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VALIDATION OF METHODOLOGY AND UNCERTAINTY ASSESSMENT OF ANTIMONY DETERMINATION IN ENVIRONMENTAL MATERIALS USING NEUTRON ACTIVATION ANALYSIS.

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ABSTRACT

Antimony is an element found in low concentrations in the environment. However, its determination has attracted great interest because of the knowledge of its toxicity and increasing application. Neutron activation analysis (NAA) is a suitable method for the determination of several elements in different types, but in case of Sb, the analysis presents some difficulties due to spectral interferences. The objective of this research was to validate the method of NAA and uncertainty assessment for Sb determination in environmental samples. The experimental procedure consisted of irradiating twelve certified reference samples of different kind of matrices. The samples were irradiated in the nuclear research reactor IEA R1 IPEN/CNEN/SP followed by measurement of induced radioactivity, using a hyperpure germanium detector coupled to a gamma ray spectrometry. The radioisotopes ^{122}Sb and ^{124}Sb were measured and the Sb concentrations with their respective uncertainties were obtained by the comparative method. Relative errors and values of Z scores were calculated to evaluate the accuracy of the results for Sb determination in certified reference materials. The evaluation of the components that contribute to uncertainty measurement of the Sb concentration, showed that the major uncertainty contribution is due to statistical counting. The results also indicated that the uncertainty value of the combined standard uncertainty depends on the radioisotope measured and the decay time used for counting.

1. INTRODUCTION

Antimony is an element which for many years was not considered as an element of great importance from the environmental point of view, probably due to its low abundance in nature and poor solubitity of its compounds [1]. Presently, the concern on Sb determination has grown considerably due to the anthropogenic processes that increase concentrations in the environment and the growing application in industry. Chronic exposure by inhalation of this element can cause pneucomoconiosis diseases associated to obstructive pulmonary disease, cardiac abnormalities, increased blood pressure, abdominal pain, ulcers, skin and eye irritation [2]. Antimony is not considered carcinogenic. Nevertheless, according to the International Agency of Research on Cancer -IARC [3], antimony trioxide can be potentially carcinogenic in humans [4].

Regarding the analytical methodologies used in Sb determination, several techniques have been suggested in order to obtain reliable results, since its concentrations are low, requiring analytical instrumentation with low detection limits. Among the techniques for Sb determination in different matrices, the most employed ones are the flame atomic absorption spectrometry (FAAS), electrothermal atomic absorption spectrometry (ET AAS), optical emission spectrometry with an inductively coupled plasma source (ICP OES), inductively coupled plasma mass spectrometry (ICP-MS) [5-6] and neutron activation analysis (NAA) [7-10].

The neutron activation analysis technique has a high metrological level for the determination of several elements in different matrices.

The determination of this element in environmental and biological samples presents some analytical difficulties due its low concentrations and gamma ray spectrum interferences, in particular of ^{134}Cs and ^{152}Eu radioisotopes [11] and from ^{76}As, that emits gamma rays with energies of 559.10 and 563.23 keV very close to 564.24 keV of ^{122}Sb.

For calculation of the Sb uncertainty, a large number of factors can influence the result of the analytical determination and these are referred to as potential uncertainty sources. Normally, the measurement result consists only of an estimate and becomes applicable only when this is associated with an uncertainty [12].

In practice, the uncertainty of a result is due to many possible sources, including sampling, matrix effects and interferences, environmental conditions, uncertainties of masses and volumetric devices, reference values, approximations, measurement procedure and random variation [13].

The EURACHEM Guide [14] indicates a measurement uncertainty estimation process divided into four stages. The first step is the measurement specification, where is clearly defined what is being measured and for the estimate evaluation a mathematical formulation is needed where all possible uncertainty sources are considered, according to the relation 1 [15].

$$Y= f\,(X_1, X_2,\ldots, X_n) \qquad (1)$$

Where:

Y= Experimental measurement result
$X_1,\ldots, X_n$ = Uncertainty sources that influence the final result

In the second stage, once the measurand is specified, the possible sources that contribute to the parameters uncertainty are defined. These factors, which influence the analytical results are usually grouped and presented in a diagram of cause and effect, known as the Ishikawa diagram, or fishbone.

