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Fast determination of ^{90}Sr in urine samples and internal dose evaluation in emergency situations

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ABSTRACT

During nuclear or radiological emergency, it is important to rapidly identify contaminated people to administer them necessary treatments as soon as possible. A simple method to project and adapt measurement techniques for a mass monitoring of internal contamination based on the concept of “minimum detectable dose” has been developed. This method has been applied to ^{90}Sr, as it has a specific significance for spread in anthropic activity and radiotoxicity, so to require a fast, precise and suitable technique for its determination in many biological samples. A chromatographic separation allowed to isolate and measure by gas proportional counting ^{90}Sr excreted into urine and to calculate the correspondent intake and committed effective dose within half a day, taking also into account an overestimation of 10 % of total activity due to ^{90}Y not eliminated or formed during the measurement. The capacity to attribute values of committed effective dose lower than the precautionary reference level of 10 mSv, which is generally considered safe and does not typically cause health observable effects, has proved the method efficacy, even for a sample collection range larger than 20 days from the intake and for the most dangerous Type S of the investigated radionuclide.

1. Introduction

Emergency situations associated with the release of significant amounts of radioactive material into the environment, whether of accidental or malicious origin, require the activation of a complex series of interventions aimed at mitigating the effects on the affected people, among which those in the field of radiation protection are obviously harmful.

Considering the general, often disproportionate perception of radiological risk, one of the most critical aspects in such situations is undoubtedly the possibility of radioactive contamination (internal contamination) of the individuals involved. Although it is highly unlikely the contamination would lead to internal dose levels high enough to cause deterministic effects in this type of scenario, it is still possible that it may result in effective doses that significantly increase the risk of long-term stochastic effects. Given that these events frequently generate a strong impact on insiders, it’s very likely or quite sure that they would have a heavy psychological impact on the public, also in the presence of low or even negligible levels of contamination.

In this context the ability to conduct sufficiently accurate evaluations of exposure levels (internal dose) in a timely manner for each person involved is a fundamental tool, not only for identifying individuals who may require health intervention, whether it be actual medical treatment or simply the inclusion in long-term follow-up lists, but also for facilitating the social management of the event. To this purpose, a quick and reliable quantification of the actual individual risk is undoubtedly of basic importance.

In a situation where radiation protection criteria must be substantially adjusted to specific needs, even adopting, if necessary, reference dose levels that are significantly higher than those used in standard radiation protection practices, as suggested by the ICRP itself (International Commission on Radiological Protection, 2009; International Commission on Radiological Protection, 2020), the aforementioned "speed" is closely related to the capacity to carry out a sufficiently

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high number of appropriate individual measurements in a short time. Achieving this goal requires not only the involvement of multiple specialized laboratories but also, and above all, the adoption of measurement techniques specifically designed and validated to this aim.

From this perspective, while simplified in vivo measurement methodologies (whole-body counting, organ counting) have been extensively tested and adopted in many countries as suitable and easy-to-use solutions for gamma-emitting radionuclides (Franck et al., 2012; Ciciani et al., 2021), internal contamination monitoring for pure alpha emitters and pure beta emitters is much more complex to address. In most cases, there are in fact no truly alternative methods useful for screening, and standard radiotoxicological analysis procedures for excreta are often too complex, time spending and costly for large-scale implementation. In these cases, the only realistically feasible solution, despite its limitations, is to reconfigure the analysis protocols to seek a compromise between the quality of the expected result and the simplification of analytical and sampling techniques. Given the hypothesized conditions (monitoring in a short time a large number of individuals of the public), this approach can only be directed toward the analysis of small volume urine samples.

The aim of this work is to present the results achieved by the ENEA Radiation Protection Institute in studying and developing a method to be used in emergency situations for the rapid monitoring of internal contamination by ^{90}Sr. It is based on the analyses of very small urine samples (30 ml) performed by counting the total beta activity immediately after the chromatographic separation of strontium, so to have the opportunity to collect and analyze many samples in a low expensive and time-consuming way.

This monitoring method was designed by adopting an original criterion founded on the concept of “minimum detectable dose” and that can be extended to all types of internal contamination measurements. It was specifically studied for the prior identification of which parameters to "play with" for defining an analysis procedure capable of detecting all contaminations leading to the exceedance of a predetermined effective dose value assumed as a Reference Level, as well as delineating the useful application times (in this case, the time interval between the moment when the contamination occurred and the collection of the urine sample).

