Annex I of Technical Volume 4 CHARACTERISTICS AND MEASUREMENT OF RADIOACTIVITY AND RADIATION LEVELS

This annex provides background information on the quantities and units used in radiation protection and an introduction to the characteristics of radioactive material. Discussed are also their dispersion and deposition in the terrestrial environment and their measurement as well as the assessment of the resulting exposure due to ionizing radiation emitted by radioactive material.

I-1. QUANTIFICATION

The aftermath of the accident at the Fukushima Daiichi nuclear power plant (NPP) has been characterized by the presence of radioactive material in the environment and by the radiation doses consequently incurred by people exposed to ionizing radiation emitted by the radioactive material. Both the amount of radioactive material and the subsequent radiation exposure require appropriate quantification. Quantifying radioactivity and radiation levels has proved to be difficult to understand, particularly for members of the public. After the accident, the use, by the authorities and other stakeholders, of different quantities for describing the same phenomena and of different units for describing the amount of the same quantity has not been helpful. A brief summary of the situation is therefore provided.

I–1.1. Activity

The most important quantity describing the amount of radioactive material is the activity. Activity is expressed and measured in the unit of becquerel (Bq), which is equivalent to the reciprocal of a second (s⁻¹). The activity of a radionuclide corresponding to 1 Bq is associated with one radioactive decay (transformation) in this radionuclide per second; this process is associated with the emission of one or more ionizing particles or photons. One Bq represents an extremely small activity. In the past, the unit curie was used for the activity, but only Bq is used in this technical volume. In the available literature, reporting of activity from the accident is mainly expressed in Bq and in its derivatives, multiples and submultiples (e.g., millibequerel (mBq or 10^{-3} Bq), petabequerel (PBq or 10^{15} Bq), etc.). Moreover, the presence of radioactive material in the environment is quite often related to the unit of area (m², km², etc.), unit of mass (in g, kg, etc.) or unit of volume (per cm³, m³, liter, etc.). In this case, such units as Bq/m², Bq/kg, Bq/kg, Bq/cm³ and Bq/m³ are used. These different ways of reporting activity have been extremely confusing for the public.

I–1.2. Dose

People may be exposed to radiation emitted by radioactive material that was released by an accident and that is present in the surrounding environment. The relevant quantity is the dose incurred by people, namely the amount of energy absorbed per unit mass of exposed material, including individual tissues and organs of the body. Doses may be delivered from radioactive material located outside the body (external exposure) or by radioactive material that may have been incorporated into the body via inhalation, ingestion, open wounds or the skin (internal exposure). The potential health consequences caused by the exposure will depend on the amount of radiation exposure received, the types of radiation involved and the organs exposed. This exposure is assessed in terms of the total radiation dose (caused by external and internal exposure) incurred by the affected individual. The radiation dose, or more accurately, absorbed dose (often only dose), is essentially the concentration of energy deposited in material (e.g. tissues, organs etc.) as a result of an exposure to ionizing radiation. The unit of absorbed dose is gray, its symbol is Gy¹.

¹ In the past, a unit termed 'rad' was used, where 100 rad = 1 Gy.

However, in order to have a more accurate description of the health risk associated with a given dose (only for purposes of radiation protection), the absorbed dose has to be weighted to take into account that different types of radiation have different levels of effectiveness in inducing biological damage. Therefore, an additional quantity, called the equivalent dose, related to a specific tissue or organ, is used for this purpose. The equivalent dose is derived from the mean absorbed dose in the tissue or organ (referred to as the organ dose) multiplied by the radiation weighting factors to take into consideration specific properties of the type of radiation involved. For the assessment of the wholebody exposure, the effective dose is used. It is defined as the tissue-weighted sum of the equivalent doses in all specified tissues and organs of the body. The effective dose reflects the overall stochastic health risk. This quantity represents the probability of cancer induction and genetic effects of ionizing radiation in an exposed individual. The tissue weighting factors take account of the different sensitivities of different organs and tissues for induction of stochastic effects. The equivalent dose and effective dose are quantified using the unit of sievert (Sv)².

