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REVIEW OF THE A_1 AND A_2 VALUES: FINAL CURTAIN?

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ABSTRACT

The A_1 and A_2 radioactive transport limit values of the Q System described in the advisory material SSG-26 have been developed to provide the 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 Monte Carlo based calculation methods, which are more representative of the physical processes.

An international working group involving NRA, PHE, GRS and IRSN was created in 2013 to discuss the improvement of calculation methods described in the Q System. This group later became part of the IAEA TRANSSC Technical Expert Group (TTEG) on Radiation Protection. The first findings and results were presented during the PATRAM 2016 conference, and the progress of the review was described during PATRAM 2019. While most trends on the potential changes in A_1 and A_2 values were presented, further discussions aimed at consolidating hypotheses and calculation methods were necessary to complete the review and propose to the TRANSSC Member States an update of the Q system consistent with the latest ICRP recommendations and methodologies.

This paper describes the work that has been performed since 2019 — especially regarding inhalation, contamination, ingestion, alpha and neutron considerations, explains the main changes in the calculation methods as well as the tools that have been developed to evaluate the Q values for any radionuclide, and shows results that could be implemented in the future revision of the IAEA SSR-6 regulations.

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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 were derived from the “Q system” radiological model [5], based on five different exposure scenarios considering upper thresholds of 50 mSv (effective dose) and 500 mSv (skin equivalent dose).

A detailed introduction on the origin of the Q system, the organization of its review by the “Working Group on A_1/A_2 ” (WG A_1/A_2), the changes in the ICRP publications and the first results of the review are detailed in the previous articles on that topic, presented at the PATRAM 2016 [3] and 2019 symposia [4]. The current paper presents the progress made since then and the conclusions of the review. The following topics are covered:

- irradiation geometry and eye lens criterion (Q_A , Q_B),
- ($\alpha, n\gamma$) reactions (Q_A , Q_B),
- inhalation and ingestion (Q_C , Q_D),
- contamination – particularly α emitters (Q_D),
- submersion and inhalation doses due to Rn isotopes (Q_E), and
- progeny radionuclides.

DOSE DUE TO EXTERNAL EXPOSURE (A_1)

Final considerations to derive Q_A and Q_B

From the last progress paper presented during PATRAM 2019 [4], two remaining topics needed to reach a conclusion: irradiation geometry considered to evaluate Q_A and Q_B and reference criterion to determine the equivalent dose to the eye lens ($Q_{B,eye}$).

The WG agreed that the irradiation field should average the exposure from a severe transport accident (where a type A package would lose its content) and that the ICRP 116 [19] dose coefficients for a parallel beam of ionizing radiation would be unrealistic for a point source 1 m away from a person, as shown in figure 1. Moreover, it was also considered unlikely that an individual would remain static for more than 30 minutes (except if they lay unconscious next to the source). Thus, the antero-posterior (AP), postero-anterior (PA) and lateral (RLAT, LLAT) geometries were eventually not considered. The rotational (ROT) and isotropic (ISO) geometries were kept in the analysis as they represent irradiation from different angles, simulating a person moving in the vicinity of the damaged package. The current Q system considers the ISO geometry; however, though it was questioned during the TC-800¹, no strong argument, in favor of this geometry, was reported in SSG-26 [2] or working documents. The WG also noted that the ROT geometry was lacking dose coefficients, in the literature, for many types of particles, for both effective and equivalent dose.

In the end, considering that the ROT effective dose rate would increase by less than 26% compared to the ISO geometry (the maximum increase for photons is obtained at 40 keV [19] – cf. figure 1), the WG decided to keep the current ISO geometry to derive the final A_1 values. It is underlined here that the new method allows for a relatively quick and easy evaluation of A_1 values for any kind of geometry, assuming the dose coefficients evaluated are similar to those stated in the ICRP 116.

¹ TC-800 was the technical committee of the IAEA SAGSTRAM (former TRANSSC) in charge of reviewing the 1985 version of the Q system [5] between 1992 and 1996.

