



# Rapid $^{90}\text{Sr}$ quantification method based on the Bateman equation for routine laboratory work

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**Abstract.** Artificially introduced into the environment  $^{90}\text{Sr}$  is highly radiotoxic, so its content levels in foodstuff and biota require constant monitoring for radiological protection. Most analytical procedures used for  $^{90}\text{Sr}$  determination are time-consuming, and therefore, a faster approach is needed. Employing the Bateman equation enables more efficient exploitation of the secular equilibrium between  $^{90}\text{Sr}$  and its daughter radionuclide  $^{90}\text{Y}$  in the calculations. This article describes a method for computing the  $^{90}\text{Sr}$  activity concentration, while accounting for  $^{90}\text{Y}$  activity. The developed approach was tested and validated in terms of its applicability in everyday analysis.

**Keywords:** Bateman equation • Liquid scintillation • Strontium-90 • Yttrium-90

## Introduction

The main sources of radiochemical contamination are nuclear weapon tests [1] and accidents at nuclear power plants (e.g., Chernobyl in 1986) [2]. It is worth noting that the accident at the Fukushima Daiichi power plant released very low levels of  $^{90}\text{Sr}$  [3]. The radioisotopes released in such accidents enter the biosphere and food chain, ultimately reaching humans. Among many fission products,  $^{90}\text{Sr}$  is considered to be long-lived (half-life = 28.8 years [4]) and has high radiotoxicity because of the released  $\beta$  radiation. The maximum radiation energies and transition probabilities of  $^{90}\text{Sr}$  and  $^{90}\text{Y}$  decay are shown in Fig. 1.

Because strontium and calcium exhibit similar chemical properties, these two elements compete with one another in human bone chemistry. However, their behaviours in organisms differ because strontium is absorbed relatively less effectively in the intestines, excreted more effectively by the kidneys and removed more rapidly from the bone surface [5]. Following acute  $^{90}\text{Sr}$  absorption, an adult person (i.e., 25–45 years old) eliminates only 2.8–3.2% of the initial  $^{90}\text{Sr}$  amount each year [5]. The  $\beta$  radiation released by  $^{90}\text{Sr}$  and its daughter isotope  $^{90}\text{Y}$  can cause leukosis, as well as other cancers that affect the bones and adjacent soft tissues. For this reason, environmental monitoring of  $^{90}\text{Sr}$  and analysis of foodstuffs for  $^{90}\text{Sr}$  content are crucial, and special emphasis should be placed on milk and dairy products [6–9].

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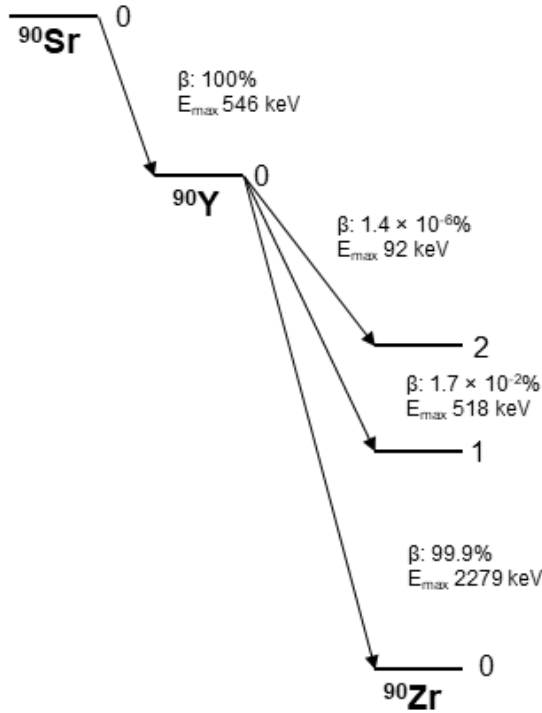


Fig. 1.  $^{90}\text{Sr}$  and  $^{90}\text{Y}$  decay scheme [4].