In compliance with the ICRP indications for emergency situations (International Commission on Radiological Protection, 2009; International Commission on Radiological Protection, 2020), the condition that must be guaranteed is that the Reference Level, expressed in terms of committed effective dose, must be included at least within the range 20 mSv–100 mSv. In this work it is conservatively set at 10 mSv.

It’s important to point out this study emphasizes the focus on contamination from pure ^{90}Sr sources or from mixed sources where significant interferences from other beta-emitting strontium isotopes, such as ^{89}Sr, are not regarded as significant. A brief analysis of this last specific case will be included in the conclusions, along with references to other publications for a more detailed treatment.

1.1. Minimum detectable activity (concentration) and minimum detectable dose

As already mentioned in the introduction, in order to choose the best way of intervention for managing a radiological emergency taking care of people involved, it is necessary to establish a *dose reference level, RL*, which could be considered as a sort of a *decision threshold* for the discrimination of the cases to be treated as relevant or not and for the adoption of appropriate actions.

In general, in internal dosimetry the measurement data cannot be immediately compared with pre-established reference levels but must firstly be interpreted in terms of intake and dose (effective committed dose). In a situation where the comparison should be immediate or, at least, very fast, it’s useful to introduce the concept of “minimum detectable dose”, MDD(t), defined as the dose corresponding to a measured activity equal to the minimum detectable activity, MDA, characteristic of the measurement technique adopted. Limiting the analysis to monitoring performed by measurement of urine samples, MDD(t) is expressed by the following formula:

$$MDD(t) = e(50)\cdot\frac{MDA_{24h}}{m(t)} \tag{3}$$

where: t is the time (days) elapsed between intake and collection of urine sample.

e(50) is the committed effective dose coefficient (mSv/Bq) for the radionuclide investigated its specific chemical and physical form and the route of intake,
MDA_{24h} is the value of the minimum detectable activity of the radionuclide normalized to 24-h excretion (Bq/24h);
m(t) is the proper urinary excretion function (fraction of the intake excreted by urine in the day t after the contamination);

Indicating with V the volume in litres of urine analyzed, with R_V the ratio between the volume of daily urinary excretion for the Reference Man (1.6 L/day (International Commission on Radiological Protection, 2002)) and the volume V, and with MDA_V the minimum detectable activity obtained by analysing that volume of urine, we obtain:

$$MDA_{24h}(Bq/24h) = MDA_V\cdot R_V = MDA_V\cdot\frac{1.6}{V} \tag{4}$$

Taking into account that the value of *MDD(T)* increases over time as *m(t)* decreases, the measurement technique adopted can therefore be considered suitable for the specific monitoring as long as the following condition is satisfied:

$$MDD(t) = e(50)\cdot\frac{MDA_{24h}}{m(t)} = e(50)\cdot MDA_V\cdot\frac{R_V}{m(t)} < RL \tag{5}$$

In order to design a measurement technique and its execution procedure so that this condition is respected, first of all it is necessary to make explicit the parameters involved, then evaluating any possible “combinations” of them in order to obtain the appropriate *MDD(t)* values.

Given that the measurement technique here considered are only those where the outcome of the detection of a radiation is expressed in terms of counts, to this end we proceed as follows.

Once assumed a pre-established Dose Reference Level, *RL*, the corresponding intake values *I(RL)* for the specific internal contamination is given by

$$I(RL) = \frac{RL}{e(50)} \tag{6}$$

where, as in equation (3), *e(50)* is the committed effective dose coefficient (mSv/Bq) for the radionuclide investigated, its specific chemical and physical form and the route of intake.

Multiplying *I(RL)* by the appropriate value of the urinary excretion function, *m(t)*, we obtain the activity $A_{24h}(t)$ expected in the urine excreted in the day, *t*, from the contamination by:

$$A_{24h}(t) = I(RL)\cdot m(t) \tag{7}$$

Indicating again with V the volume in litres of urine analyzed and with R_V the ratio between V, and the daily urinary excretion for the Reference Man, the activity $A_S(t)$ expected in the analysis sample S collected at a time t from the contamination is obtained by:

$$A_S(t) = \frac{A_{24h}(t)}{R_V} = A_{24h}(t)\cdot\frac{V}{1.6} \tag{8}$$

Multiplying $A_S(t)$ by the counting time T_m adopted and by the probability, Y (yield), of emission of the specific radiation considered, we have:

$$D_{S,T_m}(t) = A_S(t) \cdot T_m \cdot Y = I(RL) \cdot m(t) \cdot \frac{V}{1.6} \cdot T_m \cdot Y \tag{9}$$

where $D_{S,Tm}(t)$ is the expected number of radiations emitted during a measurement of duration T_m due to activity $A_S(t)$ of the radionuclide present in the urine sample used for analysis.

