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REVIEW OF THE A_1 AND A_2 VALUES: DEVELOPMENT, PROGRESS AND OUTCOMES.

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ABSTRACT

The A_1 and A_2 values of the Q System described in the advisory material SSG-26 have been developed to provide maximum allowable contents in packages not designed to withstand accidents, with the objective to limit the accidental exposure of persons below an effective dose of 50 mSv and a skin equivalent dose of 500 mSv. Current values were determined in 1996 according to specific scenarios for five exposure pathways. Since then, the ICRP has published revised radiological data. In addition, progress in computer hardware and software allow the implementation of new methods of calculation, which are more complete and accurate.

In September 2013, it was decided between NRA, PHE, GRS and IRSN to create an international working group to discuss the improvement of calculation methods described in the Q System. The first findings and results were presented during the PATRAM 2016 conference. The main items considered are the following:

1. Using new data from the latest ICRP publications for emission spectra and external dose coefficients.
2. Using Monte-Carlo methods to take into account contributions from all radiations.
3. Selecting the irradiation field geometry.
4. Selecting the calculation model for beta radiation and neutron emission from (α,n) reactions.
5. Developing a specific irradiation scenario to the eye lens and the associated reference dose.
6. Dealing with the progeny radionuclides.
7. Reviewing Q_C (inhalation) and Q_D (ingestion) values with the updated ICRP intake dose coefficients that introduced new particle sizes and chemical forms.
8. Reviewing Q_D (contamination) and Q_E (submersion) using Monte-Carlo methods.
9. Considering the multi-path cumulative dose principle where simultaneous exposures may occur.

The review of items 1 to 4 has been completed; items 5 to 9 are in progress and discussion is pending on further work to be completed in the future.

This paper will indicate the status of work that has been performed since 2016, explain the main changes in the calculation methods as well as the tools that have been developed to evaluate the Q values for any radionuclide, show results and describe the actions that are not yet completed. The WG expects the updated A values to be presented to the TRANSSC by 2021.

INTRODUCTION

The A_1 and A_2 values tabulated in the IAEA transport regulations SSR-6 [1] have been determined to limit the contents of packages so that "*the radiological consequences […] are deemed to be acceptable, within the principles of radiological protection, following failure of the package after an accident*" (para. 402.1 in SSG-26 [2]) where the package has lost its safety and radiation protection functions. These values had been derived from the "Q system" radiological model, based on five different exposure scenarios and described in the advisory material SSG-26, using reference doses of 50 mSv (effective dose) and 500 mSv (equivalent dose to skin). It is considered that exposures below these limits would not lead to significant health detriment, either deterministic or stochastic.

A detailed introduction on the origin of the Q system, the organization of its review and the changes in the ICRP publications (that drive the current work) is detailed in the previous article on that topic, presented at the PATRAM 2016 symposium [3]. Since then, new progress has been made on several topics:

- Beta and neutron radiations (Q_A, Q_B)
- Eye lens irradiation (Q_B)
- Skin dose coefficients (Q_B, Q_D)
- Inhalation (Q_C, Q_E)
- Contamination (Q_D)
- Progeny radionuclides

In 2018, TRANSSC implemented the TTEG (TRANSSC Technical Expert Groups); the "WG A_1/A_2" is a sub-group of the TTEG on Radiation Protection. CERN joined the group in 2018. More about the WG activities can be read in two other PATRAM 2019 articles [22][23].

CALCULATION MODEL FOR BETA RADIATION

The Q system considers a 150 mg·cm^{-2} absorber for the calculation of Q_B. It is an arbitrary figure originally chosen to simulate either residual shielding between the radioactive source and the bystander [2] (due to package debris or because of the capsule containing the source), or auto-shielding of the source itself [4]. This value is not properly documented in SSG-26 and was mentioned as a simple derivation of an assumption made in the 1973 edition of the IAEA Regulations.

