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Additional Information





## 37 **Introduction**

38 In emergency situations, such as nuclear and radiological accidents, terrorist attacks using  
39 radiological dispersal devices (RDDs) or “dirty bombs”, and improvised nuclear devices  
40 (INDs), numerous short- and long-lived radionuclides will be present, requiring rapid  
41 identification and quantification. Most  $\gamma$ -emitters are easily determined by direct  $\gamma$ -  
42 measurement. However, the isolation of alpha and beta emitters is time-consuming  
43 although essential for their quantification. For this reason, there is a need to develop rapid  
44 analytical procedures for alpha and beta emitters.

45 Among pure-beta emitters, radiostrontium ( $^{90}\text{Sr}$  and  $^{89}\text{Sr}$ ) are hazardous radionuclides due  
46 to their long half-life ( $^{90}\text{Sr}$ ,  $T_{1/2}=28.79$  years) and the high energy of the beta particles  
47 emitted ( $^{90}\text{Y}$ , 2.28 MeV, from  $^{90}\text{Sr}$ , 0.55 MeV; and  $^{89}\text{Sr}$ , 1.50 MeV). In radiological  
48 accidents only  $^{90}\text{Sr}$  could be present, but  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  are both released in nuclear  
49 accidents. Although the  $^{89}\text{Sr}$  activity released is higher than  $^{90}\text{Sr}$  activity, it decreases  
50 faster due to its relatively short half-life ( $T_{1/2}=50.56$  days). For example, the estimation  
51 range of the atmospheric release in the Fukushima accident was 0.04 – 13 PBq for  $^{89}\text{Sr}$ ,  
52 and 0.003-0.14 PBq for  $^{90}\text{Sr}$  [1]. The release was much higher in Chernobyl,  
53 approximately 115 PBq of  $^{89}\text{Sr}$  and 10 PBq of  $^{90}\text{Sr}$  were released into the environment  
54 [2]. Moreover, the  $^{89}\text{Sr}/^{90}\text{Sr}$  activity ratio in Fukushima surface soil after the deposition  
55 ranged from 6.5 to 15.7, with an average of 13.3 [3].

56 Strontium isotopes in the environment can migrate through the food-chain into milk and  
57 dairy products. They are absorbed into the human body due to their similar chemistry to  
58 calcium and are especially dangerous to infants. For this reason, the European regulations  
59 limit the total strontium isotopes, notably  $^{90}\text{Sr}$  activity, in dairy products (included milk)  
60 after a nuclear accident or any other radiological emergency to 125 Bq/kg, and 75 Bq/kg  
61 for infant food [4].

62 Radiostrontium separation has always been a matter of concern and many alternatives  
63 have been proposed for its separation [5]. Several rapid methods of determining  
64 radiostrontium in milk samples, which differ mainly in their pre-treatment steps, have  
65 been developed in recent years [6-9]. Brunet. al. [6] and IAEA [9] recommended the use  
66 of cation exchange resins to remove fat and proteins from milk. Maxwell et al. [7]  
67 proposed a calcium phosphate precipitation, while Kabai et al. [8] reported the use of

68 trichloroacetic acid as acidifying agent for protein precipitation. In some methods, an  
69 oxalate [6, 8] or carbonate precipitation [9] is also employed to separate alkaline-earth  
70 elements from those of other groups before the isolation of strontium by extraction  
71 chromatography resins. Gas flow proportional counting and liquid scintillation counting  
72 can be used for measurement. The time needed for strontium determination is  
73 approximately 15 hours by the Brun method [6] and 7 – 8 hours for the Maxwell, Kabai  
74 and IAEA methods [7-9].

75 The time of analysis is the most important factor to be considered in an emergency.  
76 Although there are well-established procedures in the bibliography, in emergency  
77 situations it is important to make the results available in the minimum time. This study  
78 therefore mainly focused on the separation and measurement steps by plastic scintillation  
79 resins (PS resins), which are plastic scintillation microspheres coated with a selective  
80 extractant for strontium, 4,4'(5')-di-*t*-butylcyclohexano-18-crown-6 (DtBuCH18C6) in  
81 1-octanol. Their main advantage is that this resin can be directly measured with the  
82 radionuclide immobilised onto the plastic microsphere by scintillation counting, which  
83 permits the separation and measurement preparation steps to be combined in a single step;  
84 thus achieving a considerable reduction in time, effort and reagents. A further advantage  
85 is that, unlike Liquid Scintillation (LS), no mixed waste is generated.

86 In previous studies, Bagán et al. [10] employed PS resins to determine radiostrontium in  
87 aqueous samples with good results; relative bias for the determination of  $^{90}\text{Sr}$  activity in  
88 drinking, sea and river waters was less than 4%. Plastic scintillation resins coated with  
89 different extractants have also been used for  $^{210}\text{Pb}$  and  $^{99}\text{Tc}$  determination [11, 12]. One  
90 of the goals of the present work was to adapt PS resins for application to strontium  
91 determination in milk samples in emergency situations, taking different aspects into  
92 account, such as the conditioning and washing media of the columns and matrix effects.

93 This paper proposes a novel procedure that reduces the time required to determine  
94 radiostrontium activity in milk samples by PS resins in emergency situations, in which  
95 the separation and measurement preparation stages are joined into a single step. The study  
96 focuses on adapting PS resins to milk samples, and on optimizing the pre-treatment steps  
97 of the procedure. Different types of milk samples were analysed by different proportions  
98 of  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  in order to verify the proposed method and calibration system. The  
99 method was also validated by a reference material.

## 100 **Experimental**

### 101 *Reagents and materials*

102 All the reagents used were of analytical or reagent grade. 4,4'(5')-di-*t*-butylcyclohexano-  
103 18-crown-6 (DtBuCH18C6) (>95%), lithium nitrate (>99%), methanol (99%) and 1-  
104 octanol (98%) were supplied by Sigma-Aldrich (St. Louis, USA); ammonia (25%),  
105 trichloroacetic acid (>99.5%), oxalic acid and nitric acid (69%) were supplied by Panreac  
106 (Barcelona, Spain) and strontium nitrate was supplied by Merck (Darmstadt, Germany).  
107 Double deionised water was obtained from a Milli-Q Gradient A10 from Merck Millipore  
108 (Darmstadt, Germany).

109 The plastic scintillation microspheres used had a median diameter of 60  $\mu\text{m}$  and were  
110 composed of polystyrene, 2,5-diphenyloxazole and 1,4-Bis (5-phenyloxazol-2-yl).  
111 PSresins were prepared by deposition of a solution of DtBuCH18C6 in 1-octanol on the  
112 surface of the plastic scintillation microspheres. Both plastic scintillation microspheres  
113 and PS resin were prepared following procedures previously described by the authors [10,  
114 13].

115 A  $^{90}\text{Sr}/^{90}\text{Y}$  active stock solution of 37.2(3)  $\text{Bq}\cdot\text{g}^{-1}$  containing  $\text{Sr}^{2+}$  (100  $\mu\text{g g}^{-1}$ ) and  $\text{Y}^{3+}$  (100  
116  $\mu\text{g g}^{-1}$ ) in 0.1 M HCl was prepared from a standard of 4.07(3)  $\text{kBq}\cdot\text{g}^{-1}$  supplied by  
117 Amersham International (Buckinghamshire, England), and a  $^{89}\text{Sr}$  active stock solution of  
118 1.303(3)  $\text{kBq}\cdot\text{g}^{-1}$  containing  $\text{Sr}^{2+}$  (50  $\mu\text{g g}^{-1}$ ) in 0.1 M HCl was prepared from a standard  
119 of 103(3)  $\text{kBq}\cdot\text{g}^{-1}$  supplied by Eckert Ziegler (Berlin, Germany).

120 For PS resin support, 2 mL solid-phase extraction cartridges (SPE cartridges) and frits  
121 from Triskem International (Rennes, France) were used. The cartridges were measured  
122 within 20 mL polyethylene vials from PerkinElmer (Waltham, Massachusetts, USA).