Figure 1 shows the diagram of cause and effect and the uncertainty sources associated with the comparative instrumental NAA [16-18] which was used in this study. In this figure the central vector C represents the measurement and the ramifications are the different factors contributions that affect the outcome of the analysis.

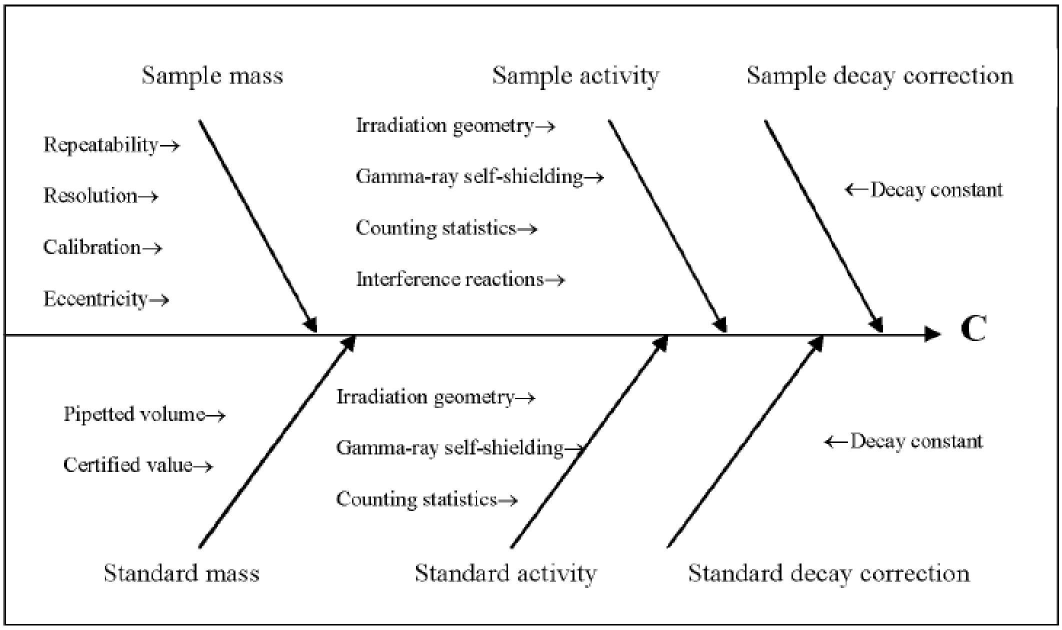

Figure 1: Cause and effect diagram for comparative NAA

In this study, the uncertainties of Sb determination results obtained by NAA in environmental and geological certified reference materials were evaluated and these analyses are discussed.

2. MATERIALS AND METHODS

2.1. Materials

Twelve reference materials were selected for Sb determination from biological and geological matrices produced by the following institutions: National Institute of Standards and Technology (NIST), USA; International Atomic Energy Agency (IAEA), Austria; Institute of Nuclear Chemistry and Technology (INCT), Poland and by the European Community Bureau of Reference (BCR), Belgium.

2.2. Procedure

Aliquots of approximately 180 mg of each CRM sample were irradiated together with the synthetic standard of Sb in sealed polyethylene bags previously cleaned using diluted nitric acid solution and Milli-Q purified water. Sb synthetic standard was prepared using a diluted solution of this element that was prepared using a certified solution of Sb, purchased from Spex Certiprep, USA. Samples and Sb standard (mass = 50µg) were wrapped separately in aluminum foil and irradiated together within sealed aluminum container suitable for irradiation inside the reactor core.

The samples and element standard were irradiated under a thermal neutron flux of about $5x10^{12}$ cm^{-2} s^{-1} for a period of 8 h for geological materials and of 16 h for biological materials in the IEA-R1 nuclear research reactor of the Nuclear and Energy Research Institute, IPEN-CNEN/SP.

The counting system consisted of a 2018GC hyperpure Ge detector coupled to a Digital Spectrum Analyzer DAS 100, both from Canberra. The nominal resolution (FWHM) of the system was 1.0 keV for the 122 keV peak of ^{57}Co and 1.78 keV for 1.332 keV peak of ^{60}Co. Samples and standard were measured twice for different decay times. The first measurement was performed after 7 to 9 days of decay time, using counting times of 3,600 s for the standard and of 36,000 s for the samples. The second measurement was performed after 13 to 16 days of decay time, using counting time of 5,400 s and 50,000 s for the standard and samples, respectively.