Indeed, a thickness of 0.2 mm of steel was considered as a reasonable assumption for ^{90}Sr, then was used to derive the Q_B values of all other radionuclides. This 0.2 mm of steel later became this shielding factor of 150 mg·cm^{-2} (it could also correspond to a thickness of glass of around 0.6 mm).

After further investigations on actual sources, it was found that, except for ^{90}Sr, no other special form radioactive source would be protected by such a thin layer of stainless steel, the minimum being between 0.4 and 0.6 mm for sources such as ^{192}Ir where the minimum thickness is sought to reach

maximum efficiency for gammagraphy. Besides, the 0.2 mm thickness is only used for the beta window protector, which represents only one face of the encapsulated source, the rest of the capsule being more than 1 mm in thickness.

Eventually, most pure sources have significant thicknesses: for strontium alone, a shielding factor of 150 mg·cm^{-2} would represent a thickness of at least 0.6 mm which is far smaller than the size of that kind of source: a A_1 value for ^{90}Sr of 0.1 TBq would lead to a sphere 12 mm in radius; for iridium alone, a shielding factor of 150 mg·cm^{-2} would represent a thickness of at least 0.07 mm which is far smaller than the size of that kind of source: a A_1 value for ^{192}Ir of 1 TBq would lead to a sphere 0.3 mm in radius (pure isotope).

However, for fluorine alone, a shielding factor of 150 mg·cm^{-2} would represent a thickness of at least 0.9 mm, which is greater than the size of an A_1 value of 1 TBq for ^{18}F, found to be equal to 0.03 mm in radius. But, in this special case (and many other of that type), radioisotopes are transported in matrices or as part of the material itself (e.g. ^{18}F as fluorodeoxyglucose) that bring much auto-shielding: in fact, active radioisotopes are not transported in such concentrated form (too small to deal with, some of them need to be injected in patients! etc.).

An issue remains when addressing the A_2 values because the radioactive material is no longer considered non-dispersible since it is conservatively assumed to be in any kind of solid form by the Q system[i]. The WG considers that a shielding thickness of 0.5 mm hypothesis will be sufficient and globally reasonable to calculate the external dose: the original assumptions (debris, remnant shielding around the damaged package, the auto-absorption of the powder grains) and the fact that the source, if not compact, will be spread around (not a point source anymore).

In the end, the WG accepted that is was reasonable to consider a shielding thickness of 0.5 mm of stainless steel for all radioisotopes when evaluating Q_B values. Moreover, for consistency, this shielding shall also be considered in deriving the Q_A values. The effects of such choice are described in another PATRAM 2019 paper [22]. The WG also calculated that this assumption would lead to equivalent results with radioisotopes distributed in a UO_2 pellet[ii].

CALCULATION MODEL FOR NEUTRON EMITTERS

The current Q System introduced a "Q_F value" for alpha emitters, as a replacement for the Q_A values dedicated to gamma radiations; dose due to neutrons from (α,n) reactions and spontaneous fissions were then considered not significant (with the exception of a few radionuclides such as ^{252}Cf for which an equivalent Q_A value was derived as a result of neutron emissions). The SSG-26 does not properly justify the choice of Q_F being equal to 10^4 Q_C to properly take alpha (and neutron) radiations into account. In 1973, a factor of 10^3 (instead of 10^4) was chosen for alpha emitters; A_1 values from neutron emissions were considered covered by those due to alpha or gamma emissions.

As the WG uses Monte Carlo methods [3], neutron doses can now be evaluated explicitly in calculations. Thus, the arbitrary Q_F value will be removed. The spontaneous fission spectra will be derived from the ICRP 107 publication [15]. However, since neutron fluxes from (α,n) reactions are dependent on the source medium (target and mass ratio), the publication does not contain any data.