### 123 *Milk samples*

124 Most milk samples were purchased in the local supermarket, including cow's, goat's and  
125 sheep's milk. The influence of fat content was also studied with whole (3.6g/100mL),  
126 semi-skimmed (1.6g/100mL) and skimmed (0.3g/100mL) cow's milk. The type of milk

127 treatment was considered by the analysis of milk powder, ultra-high temperature  
128 processed milk (UHT), pasteurized milk and raw milk.

129 A certified reference material (IAEA-473 milk powder) was analysed to validate the  
130 method.

### 131 *Equipment*

132 A Quantulus 1220 liquid scintillation spectrometer (PerkinElmer) was used with  
133 logarithmic amplification, a multichannel analyser (MCA) (4096 channels distributed in  
134 four segments of 1024), and alpha/beta discrimination and background reduction by  
135 active guard.

136 Elementary analysis was performed on an Optima 8300 ICP-OES spectrometer  
137 (PerkinElmer) in the laboratories of the University of Barcelona.

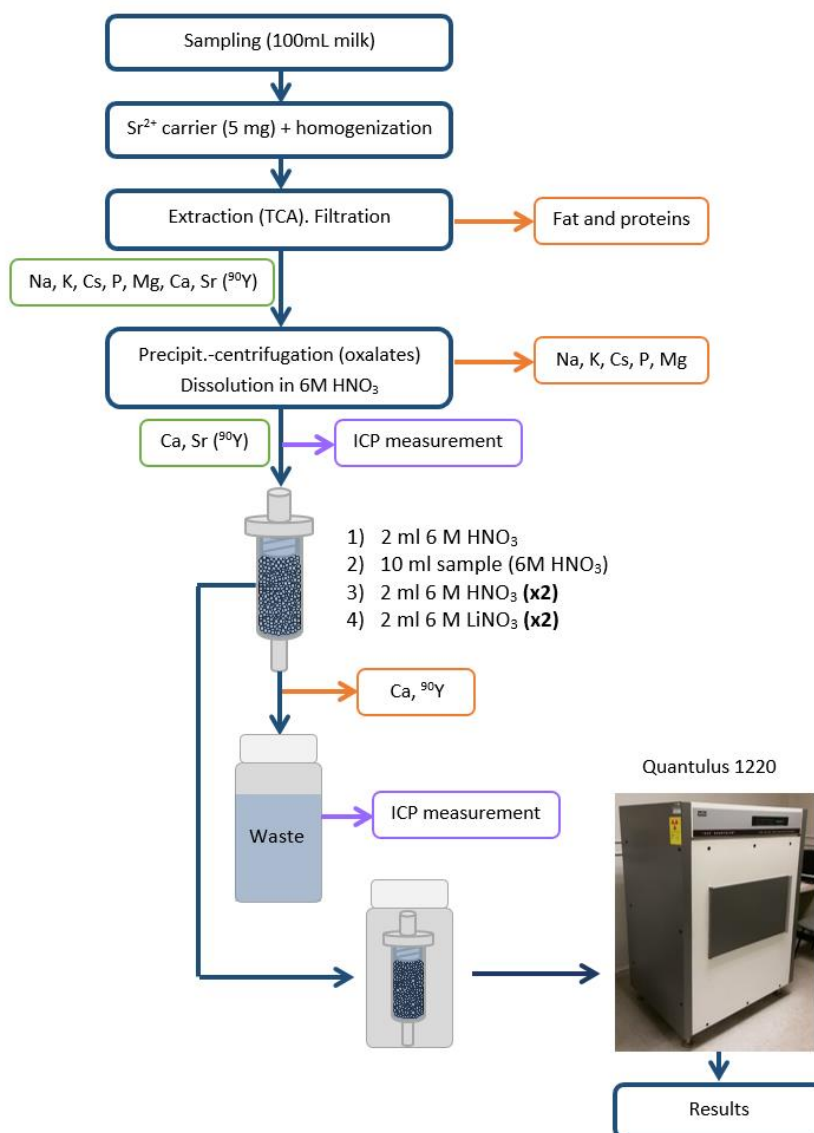
138 PS resins were prepared with an R-210 Rotavapor from Buchi (Flawil, Switzerland).

139 A MS 3 digital vortex from IKA (Staufer, Germany) was used to homogenize the PS  
140 resin in the cartridges.

141 A Beckman J2-HS (Indianapolis, Indiana, USA) was used to centrifuge 250 mL  
142 centrifuge bottles.

### 143 *Procedure*

144 The procedure used to analyse milk samples is composed of four steps: elimination of fat  
145 and protein; concentration of strontium and alkaline-earth elements; separation by PS  
146 resin; and measurement in an LS counter. A complete scheme of the method is shown in  
147 Fig. 1.



148

149 **Fig. 1** Detailed procedure for strontium determination in milk samples by PS resins

150 **Sample preparation**

151 A milk sample of 100 mL is analysed for radiostrontium determination after  
 152 homogenization. An optimized amount of strontium carrier (as  $\text{Sr}(\text{NO}_3)_2$  solution) is  
 153 added in order to obtain the chemical yield in the separation. Some of the samples are  
 154 also spiked with  $^{90}\text{Sr}/^{90}\text{Y}$  and/or  $^{89}\text{Sr}$  tracers for the validation of the procedure. The  
 155 samples are then homogenized on a hot plate by stirring with a magnetic stirrer for 15  
 156 minutes.

157 The milk is acidified for protein denaturation by mixing with 20% 2,2,2-trichloroacetic  
 158 acid (TCA) for 30 minutes, using the optimal ratio 1:1 (v/v) of TCA and milk sample.



159 After mixing, the precipitated fat and proteins are filtered through a Whatman GF/C paper  
160 filter (1.2  $\mu\text{m}$  pore size) in a Büchner funnel.

161 Oxalate precipitation is used to separate strontium and other alkaline-earth metals from  
162 the alkali metals group. After heating, the optimized amount of 5 g of oxalic acid is added  
163 to the warm filtrate solution and mixed for 5 minutes. The pH is then adjusted to 5–6 with  
164 ammonia, and the precipitate formed is centrifuged at 5000 rpm for 15 min and dissolved  
165 in 10 mL 6M  $\text{HNO}_3$  with the aid of a hot water bath to facilitate dissolution. An aliquot of  
166 the sample is taken to determine the chemical yield in the pre-treatment step by measuring  
167 strontium content by ICP-OES.

168 Strontium isotopes ( $^{89}\text{Sr}$  and  $^{90}\text{Sr}$ ) are isolated from other interferents present in the milk  
169 sample (mainly calcium and barium) and  $^{90}\text{Y}$  (in case of  $^{90}\text{Sr}$  presence) by plastic  
170 scintillation resins (PS resins). After an optimization of the separation process, the  
171 cartridge is conditioned with 2 mL 6M  $\text{HNO}_3$  before loading the sample solution. The  
172 resin is then rinsed with 2 mL 6M  $\text{HNO}_3$  (2 times) to remove the main interferents. The  
173 last rinse with 2 mL 6M  $\text{LiNO}_3$  (2 times) is performed to avoid chemiluminescence in the  
174 measurement due to the presence of nitric acid in the cartridge. The flow rate is about 1  
175  $\text{mL}\cdot\text{min}^{-1}$  in all cases. The load sample and all the rinses are collected in a 50 mL  
176 centrifuge tube and an aliquot is taken to measure the strontium concentration in the waste  
177 by ICP-OES. The difference between the strontium content in the sample solution and the  
178 waste gives the separation yield in the cartridge. Finally, the cartridge is placed in a 20  
179 mL polyethylene vial and is directly measured in a Quantulus 1220 detector.

#### 180 ***Measurement conditions***

181 All the measurements were performed in the Quantulus detector with the low coincident  
182 bias and the multichannel analyser (MCA) in the 14C configuration. Counting times were  
183 60 minutes (3 cycles of 20 minutes) for the blank and the active samples and 1 minute for  
184 the external standard gamma source. In all cases the cartridges were measured  
185 immediately after separation to avoid in growth of daughter radionuclides.