Typical analysis begins with sample mineralization by ashing in a muffle furnace, followed by dissolution of that ash in nitric acid. Further steps enable  $^{90}\text{Sr}$  or  $^{90}\text{Y}$  separation from the matrix and isolation from one another. Yttrium can be separated from strontium via liquid–liquid extraction with tributyl phosphate, which forms a complex with yttrium in the presence of nitric acid [10]. Thus, yttrium is transferred to the organic phase, while strontium remains in the aqueous phase. Yttrium is then back-extracted with 25% ammonia solution to form  $\text{Y}(\text{OH})_3$  [9, 11]. Finally,  $\text{Y}(\text{OH})_3$  dissolves in the nitric acid, and yttrium is precipitated as an oxalate [11, 12] and then further purified on ion exchange resin [11]. An alternative technique is to co-precipitate interfering radioisotopes with barium chromate, while  $^{90}\text{Y}$  is precipitated as  $\text{Y}(\text{OH})_3$  [9, 13]. In this approach, strontium is left in the solution to promote  $^{90}\text{Y}$  ingrowth. Then, yttrium is precipitated as an oxalate, and only the activity of  $^{90}\text{Y}$  is the subject of subsequent measurements. There are also other potential procedures involving ion-exchange and extraction chromatography [8, 9, 14–16]. One strategy was developed based on forming a strontium complex with crown ether (4,4'(5')-di-*t*-butyl-cyclohexano-18-crown-6), which is very selective for strontium [8, 9, 14–16]. Commonly employed detectors mostly use proportional gas flow and liquid scintillation counters [9, 10, 16].

Challenges related to  $^{90}\text{Sr}$  analysis include the presence of calcium in the matrix, overlapping  $^{90}\text{Sr}$ ,  $^{90}\text{Y}$  and  $^{89}\text{Sr}$   $\beta$ -spectra, and the required separation of other radionuclides [7, 8, 11]. These difficulties can be resolved by focusing on either  $^{90}\text{Y}$  separation and immediate radioactivity measurements, or  $^{90}\text{Sr}$  separation and waiting for secular equilibrium, at which point, half of the measured radioactivity originates

from  $^{90}\text{Sr}$ . Some methods rely on the separation of strontium from yttrium at this stage, and the measurement of  $^{90}\text{Y}$  activity, which is then equal to the  $^{90}\text{Sr}$  activity. The equilibrium between  $^{90}\text{Sr}$  and  $^{90}\text{Y}$  is reached after approximately 18 days, which makes for a long analytical process [9]. Another method is measurement of Cherenkov effect by liquid scintillation counter when scintillation cocktail is not added to the sample. This approach is preferred for samples where  $^{89}\text{Sr}$  is present. If  $^{90}\text{Y}$  is freshly separated from strontium, then the Cherenkov effect caused by  $^{89}\text{Sr}$  high energy  $\beta$  radiation ( $E_{\max} = 1495.1$  keV) can be measured. Then subsequent measurement can detect the same effect caused by  $^{90}\text{Y}$   $\beta$  radiation for further calculation of ingrowth [9]. Measuring the activity immediately after  $^{90}\text{Sr}$  separation is considered as a last-resort strategy for emergency situations because the accuracy of such analysis is compromised. However, there are mathematical approaches that can provide accurate results before secular equilibrium is reached [7, 17]. Therefore, it is not necessary to wait a long time for the equilibrium state. The aim of the present work was to develop a  $^{90}\text{Sr}$  determination method based on the Bateman equation to enable calculations of daughter radionuclide activities according to the initial activity of the parent radionuclide and the decay rate of all considered radionuclides in absence of  $^{89}\text{Sr}$ . The Bateman equation for the  $^{90}\text{Sr}/^{90}\text{Y}$  isotope pair is shown in Eq. (1), where  $A_Y$  is  $^{90}\text{Y}$  activity (Bq),  $A_{\text{Sr}_0}$  is initial  $^{90}\text{Sr}$  activity (Bq),  $\lambda_Y$  and  $\lambda_{\text{Sr}}$  are the decay constants of  $^{90}\text{Y}$  and  $^{90}\text{Sr}$  ( $\text{day}^{-1}$ ),  $t$  is time (days) and  $e$  is the Euler number:

$$(1) \quad A_Y = \frac{\lambda_Y}{\lambda_Y - \lambda_{\text{Sr}}} A_{\text{Sr}_0} (e^{-\lambda_{\text{Sr}}t} - e^{-\lambda_Yt})$$

The proposed calculation method also applies the concept of measuring the total activity and calculating the  $^{90}\text{Sr}$  fraction in a modified manner [17].