Samples and Sb standard were measured in the same geometry and the distance from sample to detector was chosen so that the dead time was less than 12%. The software Genie 2000 version 3.1 from Canberra was used for data acquisition and spectral processing. The radioisotopes were identified by its half-lives and gamma ray energies and the concentrations of elements were calculated by the comparative method, using the in-house built ESPECTRO program.

Concentrations of Sb in the materials were calculated from the counting data obtained for the peaks of 564 keV gamma ray energy of ^{122}Sb ($t_{1/2}$=2.7 d) and 1.692 keV gamma ray energy of ^{124}Sb ($t_{1/2}$=60.2 d). To express the Sb results on dry weight basis, approximately 200 mg of each certified reference material were dried at 85°C for 24 hours for moisture determination.

3. RESULTS AND DISCUSSION

3.1. Quantification of uncertainty components

Table 1 lists the contribution for calculation of the uncertainty of the sample mass. For the calculation, repeatability contribution was considered from the mass weighing measurement values (n =20) of approximately 200 mg, similar to the mass value of the subsample portions that were evaluated in the study. The other components considered were the resolution, calibration and eccentricity of the scale. Those values are provided in the scale calibration certificates.

Table 1. Contributions to the combined standard uncertainty in the sample mass

	Contributions			
	Repeatability	**Resolution**	**Calibration**	**Eccentricity**
Uncertainty (g)	1.00E-03	1.00E-05	1.00E-05	2.00E-05
Probability distribution	normal	rectangular	normal	rectangular
Factor	1	1/2√3	2	1/2√3
Standard uncertainty (g)	1.00E-03	2.89E-06	4.88E-06	5.77E-06
Combined uncertainty (g)	**1.00003E-03**			

The main contribution in the calculation of the sample standard combined uncertainty of standard was given by the uncertainty of weighing repeatability. Among the values provided in the certificate of the analytical scale, the values for the eccentricity of the scale, which measures its stability while weighing in different positions, showed the highest uncertainty value. The smallest contribution considered was for the calibration of the scale.

The results of the sample standard combined uncertainty of the standard's mass, shown in Figure 2, comprise an estimated uncertainty of various contributions.

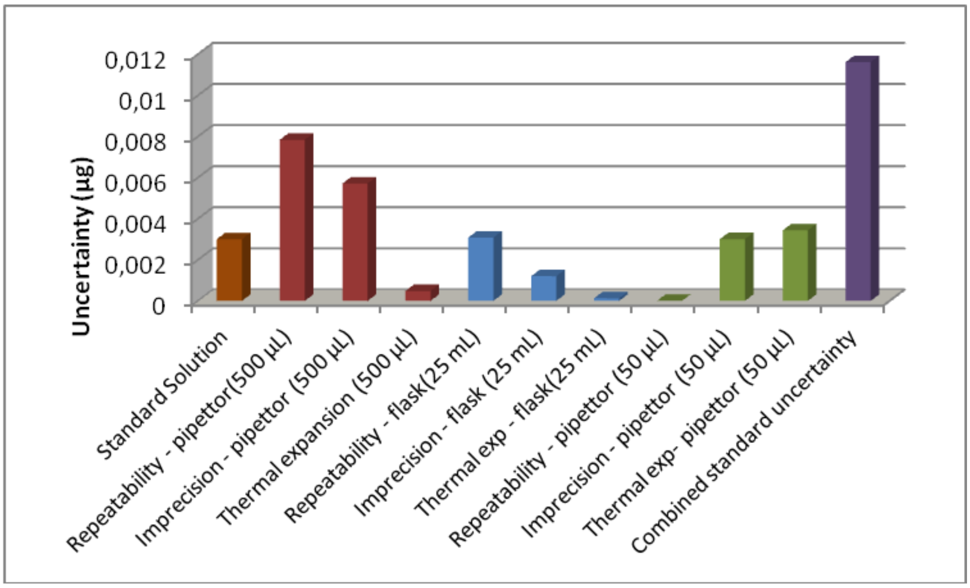

Figure 2: Contributions of the combined standard uncertainty in the mass of the standard