#### 186 ***Calibration***

187 Five water samples containing a known amount of  $^{90}\text{Sr}$  (7.5 Bq) and five water samples  
188 containing a known amount of  $^{89}\text{Sr}$  (0.5 Bq) were processed with PS resin and measured

189 in the scintillation counter for calibration of the detection efficiencies. In both cases the  
190 samples contained a variable amount of Sr<sup>2+</sup>: 2.7, 3.6, 4.4, 5.3, 6.4 mg of Sr<sup>2+</sup> in the case  
191 of the <sup>90</sup>Sr and 2.5, 3.3, 4.2, 5.0, 5.7 mg of Sr<sup>2+</sup> for the <sup>89</sup>Sr samples.

192 With the results obtained three calibration parameters were determined:

193 • Efficiency of <sup>90</sup>Sr: mean of the detection efficiencies in the total window for the  
194 samples that contain <sup>90</sup>Sr only.

195 • Efficiency of <sup>89</sup>Sr: mean of the detection efficiencies in the total window for the  
196 samples that contain <sup>89</sup>Sr only.

197 • Combined efficiency (<sup>90</sup>Sr + <sup>89</sup>Sr): mean of the detection efficiencies in the total  
198 window for all the calibration samples.

### 199 *Activity determination*

200 Total radiostrontium activity (<sup>90</sup>Sr + <sup>89</sup>Sr) in the samples was calculated by the following  
201 expression:

$$202 \quad A(^{90}\text{Sr} + ^{89}\text{Sr}) = \frac{\text{net count rate in the total window}}{\text{Eff}_{\text{comb}} \cdot R_1 \cdot R_2} \cdot 100 \quad (1)$$

203 where Eff<sub>com</sub> is the combined efficiency of <sup>90</sup>Sr and <sup>89</sup>Sr, R<sub>1</sub> being the pre-treatment  
204 recovery and R<sub>2</sub> the PS resin retention.

205 In the samples known not to contain <sup>90</sup>Sr or <sup>89</sup>Sr (i.e. the reference material) the activity  
206 of the radionuclide present in the sample can be calculated from its corresponding  
207 detection efficiency.

### 208 *Data treatment*

209 For the optimum visualization of the shape of the spectra obtained, the spectrum acquired  
210 for each measurement was smoothed using a Savitzky–Golay algorithm with an average  
211 window of 10 points for each side [14]. The normalized net spectrum was obtained by  
212 subtracting the spectrum of the equivalent blank solution and dividing the spectrum by  
213 the net count rate in the full window.

214 Detection efficiency was calculated as the ratio between the net counts in the full window  
215 and the activity passed through the PS resin.

216 The Standard External Quenching Parameter (SQP(E)) was calculated by the detector.  
217 SQP(E) corresponds to the end-point channel that limits 99.5% of the total counts from  
218 the spectrum generated by the external gamma source ( $^{152}\text{Eu}$ ).

219 Pre-treatment recovery ( $R_1$ ) was calculated from the percentage ratio between the total  
220 amount of  $\text{Sr}^{2+}$  contained in the sample after adding the  $\text{Sr}^{2+}$  carrier and the amount of  
221  $\text{Sr}^{2+}$  contained in the solution before being passed to the PS resin. PS resin retention ( $R_2$ )  
222 was calculated from the percentage ratio between the amount of  $\text{Sr}^{2+}$  contained in the  
223 solution before being passed to the PS resin and the amount of  $\text{Sr}^{2+}$  contained in the wastes  
224 eluted from the SPE cartridge. Variability was obtained from the standard deviation of  
225 the triplicate samples values.

226 The detection limit was calculated with the blank samples following the equation  
227 proposed by Currie [15]:

$$228 \quad LD = 2.71 + 4.65 \cdot \sqrt{B} \quad (2)$$

$$229 \quad MDA = \frac{LD}{\text{Eff}_{\text{comb}} \cdot R_1 \cdot R_2 \cdot V \cdot t} \quad (3)$$

230 where LD is the detection limit (counts), B is the number of counts for the blank, V is the  
231 volume of sample (L), t is the counting time (s),  $\text{Eff}_{\text{comb}}$  is the combined detection  
232 efficiency of the standard solution,  $R_1$  is the pre-treatment recovery,  $R_2$  is the PS resin  
233 recovery and MDA is the minimum detectable activity ( $\text{Bq L}^{-1}$ ).

## 234 **Results and Discussion**

235 The optimized procedure proposed here uses plastic scintillation resins to achieve rapid  
236 separation and measurement of radiostrontium in milk samples. However, this complex  
237 matrix needs a number of pre-treatment steps before the complete isolation of strontium  
238 by extraction chromatographic separation (with PS resins) and measurement by  
239 scintillation counting. The optimization of the pre-treatment steps and the separation steps  
240 with PS resins are described in the following sub-sections.

241 *Optimization of separation with PS resin*

242 In previous experiences in using PS resin to determine radiostrontium, some changes  
243 were proposed to adapt the established procedure for determining radiostrontium in water  
244 samples to the requirements of milk analysis in emergency situations, including:

- 245 • using smaller diameter PS resin to improve separation and detection efficiencies  
246 (from 190  $\mu\text{m}$  to 60  $\mu\text{m}$ );
- 247 • using 2 mL SPE-extraction cartridges in a vacuum box instead of adapting 6 mL  
248 polyethylene vials with a peristaltic pump to simplify chemical separation; plus  
249 using 1 g of PS resin instead of 3 g due to the limited amount of PS resin that can  
250 be placed in the column;
- 251 • replacing the  $\text{LiNO}_3$  sample solution by  $\text{HNO}_3$  because the oxalate precipitate is  
252 dissolved in nitric acid before passing through the PS resin (it is also cheaper and  
253 easier to obtain); reducing the conditioning and cleaning volumes from 5 and 20  
254 mL to 2 and 8 mL, respectively, for faster separation time and less waste;
- 255 • optimizing the coating of the microsphere with extractant from 1:1/4 to 1:1/8;
- 256 • studying the capacity of the resin to determine the conditions in which it can deal  
257 with the new matrix composition and adapting the amount of  $\text{Sr}^{2+}$  carrier added to  
258 reduce errors due to the natural strontium in the sample.

259 *Separation conditions and PS resin composition*

260 The first modifications of the method evaluated were those related with the diameter of  
261 the PS resin and the separation devices (SPE-cartridge and vacuum box). The PS resins  
262 used had a median diameter of 60  $\mu\text{m}$  and were covered with the extractant at a ratio of  
263 1:1/4 [10]. 1 g of PS resin was placed in a 2 mL commercial SPE-cartridge and separation  
264 was performed in a commercial twelve-position vacuum box. In this first evaluation,  
265 conditioning, sample loading and cleaning were identical to the previously used reference  
266 procedure (i.e. 5 mL of 6M  $\text{LiNO}_3$  for conditioning, 10 mL of sample in 6M  $\text{LiNO}_3$  and  
267 four cleaning steps with 5 mL of 6M  $\text{LiNO}_3$ ). The samples also contained 1 mg of stable  
268 strontium, in the form of strontium nitrate, to check the chemical recovery.

269 The results obtained (see Table 1) show good detection efficiency and SQP(E) and  
270 slightly reduced recovery (from 100% to 92%), probably because in the previous

271 experience the amount of stable strontium added was less than 0.1 mg. The values  
272 obtained confirm that the changes introduced (i.e. PS resin size, cartridge type and suction  
273 device) do not significantly change the quality parameters of the separation and  
274 measurement by PS resins.

275 The second change evaluated was reducing the amount of cleaning solution from 20 mL  
276 to 8 mL to reduce the amount of waste generated and the time required for separation.  
277 The results were similar to those obtained with 20 mL of solution, showing that the  
278 volume can be reduced.