Considering the measurement efficiency, ε, and the chemical recovery, ρ, obtained by the pre-treatment of the sample, it is possible to calculate the expected number of counts recorded by the instrument with the following:

$$C_{S,T_m}(t) = D_{S,T_m}(t) \cdot \varepsilon \cdot \rho \tag{10}$$

that is:

$$C_{S,T_m}(t) = I(R_L) \cdot m(t) \cdot \frac{V}{1.6} \cdot T_m \cdot Y \cdot \ \varepsilon \cdot \rho \tag{11}$$

Therefore, indicating with L_D the detection limit (ISO 11929, 2010) of the measurement technique expressed in terms of total counts, the measurement technique is suitable if the following minimum criterion is satisfied:

$$C_{S,T_m}(t) \geq LD \tag{12}$$

that is:

$$I(R_L) \cdot m(t) \cdot \frac{V}{1.6} \cdot T_m \cdot Y \ \cdot \varepsilon \cdot \rho \geq LD \tag{13}$$

where LD must be calculated using as a “blank” a set of samples of urine of volume V produced by surely not-contaminated persons and applying exactly the same measurement procedure and parameters (counting time, chemical recovery, detection efficiency) established for the evaluation of $C_{S,Tm}(t)$.

From a practical point of view, the condition set in Eq. (13) must be satisfied for urine samples collected for several days after the intake, especially in events involving many people. Given the dependence of the first term of the equation on $m(t)$, whose value obviously decreases over time, the duration of the time interval useful for urine collection (Δt_{UC}) is anyway limited.

The time interval for useful collection, Δt_{UC}, is a parameter related to the condition of applicability of the method with respect to the Reference Level adopted (equation (13)). It indicates the duration of the time interval in which, applying a specific monitoring procedure, sample collection is appropriate to achieve pre-established performance, that is: all contaminations giving a committed effective dose above the reference level shall be detected; in other words: the minimum detectable dose of the procedure is below the reference level if the sample is collected in the interval Δt_{UC}. It could be even not significant or appropriate in terms of management of the event, however, given the possible need to use the monitoring procedure in events that lead to a mass contamination, Δt_{UC} should anyway be long enough to allow the collection of urine for the greatest number of people involved, taking into account the almost inevitable inconveniences and errors that this situation would certainly entail, especially when dealing with members of the public. For this reason, the conditions of acceptability of the monitoring procedure will also include a condition on Δt_{UC}. The condition here assumed is that it must be not less than 5 days, if possible, although this value is not to be taken as mandatory, but as a prudential indication to be considered and evaluated on the basis of the actual situation to be faced.[1]

From this equation it is highlighted that parameters on which it is possible to directly intervene in order to define a proper measurement procedure mainly are: the value of $I(RL)$ (i.e assuming a different value of the reference level, RL), the quantity of urine analyzed (V), the counting time (T_m) and the chemical recovery (through the choice of a proper pre-treatment method); the value of the excretion function, $m(t)$, essentially defines the time interval for a useful sample collection, while the counting efficiency, which must obviously be maximized, is substantially dependent on the type of instrument used for the measurement and its set up.

Given the goal is simplifying and shortening as much as possible the monitoring method to perform the largest number of measurements per day respecting the above mentioned conditions (equation (12)), it must be underlined that.

- increasing the reference level, RL, is the simplest, most effective and fastest way to achieve the objective, taking into account that no changes to the measurement method would even be necessary; however, this choice must be carefully evaluated and balanced as, by abusing it to the point of substantially modifying pre-established monitoring conditions, it could lead to serious repercussions both from a strictly technical-health point of view and from that of the political management of the event;
- increasing the quantity of urine analyzed, V, should be a way to obtain a gain in terms of expected counts (value of $C_{S,Tm}(t)$); conversely it could have an unfavorable impact on the absolute value of the detection limit, LD, because of the variations on measurement parameters, and especially on the productivity (increasing of the duration of the procedure, lowering of the number of samples that can be processed simultaneously by a single operator);
- counting time, T_m, is a parameter that can be effectively used to optimize the procedure only when it lasts a few hours or less. It is important to consider that an increase in Tm directly impacts on the duration of the procedure. Meanwhile, the gain in terms of the ratio between $C_{S,Tm}(t)$ and LD typically follows the trend of the square root of the relative increase of T_m itself, assuming that the hypothesis of Poisson statistics is applicable;
- chemical recovery, ρ, can represent a significant factor only when it is very low and variable; in the case of small urine samples pre-treated as discussed in this work, it is generally very high (much higher than 70 %) and stable, and it hardly constitutes a parameter on which to operate to optimize the procedure.