### Bateman equation consideration

In general, the total activity of the sample ( $A_{\text{tot}}$ ) (in Bq) is equal to the sum of  $^{90}\text{Sr}$  ( $A_{\text{Sr}}$ ),  $^{90}\text{Y}$  ( $A_Y$ ) activities, as expressed in Eq. (2). Those individual activities change along with the decay of the corresponding radionuclides. However, since  $A_{\text{Sr}}$  and  $A_Y$  are unknown, they must be expressed as fractions of  $A_{\text{tot}}$ . Equation (3) defines  $A_{\text{tot}}$  as a sum of fractions changing with time ( $t$ ; days), where  $\lambda_{\text{Sr}}$  represent the  $^{90}\text{Sr}$  decay constant ( $\text{day}^{-1}$ )

$$(2) \quad A_{\text{tot}} = A_{\text{Sr}} + A_Y$$

$$(3) \quad A_{\text{tot}} = x A_{\text{tot}} e^{-\lambda_{\text{Sr}}t} + (1-x) A_{\text{tot}} e^{-\lambda_{\text{Sr}}t}$$

The fraction of  $^{90}\text{Sr}$  (i.e.,  $x$ ) is defined as shown in Eq. (4):

$$(4) \quad x = \frac{A_{\text{Sr}}}{A_{\text{tot}}} = \frac{A_{\text{Sr}}}{A_{\text{Sr}} + A_Y}$$

Because  $A_Y$  is unknown, it must be calculated using the Bateman equation. It is important to

note that  $A_{Sr}$  is assumed to be equal to  $A_{Sr_0}$  because the half-life of <sup>90</sup>Sr is much longer than the time elapsed during the preparation and measurement of the sample. Thus,  $x$  can be expressed as shown in Eq. (5). Decay constant of <sup>90</sup>Y is denoted as  $\lambda_Y$  ( $\text{day}^{-1}$ ):

$$(5) \quad x = \frac{A_{Sr}}{A_{Sr} + \frac{\lambda_Y}{\lambda_Y - \lambda_{Sr}} A_{Sr} (e^{-\lambda_{Sr}t} - e^{-\lambda_Yt})}$$

Because  $\lambda_Y$  is much greater than  $\lambda_{Sr}$ , the latter can be assumed to be equal to zero. Therefore, Eq. (5) is simplified to Eq. (6)

$$(6) \quad x = \frac{1}{2 - e^{-\lambda_Yt}}$$

To calculate  $A_{Sr}$ , Eqs. (2) and (3) are combined to give the expressions in Eqs. (7) and (8) as

$$(7) \quad A_{Sr} = x A_{\text{tot}} e^{-\lambda_{Sr}t}$$

$$(8) \quad A_Y = (1 - x) A_{\text{tot}} e^{-\lambda_{Sr}t}$$

In the case of <sup>89</sup>Sr presence the approach presented above cannot be applied because <sup>89</sup>Sr and <sup>90</sup>Sr activities are independent of each other. In such instance, the total activity of the sample can be expressed as shown in Eq. (9), where  $A_{89}$  is initial <sup>89</sup>Sr activity (Bq) and  $\lambda_{89}$  its decay constant ( $\text{day}^{-1}$ ) as

$$(9) \quad A_{\text{tot}} = A_{Sr_0} e^{-\lambda_{Sr}t} + \frac{\lambda_Y}{\lambda_Y - \lambda_{Sr}} A_{Sr_0} (e^{-\lambda_{Sr}t} - e^{-\lambda_Yt}) + A_{89} e^{-\lambda_{89}t}$$

For calculation of <sup>89</sup>Sr and <sup>90</sup>Sr activities, separate measurement of <sup>90</sup>Y activity is desired after its ingrowth. In an emergency situation one cannot presume that <sup>90</sup>Sr and <sup>90</sup>Y are in equilibrium and also cannot wait for it. The activity of <sup>90</sup>Sr is shown in Eq. (10), where  $t_i$  is time of <sup>90</sup>Y ingrowth and  $t_m$  is time from <sup>90</sup>Y purification to the midpoint of the measurement as