279 In the third change evaluated  $\text{LiNO}_3$  was partially replaced by  $\text{HNO}_3$  to reduce costs.  
280 Conditioning was done by 2 mL of 6M  $\text{HNO}_3$ , the sample medium was 6M  $\text{HNO}_3$  and  
281 the first two cleaning steps were done with  $\text{HNO}_3$ , while the last two were still done with  
282  $\text{LiNO}_3$  to prevent proton chemiluminescence. In this case the samples contained  $^{89}\text{Sr}$   
283 instead of  $^{90}\text{Sr}$ . As in the previous cases, these modifications did not cause significant  
284 variations in the recovery and measurement parameters. The detection efficiency of  $^{89}\text{Sr}$   
285 seems to be slightly higher than that of  $^{90}\text{Sr}$  due to its higher energy, although both  
286 isotopes show similar high values.

287 The last variable evaluated was reducing the PSm impregnating extractant from 1:1/4 to  
288 1:1/8 to evaluate the possibility of optimizing the microsphere coating. Again no  
289 significant differences regarding the previous conditions were observed.

290 **Table 1** Values of recovery, detection efficiency and SQP(E) parameter for the  
291 optimization of the separation and measurement of radiostrontium with PS resins

	Recovery (%)	Detection efficiency (%)	SQP(E)
Cleaning with 20 mL of $\text{LiNO}_3$	92(2)	93(5)	796(3)
Cleaning with 8 mL of $\text{LiNO}_3$	96(3)	93(3)	794(4)
Use of $\text{HNO}_3$ instead of $\text{LiNO}_3$	91(2)	96(2)*	791(7)
1:1/8 extractant proportion	93(2)	94(4)	785(3)

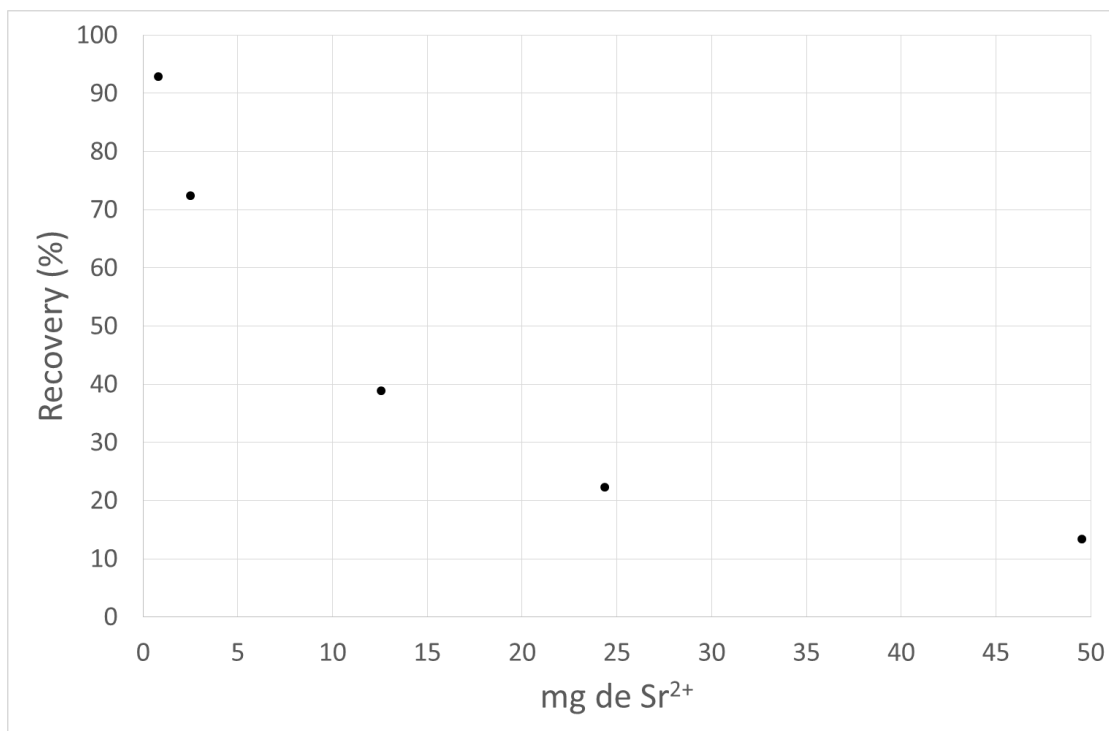
292 \* $^{89}\text{Sr}$

293 Due to the results obtained after testing all the modifications, the changes proposed were  
294 consolidated for the final separation procedure.

295 **PS resin capacity**

296 Stable strontium is added to the sample from the start of the analytical procedure to trace  
297 the chemical recovery of the whole method. The quantity of stable strontium added to  
298 emergency milk samples can vary from 1 to 10 mg, according to the method used and  
299 amount of milk measured. This quantity depends on two factors: the natural amount of  
300 strontium in milk and the effect of the amount of strontium in the chemical recovery. The  
301 content of stable strontium in milk can vary according to the animal species and what it  
302 has been fed, etc. As values of 0.5 to 3 mg·L<sup>-1</sup> are common, the amount of strontium  
303 carrier added should be high enough to avoid systematic errors due to the natural  
304 strontium present in the sample. On the other hand, it has been reported that resin recovery  
305 can decrease with the amount of strontium added, so that values between 1 and 50 mg  
306 could be critical in this regard.

307 Fig. 2 shows the recovery values obtained when samples containing from 1 to 50 mg of  
308 stable strontium and <sup>90</sup>Sr were analysed with PS resin. It can be seen that recovery  
309 declines with increasing amounts of added stable strontium, showing that PS resin is not  
310 able to retain all the strontium loaded.



311

312 **Fig. 2** Variation of chemical recovery with the amount of strontium added to the sample  
313 for columns with 1 g of PS resin

314 From the results obtained, 5 mg of stable strontium was adopted as a compromise value  
315 of the stable carrier, since it is high enough to minimize errors due to potential high values  
316 of natural strontium in milk and the chemical recovery is still high. In this situation, it can  
317 be considered that analysing 100 mL of milk sample with a natural strontium  
318 concentration of 1 mg L<sup>-1</sup> the systematic error will be lower than 1% and chemical  
319 recovery will be higher than 50%. The quantity of PS resin added to the cartridge was  
320 raised from 1 to 1.5 g to improve the column capacity and extend the exchange pathway  
321 through the cartridge. The final conditions and a comparison with the first study can be  
322 seen in Table 2 [10].

323 **Table 2** Modifications in the PS resin and separation conditions in comparison with the  
324 previous work

Conditions	Previous work [10]	Present study
Diameter PSm	190 µm	60 µm
Support	6mL PE vial	2mL cartridge
Flux control	Peristaltic pump	Vacuum box
PS resin	3 g	1.5 g
PSm: extractant	1:1/4	1:1/8
Sr <sup>2+</sup> carrier	0.1 mg	5 mg
Separation conditions	1) 5mL 6M LiNO <sub>3</sub> 2) 10mL sample (6M LiNO <sub>3</sub> ) 3) 4x 5mL 6M LiNO <sub>3</sub>	1) 2mL 6M HNO <sub>3</sub> 2) 10mL sample (6M HNO <sub>3</sub> ) 3) 2x 2mL 6M HNO <sub>3</sub> 4) 2x 2mL 6M LiNO <sub>3</sub>

### 325 *Pre-treatment optimization*

326 After an exhaustive review of the literature, two pre-treatment steps were chosen: fat and  
327 proteins were removed by acidifying with trichloroacetic acid, and the subsequent  
328 concentration of strontium and other alkaline-earth metals (mainly calcium) were  
329 performed by oxalate precipitation. These steps are typically used for calcium  
330 determination in milk samples [16, 17] and were also used by Kabai et al. [8] for  
331 determining radiostrontium due to the similar behaviour of calcium and strontium. They  
332 were chosen for the present study because of their simplicity and effectiveness in

333 emergencies. The pre-treatment steps were thus optimized, and different types of milk  
334 were studied.