Firstly, the uncertainty of the standard solution concentration of the Sb stock, obtained from the solution's certificate, was considered. The contributions to uncertainties during dilution of the standard solution prepared in the laboratory was also considered. For this, the uncertainty of the volumetric flask utilized for dilution was used. The value is supplied by the producer. The contribution of the uncertainty for repeatability of the volumetric flask was obtained by the verification of the calibration results for the flask and obtained from thermal expansion difference between the temperatures of the laboratory, at the time of calibration, using expansion coefficient of 2.1 x 10^{-4} °C^{-1} for a variation 6 ^{o}C.

The contributions of the pipettor were also estimated from the values of the repeatability The contributions of the pipettor were also estimated from the values of the repeatability uncertainty, expressed by the standard deviation of the pipettor's calibration for the volume of standard solution used for dilution, from the inaccuracy of the pipette for the pipetted volume and the thermal expansion between the differences of the laboratory temperatures during the pipetting process. For the contributions of the uncertainty of pipetting the already diluted solution, in strips of Whatman filter paper number 40, the uncertainty of repeatability for the volume of solution pipetted in paper strips was considered.

The inaccuracy of the pipette for the same volume was also considered and the values of thermal expansion between the temperature differences of the laboratory. It was observed that in the calculation of the combined standard uncertainty for the mass of the sample, the repeatability uncertainty of the pippetor's calibration had an important contribution to the combined standard mass uncertainty of the standard.

To calculate the uncertainty contribution of the decay constant, the uncertainty of the half-life for ^{122}Sb and ^{124}Sb, obtained from the National Nuclear Data Center database [19], was used. Both half-lives were converted to minutes and the uncertainty of the decay constant (λ) propagated as exponential uncertainty.

The activity uncertainty shown in Table 2 was evaluated from the uncertainty score rate, calculated from the square root of the measured activity, since the measurement follows the Poisson distribution.

Table 2. Contribution of the count rate for the combined standard uncertainty of the activity in the sample and standard

Certified Reference Material	***Gamma ray Energies*** *(keV)*	**Sample**		**Standard**	
		u_a (%)	**u_a (%)**	**u_p (%)**	**u_p (%)**
		Decay Time 7-9 d	***Decay Time 13-16 d***	***Decay Time 7-9 d***	***Decay Time 13-16 d***
NIST SRM 1515 Apple Leaves	564.24	16.0	36.0	0.6	1.0
	1690.98	4.5	12.4	3.0	2.0
NIST SRM 1633b Constituents Elements in Coal Fly Ash	564.24	4.3	2.0	1.5	0.05
	1690.98	5.4	4.5	7.0	0.03
INCT MPH-2 Mixed Polish Herbs	564.24	2,4	12.6	0.3	1.0
	1690.98	36	15.1	0.05	2.0
INCT- TL-1 Tea Leaves	564.24	2.6	2.2	0.4	1.0
	1690.98	23.3	12.1	2.3	2.0
INCT – OBTL-5 Oriental Basma Tobacco Leaves	564.24	5.2	12.0	1.0	1.5
	1690.98	35.3	26.5	3.0	3.0
INCT-PVTL-6 Polish Virginia Tobacco Leaves	564.24	5.2	38.0	1.0	1.3
	1690.98	49.4	27.0	3.0	2.6
NIST SRM 1573a Tomato Leaves	564.24	17.4	32.0	1.0	1.0
	1690.98	32.0	32.2	3.2	2.0
IAEA SL-1Trace and Minor Elements in Lake Sediment	564.24	2.2	4.2	1.0	1.5
	1690.98	19.6	15.2	5.0	3.0
IAEA-140/TM Trace Elements and Methyl mercury in Seaweed (Focus sp)	564.24	25.0	9.5	1.0	1.0
	1690.98	--	25.6	3.2	2.2
IAEA 336 Trace and Minor Elements in Lichen	564.24	2.3	6.6	0.5	1.0
	1690.98	29.3	15.2	2.5	2.1
BCR 679 Trace Element on White Cabbage	564.24	43.4	17.4	1.1	1.4
	1690.98	17.1	21.0	5.0	3.0
CTA VTL-2 Virginia Tobacco Leaves	564.24	1.2	2.6	0.5	1.0
	1690.98	14.0	8.0	3.0	2.2

u_a= component of the combined standard uncertainty in the activity in the sample

u_p= component of the combined standard uncertainty in the activity in the standard