$$(10) \quad A_{Sr} = \frac{A_Y}{(1 - e^{-\lambda_Yt_i}) e^{-\lambda_Yt_m}}$$

In order to calculate the <sup>89</sup>Sr activity the sum of <sup>90</sup>Sr and <sup>90</sup>Y activities must be subtracted from the total activity. It is described in Eq. (11) where  $t_{89}$  is the time from start to the midpoint of the activity measurement and  $t_Y$  is the time from yttrium separation to the midpoint of activity measurement,  $A_{Sr+Y}$  is the sum of <sup>90</sup>Sr and <sup>90</sup>Y activities and  $A_{Y1}$  is <sup>90</sup>Y activity measured after its ingrowth. This approach is described in detail in [18]:

$$(11) \quad A_{89} = \frac{A_{\text{tot}} - A_{Sr+Y} - [A_{Y1} (1 - e^{-\lambda_Yt_Y})]}{e^{-\lambda_{89}t_{89}}}$$

## Radiometry experiments

All reagents used in these experiments were of analytical grade or higher. The strontium carrier was prepared from 15.21 g of  $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$  (Sigma-Aldrich)

dissolved in 500 mL of distilled water. Nitric acid, aqueous ammonia solution (25%), oxalic acid, ammonium carbonate, sodium hydroxide, and disodium ethylenediaminetetraacetate ( $\text{Na}_2\text{EDTA}$ ) were supplied by Chempur, Sr-resin (SR-B100-A; particle size = 100–150  $\mu\text{m}$ ) was supplied by Triskem, the scintillation cocktail Insta-Gel Plus was purchased from PerkinElmer, and <sup>90</sup>Sr and <sup>85</sup>Sr standard solutions were obtained from Polatom, Świerk (Poland).

Radiometry was performed on the investigated radionuclides using a low-background scintillation counter (Quantulus 6220, PerkinElmer). The counting protocol was set to a normal counting mode, with the transformed spectral index of the external standard (tSIE) as the quench indicator and the static control switched on (coincidence time = 18 ns; delay before burst = 500 ns; pulse amplitude comparison strength = 50). Guard compensation technology (GCT) optimization was conducted during the procedure development stage and was stored in the equipment memory; the GCT mode was set to 'low'. The counting process proceeded for 60 min over a counting range of 0.5–1200 keV in low-diffusion plastic vials (PerkinElmer). Background subtraction '1st vial' mode was used, where a 'blank' vial containing a sample without the analyte was placed in the first position on the rack. Then, its spectrum was considered as the background and was subtracted from the subsequent spiked sample spectrum. The Quantulus instrument was calibrated by measuring a <sup>90</sup>Sr/<sup>90</sup>Y source in secular equilibrium. The counting efficiency was determined to be >98% and assumed to be 100% in all sample measurements.

In addition, samples spiked with <sup>85</sup>Sr were analysed to verify the strontium recovery independently from the proposed computational method. These analyses were performed using a high-purity germanium gamma spectrometer (Tennelec CPVDS-30 20190), which had 18% counting efficiency. The measurements were conducted in 100-mL cylindrical containers that are routinely used for <sup>85</sup>Sr determinations.

The linearity was determined by measuring four <sup>90</sup>Sr/<sup>90</sup>Y sources in secular equilibrium at different activity levels. Then the counts were plotted as a function of the source activity, which enabled calculation of the determination coefficients.

The reliability of the applied procedure was determined based on <sup>90</sup>Sr recovery. Each subsample's radioactivity was measured immediately after preparation, and also later at a random time. This was necessary to evaluate the accuracy at any given moment.

## Analytical methodology

Milk powder samples were analysed in terms of their <sup>90</sup>Sr content with an accredited method [13]. Specifically, the activity concentration was confirmed to be below the minimal detectable activity (MDA), and then a large sample of the material was ashed in a muffle furnace. The ash was divided into subsamples representing about 50 g of the original material. Sub-

samples were placed in 250-mL beakers and spiked with  $^{90}\text{Sr}$  standard solution by weight; there were two fortification levels ( $5 \text{ Bq}\cdot\text{kg}^{-1}$  and  $50 \text{ Bq}\cdot\text{kg}^{-1}$ ) and 12 subsamples in each level. Finally, the analytical results were compared with previously calculated activity concentrations as a recovery measure.