### 335 ***Fat and protein separation***

336 The proposed method uses trichloroacetic acid (TCA) for fat and protein removal. The  
337 influence of different volumes of TCA was evaluated for this step and also for oxalate  
338 precipitation. 5 mg of Sr<sup>2+</sup> carrier were added to the sample, and Sr content was measured  
339 in the aqueous solution after fat filtration and in the oxalate precipitate after dissolution  
340 in 6M HNO<sub>3</sub>. Table 3 shows strontium recoveries after both separation steps using 50  
341 mL, 75 mL and 100 mL of TCA for 100 mL of whole cow's milk (three replicates were  
342 analysed in each case). Both steps achieved very high strontium recovery in all cases (96  
343 – 100%), showing that TCA volume does not greatly affect fat and protein separation or  
344 oxalate precipitation. A ratio of 1:1 (v/v) (100 mL of TCA) was chosen for protein  
345 denaturation, although lower volumes can also be used with good strontium recovery.

346 **Table 3** Strontium recoveries after fat and protein separation and oxalate precipitation  
347 step using 50 mL, 75 mL and 100 mL trichloroacetic acid (TCA) (conditions: 5 mg of  
348 Sr<sup>2+</sup>; 1.5 g of PS resin; Cow's milk (whole); 5 g oxalic acid)

	Recovery (%) (SD)		
	50 mL TCA	75 mL TCA	100 mL TCA
Fat and protein step	97 (4)	98 (1)	101 (4)
Oxalate precipitation step	96 (1)	98 (2)	100 (1)

### 349 ***Oxalate precipitation***

350 The alkaline-earth elements were separated from other groups by the same procedure,  
351 using oxalic acid as precipitation agent (in form of insoluble salts of strontium oxalate)  
352 and different amounts of oxalic acid were evaluated. Table 4 shows the strontium  
353 recovered after fat and protein separation and oxalate precipitation for 100 mL of whole  
354 cow's milk. 50 mL of TCA were used in the first step, and 1 g, 2 g and 5 g of oxalic acid  
355 were tested in the second. Three replicates were analysed in each case. High recoveries  
356 were also obtained in all cases (97 – 99%) due to the oxalic acid content being higher  
357 than strontium in the solution. In this step, as calcium and strontium oxalate salts



358 precipitate, 5 g of oxalic acid were used in the final procedure to avoid strontium losses  
 359 in milk samples with high calcium content.

360 **Table 4** Strontium recoveries after fat and protein separation and oxalate precipitation  
 361 with 1 g, 2 g and 5 g oxalic acid (conditions: 5 mg of Sr<sup>2+</sup>; 1.5 g of PS resin; Cow's milk  
 362 (whole); 50 mL of TCA)

	Recovery (%) (SD)		
	1 g oxalic acid	2 g oxalic acid	5 g oxalic acid
Fat and protein step	99 (3)	98 (4)	97 (4)
Oxalate precipitation step	94 (2)	96 (2)	96 (1)

363 *Influence of type of milk*

364 Milk composition can change according to the type of animal involved and the fat content,  
 365 and different concentrations of calcium, potassium, magnesium, fat, etc. can affect  
 366 strontium recovery in the separation. In order to study the influence of the pre-treatment  
 367 on the different steps, the procedure was tested for whole, semi-skimmed and skimmed  
 368 cow's milk, and also for cow's, goat's and sheep's milk. Three replicates were analysed  
 369 in each case. Table 5 shows the strontium recoveries of cow's milk with different fat  
 370 contents. As the results are quite high in both steps in all the types, the pre-treatment  
 371 method is robust and can be used regardless of fat content. However, the higher the fat  
 372 concentration, the longer the time needed for filtration (from 15 to 30 minutes).

373 **Table 5** Strontium recoveries for whole, semi-skimmed and skimmed cow's milk  
 374 (conditions: 5 mg of Sr<sup>2+</sup>; 1.5 g of PS resin; 100 mL of TCA; 5 g oxalic acid)

	Recovery (%) (SD)		
	Whole	Semi-skimmed	Skimmed
Fat and protein step	101 (4)	95	100 (1)
Oxalate precipitation step	100 (1)	91	100 (1)

375

376 Moreover, human consumption of milk is not limited to cow milk but also mainly goat  
377 and sheep milk. As exposed in literature, cow's, goat's and sheep's milk have different  
378 compositions. For example, sheep's milk is higher in calcium, sodium and phosphorus  
379 than the other types (see Table 7). Strontium concentration ranges from 0.5 to 2.7 mg L<sup>-1</sup>,  
380 so that there is a less than a 5% relative error when 5 mg of stable Sr<sup>2+</sup> are used in the  
381 determination. Cow's, goat's and sheep's milk was analysed following the pre-treatment  
382 procedure to evaluate the influence of different types of milk on the effectiveness of the  
383 proposed procedure (see table 6). Both fat and protein elimination were slightly reduced  
384 in some types, although high recoveries were obtained in the oxalate precipitation step in  
385 all cases, regardless of different calcium contents.

386 **Table 6** Strontium recoveries for cow's milk (whole), sheep's and goat's milk (semi-  
387 skimmed) (conditions: 5 mg of Sr<sup>2+</sup>; 1.5 g of PS resin; 75 mL of TCA; 5 g oxalic acid)

	Recovery (%) (SD)		
	Cow	Goat	Sheep
Fat and protein step	98 (1)	86 (1)	78(1)
Oxalate precipitation step	98 (2)	102 (3)	94(2)

388 This shows that the pre-treatment procedure obtains good and reproducible results on  
389 different milk types and fat contents.

### 390 *Elementary composition analysis.*

391 To complete the analysis of the pre-treatment steps and PS resin separation, the  
392 elementary composition was analysed after each step to prove the effectiveness of the  
393 proposed separation scheme for the different elements; with special attention to potential  
394 active monovalents and divalents contained in milk, which are the main strontium  
395 interferences. Differences in the composition of milk samples are mainly due to the type of  
396 animal. Table 7 shows the average elemental composition of the different types (cow's,  
397 sheep's and goat's milk) after fat and protein elimination, oxalate precipitate dissolution,  
398 and in the PS resin waste after separation. The % interferent removal was calculated for  
399 the oxalate precipitation step by comparing the elemental composition in this step with  
400 the fat elimination step. The % PS resin retention was also calculated taking into account

401 PS resin waste composition and the previous step. As can be seen, the oxalate  
 402 precipitation can remove monovalent elements (Na, K, P) and some soluble divalent  
 403 elements, such as magnesium and zinc, with average values between 85 – 100%. Other  
 404 divalent elements totally or partially precipitate and form oxalate insoluble salts, such as  
 405 calcium and barium, with an average of 6-10% and 42-48% elimination, respectively.  
 406 Regarding PS resin separation, the results show that only 13-16% of the remaining  
 407 calcium is retained in the cartridge and only 20-24% of barium is retained. This is a  
 408 smaller percentage than the initial and achieves the complete isolation of strontium from  
 409 the rest of the interferents.