It should be noted that both equivalent dose and effective dose are not 'physical quantities' that could be defined in relation to invariable physical parameters. They have been formally defined as radiation protection quantities to be used for normal radiation protection purposes, i.e. for situations associated with the relatively low doses normally encountered. They cannot be formally used to express high exposure, when the use of the absorbed dose is more appropriate. The stochastic effects, typical for relatively low exposures, appear with some delay (several years) following the exposure. On the other hand, the deterministic effects, which are caused by high doses, can cause health harm within a rather short period of time after the exposure (hours or days).

A further complication is the fact that these radiation protection quantities, i.e. equivalent dose and effective dose, are not directly measurable. Therefore, instruments for the assessment of these quantities are calibrated in terms of the so-called operational quantities (measurable) which are approximating radiation protection quantities. In assessment of the exposure to external radiation, the quantity of the ambient dose equivalent and personal dose equivalent are widely applied. The unit of the operational quantities is also Sv. The contribution of the internal exposure to the overall exposure of persons depends on the activity and the type of inhaled or ingested radioactive material.

In general, the exposure and the impact due to the accident have been reported in all these units, i.e. Bq, Gy and Sv, and in their multiples and submultiples. Moreover, when dose rates (the dose per unit of time) are reported, the unit time may vary: sometimes rates are reported per year or annum, sometimes per hour, etc.

I-2. RADIONUCLIDES: TYPES, TRANSPORT AND MEASUREMENT [I-1]

I–2.1. Significant radionuclides

As discussed in Section 4.1 of this volume, the extent to which different radionuclides are released under severe accident conditions depends on the specific nature of the accident in question and on the volatility and other physical and chemical properties of these radionuclides. The extent to which they affect human health depends on their amount released into the environment, their biological behaviour and the duration of exposure. The radionuclides which can generally be expected to be released from an NPP under severe accident conditions can be categorized especially according to their volatility (physical half-lives in parentheses).

Fission noble gases. This category includes krypton, mainly 85 Kr (10.8 years) and xenon, mainly 133 Xe (5.25 days). The noble gases are generally the first radionuclides to be released in a nuclear

² In the past, a unit termed 'rem' was used, where 100 rem = 1 Sv.

accident due to the low potential of these elements for retention in the damaged reactor. An important characteristic behaviour of noble gases is that they do not deposit following release to the atmosphere.

Volatile fission products. This category includes key radioisotopes of the elements which would be expected to be most widely dispersed and deposited in the environment from a reactor under severe accident conditions. These include:

- Caesium: most significantly 134 Cs (2.06 years), 137 Cs (30.1 years), 135 Cs (2 300 000 years) and 136 Cs (13.2 days).
- Iodine: most significantly, from a radiation protection perspective, ¹³¹I (8.02 days); short-lived isotopes ¹³²I (2.30 hours), which is due to ingrowth from ¹³²Te, and ¹³³I (20.9 hours); and additional radionuclides, including the long-lived but radiologically insignificant isotope ¹²⁹I (16 100 000 years).
- Tellurium: including ^{129m}Te/¹²⁹Te (33.6 days/1.16 hours, respectively) and ¹³²Te/¹³²I (3.23 days/2.30 hours, respectively). ¹³²Te decays to ¹³²I in the environment or the human body after intake, and would therefore also contribute to thyroid dose.
- Other elements, including bromine, rubidium and antimony, e.g. ¹²⁵Sb (2.76 years).

Almost all of these volatile fission products can be released at temperatures lower than 2300°C, even before the reactor reaches molten pool conditions. The releases of these elements are accelerated under oxidizing conditions [I–2, I–3]. Radiocaesium, especially ¹³⁴Cs and ¹³⁷Cs, and radioiodine, especially ¹³¹I, are key radionuclides from a radiological protection perspective.

Semi-volatile fission products. This category includes molybdenum and technetium, e.g. $^{99}Mo/^{99m}Tc$ (2.75 days/6.01 hours, respectively); rhodium, and palladium. These products may reach high release fractions depending on the nature of the accident and are characterized by a release that is highly sensitive to the oxidizing-reducing conditions.