The procedure began with the addition of the strontium carrier solution to the ash. In the next step, the ashed sample was dissolved in 60 mL (1:1 v/v) nitric acid in water and filtered. The solution volume was increased to 200 mL with distilled water, and then, 200 mL of 8% oxalic acid was added, with five drops of bromocresol green. Strontium oxalate precipitation was achieved upon adding ammonia solution to reach pH 4–4.5. The precipitate was ashed in a muffle furnace at  $830^\circ\text{C}$  for approximately 4 h. The subsequent steps are presented in Fig. 2. It is worth to point that elutes which here are considered as waste contain  $^{90}\text{Y}$  which could be used for  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  calculation as described in Eqs. (10) and (11). This allows to calculate the results much quicker in comparison to methods which require  $^{90}\text{Sr}/^{90}\text{Y}$  equilibrium.

Prior to the analysis, 3 g of Sr-Resin was conditioned in distilled water for 12 h and then poured into a glass chromatographic column (inner diameter = 10 mm) and further conditioned with 20 mL of 65% nitric acid. The column was cleaned and refilled with Sr-Resin after each third analysis. After each first and second analyses, the resin was regener-

ated with 0.1 M  $\text{Na}_2\text{EDTA}/0.1 \text{ M NaOH}$  solution, as advised by the manufacturer.

The  $^{90}\text{Sr}$  concentrations ( $C_{\text{Sr-90}}$ ; Bq) were calculated using Eq. (12) as

$$(12) \quad C_{\text{Sr-90}} = \frac{x \frac{\text{CPM}}{60} e^{-\lambda_{\text{Sr}} t}}{r_{\text{Sr}} m}$$

where  $x$  is determined from Eq. (6), CPM represents the count rate per minute,  $r_{\text{Sr}}$  is the strontium recovery,  $m$  is the sample mass (kg) and the time ( $t$ ) was measured from the 0.05 M nitric acid addition until the end of the liquid scintillation counting (LSC) measurement. The chemical recovery of strontium was calculated with Eq. (13), where  $m_{\text{carb}}$  is the mass of the dry carbonate (g),  $C_{\text{car}}$  is the concentration of the carrier solution ( $\text{g}\cdot\text{g}_{\text{sol}}^{-1}$ ) and  $m_{\text{car}}$  is the mass of the added carrier solution (g):

$$(13) \quad r_{\text{Sr}} = \frac{m_{\text{carb}}}{1.685 C_{\text{car}} m_{\text{car}}}$$

The matrix effect was assessed by analysing  $^{85}\text{Sr}$ -spiked samples. The analytical procedure was the same as that used for  $^{90}\text{Sr}$  determination until the end of the chromatography step. After collecting the strontium fraction, the solution was diluted to 100 mL and measured using a gamma spectrometer to determine the  $^{85}\text{Sr}$  recovery.

The MDA ( $\text{Bq}\cdot\text{kg}^{-1}$ ) [19] was calculated using Eq. (14) for each individual sample

$$(14) \quad \text{MDA} = \frac{2.71 + 4.65 \sqrt{\frac{\text{CPM}}{60}}}{3600 m}$$

The combined standard uncertainty was estimated using the Nordtest method; this approach has previously been described in [20].

## Results and discussion

Table 1 presents the recovery statistics as the standard deviation (SD) for both fortification levels. The testing revealed no significant difference between the immediate and delayed results.

The mean  $^{90}\text{Sr}$  recovery range indicates the negative bias of the procedure. The matrix effect was evaluated based on  $^{85}\text{Sr}$  recovery measurements, and the results also indicate 83% mean recovery of strontium. Neither LSC detection nor calculations based on the Bateman equation were used in this experiment. The recovery results from both experiments are consistent, which indicates that the bias originates from the chemical treatment of the sample. Further optimization should improve this procedure and minimize the bias.

Table 2 presents the performance characteristics of the applied method.

