

## Investigation of Contaminated Environmental Samples; US DOE EML Laboratory's Quality Assessment Programme

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**Summary:** A Quality Assessment Programme (QAP) was initiated by the US, Department of Energy (DOE) in 1998 to establish reliability in the measurement of radionuclides in contaminated environmental samples, *i.e.* soil, vegetation and exposed air filters. For this purpose, recognized relevant laboratories around the world were contacted and invited to participate in a regular proficiency programme. The Neutron Activation Analysis (NAA) Miniature Neutron Source Reactor (MNSR) Laboratory at the Chemistry Division (CD), Directorate of Science, Pakistan Institute of Nuclear Science and Technology (PINSTECH), an IAEA Regional Resource Unit (IAEA-RRU), along with 76 other international laboratories participated in this exercise. In this paper, the performance of the NAA MNSR Laboratory throughout the QAP Programme (1998-2004) is presented in detail, describing the procedures employed, the problems encountered and the improvements and expertise gained from participating in this exercise.

### Introduction

After the cold war, many former production sites in the USA were converted for environmental restoration and long term monitoring [1]. Therefore, to monitor the degree of radionuclide contamination in the environment originating from nuclear activities became a requirement. As most of the waste found at the US Department of Energy (DOE) facilities is a mixture of anthropogenic radionuclides in complex organic and inorganic matrices; they were extremely difficult to characterize. To avoid any discrepancy between the results reported by various laboratories analyzing these samples and to obtain reliable results, the Environmental Measurement Laboratory (EML) of U.S. DOE initiated a Quality Assessment Programme (QAP) in March 1998 (QAP 48) which ended in May 2004 (QAP 60) [2].

Two proficiency exercises were performed annually during the QAP programme; first in April/May and the second in September. The samples analyzed are subjected to regulatory protocol and include soil, vegetation, water and exposed air filters. Details of these are given in Table-1. The participating laboratories transmitted the results of these exercises, for any of the radionuclides listed in Table-2, online to EML where they were assessed and compiled into a report. These reports were published on the EML website [3] in June/July and

December of each year, respectively. Complete result files in pdf format with the performance evaluation and ratings for participating laboratories throughout the QAP programme are available on this website and hardcopies are available from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Road, Springfield, Virginia 22161.[4-14] The NAA MNSR laboratory has participated in this programme on a regular basis, from its beginning and over the years has gained experience in the measurement of radionuclides in contaminated environmental samples. An overview is given in this paper of the employed analytical procedures, the results obtained and the knowledge and experience gained by taking part in this exercise.

### Results and Discussion

The results obtained during the QAP Programme have been published in the series of Semi-Annual Reports of the Department of Energy, Office of Environmental Management, Quality Assessment Programme [EML-596, EML-600, EML-604, EML-605, EML-608, EML-611, EML-613, EML-615, EML-617, EML-618 and EML-621 for the QAP Exercises 48 (9803) to 58 (0303)] [4-14]. The results submitted by the NAA MNSR laboratory have been reproduced here along with our initial

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Table-1: Details of samples analyzed during the U.S. DOE's QAP programme (1998-2004).