Low-volatile fission products. This category includes strontium and yttrium, most significantly ⁸⁹Sr (50.6 days) and 90 Sr/ 90 Y (28.8 years/2.67 days, respectively); niobium, e.g. 95 Nb (35.0 days); ruthenium; lanthanum, e.g. 140 La (1.68 days); cerium, including 144 Ce (285 days) and 141 Ce (32.5 days); and europium (154 Eu and 155 Eu).

These products are characterized by low but possibly significant levels of release during a fuel rod degradation phase that can be affected by specific accident conditions inside the reactor core. For instance, the release of ruthenium can be very high in case of air ingress into the reactor (as during the accident at the Chernobyl NPP in 1986). Significant retention of these elements is nevertheless expected in the upper core structures and hence limited releases to atmosphere.

Refractory elements (non-volatile fission products): This category includes zirconium, neodymium, and praseodymium. No significant release of these highly refractory fission products has been demonstrated experimentally.

Actinides: These can be subdivided into two categories. The first includes uranium and neptunium, which behave similarly to the low volatile fission products, with uranium releases higher under oxidizing conditions. The second group includes plutonium, most significantly ^{239,240}Pu (24 100 and 6560 years, respectively); ²³⁸Pu (87.7 years); and ²⁴¹Pu (14.3 years), which gives rise by ingrowth to ²⁴¹Am (433 years). Plutonium radionuclides tend to behave more like the non-volatile fission products.

Other products: This category is comprised mainly of activation products including ³H (12.3 years) from the activation of water and additives; and ⁵⁴Mn (312 days), ^{110m}Ag (250 days), and ⁵⁹Fe (44.5 days) from the activation of materials in reactor structures.

I-2.2. Dispersion and deposition of radionuclides

The fate of these radionuclides in the environment following release, and the effectiveness with which they can be measured, depends on a variety of physical and chemical factors. The dispersion of radionuclides released to the atmosphere is dependent on the meteorological conditions which prevail at the time of release, especially the wind speed and direction. Radionuclides in different chemical forms (e.g. either in gaseous form or condensed onto particulate matter) can be subject to different rates of advection and diffusion in the atmosphere. Rates of deposition of airborne radionuclides onto the ground are also highly dependent on the chemical form of the radionuclide as well as on the occurrence and intensity of rainfall during transport. Variations in topography and land cover also influence the rates of deposition. The combined temporal variation in meteorological conditions, especially wind and rainfall, and in emission rates can result in complicated temporal and spatial distributions of activity concentrations in air and deposition densities of radionuclides.

Deposition is the process by which aerosol particulates and gases are retained by, or deposited onto, surfaces. There is a concomitant decrease in airborne activity concentrations as a result. Deposition of airborne radionuclides occurs via numerous mechanisms which can be broadly categorized as dry or wet, depending on whether or not precipitation has occurred, as follows:

- Dry deposition deposition of airborne gases or aerosols on the surface by gravitational settling (for large particles) and turbulent diffusion depend on the type of surface cover: soil, vegetation and other material. Rates of dry deposition strongly depend on the physico-chemical form of airborne material, meteorology and the physical, chemical and biological properties of surface matter.
- Wet deposition atmospheric hydrometeors scavenge gas or aerosol particulates. Wet deposition depends on multiple processes, such as nucleation, collision, dissolution and evaporation of materials. The types of wet deposition include below-cloud scavenging (washout) and in-cloud scavenging (rainout) for rain and ice phases, and cloud/fog deposition.

A schematic view of the main deposition processes is presented in Fig. I-1.

In general, rates of dry deposition tend to decrease with distance from the release location. Wet deposition mechanisms depend upon the pattern of precipitation and generally lead to more heterogeneous distributions of radionuclides in the environment. Rates of wet deposition are generally higher due to the fact that the vertically integrated air concentration of radionuclides contributes to wet deposition, while only surface concentrations contribute to dry deposition.

Rates of dry and wet deposition can be quantified by deposition velocities, although other parameters such as washout coefficients are often used for wet deposition. Deposition velocities can vary over two orders of magnitude. This influences both the amount of material deposited on the ground and the long range dispersion in the atmosphere due to depletion of the plume of airborne radionuclides.