For the calculation of the statistical data counting uncertainty, data from four determinations of Sb were considered, using the ^{122}Sb peak of 564.24 keV and the ^{124}Sb peak of 1690.98 keV. The average activities for ^{122}Sb and ^{124}Sb peaks were estimated, for the four determinations, in each certified reference material and for the Sb standard, through error propagation.

From results obtained in Table 2, high uncertainty values for peak 1690.98 keV were obtained, in the first counting, with 7 days of decay time, due to low counting statistics. However, for the 15 days decay counting, the uncertainty results are smaller due to the decay of interfering activities of radionuclides that were present in the first counting of 7 days decay time.

Although the counting uncertainty results for the 1690.98 keV peak are low after 15 days of decay time when compared to the values obtained in the first counting, the results for the ^{124}Sb peak showed higher uncertainty values then for the 564.24 keV ^{122}Sb peak.

The results calculated for the standard uncertainty are lower when compared to the values of the uncertainties in the sample. In the first counting, the values ranged from 0.05% for the material INCT MPH-2 Mixed Polish Herbs to 5.0% for the material IAEA SL-1 Minor and Trace Elements in Lake Sediment using the 1690.98 keV ^{124}Sb peak. The same behavior was observed for the uncertainties in the statistical counting of the sample, where the standards showed high uncertainty values in the second counting, for the 1690.98 keV peak and lower for the 564.24 keV peak.

Table 3 contains the results of the other factor of uncertainty in the activity of the sample, the uncertainty in the attenuation range, which was evaluated from a test conducted with a synthetic standard sample and two certified reference materials.

Table 3. Contribution of gamma attenuation for the combined standard uncertainty of the activity in the sample and standard

Certified Reference Material	***Decay Time 7-9 d***				
	Energy (keV)	Activity (***Standard)*** (cps)	Activity ***(Standard + Sample)*** (cps)	Ratio	u_{at}(%)
INCT MPH-2	564.24	30.664	26.852	0.9	12.4
	1690.98	1.630	1.591	1.0	2.4
IAEA SL	564.24	30.664	28.285	0.9	7.7
	1690.98	1.630	1.578	1.0	3.2
Certified Reference Material	***Decay Time 13-16 d***				
	Energy (keV)	Peack Area (***Standard)*** (cps)	Peack Area ***(Standard + Sample)*** (cps)	Reason	u_{at}(%)
INCT MPH-2	564.24	5.119	4.225	0.8	17.5
	1690.98	1.496	1.425	0.9	4.7
IAEA SL-1	564.24	5.119	4.701	0.9	8.2
	1690.98	1.496	1.462	1.0	2.3

The experimental procedure for the test consisted in irradiating a synthetic Sb standard under the same irradiation conditions of the standards and samples of this study.

There were two measurement series, the first with about 7 days of decay for a period of 36.000 seconds and the second with approximately 15 days of decay, with a counting time of 50.000 seconds. In the first stage of the test, the irradiated synthetic standards counting was made, fixed in the external side of the stainless steel support, alongside another empty stainless steel support. After the activity measurement of the Sb standard, the counting of the synthetic standard was made alongside the non-irradiated reference material sample.

From the obtained difference between the activities (cps) from the Sb peaks in both measurements series, the ratio between the activities and its were determined.

The results of the obtained uncertainties showed superior results for the 7 and 15 days decay counting considering the 564.24 keV peak. It is possible to observe the decrease of the uncertainty value, considering the 1690.98 keV peak, due to the spectrum resolution, significantly better for higher energy peaks and it was also observed that the uncertainty values were superior for the reference material of geological origin IAEA SL-1, possibly due to their density. The standard uncertainties of the contributions considered in the research were combined by the relative method.

The estimated contributions for the calculation of the combined standard uncertainties are presented on Figure 3 for the CRM NIST SRM 1515. These uncertainties results are given for the two Sb peaks, the ^{122}Sb 564.24 keV peak and the ^{124}Sb 1690.98 keV peak, for the 7 and 15 day decay times.