For this issue, the WG agreed to use the SOURCES-4C calculation code [5], currently seen as the best database available to reasonably evaluate neutron spectra due to (α,n) reactions. The ratio of target/α source is an important factor in the flux per unit volume that is produced. The higher the mass

of the target, the higher the total flux, until it seems to reach a maximum limit; e.g. for an ideal ^{244}Cm-Be source of 50 Ci, the optimum mass ratio was found to be around 0.13 (with a neutron emission rate of 2.2 10^7 n/s/cm^3) but the total flux was only 2.5 10^6 n/s, while the maximum flux (~99%) is 6.1 10^6 n/s for a mass ratio of 370 (meaning the mass of Be is 230 g for less than 1 g of Cm). The mass ratios of many actual sources were found to be within the range of 0.1 to 2.

The WG will also consider the γ emissions accompanying the (α,n) reactions: their high emission rate and energies could significantly contribute to the dose (e.g. 4,4 MeV for ^{9}Be → ^{12}C reaction at a rate of 0.59 per neutron and 6,1 MeV for ^{13}C → ^{16}O reaction).

In the end, the WG will evaluate the Q_A and Q_B values for beryllium (most penalizing) and oxygen (most common, e.g. in spent fuel) targets with a mass ratio of 10[iii] and propose them to the TRANSSC. It is possible that two lists will be presented in the revision of the SSR-6 to account for the transport of materials with targets other than oxygen. Another solution would be to introduce a factor to the A_1 values that will consider the multiplication effect of such target (if it does not influence the A_2 values, which is mostly the case for alpha emitters).

Those elements are still discussed within the WG. A clear and common position will be established by the end of 2019.

EYE LENS IRRADIATION

A new limit for the equivalent dose to the eye lens was recommended by ICRP 118 [17] for workers. It was decreased from 150 mSv per year to 20 mSv per year averaged on 5 years with a maximum of 50 mSv per year. Since the accident doses considered in the Q system are based on the former annual dose limits for workers, this reduction by a factor of 3 to 7.5 may call into question the appropriateness of the statement in SSG-26, para. I.28, that "*the dose to the skin is always limiting for maximum beta energies and that specific consideration of dose to the lens of the eye is unnecessary*".

ICRP 103 and 118 now include a value of 0.5 Gy as being the deterministic effect threshold of the lens of the eye for a short-time irradiation (cataract). Such value could be used as a reference dose limit for the Q system. However, nothing in the regulations or in the ICRP recommendations address reference equivalent dose limit in case of an accident (while the range of 20 – 100 mSv exists for effective dose); besides, as noted earlier, the current dose references used in the Q system are identical to the former annual dose limits; in the end, while an eye equivalent dose reference of 50 mSv (following the same pattern as the annual maximal effective dose limit to workers) would seem too penalizing, a reference of 500 mSv would not seem reasonable for the sake of safety.

It is important to underline that, with a 500 mSv reference dose, the Q value for the eye is never under that for the skin, and with a reference dose of 150 mSv it is lower for 8 radionuclides included in the SSR-6.

The decision on the appropriate reference dose limit will be taken by the end of 2019.

SKIN DOSE COEFFICIENTS

The ICRP data are mostly complete to deal with the review of the Q System. However, three sets of skin dose coefficients cannot be found ICRP 116 publication [16]:

- local skin-equivalent dose coefficients for positrons;
- local skin-equivalent dose coefficients for neutrons;
- local skin-equivalent dose coefficients for photons.

The skin dose coefficients due to positrons were derived using the same methodology presented in the ICRP 116 [16]. The results are in accordance with the literature [6]: the dose coefficients are only different from those derived for electrons at low energies (below 60 keV) and the results are impacted by a factor of less than 10% for Q_A values and less than 3% for Q_B values. For the sake of consistency, the WG will use the new positron skin dose database.

An important question was also raised: using ICRP 116 coefficients means that all $Q_{B,skin}$ values mix mean skin dose coefficients for photons and neutrons and local skin-equivalent dose coefficients for electrons and positrons, which are not derived from the same calculation method. The WG then decided to homogenize the calculation method by deriving local skin-equivalent dose coefficients for photons and neutrons; it meant creating new dose coefficient databases (that are presented in another PATRAM 2019 article [22]). The issue is important as it also concerns the Q_D calculations.