All must be commensurate with the need to be able to carry out as many measurements as realistically possible in a short time, which is why in the planning phase the sample pre-treatment methodology adopted in this work has been carefully selected and evaluated, through many tests, not only in terms of duration and capacity of achieving a high and stable chemical yield, but also in terms of the number of samples that can be processed by a single operator in a fixed, reasonable time (for example, the duration of a work shift).

2. Experimental

The standard methodologies (Horwitz et al., 1992; Water quality) commonly adopted in monitoring internal contamination by ^{90}Sr are based on the chromatographic extraction and separation of the radionuclide in urine samples (volumes of a few hundred mL, typically 500 mL) and on the evaluation of its activity by measuring the ingrowth of the daughter, ^{90}Y (Water quality; Ridone et al.), or following its decay after it has reached equilibrium in activity with the parent. Both processes require many days to obtain reliable measurements and are therefore not suitable for use in emergency situations or any case where a large number of people are involved. Gross beta counting with multiple channel proportional counters can be an effective and rapid way to measure simultaneously many samples together, after the removal of interfering alkali metals during the phosphate co-precipitation in aqueous solutions. However, as ^{90}Y, both excreted and produced by its parent nuclei, co-precipitates with ^{90}Sr, it is impossible for the

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[1] It’s important to point out that $\Delta\tau_{UC}$ must not be confused with the duration of sample collection (of urine) which can be a spot collection or extended up to 24 h as a maximum.

instrument to distinguish the contribution of the two radionuclides during the measurements, so that the overestimation of the activity could even be of 50 %.[2]

2.1. Reagents and materials

Ultrapure grade HNO_3, $Ca(NO_3)_2.4H_2O$, NH_4OH, and H_3PO_4 were used in the chemical pre-treatment of samples. All the solutions were prepared with bi-distilled water, obtained by an Aquatron Automatic Water Stills A4000D. A standard solution of $^{90}Sr/^{90}Y$ in 0.1 M HCl, supplied by ENEA National Institute of Ionizing Radiation Metrology (INMRI), was used for the preparation of spiked samples at different activity. Chromatography columns for radionuclide separation were prepacked with 1 M 4,4′(5')-di-t-butylcyclohexano 18-crown-6 in 1-octanol resin (Sr Resin™) and provided by TrisKem International (France).

2.2. Instrumentation

Sample activity was measured in steel planchets, placed in a 10-channel Low-level Proportional Counter LB 770, a proportional gas flow counter with two separate measuring channels for alpha and beta activities and manufactured by Berthold Technologies (Germany). The counting efficiency for $^{90}Sr/^{90}Y$ in secular equilibrium was determined in all detectors by the use of $^{90}Sr/^{90}Y$ certified solid sealed sources supplied by Amersham. For the purposes of this work, the counting efficiency of the measuring system for only ^{90}Sr and only ^{90}Y were experimentally evaluated using a certified $^{90}Sr/^{90}Y$ liquid source (Strok et al., 2006, 2008).