410 **Table 7** Elemental composition in different steps of the procedure for cow's, sheep's and  
 411 goat's milk

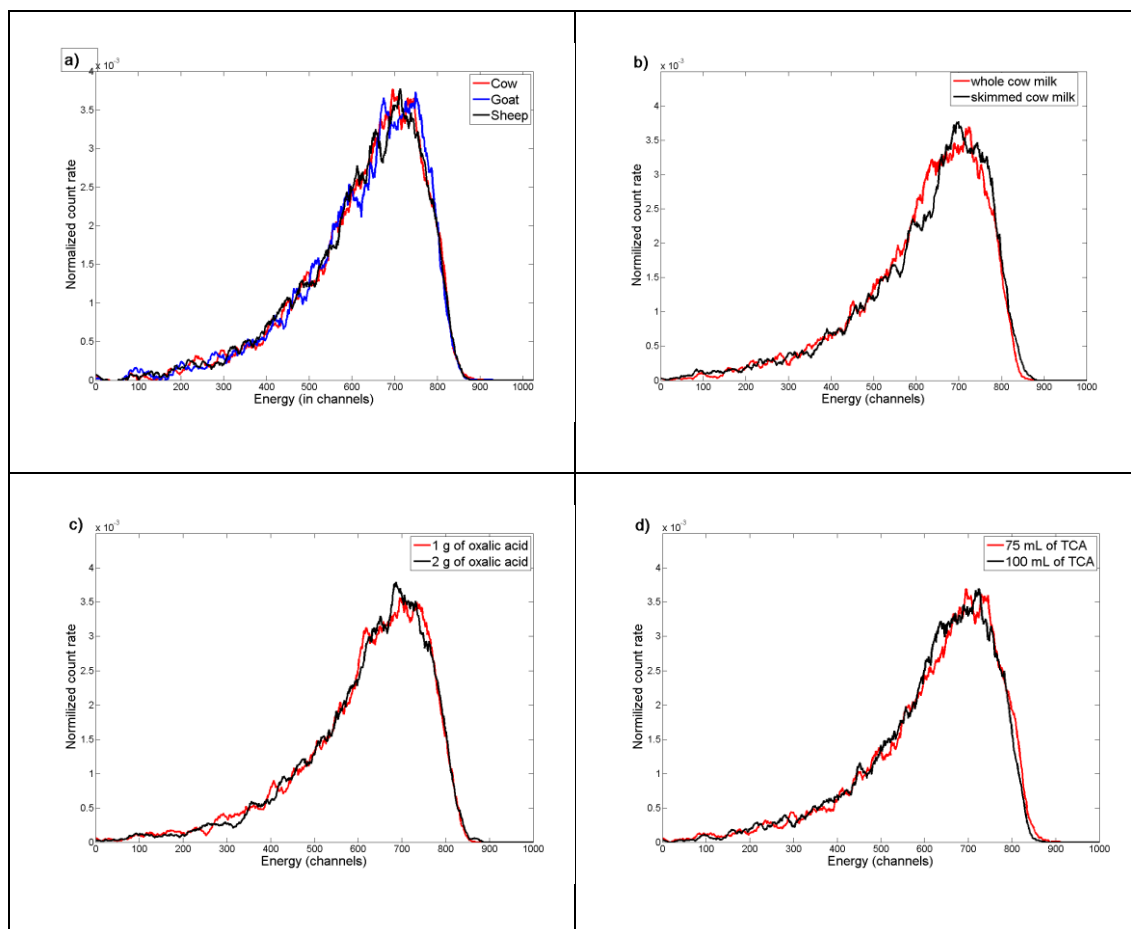
Milk		Ca	Mg	Zn	Na	K	P	Ba
Cow	fat removed (mg L <sup>-1</sup> )	1073	101	4.88	460	1526	715	1.74
	Oxalate dissolved (mg L <sup>-1</sup> )	1014	1.5	0.52	15	9.0	4.2	0.90
	Oxalate removal (%)	5.6	98	89	96	99	99	48
	PS resin waste (mg L <sup>-1</sup> )	881	1.4	0.45	18	8.0	6.4	0.72
	PS resin retention (%)	13.1	-	12	-	-	-	20
Sheep	fat removed (mg L <sup>-1</sup> )	1746	158	6.4	1121	1127	1532	2.24
	Oxalate dissolved (mg L <sup>-1</sup> )	1599	2.9	0.97	46	10	14.9	1.28
	Oxalate removal (%)	8.4	98	85	96	99	99	42
	PS resin waste (mg L <sup>-1</sup> )	1347	2.3	0.80	49	8	15.3	0.98
	PS resin retention (%)	15	-	18	-	-	-	24
Goat	fat removed (mg L <sup>-1</sup> )	1255	129	4.7	627	1783	884	1.87
	Oxalate dissolved (mg L <sup>-1</sup> )	1125	1.7	0.47	22	14	5.6	1.07
	Oxalate removal (%)	10	99	89	97	99	99	43
	PS resin waste (mg L <sup>-1</sup> )	973	1.4	0.43	27	12	6.5	0.85
	PS resin retention (%)	14	-	9.2	-	-	-	20

412

413 Calibration of  $^{90}\text{Sr}$  and  $^{89}\text{Sr}$  detection efficiency

414 Influence of sample matrix and sample pre-treatment on detection efficiency

415 Several of the samples used in the pre-treatment optimization were also analysed with PS  
 416 resin after being spiked with  $^{90}\text{Sr}$  or  $^{89}\text{Sr}$  to identify the parameters that could affect their  
 417 detection efficiencies. Fig. 3 shows the spectrum of  $^{89}\text{Sr}$  obtained from cow's, goat's and  
 418 sheep's milk; whole, semi-skimmed and skimmed cow's milk with different fat contents;  
 419 samples treated with different amounts of oxalic acid (1 g and 2 g) and samples treated  
 420 with different amounts of TCA (75 mL and 100 mL). The spectra obtained were always  
 421 in the same position, which shows the absence of any quenching effect that could  
 422 influence detection efficiency.

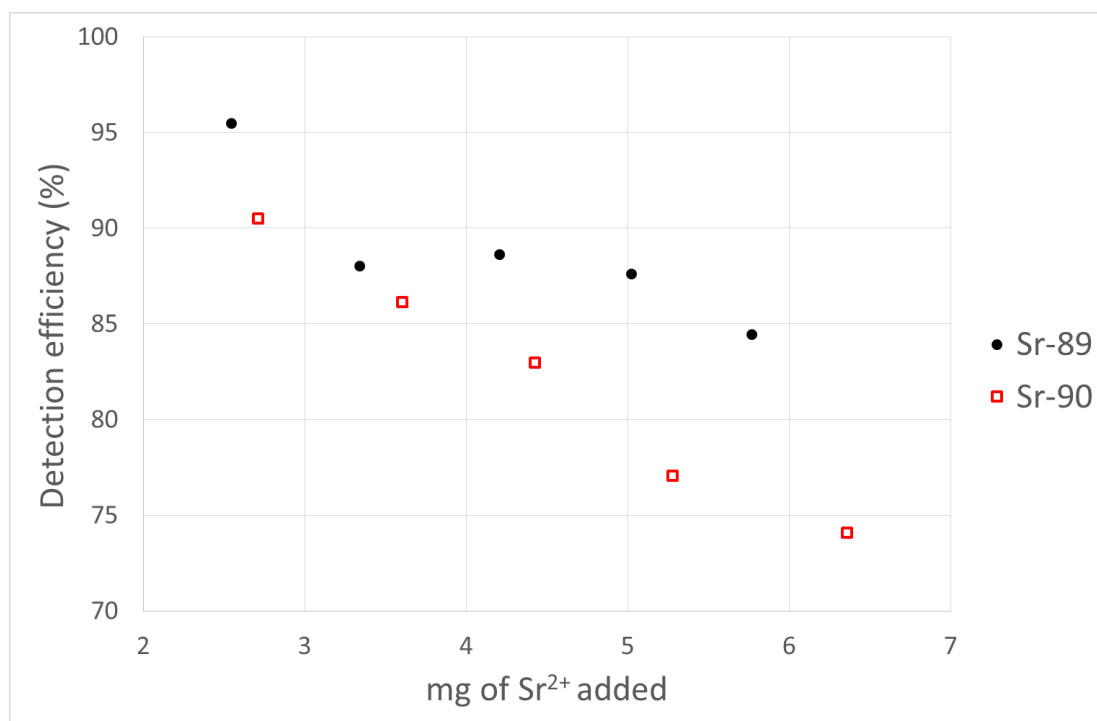


423 **Fig. 3**  $^{89}\text{Sr}$  of milk sample measured with PS resins. a) cow's, goat's and sheep's milk  
 424 samples; b) whole, semi-skimmed and skimmed cow's milk sample; c) cow's milk  
 425 sample pretreated with 1 and 2 g of oxalic acid; d) cow's milk sample pretreated with 75  
 426 and 100 mL of TCA

427 The mean values of the quenching parameter (SQP(E)),  $^{90}\text{Sr}$  detection efficiency and  $^{89}\text{Sr}$   
428 detection efficiency were 786(6) (n=30), 86(6)% (n=11) and 91(6)% (n=11), respectively.  
429 An inspection of the values obtained for the different parameters evaluated (i.e. animal,  
430 fat content, industrial pre-treatment, TCA amount used and quantity of oxalic acid) do  
431 not show any correlation between these parameters and detection efficiency.