CALCULATION MODEL FOR INHALATION

The WG agreed to keep the inhalation scenario as it is currently described in the Q system (total release of radioactive material with an airborne fraction of 10^{-3} in a 300 m^3 warehouse, exposure of 30 minutes). ICRP 68 [13] inhalation dose coefficients are currently used to derive the Q_C values.

Since 2015, ICRP has been publishing new intake dose coefficients to replace those of ICRP 68. As of today, only publications 130 [18], 134 [19] and 137 [20] have been released (five publications are expected). New worker inhalation dose coefficients for aerosols of particle size from 0.001 µm to 20 µm were calculated and new chemical forms were introduced; the highest dose coefficients are often those for nanoparticles. It is unclear if the particle size of the materials normally transported is comparable to that of nanoparticles and whether, dose coefficients for nanoparticles should therefore be used instead of those for an activity median aerodynamic diameter (AMAD) of 1 µm as in the current Q system. To illustrate the issue, table 1 gives a sample of the expected changes:

Table 1. Comparison of inhalation dose coefficients between ICRP 68 and ICRP 130

Isotope	Current values			Worst case form			
	Form	Coeff. Sv/Bq	Description	1 µm Sv/Bq	5 µm Sv/Bq	Max (size) Sv/Bq	
^{3}H	Organic	5,0E-11	Carbon tritide	5,2E-10	2,6E-10	2,3E-09	(0,01 µm)
^{14}C	Vapour	5,8E-10	Elemental carbon	1,2E-08	6,8E-09	5,3E-08	(0,01 µm)
^{45}Ca	Type M, 1 µm	2,7E-09	Type S	1,8E-09	1,1E-09	9,0E-09	(0,01 µm)
^{60}Co	Type S, 1 µm	2,9E-08	Type S, cobalt oxide	5,9E-08	3,1E-08	2,6E-07	(0,01 µm)
^{90}Sr	Type S, 1 µm	1,5E-07	Type S, FAP, PSL	3,8E-07	2,0E-07	1,7E-06	(0,01 µm)
^{95}Zr	Type S, 1 µm	5,5E-09	Type S, oxide, tritide	4,5E-09	2,6E-09	2,1E-08	(0,01 µm)
^{99m}Tc	Type S, 1 µm	5,0E-11	Type S	9,5E-12	1,2E-11	5,8E-11	(0,003 µm)

ICRP 130 suggests that only AMAD particles of 1 to 5 µm should be taken into account for environmental and occupational exposure as they are characteristic of aerosols produced by dispersion mechanisms, except for daughter radionuclides of gases (namely radon) for which nanoparticles are created as a result of radioactive decay It was also noted that significant amounts of nanoparticles are unlikely to be produced in an accident (10^6 particles of 10 nm are necessary to have the same mass as a 1 µm particle, which seems far above the likely distribution of particles of that size in powders usually transported).

Whatever the decision, from the first investigations made by the WG with data at their disposal, the Q_C values will inevitably change.

CALCULATION MODEL FOR SKIN CONTAMINATION

The current Q system evaluates the skin contamination Q_D values with the dose coefficients taken from Cross et al. [7] which uses Monte Carlo calculations for an air/water interface. While this method is close to the one used for the current review of the Q system, the WG also agreed to move to the air/skin cube model detailed in ICRP Publication 116. In the end, two models will be compared:

- the Cross et al. model:
  - cylinder of water of 100 cm^2;
  - isotropic source of 100 cm^2;
  - target of 1 cm^2;
  - integration of the dose between 60 and 80 µm;
- the ICRP 116 model:
  - cube of skin of 10x10x10 cm;
  - isotropic source of 38.5 cm^2 (i.e. radius of 7 cm);
  - target of 1 cm^2;
  - integration of the dose between 50 and 100 µm.

As for the external dose to the skin, the local skin equivalent dose coefficients will be used to better represent the most exposed area (and not the dose to the entire skin of the body).