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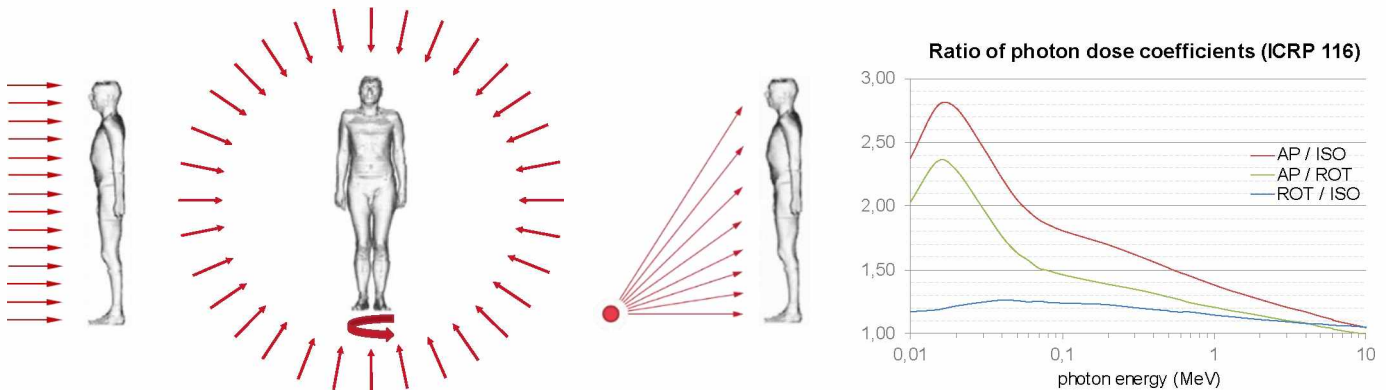


Figure 1. ICRP 116 AP & ISO exposure geometries vs. a realistic AP exposure geometry, and comparison of photon dose coefficients between AP, ISO and ROT geometries

Based on the information and recommendations in ICRP Publications 103 and 118, it was decided to keep the dose criteria of 50 mSv (effective dose) and 500 mSv (skin equivalent dose) as being reasonable. Even if the dose criteria are originally based on annual dose limits for workers, they represent a fraction of the thresholds or reference values stated in both ICRP Publications 103 [16] and 59 [14] (still validated in ICRP Publication 118 [20]) for acute exposures with deterministic effects (namely effective dose from 100 to 500 mSv and equivalent dose to the skin from 1 to 20 Gy). Similarly, following a preliminary review of ICRP Publication 118 for the dose limit for the lens of the eye [20], the WG suggested to use a criterion of 250 mSv based on half of the dose threshold of 0.5 Gy stated in the publication (e.g. para. 4.13) for radiation-induced eye cataracts due to acute exposure. A $Q_{B,eye}$ value was then derived, together with Q_A and $Q_{B,skin}$. This new $Q_{B,eye}$ value has limited impact on A_1 .

Calculation model for (α,n) reactions

The current Q System introduced a “ Q_F value” for α emitters that replaced the Q_A value which only considered the dose from photon radiation. The dose due to neutrons from (α,n) reactions and spontaneous fission were then considered not significant (with the exception of ^{252}Cf , ^{254}Cf and ^{248}Cm for which an equivalent Q_A value was derived as a result of neutron emissions). SSG-26 does not properly justify the choice of Q_F being equal to $10^4 Q_C$ to properly take α and neutron radiations into account. In 1973, a factor of 10^3 instead of 10^4 , was chosen for α emitters; A_1 values from neutron emissions were considered negligible and covered by those due to α or γ emissions.

As recalled in [4], the new method now considers neutron emission spectra from ICRP Publication 107 [17] and neutron dose coefficients from ICRP Publication 116 and [9]. However, ICRP does not provide any data related to (α,n) reactions, as they depend, on the one hand, on the interactions of the α particles (following the α -decay) with the source medium, on the other hand, on the mass ratio between the radioactive compound and the stable element target. For this issue, the WG agreed to use the SOURCES-4C calculation code [6], currently considered as the best dataset available to reasonably evaluate neutron spectra due to (α,n) reactions. The WG also considered the γ emissions accompanying those reactions.

The WG performed parametric and sensitivity analyses including all possible α emitting radionuclides. Two commonly used targets were considered: beryllium (mass ratio of 5) because this is the most penalizing, and oxygen (molar ratio of 5) because this is the most common (oxide forms).