#### 432 $^{90}\text{Sr}$ and $^{89}\text{Sr}$ detection efficiency calibration

433 To calibrate  $^{90}\text{Sr}$  and  $^{89}\text{Sr}$  detection efficiencies five water samples containing between 2  
434 and 6 mg of stable strontium and  $^{90}\text{Sr}$  or  $^{89}\text{Sr}$  were analysed. Fig.4 shows the values of  
435 detection efficiency obtained.



436

437 **Fig. 4** Values of detection efficiency for  $^{90}\text{Sr}$  and  $^{89}\text{Sr}$  in samples containing a variable  
438 amount of  $\text{Sr}^{2+}$

439 From the values obtained (Fig. 4), the detection efficiency is seen to drop for  $^{90}\text{Sr}$  and a  
440 linear correlation can be applied. This is not so evident in the case of  $^{89}\text{Sr}$ , in which the  
441 drop is not so clear, probably because of the uncertainty associated with the experimental  
442 process being in the same order of the variation of the efficiency in the stable strontium  
443 range studied. On the other hand, values of SQP(E) do not show any trend and the mean  
444 values are very similar to those obtained by varying sample matrix and sample pre-

445 treatment. If the value of mean detection efficiencies for  $^{90}\text{Sr}$  and  $^{89}\text{Sr}$  are calculated, they  
446 are found to be similar to those of the pre-treatment: 82(7) for  $^{90}\text{Sr}$  and 89(4) for  $^{89}\text{Sr}$ .

447 Several calibration procedures have been developed for quantifying mixtures of  
448 radionuclides and, in the specific case of  $^{90}\text{Sr}$  and  $^{89}\text{Sr}$  mixtures, several windows systems  
449 and deconvolution methods combining one or more than one measurement are described  
450 in the literature [18-21]. Since our aim was to develop a fast and easy method of analysing  
451 the risk associated with radiostrontium ( $^{90}\text{Sr} + ^{89}\text{Sr}$ ) in milk samples in cases of  
452 emergency, and since the differences between  $^{90}\text{Sr}$  and  $^{89}\text{Sr}$  efficiencies are not important,  
453 the combined detection efficiency of 86(6) obtained from the mean of all the calibration  
454 sample values was applied. This combined detection efficiency value gives the total  
455 amount of radiostrontium ( $^{89}\text{Sr} + ^{90}\text{Sr}$ ). However, the  $^{89}\text{Sr}$  or  $^{90}\text{Sr}$  detection efficiency  
456 values can be used for samples with only one known isotope present.

457 **Table 8** Relative bias in the determination of radiostrontium ( $^{89}\text{Sr} + ^{90}\text{Sr}$ ) in a water  
458 sample

Ratio $^{89}\text{Sr}/^{90}\text{Sr}$	Relative bias $^{89}\text{Sr} + ^{90}\text{Sr}$ (%)
1:1	1.4
2:1	5.3
1:2	5.9

459 The calibration procedure was validated by the quantification of three water samples  
460 spiked with  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  at ratios of (1:1; 2:1 and 1:2). The deviations obtained in the  
461 quantification of total radiostrontium activity ( $^{89}\text{Sr} + ^{90}\text{Sr}$ ) are shown in Table 8. The  
462 relative bias of total radiostrontium are in all three cases less than 10%, so that the  
463 approach can be used to determine  $^{89}\text{Sr} + ^{90}\text{Sr}$ .

#### 464 *Validation of the method*

465 The final procedure was tested with different types of milk samples: different fat content  
466 (whole, semi-skimmed and skimmed), treatment (UHT, powdered, pasteurized and raw  
467 milk) and animal species (cow, sheep and goat). A total of 21 samples were analysed, 5  
468 of which were blanks and 16 were spiked in the laboratory with certified standards of  $^{89}\text{Sr}$

469 and  $^{90}\text{Sr}$ . Two samples were spiked only with one standard, and in the others, different  
 470 ratios of  $^{89}\text{Sr}/^{90}\text{Sr}$  were considered to determine the robustness of the method. As it is  
 471 known that the amount of  $^{89}\text{Sr}$  release to the environment after a nuclear emergency is  
 472 higher than  $^{90}\text{Sr}$ ,  $^{89}\text{Sr}/^{90}\text{Sr}$  ratios of 1:1, 2:1 and 10:1 were tested. The highest activity  
 473 considered was 125 Bq/L, since it is the limit for milk and dairy products in the European  
 474 regulations [4].

475 Table 9 shows strontium recoveries after the pre-treatment step and strontium retentions  
 476 in PS resin. The total recovery is obtained by multiplying both values. As can be seen,  
 477 pre-treatment recoveries are high, ranging from 87 to 96%, and the average value is 93%  
 478 with a relative standard deviation of 4%. Strontium retention in PS resin ranges between  
 479 64 to 72% because of the capacity of the resin, with an average value of 70% (6% RSD).  
 480 This shows that good strontium recoveries can be obtained for emergency situations, with  
 481 an average total recovery of 65% (7% RSD).

482 **Table 9** Strontium recoveries for different types of milk analysed with the procedure  
 483 proposed.

Type of milk	Reference	Pre-treatment Recovery (%)	PS resin Retention (%)	Total Recovery (%)
Cow/whole/UHT	M1 - M7	93.0 (2.9)	69.1 (2.0)	64.3 (3.4)
Cow/semi-skimmed/UHT	M8	86.5	71.7	62.0
Cow/skimmed/UHT	M9	90.2	71.1	64.1
Cow/whole/powder	M10 - M11	95.3 (1.3)	68.3 (0.5)	65.1 (1.4)
Cow/whole/pasteurized	M12	94.8	68.2	64.7
Goat/whole/raw	M13	94.0	70.8	66.6
Sheep/semi-skimmed/UHT	M14 - M17	93.6 (6.1)	64.3 (2.7)	60.3 (6.0)
Goat/semi-skimmed/UHT	M18 - M21	96.2 (2.7)	69.6 (1.8)	66.9 (3.1)
Average total samples		93.3 (3.7)	69.7 (4.3)	65.0 (4.5)

484 Radiostrontium activities were calculated applying the value of combined detection  
 485 efficiency. Table 10 shows the relative bias for total radiostrontium activity ( $^{89}\text{Sr} + ^{90}\text{Sr}$ )  
 486 calculated for different spiked milk samples. Blank count rates were subtracted from the  
 487 sample count rate in the activity calculation. The values of the blank samples for blank  
 488 cow's (M1, M2, M10), sheep's (M14) and goat's milk (M18) range from 1.8 to 3 cpm in

489 the total window (1 – 1024). The samples spiked with one standard (M3, M4) obtained a  
 490 relative bias below 7 % for  $^{90}\text{Sr}$  and  $^{89}\text{Sr}$ , respectively, and below 5% if the corresponding  
 491 detection efficiency value is used. In the samples spiked with both isotopes, total relative  
 492 bias was below 7% in all cases except one and in all the  $^{89}\text{Sr}/^{90}\text{Sr}$  ratios studied.

