</sup>). This table gives, for the radionuclide shown, the overall limiting dose, and the limiting scenario of worker, pathway, package type; and for the public, the limiting sub-step. The calculations were then carried out for all 356 radionuclides in the Regulations [4]. The numbers of radionuclides within each range of limiting dose per unit level of contamination for these radionuclides are shown graphically in Figure 1.

SM and SR packages are the limiting package types for all radionuclides for worker tasks – this is due to the high transport volumes considered together with the closer handling that occurs for these package types. The limiting workers for SM packages are worker C and worker F; the delivery driver who may also be involved in loading and unloading operations and the transfer site worker carrying out loading and unloading operations. SR packages are limited by the doses to the package preparation worker and the transfer site worker.

The following observations can be made with regard to these limiting values:

- The values of the annual dose per unit surface contamination span nearly 7 orders of magnitude for workers and nearly 8 orders of magnitude for members of the public.
- The highest of all values of annual dose per unit surface contamination (both for workers and the public) is calculated for 227Ac. This radionuclide produced doses of at least an order of magnitude higher than all other radionuclides assessed in this study. However, 227Ac is not normally associated with practical contamination problems in transport.
- The lowest range of values for the annual dose per unit surface contamination is calculated for very weak beta/gamma emitters such as <sup>59</sup>Ni that have low radiological relevance.
- Special consideration should be given to tritium, since this radionuclide raises important difficulties for prevention of contamination as tritium is present in water or steam.
- Worker doses were found to be about three orders of magnitude higher than doses for critical members of the public for all radionuclides. Therefore, in deriving revised limits for non-fixed surface contamination, workers are likely to be more limiting than members of the public, but this can depend on the relevant dose criteria used.
- The limiting package type for the majority of radionuclides was SM packages, although there is little difference between the maximum doses obtained for all package types, assuming the same level of surface contamination.

Nuclide	Worker				Public				Limiting
	Dose <sup>a</sup>			Package Worker Pathway	Dose <sup>a</sup>	Package	Step	Pathway	person
$^{227}$ Ac	$6.1810^{1}$	SΜ	С	inh	$5.4310^{-2}$	<b>SR</b>	4	inh	Worker
$1^{241}$ Am	$2.6810^0$	<b>SM</b>	С	inh	$4.14 10^{-3}$	<b>SR</b>	4	inh	Worker
14C	$1.97 10^{-4}$	<b>SM</b>	С	ing	$1.97 10^{-7}$	<b>SR</b>	4	inh	Worker
${}^{60}Co$	$1.84 10^{-2}$	<b>SR</b>	F	ext	$1.10 10^{-4}$	<b>SM</b>	3.1 <sup>b</sup>	ext	Worker
$51$ <sup>51</sup> $Cr$	$2.3310^{4}$	<b>SR</b>	F	ext	$1.36 10^{-6}$	<b>SM</b>	3.1 <sup>b</sup>	ext	Worker
$134$ Cs	$1.58 10^{-2}$	SR	F	ext	$7.35 10^{-5}$	SΜ	3.1 <sup>b</sup>	ext	Worker
137 <sub>Cs</sub>	$6.7810^{-3}$	<b>SR</b>	F	ext	$2.6710^{-5}$	<b>SM</b>	3.1 <sup>b</sup>	ext	Worker
$\rm ^3H$	$1.76~10^{-3}$	<b>SM</b>	С	inh	4.44 $10^{-6}$	<b>SR</b>	4	inh	Worker
131	$8.08 10^{-3}$	<b>SM</b>	C	ing	$1.9310^{-5}$	<b>SM</b>	3.1 <sup>b</sup>	ext	Worker
<sup>59</sup> Ni	$3.42 10^{-5}$	<b>SM</b>	С	inh	$1.28~10^{-8}$	<b>SR</b>	4	inh	Worker
$^{239}$ Pu	$3.1810^{0}$	<b>SM</b>	С	inh	$4.9310^{-3}$	<b>SR</b>	4	inh	Worker
$^{226}$ Ra	$2.82~10^{-1}$	SΜ	C	inh	$3.45110^{-4}$	<b>SR</b>	4	inh	Worker
35S	$1.60 10^{-4}$	<b>SM</b>	С	inh	$1.38 10^{-7}$	<b>SR</b>	4	inh	Worker
$\overline{^{90}Sr}$	$1.0310^{2}$	<b>SM</b>	С	ing	$9.6910^{-6}$	<b>SR</b>	4	inh	Worker
$^{99m}$ Tc	$9.4410^{-4}$	<b>SR</b>	F	ext	$5.82~10^{-6}$	<b>SM</b>	3.1 <sup>b</sup>	ext	Worker
$^{232}$ Th	$2.8810^0$	<b>SM</b>	C	inh	$2.47 10^{-3}$	<b>SR</b>	4	inh	Worker
$238$ U	$15.67 10^{-1}$	<b>SM</b>	C	inh	$7.89 10^{-4}$	<b>SR</b>	4	inh	Worker
$a_{\rm m}$ C $\cdot \cdot \cdot$ <sup>1</sup> $n \times Dg$ am <sup>-2</sup> $Mir$ mode									