After preliminary calculations for both targets and some common actinides (^{241}Am , ^{239}Pu and ^{244}Cm), it was decided that, for the sake of safety, only the beryllium target would be considered in the complete analysis – but the method described hereafter could be reproduced for oxygen or any other

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target. Based on SOURCES-4C, the neutron dose as a function of the α mean energies was determined, while the photon contribution has been calculated with the TALYS code [7] as a function of monoenergetic α energies. From these calculations, a database was derived allowing for the calculations of neutron and γ emission rate and dose contributions for all α emitters (i.e. 182 radionuclides stated in ICRP Publication 107 [17]). A complete description of the method and results can be found in [10].

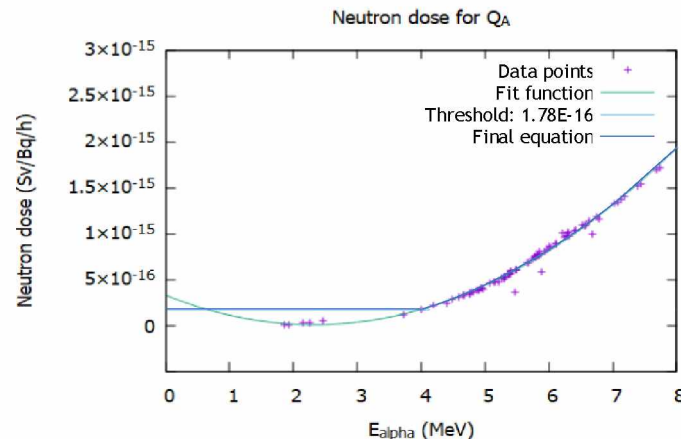


Figure 2. (α ,ny) effective dose for Q_A per primary α particle

During the revision of the current Q System on that matter, it became apparent that the “ Q_A or Q_F ” value mentioned in SSG-26 were determined as the minimum between $Q_A(\gamma)$ and $10^4 Q_C$. The new external effective and skin equivalent dose coefficients due to (α ,ny) reactions determined by the WG show that the former Q_F was too penalizing for most α emitters considered in the current Q system. Therefore, considering (α ,ny) reactions never decreases the A_1 values driven by the former Q_F (e.g. ^{244}Cm for which the current “ Q_A or Q_F ” is 16 TBq and the updated Q_A is 130 TBq). It has limited impact on A_1 values originally driven by their Q_A (e.g. ^{225}Ac) [10]. Eventually, the Q_F value can be discarded.

DOSE DUE TO INTERNAL EXPOSURE (Q_C , $Q_{D,ing}$)

As mentioned in previous WG progress papers [3][4], it was agreed to keep the ingestion and inhalation scenarios as they are currently described in the Q system. Inhalation and ingestion dose coefficients from ICRP Publication 68 [15] are currently used to derive the Q_C and $Q_{D,ing}$ values. Between 2015 and 2022, ICRP published new intake dose coefficients [21][22][23][24][25].

The WG then derived the new Q_C and $Q_{D,ing}$ values. Q_C values were derived for AMAD (Activity Median Aerodynamic Diameter) particles of 1 and 5 μm , representative of environmental and occupational exposure [21], while only 1 μm particles were considered in the current Q system. The most penalizing chemical form was considered, but the distinction between S (slow), M (moderate) and F (fast) absorption were kept for the nuclides currently concerned in table 2 of SSR-6. $Q_{D,ing}$ values were not originally derived, as it was then considered that the Q_C scenario would always be the most limitative one regarding internal exposure [2]. Looking at the new $Q_{D,ing}$ values, this statement eventually appears to be true as only some iodine isotopes would give a $Q_{D,ing}$ value lower than their Q_C .

The changes in Q_C values, though significant, are not as dramatic as expected. However, considering the current reference intake mass threshold above which a Q_C value would be considered as unlimited (10 mg, i.e. $10^{-4} Q_C/\text{g}$, which is the basis to define LSA-II material), the most noteworthy change concerns enriched uranium. Currently, the ^{235}U enrichment level under which Q_C is considered unlimited is 20%. The new enrichment level should be around 11%. The WG also noted that the

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composition of enriched uranium is not clear: SSG-26 refers to the ASTM C996-90 definition which is limited to 5% in the latest C996-20 edition, while SSR-6 [1] has a slightly different one in para. 246 ("unirradiated uranium" to which "U(enriched to 20% or less)" refers in table 2) and para. 247 (enriched uranium). However, those differences eventually do not lead to significant changes in the Q_C value.