2.3. Chemical treatment and measurements

After the addition of 2 mg of Sr carrier as a tracer to a 30 ml urine sample directly in a 50 mL centrifuge tube, 1 mL of 3M $Ca(NO_3)_2$ and 5 mL of concentrated H_3PO_4 were added, and the pH adjusted to 8.5–9 with 10 mL of concentrated NH_4OH to allow the formation of calcium phosphate precipitate with the co-precipitation of strontium. The precipitate was let to settle down for 10 min and the sample was centrifuged at 3000 rpm for 15 min. The supernatant was discarded, the precipitate was rinsed with 4M NH_4OH and centrifuged again for 15 min. Finally, it was dissolved and transferred in a glass beaker with 5 mL of concentrated HNO_3 together with 3 rinses (5 mL conc. HNO_3 each) of the centrifuge tube and it was evaporated to dryness on hot plate with some drops of H_2O_2. The residue was dissolved in 10 mL of 8M HNO_3 and loaded on a Sr-spec column previously conditioned with 5 mL of 8M HNO_3. After the sample was drained, a rinse solution of 8M HNO_3 (5 mL) was used to wash the sample beaker and the column. Strontium was then eluted with 10 mL of 0.05M HNO_3 and an aliquot of 9 mL (90 %) of this solution was quantitatively transferred to a stainless steel disc, heated to dryness on hot plate, transferred into the proportional counter and measured for 6 h. The remaining 1 mL aliquot was diluted to fall within the linear range of Atomic Absorption Spectroscopy (AAS), which was used to evaluate chemical recovery, that was performed just during gross beta measurement in order not to prolong the overall time of the procedure.

In Fig. 1 a schematic representation of this procedure is shown.

2.4. Measurement

The measurement of gross beta emission was performed using a 10-channel low-level planchet Gas Proportional Counter (Berthold LB 770) for 6 h.

The activity per unit volume A_V(Bq/L) of each sample is given by the following formula:

$$A_V = \frac{CR_S - CR_B}{\varepsilon_{90_{Sr}} \cdot \rho \cdot V \cdot k_V} \tag{14}$$

where: CR_S is the count rate of the sample (cps)

CR_B is the count rate of the blank (cps)[3]
ε_{90Sr} is the counting efficiency for ^{90}Sr
ρ is the chemical recovery
V is the volume of the sample (L)
k_V is the correction factor for the reduction of the analyzed sample volume due to the aliquot of eluate used for the chemical recovery evaluation (in this case K = 0,9)

The daily urinary excretion of ^{90}Sr activity (with the approximation that all counts are due only to strontium) is given by:

$$A_d = R_V \cdot A_V = 59,3 \cdot A_V$$

where R_V is the ratio between the volume of daily urinary excretion for a Reference Man (1600 mL/day (International Commission on Radiological Protection, 2002)) and the volume of urine effectively analyzed (in this case 30 mL x 0,9).

The evaluation of the chemical recovery was made in AAS by comparison of the concentration of Sr-carrier measured in two standard solutions prepared for the calibration of the spectrophotometer.

3. Results and discussion

This method has been tested on two sets of 8 samples each by two different laboratories (ENEA CR Saluggia and ENEA CR Trisaia) and the mean chemical yield obtained is 70 %. The Minimum Detectable Activity per unit volume MDA_V(Bq/L) has been also determined with the following formula:

$$MDA_V = \frac{2.71 + 4.65 \cdot \sqrt{CR_B \cdot T_M}}{\varepsilon_{90_{Sr}} \cdot \rho \cdot V \cdot k_V \cdot T_M} \tag{15}$$

where T_M(s) is the measuring time[4] and the other quantities defined as in eq. (1).

The MDA_V obtained with this method is 2.48E-01 Bq/L and the MDA_{24h} is 3.97E-01 Bq/d considering the daily urinary excretion of 1.6 L/d. In this case we consider small values of the relative systematic uncertainty, so the correction that ISO 11929 should make to Currie formula can be considered negligible (Kirckpatrick et al., 2015)

This procedure is based on the hypothesis that the entire activity present, net of the blank, can be attributed to ^{90}Sr. This hypothesis needs that the above-mentioned treatment leads to a complete purification of the sample from other beta-emitting radionuclides, in particular radiocesium[5] and ^{40}K, and an almost total elimination of ^{90}Y that should be present because of its excretion and its ingrowth due to ^{90}Sr decays from the time of urine collection. In any case, the method involves an overestimation of the activity due to.

- the ^{90}Y possibly not eliminated by treatment[6]

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[2] Experimentally evaluated through gross beta analyses (Attrep and Kahn).

[3] Mean value of 5 urine samples surely not contaminated.

[4] The measuring time of 6 h garantees to get a sufficient low MDA_{24h} given the small sample volumes.

[5] In ^{90}Sr separation with Sr Resin™, cesium is not considered retained, or at negligible rate, by the resin if loaded in 8M HNO_3, as reported by Eichrom methods (triskem).

[6] In this work, the effective removal of ^{90}Y from the column was experimentally evaluated via the decay curve of yttrium present in the waste solution, and it was found to be equal to 96 %.