Another aspect of the validation involved proficiency tests, which were organized by Procorad (an experienced proficiency test provider; <http://procorad.org/>). To test the method, two spiked (21SRB and 21SRC) and one blank sample of urine were analysed. Table 3 shows the obtained test results

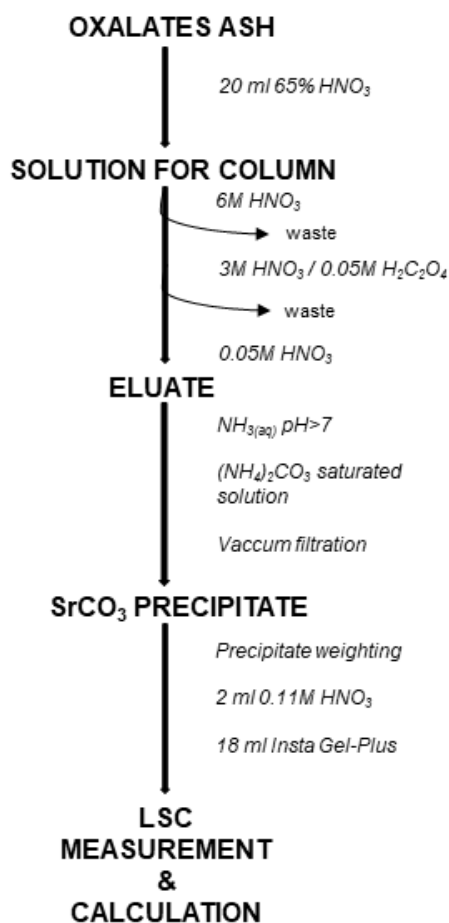


Fig. 2. Flow chart of the  $^{90}\text{Sr}$  determination procedure.

**Table 1.** Statistical evaluation of the <sup>90</sup>Sr recovery in immediate and delayed measurements

Fortification level (Bq·kg <sup>-1</sup> )	Immediate				Delayed			
	Mean (%)	Median (%)	SD (%)	<i>n</i>	Mean (%)	Median (%)	SD (%)	<i>n</i>
5	85	83	10	12	84	83	4	28
50	82	82	2	12	80	80	3	31
Total	83	82	7	24	82	81	4	59

SD, standard deviation.

**Table 2.** Validation characteristics of the applied method

Characteristics	Result
Linearity	100%
MDA	0.017 Bq·kg <sup>-1</sup>
Recovery of <sup>90</sup> Sr	82%
Relative bias	-18%
Repeatability (CV) <sup>a</sup>	8.5%
Reproducibility (CV) <sup>a</sup>	9%
Combined standard uncertainty ( <i>k</i> = 2) <sup>b</sup>	17%

<sup>a</sup> CV, coefficient of variation.

<sup>b</sup> *k*, coverage factor.

MDA, minimal detectable activity.

**Table 3.** Results from the Procorad 2021 proficiency test

Sample	Reference value (Bq·L <sup>-1</sup> )	Bateman equation-based procedure (Bq·L <sup>-1</sup> )
21SRB	3.11 ± 0.17	3.28 ± 0.56
21SRC	6.06 ± 0.34	6.06 ± 1.03

alongside reference values for comparison. The developed method detected a <sup>90</sup>Sr concentration in the blank sample below the MDA. Analysis of sample 21SRB resulted in a *z*-score of 0.5 and a normalized error of 0.3; acceptable values for *z*-score range from -2 to 2, and for normalized error, from -1 to 1. The result for the 21SRC sample obtained from analysis via the proposed method was not submitted for evaluation.

## Conclusions

To evaluate radiological hazards due to the accidental intake of radionuclides, it is necessary to employ analytical methods that can rapidly determine the levels of specific radionuclides, especially pure β-emitters, with sufficient accuracy. This report confirms that the proposed approach satisfies this requirement and achieves accuracy comparable with slower methods currently being used in laboratory practice. However, it must be stressed that the proposed method where <sup>90</sup>Sr fraction of total activity is calculated should not be used immediately after a nuclear accident, because it does not take <sup>89</sup>Sr into account.

Overall, the time required for analysis was shortened significantly thanks to the Bateman equation, which is beneficial for commercial services and scientific research regarding <sup>90</sup>Sr in any matrix. Whole analysis takes 8 days and chromatographical separation with LSC sample preparation can be done in 8 h. The analytical precision was not influenced

by implementing these calculations based on the Bateman equation. However, further optimizations of the sample treatment and chromatography stages are needed for matrices that are notorious for high Ca concentrations and those artificially fortified with mineral salts.

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**Competing interests.** The authors have no conflicts of interest to declare that are relevant to the content of this article.

**Data availability.** The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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