Upon release as a vapour under severe accident conditions, isotopes of caesium form compounds and condense onto airborne particulates and, hence, are almost entirely dispersed and deposited in particulate form.



FIG. I-1. Deposition mechanisms for airborne radionuclides.

The behaviour of iodine in the atmosphere is more complex, as the chemical forms of iodine following release are highly dynamic and complex. Iodine can be condensed onto particulate matter; common chemical species include iodide (Γ) and iodate (IO_3^{-}). It can also be released in gaseous forms, particularly I_2 and CH₃I. The chemical form can also transform during transport in the atmosphere or following deposition. As reactive gases, the latter contribute to deposited levels of isotopes of iodine. The different forms of iodine behave very differently. The dry deposition velocity of elemental iodine is estimated to be greater than that of aerosols by an order of magnitude, while the deposition velocity of organic iodine is less than that of aerosols by an order of magnitude. Particulate iodine (and caesium) will be deposited strongly by wet deposition, while organic forms of iodine are less affected by wet deposition [I–4]. The different chemical forms of radioiodine therefore result in different degrees of dispersion and deposition compared to a purely particulate phase material such as radiocaesium.

The rate at which airborne radionuclides are deposited is also influenced by interception with features on the earth's surface and by the underlying topography. Different types of land cover (forest, vegetation, grassland, buildings, sea and large water bodies), which are generally correlated with land use (e.g. forest, agricultural land, towns and cities, etc.), are associated with varying retention or interception rates. Hence, the total deposition of radionuclides differs depending on the land use.

Due to their large surface area, forest canopies are particularly effective in intercepting airborne radionuclides compared with bare soil. A large proportion of material deposited from the atmosphere can therefore be retained in forests, particularly from dry deposition processes. For complex surfaces,

such as forests (and also cities), the lateral and vertical distributions of deposited radionuclides can be very inhomogeneous. This complicates efforts to perform radiological assessments of these areas.

I–2.3. Environmental transfer

Following deposition, radionuclides can migrate and transfer into different components of the environment due to factors such as weathering, migration in the soil, transfer to plants and animals and resuspension.

The migration of radionuclides in the soil column is particularly important and can lead to a reduction in external radiation exposures with time. The migration in soil depends on the type of radionuclides, their chemical behaviour as well as soil characteristics, in addition to bioturbation.

Clay minerals or mica in soil and sediment are major sorbents of caesium, contributing significantly to the migration behaviour of caesium and particularly affecting the uptake by plants.

The transfer of radionuclides through the food chain is an important route of potential exposure of people and non-human organisms. The levels of radionuclides in plants and animals following an accidental release are highly dependent on the time of year when the release occurs. Higher levels are found for releases during the summer, when crops are growing and animals are grazing outdoors, compared to winter, when there is limited plant growth and animals tend to be fed stored feed. Higher levels are also generally observed in plants when there is direct deposition onto foliage compared to transfer by uptake of radionuclides by the roots from the soil. For this reason, ingestion of leafy vegetable and of milk from animals raised on pasture are exposure pathways of concern in the early phase.

Another mechanism by which radionuclides can be transferred in the environment is the resuspension of deposited material by the action of wind or mechanical disturbance. The resuspension factor (the ratio of the activity concentration in air to the activity concentration deposited on ground) depends on meteorological factors such as wind velocity and humidity, land conditions (climatological factors, industrial and agricultural activities) and chemical and physical properties of radionuclide bearing particulates.

I–2.4. Monitoring of radionuclides in the environment

Fixed post and portable measurements — generally sodium iodide (NaI) scintillation counter for low level dose rate and for high dose rate a simple Geiger-Müller or proportional counters — are useful for measuring the gross signal airborne and deposited radionuclides within specific time intervals. Car-borne and airborne instruments — generally sodium iodide (NaI) scintillation counters coupled with GPS location trackers — offer a useful means of determining the spatial distribution of deposited radionuclides. Deposition densities of key radionuclides may also be derived from the latter measurements.