Figure 3 summarizes the changes in Q_C values. For comparison purposes, Q_C values from both current Q system and the new method were rounded the same way as A_2 values (i.e. 1 significant figure); unlimited values are included.

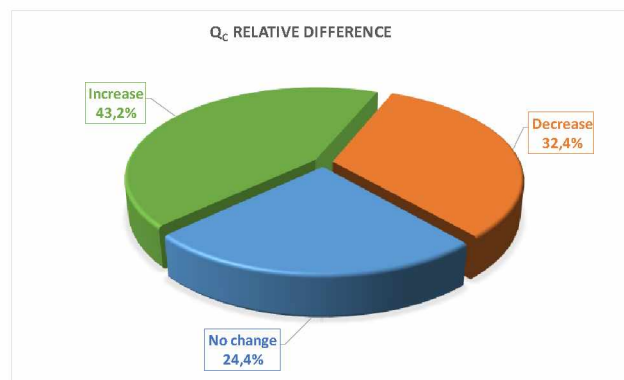


Figure 3. Relative modifications in Q_C values between current and new methods

DOSE DUE TO CONTAMINATION ($Q_{D,skin}$)

In the previous report, the WG mentioned they had to compare two contamination models: the former model from Cross et al. [8], and the model derived from ICRP Publication 116 [19]. For the sake of consistency with the global method decided at the beginning of the review of the Q system, and since the differences were not so significant and that Cross' data are limited to energy below 4 MeV, the WG eventually decided to use ICRP 116:

- cube of skin of $10 \times 10 \times 10 \text{ cm}^3$;
- isotropic source of 38.5 cm^2 (i.e. diameter of 7 cm)
- target of 1 cm^2
- integration of the dose between 50 and 100 μm (scoring region)

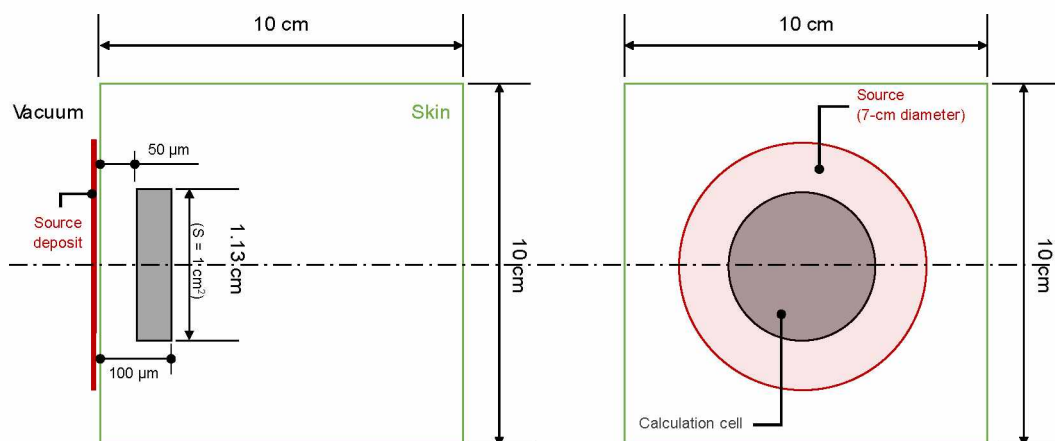


Figure 4. ICRP-116-based model to evaluate skin dose due to contamination

The most significant changes in $Q_{D,skin}$ values relate to the effects of α particles, which were not previously taken into account. The method used to derive the contamination dose coefficients and the

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corresponding results are detailed in ref. [10]. Regarding this special case, when α particles are emitted toward the skin slab with an incident energy in the [5-7] MeV range, considerable discrepancies in dose between the different particle transport codes used were observed. α particles with an energy below 5 MeV do not reach the scoring region; therefore only the secondary particles are responsible for the dose. α particles with energy exceeding 7 MeV are the main contributors to the dose as they penetrate deeper in the skin (cf. figure 5 [10]). Due to differences in the stopping power corrections of the Monte-Carlo codes, the energy range between 5 and 7 MeV provides strong relative variations of the dose to the skin when comparing different Monte Carlo codes.