493 **Table 10** Relative bias obtained for different spiked milk samples analysed with the  
 494 procedure and calibration system proposed

	Type of milk	Activity $^{89}\text{Sr}$ (Bq/L)	Activity $^{90}\text{Sr}$ (Bq/L)	Ratio $^{89}\text{Sr}/^{90}\text{Sr}$	Relative bias $^{89}\text{Sr} + ^{90}\text{Sr}$ (%)
M3	Cow/whole/UHT	-	27	0:1	-2.2 (1.9*)
M4	Cow/whole/UHT	15	-	1:0	6.6 (3.0*)
M5	Cow/whole/UHT	27	27	1:1	-1.9
M8	Cow/semi-skimmed/UHT	27	27	1:1	4.5
M9	Cow/skimmed/UHT	27	27	1:1	6.1
M11	Cow/whole/powder	27	27	1:1	1.7
M12	Cow/whole/pasteurized	27	27	1:1	2.6
M13	Goat/whole/raw	27	27	1:1	-1.9
M15	Sheep/semi-skimmed/UHT	27	27	1:1	7.0
M19	Goat/semi-skimmed/UHT	27	27	1:1	5.2
M6	Cow/whole/UHT	27	13.5	2:1	1.6
M15	Sheep/semi-skimmed/UHT	27	13.5	2:1	20.2
M19	Goat/semi-skimmed/UHT	27	13.5	2:1	6.5
M7	Cow/whole/UHT	125	13.5	10:1	-4.1
M17	Sheep/semi-skimmed/UHT	125	13.5	10:1	-3.9
M21	Goat/semi-skimmed/UHT	125	13.5	10:1	-4.1

495 \*calculated using the corresponding detection efficiency ( $^{90}\text{Sr}$  or  $^{89}\text{Sr}$ )

496 A certified reference material (IAEA-473 milk powder) was also analysed to validate the  
 497 method. The sample contained  $^{90}\text{Sr}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , with 197.2, 158 and 212 Bq kg<sup>-1</sup> of  
 498 dry mass, respectively, at the time of separation. As can be seen in Table 11, pre-treatment  
 499 recoveries and PS resin retentions are high, obtaining a good total strontium recovery. In



500 addition, relative bias for  $^{90}\text{Sr}$  is below 7% in the three replicates performed and below  
 501 3% if the  $^{90}\text{Sr}$  detection efficiency is used, which confirms the validity of the method.

502 **Table 11** Strontium recoveries (%),  $^{90}\text{Sr}$  experimental activity ( $\text{Bq kg}^{-1}$  dry mass) at the  
 503 time of separation, and relative bias (%) obtained

Type of milk	Pre-treatment recovery (%)	PS resin retention (%)	Total recovery (%)	Activity $^{90}\text{Sr}+^{89}\text{Sr}$ ( $\text{Bq/kg}$ dry mass)	Relative bias $^{90}\text{Sr}+^{89}\text{Sr}$ (%)
MR1	88.6	79.6	70.5	207.0 (198.0*)	-3.5 (0.4*)
MR2	93.1	78.7	73.3	204.6 (195.7*)	-4.7 (-0.8*)
MR3	92.1	76.7	70.6	203.3 (194.4*)	-5.2 (-1.4*)

504 \*calculated using the corresponding detection efficiency

### 505 *Comparative evaluation of the procedure*

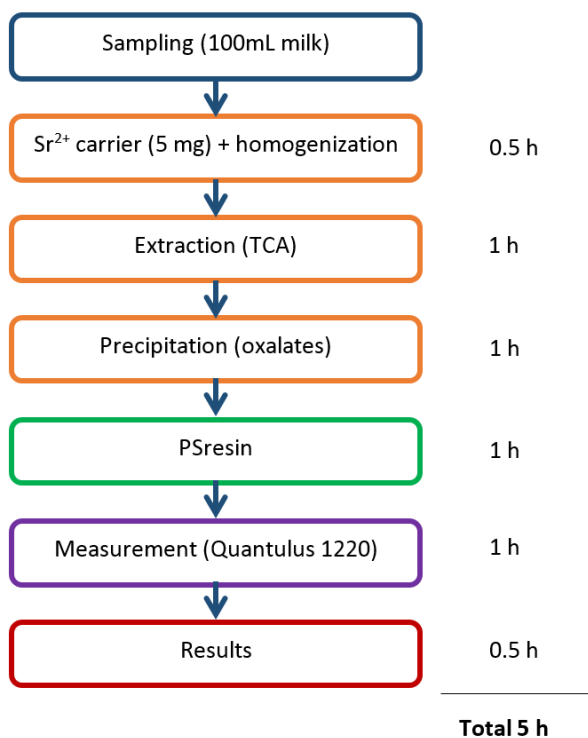
506 In order to compare the proposed procedure with other rapid methods of determining  
 507 radiostrontium in milk samples in the literature, their main characteristics are shown in  
 508 Table 12. The recoveries range between 62 and 94% for the different separation  
 509 techniques. Good results were also obtained in the present study, with an average value  
 510 of 65% and the lowest variability of all the methods considered. As in the present study,  
 511  $^{90}\text{Sr} + ^{89}\text{Sr}$  activity relative bias was below 15% in all cases. Minimum detectable activity  
 512 ranges between 0.1 and 5  $\text{Bq L}^{-1}$  according to measurement technique, sample volume  
 513 and counting time, while the proposed method achieves an MDA of 0.34  $\text{Bq L}^{-1}$ , which  
 514 is suitable for emergencies.

515 The response time is the decisive factor to consider in a rapid method. The fastest methods  
 516 in the literature need 7 – 8 hours to obtain a reliable result on radiostrontium activity,  
 517 while the method proposed here reduces the analysis time to only 5 hours. Fig.5 shows  
 518 the necessary length of each step in the procedure. The improved time is mainly due to  
 519 the separation step with PS resin, which usually requires 2 or 3 hours to separate and  
 520 prepare the sample for measurement. However, PS resin only needs 1 hour because the  
 521 cartridges can be directly measured in a 20 mL polyethylene vial. This step is also easier  
 522 to perform than in other methods.

523 **Table 12** Characteristics of different rapid methods for radiostrontium determination in  
 524 milk samples

Procedure	Sample (L)	Sr <sup>2+</sup> carrier (mg)	Recovery % (SD)	Measurement	Relative Bias <sup>90</sup> Sr % (SD)	MDA (Bq L <sup>-1</sup> )	Time (h)
Brun et al., 2002 [6]	0.5	10	62.2 (6.8)	Gas Flow Proportional Counting	-1.44 (5.26)	0.09	14 - 15
Maxwell et al., 2009 [7]	0.1	4.19	75.1 (17.0)	Gas Flow Proportional Counting	0.52 (5.70)	0.5	7 – 8
Kabai et al., 2011 [8]	0.1	10	93.5 (6.6)	LSC	-	0.8	7 – 8
IAEA, 2013 [9]	0.25	10	70 - 75	LSC	<15%	2 - 5	7 – 8
This study	0.1	5	65.0 (4.5)	Plastic Scintillation Counting	3.97 (0.96)	0.34	5

525



526

527 **Fig. 5** Analysis time of the proposed procedure for radiostrontium determination in milk  
528 samples in emergency situations

## 529 Conclusions

530 A new and rapid method is proposed for radiostrontium determination in milk samples  
531 by plastic scintillation resins (PS resins). This procedure combines the separation and  
532 measurement preparation steps to reduce manpower and reagents and avoid the  
533 production of mixed waste. The method has been shown to be faster than others in the  
534 literature, the time necessary for complete radiostrontium separation and quantification  
535 being only 5 hours.

536 After optimization of the pre-treatment and separation steps with PS resins, the procedure  
537 described here obtains good reproducible total recoveries (65.0 (4.5) % on average) for  
538 different types of milk (fat content, animal species, treatment, etc.). Relative bias for total  
539 radiostrontium activity (<sup>89</sup>Sr + <sup>90</sup>Sr) was below 7% in all the analyses except one and all  
540 the <sup>89</sup>Sr/<sup>90</sup>Sr ratios studied (1:1, 2:1 and 10:1). The minimum detectable activity of 100mL  
541 of milk sample measured in 60 minutes is about 0.34 Bq L<sup>-1</sup>.

542 The proposed procedure has thus been shown to be suitable for determining  
543 radiostrontium in emergency situations.

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