The measured ambient dose equivalent rate at a particular location may be due to radionuclides in the air or those deposited on the ground. The dose rate due to radionuclides in the air will decrease once the cloud of released radionuclides has passed but the dose rate resulting from deposited radioactive material will continue for some time. Dry and wet deposition as the plume passes give rise to separate distinct patterns in time series of ambient dose equivalent rate measurements.

Noble gases, as they are not deposited, give rise to a characteristic short term peak in time series of ambient dose equivalent rate and air concentration measurements. The width of the peak depends on the duration of plume passage. Radionuclides in the particulate form give rise to longer term peaks of ambient dose equivalent rate. In this case, a longer term contribution comes from deposited

radionuclides, so the width is dependent on the combined half-lives of the contributing radionuclides. Many radionuclides emit gamma radiation and can be measured easily by in situ monitors (e.g. ambient dose equivalent monitors and radionuclide specific gamma detectors) and by laboratory based techniques, primarily gamma spectrometry. Significant exceptions are radioisotopes of strontium and plutonium, which require more labour intensive analyses.

I-3. MEASUREMENTS OF EXTERNAL DOSES

Table I–1 details the dosimeters used during the response to the Fukushima Daiichi accident. For monitoring external exposure of the on-site workers, electronic personal dosimeters (EPDs) were used primarily. Some workers wore passive type glass dosimeters in parallel. This provided the workers with real time information to guide their actions and gave a more accurate dose of record reading. If an EPD failed or a worker noticed that the value was suspicious, the supervisor was informed, and a combination of environmental measurements and co-worker doses was used to estimate a worker's dose.

Dosimeter make and model	Category (EPD/passive)	Function	Organizations
Fuji NRN gamma beta	EPD	Dose of record and real- time tracking*	TEPCO
Panasonic gamma ZP-1460P	EPD	Dose of record and real- time tracking*	TEPCO
Panasonic gamma beta ZP-1461P	EPD	Dose of record and real- time tracking*	TEPCO
Mirion DMC2000S	EPD	Dose of record and real- time tracking*	TEPCO
Technol fluorglass dosimeter	passive	Dose of record**	TEPCO
Thermo MK-2	EPD	Dose of record and real- time tracking	IAEA
SAIC PD-10i	EPD	Real-time tracking	US Department of Energy
Panasonic UD-802 TLD	passive	Dose of record	US Department of Energy
Thermo MK-2	EPD	Dose of record and real- time tracking	US Military
Navy DT-702 TLD	passive	Dose of record	US Military
MGP dcm 2000s; RADOS Rad-60s; Isotrak DoseGuard dosimeter	EPD	Dose of record and real- time tracking	AUS US Urban Search and Rescue
Canberra UltraRadiac	EPD	Dose of record and real- time tracking	US Urban Search and Rescue
Hitachi-Aloka Medical K. K. ADM-112	EPD	Dose of record and real- time tracking	Kyoto University

TABLE I-1. MODELS AND USE FOR DOSIMETERS

*Dose of record until Nov. 2011

*Dose of record after Nov. 2011

Following the accident, members of the public living in various locations in Fukushima Prefecture have also been provided with personal dosimeters (termed glass badges) to provide personal estimates of external doses received. These badges, illustrated in Fig. I–2, are typically worn continuously for a period of three months. Some of the results of these measurement campaigns are presented in Section 4.2 of this volume and in Appendix II.



FIG. I–2. Glass badge used for personal measurement of external dose [I–5]. (Photograph courtesy of Chiyoda Technol Corporation).

I-4. DIRECT MEASUREMENTS OF LEVELS OF RADIOACTIVITY IN PEOPLE

The primary evaluations made in closest proximity to the Fukushima Daiichi NPP were conducted at the Onahama Coal Center Access Point (about 60 km from the Fukushima Daiichi site) using a sodium iodide based mobile whole-body counter (WBC). Later, a sodium iodide based fixed WBC was provided at Hirono soccer ground near J-Village [I–6]. If the estimated effective dose was greater than 20 mSv, then the individual proceeded to Japanese Atomic Energy Agency (JAEA) facilities in Tokaimura, where a high purity germanium (HPGe) based whole-body counter (WBC) was used for more accurate measurements. Another location at which personnel surveys were conducted was the Kashiwazaki-Kariwa NPP, at which hand held survey monitors and a WBC were used. JAEA had a systematic and well documented approach using calibrated hand held survey monitors to measure dose rates close to the thyroid. Some of these facilities were far from the Fukushima Daiichi NPP. Between July and October 2011, eleven WBCs were installed at Hirono soccer ground [I–6], and several personnel surveys were conducted there.