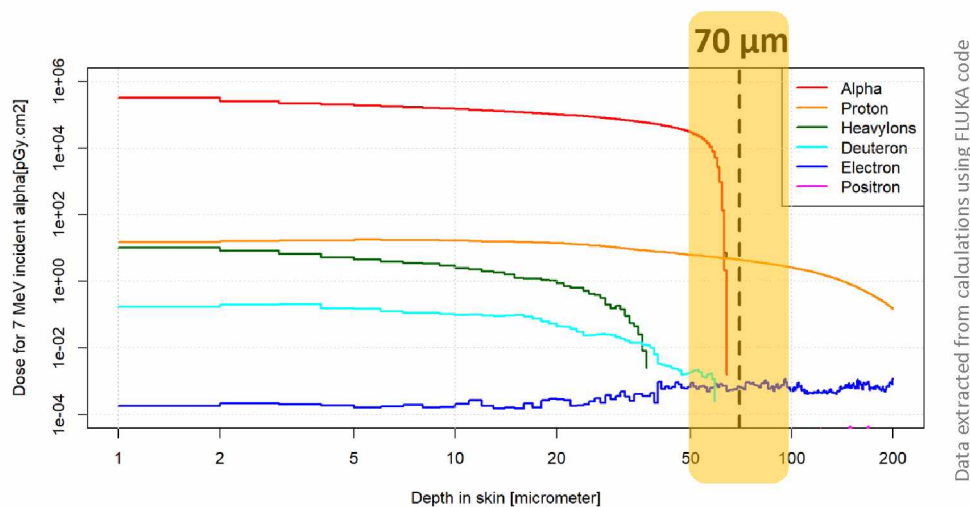


Figure 5. Penetration depth in skin of primary & secondary particles (7-MeV α)

The discrepancies between codes were explained by the corrections implemented in the Bethe theory for low energy α that differ among those codes. The WG then agreed to consider the maximal dose coefficients among the three different evaluations; using an average of those coefficients would, at most, divide the dose coefficients by a factor of 3 for an energy of 5 MeV and would not significantly affect anything from 7 MeV. None of the radionuclides producing α particles in the 5-6.5 MeV range have A_2 values dominated by this pathway: in those cases, Q_c values are lower.

Figure 6 summarizes the changes in $Q_{D,skin}$ values. For comparison purposes, $Q_{D,skin}$ values from both current Q system and the new method were rounded the same way as A_2 values (i.e. 1 significant figure); unlimited values are considered. While the chart seems similar to that of Q_c (cf. figure 3), the distribution among radionuclides is not correlated, as 59% of the radionuclides Q_c and $Q_{D,skin}$ values do not follow the same trend (increase, decrease, no change).

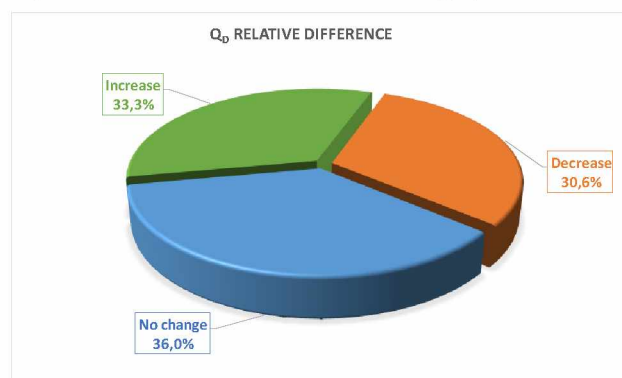


Figure 6. Relative modification in $Q_{D,skin}$ values between current and new methods

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DOSE DUE TO SUBMERSION IN NOBLE GASES (Q_E)

In the proposed revised Q system, the parameters for calculating Q_E remain unchanged. Only the dose coefficients are updated from ICRP Publication 144 [26] for effective and skin equivalent dose coefficients, and ICRP 137 Publication for ^{220}Rn and ^{222}Rn . Though the U.S. Federal Guidance Report No. 12, currently in use, was updated in Report No. 15, the ICRP publications were retained for the sake of consistency with the method considered to review and update the Q system.