**Table 2. Limiting effective doses and scenarios for selected radionuclides** 

mSv y<sup>-1</sup> per Bq cm<sup>-2</sup>. <sup>b</sup>Air mode.

# **Figure 1. Distribution of limiting effective dose per unit contamination**



### **8. DISCUSSION**

It should be noted that the Basic Model has a number of uncertainties, such as the resuspension rate, and some assumptions may not be conservative. External dose from beta and neutron radiation is not taken into account, nor are the effects of "hot particle" contamination. However as a whole the results are considered to provide the best estimate of an upper limit for the annual effective dose to workers and members of the public from non-fixed contamination.

It is possible to derive surface contamination limits from the annual dose per unit surface contamination calculated from the Basic Model by simply dividing a chosen dose criterion by this dose value for each nuclide. However, it was agreed that the scope of this CRP did not cover making any decisions on the appropriate dose criterion for workers and for the general public. The Basic Model results are linear and can be simply scaled for any given dose criterion.

The current two limits, for beta emitters and low toxicity alpha emitters, and for other alpha emitters, does not represent the span of 7 orders of magnitude of dose from the range of radionuclides. Furthermore, the distinction between low toxicity and other alpha emitters is not supported by the results, as some of the low toxicity alpha emitters give some of the highest doses. Regardless of the choice of dose criteria as the basis for deriving surface contamination limits, consideration must be given to the way in which any revised limits are specified. There are at least three basic options:

- 1. Values rounded to 1 or 2 significant figures for each radionuclide;
- 2. Values rounded to orders of magnitude for each radionuclide;
- 3. Values grouped into several categories; the grouping process may be based either on properties of the nuclides (i.e. "alpha emitters with a high radio toxicity", "strong beta/gamma emitters", "weak beta emitters" etc.) or simply according to the assessed exposures.

# **9. CONLUSIONS**

A model has been developed which enables the calculation of annual effective doses from non-fixed contamination and packages and conveyances in the transport of radioactive materials. The model includes scenarios covering generalised transport processes for four packages types that represent the range of transport operations. Annual effective doses per unit surface contamination have been derived for 356 radionuclides and these span 7 orders of magnitude. Further discussion is required on the best way to use these results in specifying new limits for non-fixed contamination in transport. A full description of the model and the results of the calculations will be published in an IAEA Tecdoc [5].

# **10. REFERENCES**

1. International Atomic Energy Agency. Regulations for the Safe Transport of Radioactive Materials, Safety Series No 6, IAEA, Vienna (1961).

2. Fairbairn, A. The derivation of maximum permissible levels of radioactive surface contamination of transport containers and vehicles. In International Atomic Energy Agency, Regulations for the Safe Transport of Radioactive Materials: Notes on Certain Aspects of the Regulations, p.79 ff., Safety Series No 7, IAEA, Vienna (1961).

3. International Commission on Radiological Protection. Report of Committee 2 on Permissible Dose from Internal Radiation, ICRP, Pergamon Press, Oxford (1959).

4. International Atomic Energy Agency. Regulations for the Safe Transport of Radioactive Material, 1996 Edition, No. TS-R-1 (ST-1 Revised), IAEA, Vienna (2000).

5. International Atomic Energy Agency. The Radiological Aspects of Package and Conveyance Non-Fixed Contamination. Final report of a co-ordinated research project, IAEA-TECDOC (In preparation), IAEA, Vienna, 2004.

6. Negin CA (1986). MICROSHIELD - A Microcomputer Program for Analyzing Dose Rate and Gamma Shielding, CONF-861102, ISSN 0003-018X CODEN TANSA, Trans. Am. Nucl. Soc., Vol. 53, Pages 421-422.