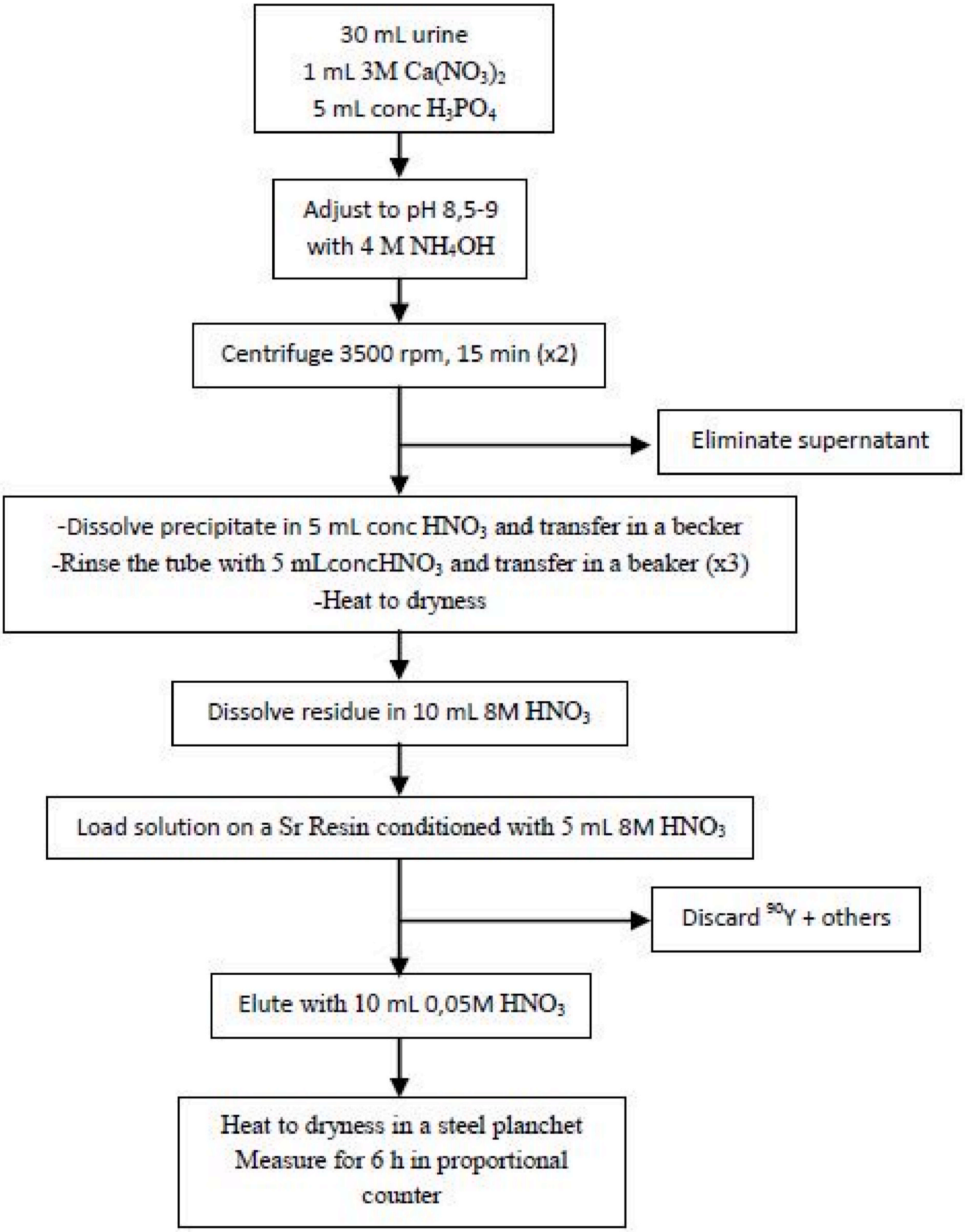

Fig. 1. Scheme of isolation of ^{90}Sr by chromatographic extraction with Sr-Resin™.

- the quantity of ^{90}Y formed in the sample from the end of ^{90}Sr extraction and separation procedure to the completion of the counting (no more than 7 h)
- the use of the counting efficiency for ^{90}Sr even for the interfering ^{90}Y whose appropriate efficiency is much higher.