QAP Exercise No.	Samples Analyzed	Soil/ Vegetation Sample Height	Standards Used	
QAP 48 (9803)	Air Filter	1 cm	IAEA-326	IAEA-327 (Soil)
	Soil		IAEA-375 (Soil)	
	Vegetation		IAEA-152 (Milk Powder)	
QAP 49 (9809)	Air Filter	1 cm	SO9803	AI9803 VE9803
	Soil		IAEA-326	IAEA-327 (Soil)
	Vegetation		IAEA-375 (Soil)	
QAP 50 (9903)	Air Filter	1 cm	SO9803	AI9803 VE9803
	Soil		SO9809	AI9809 VE9809
	Vegetation		IAEA-326	IAEA-327 (Soil)
QAP 51 (9909)	Air Filter	1 cm	SO9803	AI9803 VE9803
	Soil		SO9809	AI9809 VE9809
	Vegetation		SO9903	AI9903 VE9903
QAP 52 (2003)	Air Filter	1 cm	SO9809	AI9803 VE9803
	Soil		SO9803	AI9809 VE9809
	Vegetation		SO9909	AI0003 VE9903
QAP 53 (2009)	Air Filter	1 cm	SO9903	AI9909 VE9909
	Soil		SO9809	AI9909 VE0003
	Vegetation		SO9909	AI0009 VE9803
QAP 54 (0103)	Air Filter	1 cm	SO9903	AI0003 VE9903
	Soil		SO0003	AI9803 VE0003
	Vegetation		SO0009	AI9809 VE9809
QAP 55 (0109)	Air Filter	1 inch	SO9909	AI0009 VE0009
	Soil		SO0103	AI0103 VE0103
	Vegetation		SO0003	AI0003 VE0009
QAP 56 (0203)	Soil	1 inch	SO0009	AI0009 VE0009
			SO0103	
			SO0103	
QAP 57 (0209)	Air Filter	1 inch	SO0103	AI0103
	Soil		SO0203	AI0203
			SO0109	AI0109
*QAP 58 (0303)	Soil	1 inch	SO0109	
			SO0209	
			SO0209	AI0209
QAP 59 (0309)	Air Filter	1 inch	SO0203	AI0203
	Soil		SO0109	AI0109
			SO0109	AI0109
QAP 60 (0403)	Air Filter	1 inch	SO0209	AI0103 VE0203
	Soil		SO0203	AI0109 VE0209
	Vegetation		SO0109	AI0203 VE9809
			AI0209	

\*Results not submitted to EML

unpublished results of QAP 59 and QAP 60. The recalculated results of QAP 50 are also shown, as a calculation error was identified in the initial results submitted to EML. These results are presented in Tables-3 to 14. The data presented in these Tables consists of all values obtained with our  $\gamma$ -spectroscopy system for each matrix.

The samples investigated are contaminated environmental samples. However, as discussed later in the experimental section, the activity level in these samples is extremely low. Therefore the count rates obtained from these samples were only slightly higher than those obtained from similar un-exposed or inactive environmental samples. A study of the statistical summaries of any QAP Programme given

Table-2:  $\gamma$  Energy peaks used to quantify radionuclides in environmental matrices during the QAP programme (1998-2004).

Radionuclide	Half-life	E $\gamma$ (keV)	% Abundance
<sup>241</sup> Am	433 A	59.5	36.3
<sup>234</sup> Th	24.1 D	63.3	3.9
		92.8	3
		92.38	2.57
<sup>212</sup> Pb	10.64 H	238.63	43.1
		77.11	17.5
		74.8	9.6
<sup>214</sup> Pb	26.8 M	351.99	37.1
		295.22	19.2
		604.66	97.6
<sup>134</sup> Cs	2.062 A	795.76	85.4
<sup>214</sup> Bi	19.9 M	609	46.09
		1729.6	15.92
<sup>137</sup> Cs	30.1 A	1120.28	15.04
		661.62	84.6
		32.19	3.7
<sup>212</sup> Bi	60.55 M	727.17	11.8
		1620.56	2.75
		785.42	2.2
<sup>54</sup> Mn	312.2 D	834.81	99.978
		911.07	29
<sup>228</sup> Ac	6.13 H	968.9	17.46
		338.4	12.01
		122.07	85.6
<sup>57</sup> Co	274.74 D	136.43	10.6
<sup>60</sup> Co	5.2721 A	1173.23	99.86
		1332.51	99.98
<sup>40</sup> K	1.28X10 <sup>9</sup> A	1460.75	10.7

\*where A = years, D = Days, H = Hours and M = Minutes

in QAP reports EML-596 to EML-621[4-14] shows that via  $\alpha$ ,  $\beta$  and  $\gamma$ -spectrometry, consensus values for around 10, 14 and 8 (32 in total) radionuclides were obtained in air filter, soil and vegetation matrices, respectively. The numbers of radionuclide which could be measured in air filter, soil and vegetation samples with  $\gamma$ -spectrometry are about 5, 9 and 5 (19 in total), respectively. As most participating laboratories used one or more spectroscopic techniques and may not have analyzed all three matrices they generally reported fewer than 19 radionuclides. Thus the number of radionuclides presented in Tables 3 to 14 is close to the possible number of radionuclides that can be measured in these samples using  $\gamma$ -spectrometry for individual matrices.