The results of Figure 3 indicate that the main contribution for the combined standard uncertainty for sample is caused from the activity uncertainty, resulted by the counting ratio. The analysis showed that the combined standard uncertainty values are higher for the ^{122}Sb 564.24 keV peak for the 15 days decay counting and for ^{124}Sb 1690.68 keV peak for the 7 days decay counting, following expected trend, of the counting ratio reduction with the ^{122}Sb decay and the complexity of the ^{124}Sb spectrum, with 7 days of decay.

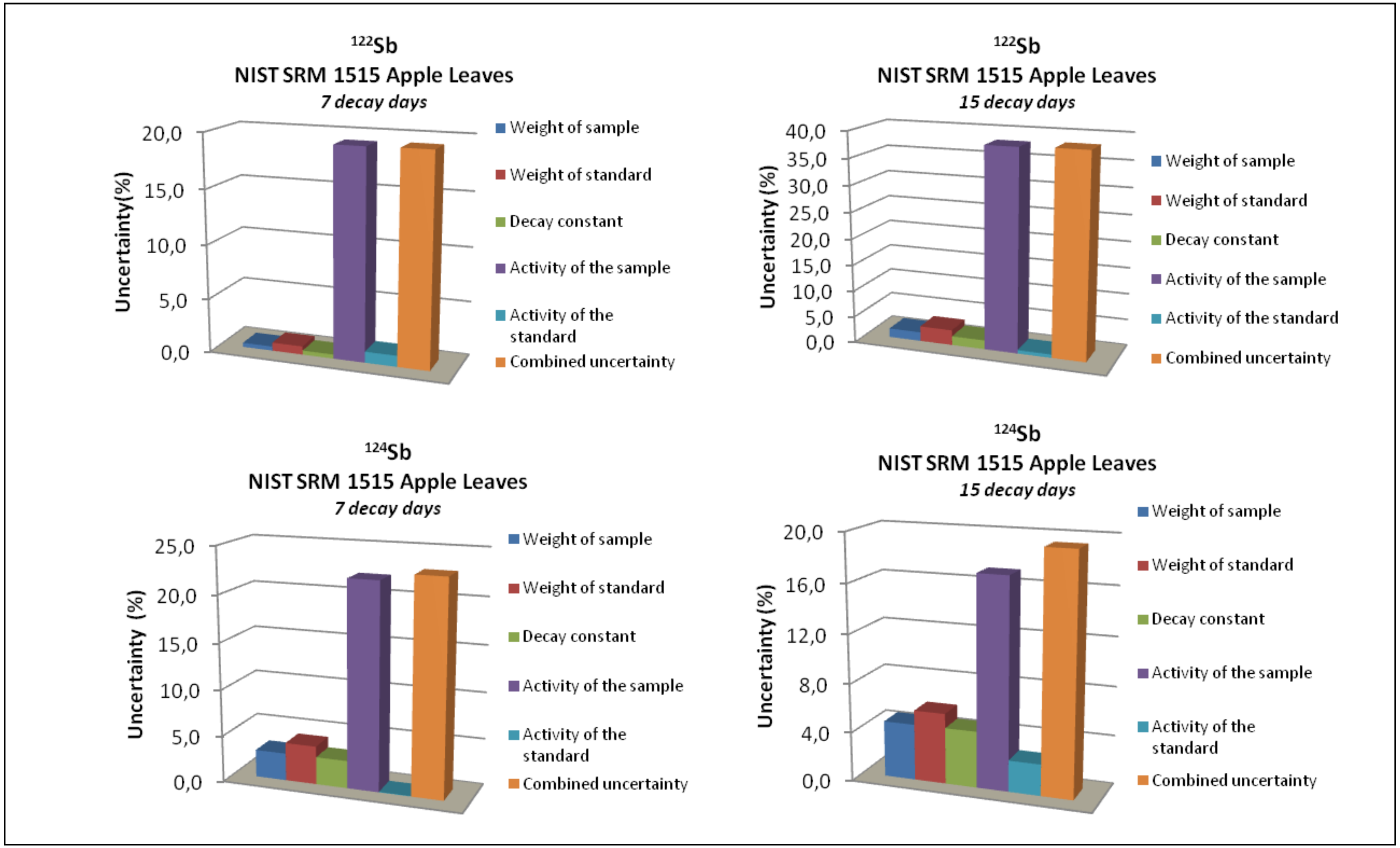

Figure 3: Percentages of uncertainty contributions and the combined standard uncertainty in the determination for Sb in the material NIST SRM 1515