PARENT AND PROGENY RADIONUCLIDES

The Q system introduced a “10-day rule” to account for the progenies in the evaluation of the A_1 and A_2 values. However, nothing is clearly explained in the SSG-26 and hypotheses were considered by members of the WG to explain the current values. For example, with the ^{47}Ca / ^{47}Sc couple, it appears that the current value was calculated after 10 days of in-growth, instead of assuming secular or transient equilibrium.

It was underlined that Q-values can be affected by the transport time and that dose calculations need to be consistent with the scenario. Participants noted the example of irradiated targets for medical isotopes extraction that may be transported only hours after their irradiation (mainly due to the limited half-life of the radionuclides). It was then suggested that the values should be given without progeny and that the consignor should work out the value for mixture transported.

In the end, two proposals will be brought to the TRANSSC: either using a properly justified “10-day rule” (or something similar), or deriving the A values for all pure radionuclides.

FIRST A_1 RESULTS FOR PURE RADIONUCLIDES

The WG calculated the A_1 values for all radionuclides listed in the ICRP 107 publication (i.e. more than 1 000 radionuclides). The following figures represent the evolution of the new A_1 values for 275 pure radionuclides listed in the SSR-6. Nuclides not included here are:

- isotopes with progenies (e.g. ^{137}Cs, ^{238}U, ^{90}Sr, etc.);
- alpha emitters, as (α,n) reactions are not currently evaluated (e.g. ^{244}Cm, ^{241}Am, etc.);
- dual β+/β- emitters, as ICRP 107 does not separate the spectra (e.g. ^{106}Ag, ^{102}Rh, etc.); the WG will use the spectra listed in the JANIS 4.0 database [8].

The detailed results and origins of the changes are thoroughly presented in another PATRAM 2019 article [26].