In total, the overestimation has been evaluated to be no more than 10 %, on average, where the maximum contribution is due to the newly formed ^{90}Y (about 6 %). This overestimation can be considered acceptable given the capabilities of the method, both in terms of the number of measurements that can be performed per day and the validation of the results obtained. It is quite clear that it is, however, possible to subsequently repeat the counting of the sample, even several times, to evaluate the regrowth of ^{90}Y, in order to be able to perform a more accurate evaluation of the intake and dose. This may be necessary especially when the level of contamination appears as critical with respect to the adopted Reference Level.

With adequate supplies of expert personnel and the organization of a proper work shift, this method, or an alternative one with comparable performances, allows a single, properly equipped “working line”, to complete up to 90–100 measurements of ^{90}Sr in 3 days.

3.1. Acceptability of the monitoring procedure and dosimetry evaluation of the method

The method proposed here is designed on the basis of the considerations set out in the previous paragraph applied specifically to ^{90}Sr as unique radionuclide present in the sample, keeping in mind that this approximation leads to an acceptable overestimation of the activity (10 % as a maximum).

The typical *LD* value obtained with this monitoring procedure is 35 counts, so considering all the experimental parameters discussed in previous paragraphs, equation (12) is clearly satisfied for all strontium compound as it is reported in the following Table 1 for urine collected at the first seven days following the intake.

From the analysis of data in Table 1 it follows that the proposed procedure for the detection of ^{90}Sr contaminations giving committed effective dose values equal to the proposed reference level (10 mSv) is largely acceptable for all chemical forms and all compound, both for ingestion and inhalation of aerosol of AMAD (Activity Median Aerodynamic Diameter) of 1 µm and 5 µm.

Although the method obviously allows us to evaluate intake and dose for all the chemical forms of strontium considered in the ICRP publications, in emergency situations where it’s very probable not to have sudden information on the nature of the contaminating agent, it is appropriate as a first step to focus preliminarily on the worst case to be monitored, here represented by inhalation of S-type compound.

In the following tables, the main dosimetry data (excretion function *m(t)* (OIR Data Viewer), intake, minimum detectable dose (MDD_E) are shown for contamination by inhalation of ^{90}Sr in form of Type S compound both for 1 µm AMAD (Table 2) and 5 µm AMAD (Table 3) in the hypothesis that the activity of the radionuclide measured in urine sample (A_m) is equal to *MDA*. Data are given for collection of urine performed up to the seventh day after the contamination (the fifth day as a minimum) and allows to establish, applying the monitoring procedure, the maximum internal dose attributable when no contamination is detected. The last day from intake useful for urine collection (upper end of Δt_{UC}) is also reported.

Analysing data in the tables, it’s quite clear that the application of the method, even in the worst-case scenario of intake by inhalation of S-type aerosol, allows for the measurement of all doses above the reference level of 10 mSv even by urine samples collected after many days following the internal contamination event, particularly up to 20 days for AMAD = 1 µm and up to 35 days for AMAD = 5 µm.

Table 2
^{90}Sr: inhalation of type S aerosol, AMAD = 1 µm.

t (days)	m(t) (AMAD = 1 µm)	A_m = MDA (a) (Bq/24h)	Intake (Bq)	E_{50} = MDD_E(t) (b) (mSv)	Ratio E_{50}/L_R
1	3,4E-04	4,0E-01	1,2E+03	4,4E-01	4 %
2	1,9E-04	4,0E-01	2,1E+03	7,9E-01	8 %
3	1,2E-04	4,0E-01	3,3E+03	1,3E+00	13 %
4	8,8E-05	4,0E-01	4,5E+03	1,7E+00	17 %
5	6,9E-05	4,0E-01	5,8E+03	2,2E+00	22 %
6	5,7E-05	4,0E-01	7,0E+03	2,6E+00	26 %
7	4,8E-05	4,0E-01	8,3E+03	3,1E+00	31 %
22 (c)	1,6E-05	4,0E-01	2,5E+04	9,4E+00	94 %

(a) Both values of measured activity (A_m) and MDA are normalized to 24h excretion.
(b) Committed effective dose.
(c) Last day from intake useful for urine collection (upper end of Δt_{UC}).