For the emergency workers who had a high potential for intake, whole body counting was performed soon after the work activity. For the other workers, whole body counting was carried out every three months before the initial involvement and every month thereafter. The measured activities in the body were interpreted using spectral analysis software (e.g. ABASCO, GENIE) and software for intake and dose calculation (e.g. MONDAL3).

WBCs are the most commonly used instruments to measure radionuclide uptakes at nuclear power plants. As indicated above, they were made widely available at several locations after the Fukushima Daiichi NPP accident. The advantages of whole body counting are that it measures body contents directly, and does not rely on indirect methods (such as analysis of urine). It can be used to quickly and accurately monitor people for internal contamination of radionuclides with energies typically between 300 keV to 1.8 MeV. WBCs are used in nuclear power plants and other facilities where the internal radioactive contamination of persons may occur. Measurements can be conducted for varying times from as little as one minute to several minutes or longer depending on the minimum detectable activities required.

As shown in Fig. I–3 below, the operator positions the person to be monitored inside the WBC (which is shielded against external ambient radiation) and in front of the detectors, and then begins the count. The instrument software stores and displays the spectral data, performs the analysis of activities of selected radionuclides and prints the report. There are many ways a person can be positioned for this measurement, and following the Fukushima Daiichi accident, measurements were taken for people who were lying, sitting or standing. WBCs are used for gamma emitters, and following the releases from the Fukushima Daiichi NPP, WBC examinations were used primarily to detect ¹³⁴Cs and ¹³⁷Cs. These counters cannot differentiate between internal and external contamination, so care must be taken to ensure that individuals are free from external contamination before measurement.



FIG. I–3. Examples of whole body counters (WBCs) [I–5] (Photographs courtesy of National Institute of Radiological Science).



FIG. I–3. Examples of whole body counters (WBCs) [I–5] (Photographs courtesy of National Institute of Radiological Science) (cont.).

To estimate doses from incorporated activities obtained from a WBC requires a knowledge or assumption about the time and route of intake (whether by inhalation or ingestion). Some uncertainty arises in the dose estimate if the mode and timing of intake are not well known.

For accidental releases of radioiodine, such as those occurring during the Fukushima Daiichi accident, thyroid monitoring can (and was) also be conducted to directly measure ¹³¹I content in the thyroid. As shown in Fig. I–4, measurements can be taken with a gamma probe as close as possible to the subject's neck near the thyroid gland without touching the skin, or at a distance of 10 cm from the skin. Having the detector close to the neck maximizes the measurement sensitivity which would be needed for most screening measurements.



FIG. I–4. Example of a thyroid measurement device [I–5] (Photograph courtesy of National Institute of Radiological Science).

Calibration factors are developed to convert measured count rates to activity in the thyroid for children and adults. Factors are also developed to convert the measured count rate to committed effective dose and thyroid dose equivalent for various age groups; as for whole body counting, it is necessary to take into account the time and route of intake. Because of the short half-life of ¹³¹I of 8 days, it is desirable for measurements to be taken as soon as possible after intake.

Laboratory measurements may also be performed to determine if radioactive materials have been absorbed, ingested or inhaled. This typically involves collecting samples of urine to determine the presence of a particular radionuclide, such as ¹³¹I, and can be helpful in making treatment decisions, if needed. For individuals who received higher radiation exposures (for example, emergency radiation workers) in a very short period of time, blood tests such as chromosome analyses may be conducted to determine if any biological effects from exposure, such as a drop in the number of blood cells (red blood cells, white blood cells, platelets) or abnormalities in chromosomes, are detected. Such techniques are only appropriate for relatively high dose, typically more than 500 mGy delivered in a matter of minutes or hours, or if the magnitude of the exposure is uncertain.

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