ICRP Publication 151 [25] proposes coefficients for different room sizes, that would be closer to the original intent of considering a limited room volume (300 m^3); however, those coefficients only exist for effective dose, not equivalent dose to the skin. In the end, the WG decided to consider the semi-infinite cloud model as in the current Q system (i.e. in the U.S. Federal Guidance Report No. 12), justifying the use of the dose coefficients from ICRP Publication 144 [26]. It is important to underline that those coefficients are calculated with polygon mesh skin models of the adult phantoms (male and female) from ICRP Publication 145 [27], which significantly refines the phantoms used in ICRP Publication 110 [18]. The equivalent skin dose is now estimated at depth between $50\text{ }\mu\text{m}$ and $100\text{ }\mu\text{m}$, no longer on the entire “skin organ” as it used to be.

As for Rn isotopes, it is reminded that special consideration is given to ^{220}Rn and ^{222}Rn as their decay products are solid radioisotopes that can be deposited in the lung while breathing, thus delivering an inhalation dose. ICRP Publication 32 [13] was used to only address the ^{222}Rn case. ^{220}Rn , which faces the same issue, is not considered in the current Q system though it can be found e.g. in the ^{232}Th decay chain. Since inhalation pathway corresponds to the Q_C scenario, that Rn is a gas and that the dose coefficients are given in the same unit (Sv/Bq) as those considered in the formula to derive Q_C values, this formula was used considering a 100% release fraction; while the current Q system considered it is a Q_E value because the release of Rn corresponds to the Q_E scenario, the calculation method used was that of Q_C . In those Rn special cases, the inhalation pathway delivers much higher doses than the external exposure pathway considered in Q_E .

Eventually, none of the new Q_E values decrease compared to those derived in the current Q system.

10-DAY RULE: CONSIDERING 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. This rule states that:

- if the half-life of daughter radionuclide is less than 10 days and lower than that of the parent radionuclide, then the mixture is considered in equilibrium;
- in all other cases, the radionuclides should be considered in a mixture law by the consignor / designer.

This rule was first introduced in the 1973 edition of the regulations as follows: if the daughter radionuclide is assumed to come into equilibrium with the parent during a transport of a maximum period of 50 days, A_1 is calculated for both the parent and the daughter, and the most limiting of the two values is assigned to the parent nuclide, with the mixture then considered as a “single radionuclide”. The same rule was applied to parent radionuclides with a short-lived daughter with a half-life not greater than 10 days. In the current Q system, a mixture rule is applied considering the activity of the parent only.

While the principles seem simple, the WG noted that for many radionuclides considered in “transient” equilibrium, the stated rules seem to have inconsistencies in SSG-26, as different deviations to those rules had to be considered by members of the WG to retrieve the current values. For example, with the $^{47}\text{Ca} / ^{47}\text{Sc}$ couple stated in SSG-26, it appears that the current value was calculated after 10 days of in-growth using a mixture rule instead of assuming equilibrium, with the parent and the daughter

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radionuclides considered as a single radionuclide “Ca-47+”. Because of the assumption that the parent radionuclide includes decay radiations of its daughters, the resulting Q value should always be lower than that of the parent alone. Of the 21 radionuclides from SSR-6 Table 2 likely to be in transient equilibrium, at least six do not respect that principle when Q_A is derived.

Another question was raised: how to deal with complex decay chains with several branches, some of them having half-lives higher than that of the parent? The simple and practical answer is to consider the full chain in equilibrium (though this would be conservative). The issue was especially clear for ^{230}Pa for which a significant part of the decay chain was not considered in the current Q system. The WG recognized that a risk of error appears in the calculation, by a consignor, of the quantity of A_1 or A_2 to be put in a package for the theoretical situation in which a mixture contains radionuclides that can belong to two or more different decay chains with different parents.

In the end, for all radionuclides in which the 10-day rule applies, the WG evaluated the Q and A values at equilibrium, whether “transient” or “secular”, using the current criteria.

The WG then suggested that the A values could be given without progeny and that the user should work out the value for mixture transported. While this possibility would clarify the use of Table 2 [1], be more accurate for the user and remove the risk of error when dealing with radionuclides coming from different chains, this would also transfer the burden to evaluate the A value to the consignor, determining a transport time at which this value is the lowest, which is a different method from that applied for more than 50 years. The final decision will be taken during the revision cycle.