3.2. Results of Sb Concentrations With the Expanded Uncertainty Obtained in Reference Materials

The expanded uncertainties of the Sb determination in reference materials were obtained from the combined standard uncertainties, calculated using the 564.24 keV peak for ^{122}Sb and the 1690.98 keV peak for ^{124}Sb for the 7 and 15 day decay countings, the k=2 coverage factor for a confidence level of 95% [20].

Table 4 presents the results of the Sb concentrations obtained with values of the certificates for comparison. This Table shows the weighted averages (C) of the Sb concentrations results obtained in four determinations with the expanded uncertainty. The relative error (RE) and Z score value were also calculated.

The results that showed a good agreement with the values of certificate are present in bold, for these results (Table 4) the relative errors ranged from 0.1 to 12.6% with | Z | smaller than 2, indicating that these results are within the certified values range at a confidence level of 95%.

For reference materials INCT-MPH-2 Mixed Polish Herbs and IAEA-336 Lichen, the obtained results were consistent with the certificate data, using the 564.24 keV peaks for ^{122}Sb and the 1690.98 keV peak for ^{124}Sb for the 7 and 15 days decay time. For the material NIST SRM 1573rd Tomato Leaves results consistent with the certificate value were also obtained, using the ^{122}Sb and ^{124}Sb peaks and decay time of the 15 days.

Table 4: Concentrations of Sb (mg.kg^{-1}), Relative Errors and Z score value obtained of CRM analyses