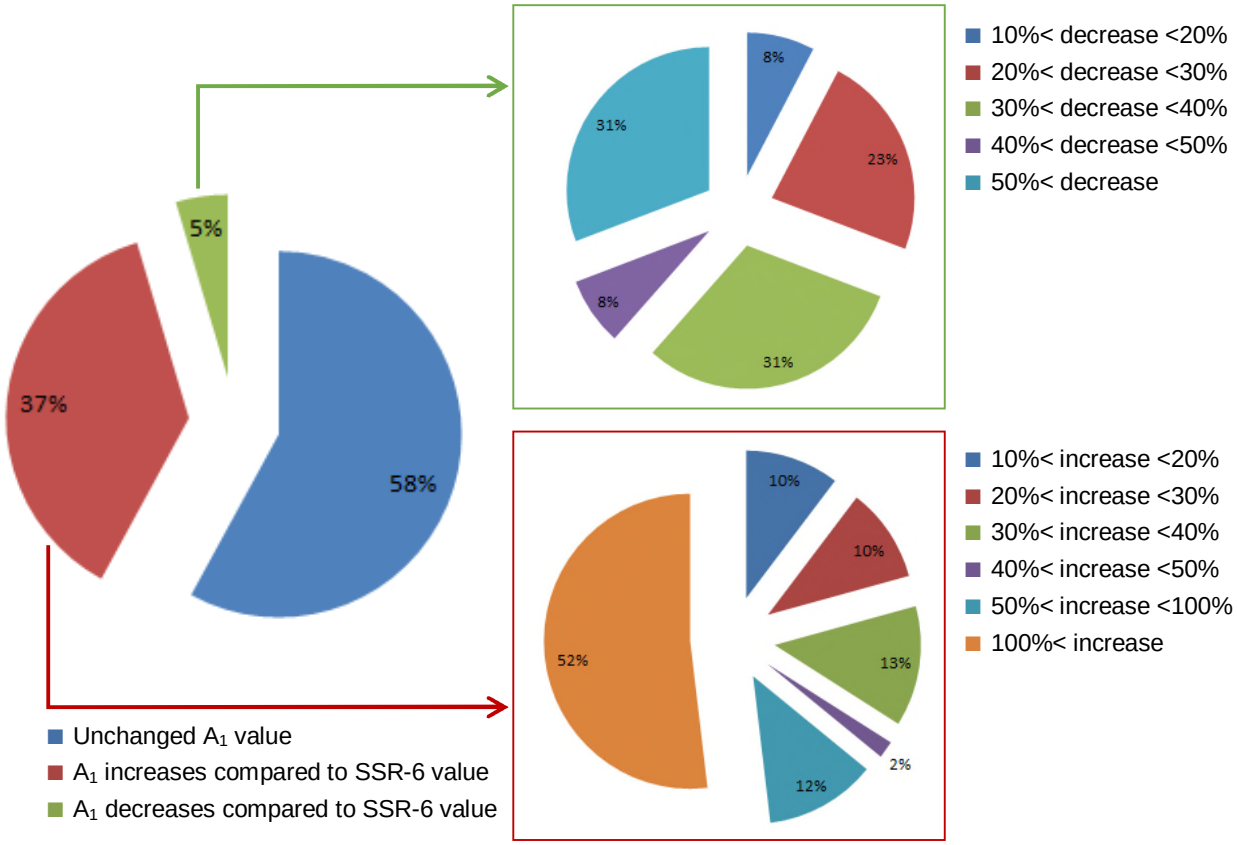

Figure 1. First A_1 results for 275 pure radionuclides listed in Table 2 of SSR-6.

TOOLS DEVELOPED BY THE WG

The new calculation method developed in 2016 [3] is only based on the use of several databases either produced from the working group or from other sources, such as ICRP publications:

- source energy to flux, for each energy bin and each particle (Q_A, Q_B, $Q_{D,c}$, Q_E)[iv]
- flux to dose, for each energy bin (Q_A, Q_B, $Q_{D,c}$, Q_E)
- source spectra (all Q values)
- intake dose coefficient for each radionuclide (Q_C, $Q_{D,i}$, Q_E)

Compared to the previous method, it is no longer necessary to evaluate the Q values with direct calculations (from the spectrum to the Q value). In this regard, it is possible to develop tools dealing with those databases. As such, they can easily be updated with future databases [22]. An example is

the MCBAS interface introduced by GRS in 2016 [9]. NRA/MHI NS ENG, CERN and IRSN also developed complementary and similar interfaces [10][11][12] ; these tools will mainly serve the purpose of comparing the results from different sets of code/library [3]. The WG intends to release such a tool to facilitate the evaluation of any Q and A_1/A_2 values among transport stakeholders.