NEW A_1 AND A_2 VALUES

The WG calculated updated A_1 and A_2 values. The main changes are presented in the charts of figure 7 hereafter with the same assumptions as the Q system. In particular, the WG considered: the upper threshold of 40 TBq, no multiple pathway summation [4], the same radionuclides in equilibrium as in SSR-6 Table 2. Both the databases and the calculation tool (CORAL, cf. ref [4]) underwent a successful validation process. A more detailed analysis of the decreasing A values is provided in [12].

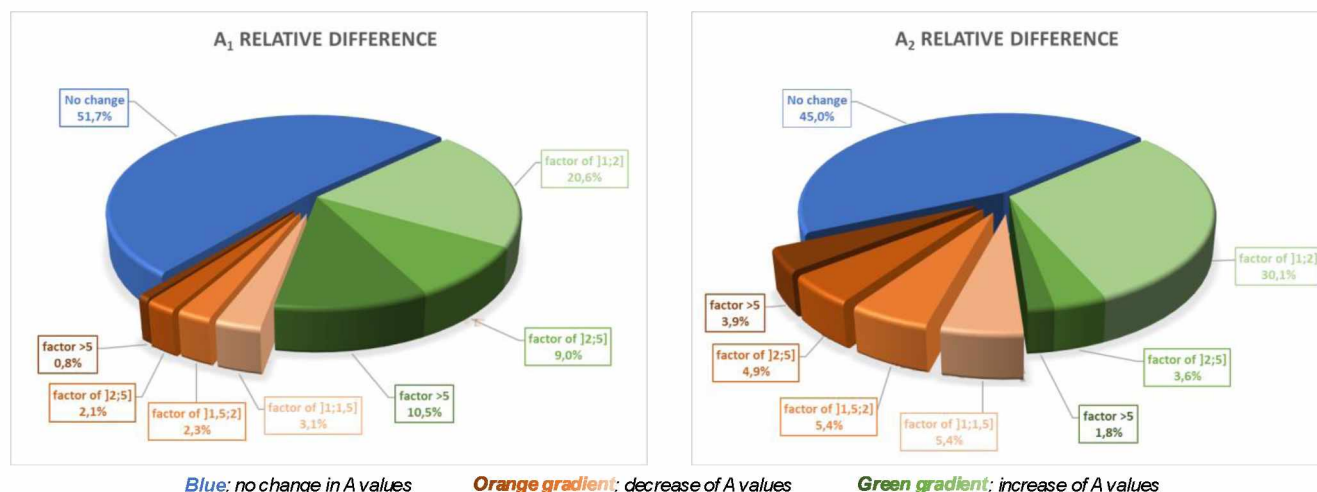


Figure 7. Relative modification in A_1 and A_2 values between current and new methods

Considering multiple pathway exposure [4], the A_2 values that would decrease would be about 35% instead of 20%.

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CONCLUSIONS

Six years after deciding on the final method to derive the new Q and A values, the WG A₁/A₂ could produce new values and clarify the Q system through a rigorous scientific approach and validation process (data, calculations and software). This method, implemented in the CORAL software developed by the WG, allows for a quick update of the values as long as the exposure scenarios do not change, and for producing Q and A values for all 1252 radionuclides of the ICRP publications – with or without their progenies.

The consideration of the latest ICRP data and recommendations, in addition to the new calculation approach by Monte Carlo method, leads to decrease 8,3 % of A₁ values and 19,5 % of A₂ values.

The WG will produce a fully documented report including all data to be used in other situations related to radiation protection. The interim report describing the method and results was made available to the TRANSSC member states before their 45th meeting; the final report will also include the comments from the members states. The decision to update the A₁ and A₂ values will be taken by the TRANSSC during the revision cycle, considering different aspects (practices, financial consequences, use of new radionuclides – especially medical isotopes, industrial aspects, etc.).

The proposed values should remain stable for a fairly reasonable period of time. As ICRP updates data and recommendations on a regular basis (e. g. new phantom recently published), those new values will likely be challenged in the future. More precise models could also be tested. However, now that the method is fully documented, the change in A₁ and A₂ values should be easier to assess before taking the decision to update them, considering the philosophy of the Q system.

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