Reference Materials	**Gamma ray energy (keV) measured**	***Decay Time 7-9 d***				**Gamma ray energy (keV) measured**	***Decay Time 13-16 d***				**Values of the Certificates** *(mg kg^{-1})*
		C^a ± U^b *(mg kg^{-1})*		***RE(%)***	***Z***		***C^a ± U^b*** *(mg kg^{-1})*		***RE(%)***	***Z***	
NIST SRM 1515 Apple Leaves	564.24	0.03 ± 0.018	(n=4)	--	--	564.24	0.04 ± 0.022	(n=4)	--	--	[0.013]^c
	1690.98	<0.040	(n=1)	--		1690.98	0.03 ± 0.017	(n=1)	--		
NIST SRM 1633b Constituents Elements in Coal Fly Ash	564.24	5.0 ± 1.032	(n=4)	--	--	564.24	5.9 ± 0.316	(n=4)	--	--	[6]^c
	1690,98	4.8 ± 1.0	(n=4)	--		1690,98	**4.5 ± 0.6**	(n=4)	--		
INCT MPH-2 Mixed Polish Herbs	564.24	**0.065 ± 0.003**	(n=4)	0.8	-0.1	564.24	**0.07 ±0.016**	(n=4)	12.7	0.5	0.0655±0.0091
	1690,98	**0.06 ± 0.013**	(n=4)	9.3	-0.2	1690,98	**0.068 ± 0.044**	(n=4)	3.8	0.1	
INCT- TL-1 Tea Leaves	564.24	0.081 ± 0.005	(n=4)	--	--	564.24	0.37 ± 0.018	(n=4)	--	--	[0.050]^c
	1690,98	0.09 ± 0.06	(n=3)	--		1690,98	0.04 ± 0.02	(n=4)	--		
INCT – OBTL-5 Oriental Basma Tobacco Leaves	564.24	0.60 ± 0.01	(n=4)	694.7	27.5	564.24	0.50 ± 0.01	(n=4)	602.0	7.7	0.0755 ± 0.0125
	1690,98	**0.07 ± 0.16**	(n=4)	7.3	-0.03	1690,98	**0.08 ± 0.12**	(n=4)	6.0	0.04	
INCT-PVTL-6 Polish Virginia Tobacco Leaves	564.24	**0.03 ± 0.017**	(n=4)	11.3	-0.2	564.24	0.15 ± 0.037	(n=4)	303.2	3.1	0.0372 ± 0.0039
	1690,98	0.09 ± 0.08	(n=4)	136.6	0.6	1690,98	**0.03 ± 0.03**	(n=4)	11.3	-0.1	
NIST SRM 1573a Tomato Leaves	564.24	0.04 ± 0.031	(n=1)	36.5	-0.8	564.24	**0.07 ± 0.04**	(n=3)	11.1	0.2	0.063±0.006
	1690,98	0.08 ± 0.13	(n=1)	27.0	0.1	1690,98	**0.07 ± 0.05**	(n=4)	6.3	0.1	
IAEA SL-1Trace and Minor Elements in Lake Sediment	564.24	**1.4 ± 0.302**	(n=4)	8.3	0.2	564.24	2.12 ± 0.183	(n=4)	58.8	1.7	[1.31(1.19- 1.43)]); n=15, p=0.05
	1690,98	**1.1 ± 0.537**	(n=4)	12.6	-0.2	1690,98	0.5 ± 0.379	(n=4)	59.5	-1.4	
IAEA-140/TM Trace Elements and Methyl mercury in Seaweed (Focus sp)	564.24	0.06 ± 0.013	(n=2)	38.8	-0.6	564.24	0.12 ± 0.023	(n=4)	16.5	0.2	0.103(0.081-0.125) n=12, p=0.05
	1690,98	0.02 ± 0.021	(n=1)	76.7	-0.4	1690,98	**0.10 ± 0.07**	(n=4)	3.0	-0.02	
IAEA 336 Trace and Minor Elements in Lichen	564.24	**0.079 ± 0.018**	(n=4)	8.2	0.3	564.24	0.08 ± 0.011	(n=4)	13.7	0.5	0.073(0.063-0.083); n=12; p=0.05
	1690.98	**0.08 ± 0.055**	(n=4)	4.1	0.1	1690.98	**0.07 ± 0.032**	(n=4)	0.1	0.003	
BCR 679 Trace Element on White Cabbage	564.24	0.022 ± 0.020	(n=4)	7.0	0.07	564.24	0.03 ± 0.024	(n=2)	31.0	0.3	0.0206±0.0014
	1690,98	<0.139	(n=1)	--	--	1690,98	0.02 ± 0.10	(n=2)	16.5	0.03	
CTA VTL-2 Virgínia Tobacco Leaves	564.24	0.25 ± 0.062	(n=4)	20.0	-0.9	564.24	**0.35 ± 0.020**	(n=4)	12.2	0.5	0.312±0.025
	1690,98	**0.3 ±0.127**	(n=4)	0.6	-0.02	1690,98	**0.32 ± 0.080**	(n=4)	2.9	0.3	

^{a}= weighted average concentrations ^{b}= expanded uncertainty, calculated using a factor k=2, for a confidence level of 95%; ^{c} Informative value

4. CONCLUSIONS

The results obtained in this study demonstrated the possibility of determination of Sb in biological and geological samples using neutron activation analysis, depending on its content in the sample and establishing appropriate experimental conditions for the detection of ^{122}Sb and ^{124}Sb radioisotopes. Among the twelve reference materials analyzed by INAA method, the most precise and accurate results were obtained for the reference materials INCT MPH-2 Mixed Polish Herbs, IAEA SL-1 Minor and Trace Elements in Lake Sediment, IAEA-140/TM Trace Elements and Methyl mercury in Seaweed (Focus sp), IAEA 336 Trace and Minor Elements in Lichen, CTA-VTL-2 Virginia Tobacco Leaves.

For other certified reference materials, the data showed the need to eliminate the problem of spectral interferences by subtracting scores, or by radiochemical separation. In the evaluation of the contribution sources for uncertainty in the concentration of Sb, the major contribution that was found is due to the counting rate in the sample. The results of this study indicated that the values of the combined standard uncertainties depend on the radioisotope measured and the decay time.

For the ^{122}Sb peak, the 15 days decay counts and the ^{124}Sb for 7 days decay counts, high values of the combined uncertainties were obtained, due to the low counting rates obtained for the ^{122}Sb peak and the complexity of the spectrum for detecting ^{124}Sb with 7 days of decay

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