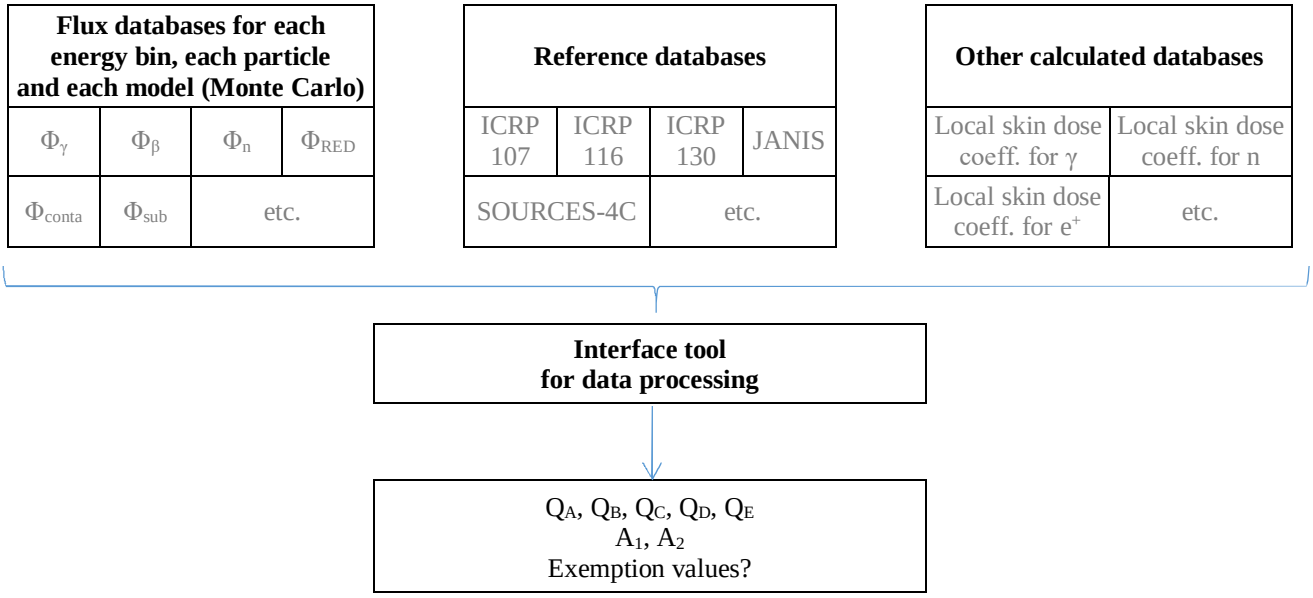

Figure 2. Overview of the tool principles to evaluate the radionuclide basic values.

OTHER MATTERS: NATURAL URANIUM AND THORIUM

In 2017, WNTI wanted to assign footnote "(a)" to U(natural) and Th(natural) in the first column of Table 2 [1]. The proposal was rejected with the motivation that "*more detailed discussion and a conclusion in A_1/A_2 WG of experts is needed before changing current status of Table 2*". In 2015, the WG had already stated to the TRANSSC that "*the WG assumes that the progeny have been taken into account properly. […] The WG will calculate A_2 values for U(nat) and Th(nat) using the current Q system for the proposed nuclides in order to investigate the possible impact of the progeny (e.g. ^{226}Ra)*".

In 2018, the conclusions of the WG were the following:

- SSR-6 mentions U(nat), U-natural and natural uranium. "Natural uranium" is clearly defined in section II of SSR-6. "U-natural" is quoted in footnote (b) of Table 2 only for the exemption values of U(nat) and is defined in RP 65 [21]. In 2015, the WG expected U(nat) to be the same material;
- the Q System gives Q_A and Q_B values to U(nat), and not ^{238}U; adding the fact that they are very similar to the values for ^{226}Ra, the WG assumed that secular equilibrium was considered for U(nat). Evidence of that hypothesis, including Th(nat), could be found in working papers presented during SAGSTRAM TC-800 on the previous review of the radionuclide basic values in 1995;
- whatever the method used to derive the A_1/A_2 values of U(nat) and Th(nat), they are still evaluated as "unlimited".

Under these findings, the WNTI proposal was accepted and is now included in the 2018 edition of the SSR-6.

CONCLUSION AND FURTHER DEVELOPMENT

The state of the current work (as of November 2018) was presented to the TRANSSC 37. That workshop was an important step in the process as it clarifies the position of the WG and prepares the committee for the resolution related to the new values that will be proposed to update the regulations.

In this regard, the review of the Q System by the WG is intended to be finished by 2021 for the new values to be considered in the next revision of the SSR-6. Many decisions are yet to be taken (treatment of progenies, choice of irradiation geometry, dose limit for the eye lens, cumulative dose principle, etc.) but most time-consuming calculations are finished (Q_A, Q_B, Q_D) or underway (Q_C, Q_E). The final decisions can be quickly processed in the tools developed by the members of the WG.

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[i] Q System only considers solids or noble gases; as a result, there are additional requirements for type A packages transporting gases (non-noble) and liquids.
[ii] Uranium is denser than steel, thus leading to more energetic bremsstrahlung. However, uranium is mostly found in the transport of spent fuel, hence the calculation for a UO_2 pellet.
[iii] This value is still under discussion, as the WG does not yet know the extent of the SOURCES-4C validation domain.
[iv] In the Q system, Q_D is either due to ingestion $Q_{D,i}$ or contamination $Q_{D,c}$.