Table 3
90Sr: inhalation of type S aerosol, AMAD = 5 µm Structured.

t (days)	m(t)(AMAD = 5 µm)	AM = MDA (a) (Bq/24h)	Intake (Bq)	E50 = MDDE (t) (b) (mSv)	Ratio E50/LR
1	3,6E-04	4,0E-01	1,1E+03	2,2E-01	2 %
2	2,2E-04	4,0E-01	1,8E+03	3,6E-01	4 %
3	1,4E-04	4,0E-01	2,8E+03	5,7E-01	6 %
4	9,7E-05	4,0E-01	4,1E+03	8,2E-01	8 %
5	7,5E-05	4,0E-01	5,3E+03	1,1E+00	11 %
6	6,1E-05	4,0E-01	6,5E+03	1,3E+00	13 %
7	5,1E-05	4,0E-01	7,8E+03	1,6E+00	16 %
36(c)	8,0E-06	4,0E-01	5,0E+04	9,9E+00	99 %

(a) Both values of measured activity (Am) and MDA are normalized to 24h excretion.
(b) Committed effective dose.
[c] Last day from intake useful for urine collection (upper end of ΔtUC).

4. Conclusions

The method for the rapid analysis of ^{90}Sr in very small urine samples (30 ml) described in this work looks a reliable and effective tool to be used to deal with massive internal contaminations involving ^{90}Sr. Infact.

- it allows for the detection of all internal doses by intake of ^{90}Sr above a reference level of 10 mSv for any kind of physical and chemical form of the contaminant and any route of intake,
- it can be performed using urine samples collected even many days after the contamination,
- it’s not affected by significant variability of the measurement parameters,
- it guarantees high and stable chemical recovery (70 %) and a good separation of ^{90}Sr from the most common interfering radionuclides such as ^{40}K and ^{137}Cs,
- it is only affected by a quite systematic but anyway conservative small overestimation (10 % as a maximum) of the measured activity of ^{90}Sr due to residual and in-growing ^{90}Y present in the measurement sample,

Table 1
Values of the parameter $C_{S,Tm}$ obtained for ^{90}Sr applying the proposed monitoring procedure to urine collected in the first seven days following the intake.

$C_{S,Tm}$ (counts) - intake of ^{90}Sr								
t	Inhalation of aerosol						Ingestion	
(days from intake)	Type S		Type M		Type F		Titanate	other compounds
(d)	1 µm	5 µm	1 µm	5 µm	1 µm	5 µm		
1	776	1560	197000	352000	1630000	1520000	1420000	1700000
2	434	954	113000	212000	694000	759000	599000	687000
3	274	607	72300	130000	451000	461000	386000	434000
4	201	421	52000	96300	330000	325000	284000	322000
5	158	325	43400	72300	257000	260000	221000	253000
6	130	265	34700	62600	208000	211000	181000	206000
7	110	221	29000	53000	173000	176000	150000	170000

- makes it possible to provide up to 90–100 measurements of ^{90}Sr in 3 days for each single, appropriate "working line" involved.

By optimizing several parameters, such as sample volume, counting time, minimum detectable activity (MDA) and minimum detectable dose (MDD), the procedure ensures timely individual dose assessments and can form the basis for effective radiation protection responses to face and mitigate the impact, at least psychological, that events of this kind most likely would cause, especially in the general population.

As previously mentioned, the main critical issue of the method is related to the presence of interfering substances in the contaminant mixture that cannot be eliminated through radiochemical treatment of the sample, specifically pure beta-emitting strontium radioisotopes, among which the most significant is ^{89}Sr. Referring to specific publications for an in-depth discussion of the analytical and measurement techniques for samples contaminated by a mix of ^{90}Sr and ^{89}Sr (Holmgren et al., 2014; Maxwell, 2008), it should be emphasized here that, due to its relatively short half-life (52 days), the presence of ^{89}Sr is highly unlikely in accidental events involving the release of aged nuclear waste or in dirty bomb attacks. Instead, it may be relevant primarily in cases of radiological emergencies caused by accidents at active nuclear facilities. In such cases, considering the substantial overlap in environmental dispersion of the different strontium isotopes, the evaluation of the respective contributions of ^{90}Sr and ^{89}Sr to the dose can be inferred by correcting the measured total beta activity on the basis of the release inventory, supported by appropriate measurements conducted on environmental samples.

CRediT authorship contribution statement

D. Arginelli: Writing – review & editing, Writing – original draft, Supervision, Data curation, Conceptualization. **M.C. Botta:** Writing – original draft, Formal analysis, Data curation. **S. Ridone:** Writing – review & editing, Writing – original draft, Data curation, Conceptualization. **S. Zicari:** Data curation. **P. Battisti:** Writing – review & editing, Writing – original draft, Validation, Supervision, Methodology, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors are unable or have chosen not to specify which data has been used.

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