During each QAP exercise, the results had to be submitted to EML within 90 days of the start of the exercise. Due to time constraints and other unforeseeable reasons the soil and air filter samples were analyzed first and if sufficient time was available, the vegetation sample was also analyzed. Therefore vegetation samples were not analyzed after QAP 55 (0109) exercise and the results presented in Table-14, for the vegetation sample in the QAP 60 (0403) exercise, were obtained after the QAP 60

Table-3: Results of QAP 48 (9803) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	EML Evaluation	QAP 47 Evaluation
Matrix: AI Air Filter Bq / filter								
1	<sup>134</sup> Cs	346.800	37.400	19.740	1.380	17.568	N	
1	<sup>137</sup> Cs	218.900	9.500	11.860	0.957	18.457	N	
Matrix: SO Soil Bq / kg								
1	<sup>40</sup> K	362.400	28.300	313.500	10.150	1.156	A	
1	<sup>137</sup> Cs	380.200	8.800	329.500	9.260	1.154	A	
Matrix: VE Vegetation Bq / kg								
1	<sup>40</sup> K	783.300	9.400	707.500	24.987	1.107	A	
1	<sup>137</sup> Cs	189.600	8.600	181.500	7.141	1.045	A	

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

Table-4: Results of QAP 49 (9809) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	EML Evaluation	QAP 48 Evaluation
Matrix: AI Air Filter Bq / filter								
1	<sup>60</sup> Co	9.035	0.380	9.160	0.580	0.986	A	
1	<sup>137</sup> Cs	21.190	0.380	22.470	1.030	0.943	A	N
1	<sup>54</sup> Mn	4.990	0.520	4.920	0.400	1.014	A	
1	<sup>125</sup> Sb	8.081	0.300	8.890	0.550	0.909	A	
Matrix: SO Soil Bq / kg								
1	<sup>137</sup> Cs	946.260	39.350	954.000	38.000	0.992	A	A
Matrix: VE Vegetation Bq / kg								
1	<sup>137</sup> Cs	430.240	12.230	390.000	20.000	1.103	A	A
1	<sup>40</sup> K	449.980	50.150	460.000	20.000	0.978	A	A

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

Table-5a: QAP 50 (9903) preliminary results submitted to EML by NAA MNSR laboratory PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	EML Evaluation	QAP 49 Evaluation
Matrix: AI Air Filter Bq / filter								
1	<sup>57</sup> Co	6.560	0.340	3.010	0.140	2.179	N	
1	<sup>60</sup> Co	6.290	0.190	4.960	0.280	1.268	W	A
1	<sup>137</sup> Cs	6.480	0.290	6.050	0.300	1.071	A	A
Matrix: SO Soil Bq / kg								
1	<sup>137</sup> Cs	712.790	49.870	659.500	24.950	1.081	A	A
1	<sup>40</sup> K	390.460	120.210	362.750	20.156	1.076	A	
Matrix: VE Vegetation Bq / kg								
1	<sup>60</sup> Co	38.970	11.200	21.450	1.000	1.817	N	
1	<sup>137</sup> Cs	572.070	27.380	467.000	20.000	1.225	A	A
1	<sup>40</sup> K	625.840	86.680	656.500	20.000	0.953	A	A

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

Table-5b: Corrected results of QAP 50 (9909) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	EML Evaluation	QAP 49 Evaluation
Matrix: AI Air Filter Bq / filter								
1	<sup>57</sup> Co	2.569	0.134	3.010	0.140	0.853	W	
1	<sup>60</sup> Co	5.512	0.165	4.960	0.280	1.111	A	A
1	<sup>137</sup> Cs	6.330	0.280	6.050	0.300	1.046	A	A
Matrix: SO Soil Bq / kg								
1	<sup>137</sup> Cs	696.8	120.2	659.500	24.950	1.057	A	A
1	<sup>40</sup> K	390.45	48.75	362.750	20.156	1.076	A	
Matrix: VE Vegetation Bq / kg								
1	<sup>60</sup> Co	27.20	89.21	21.450	1.000	1.268	W	
1	<sup>137</sup> Cs	519.25	3.99	467.000	20.000	1.112	A	A
1	<sup>40</sup> K	622.71	50.87	656.500	20.000	0.947	A	A

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

exercise ended. None of the QAP 58 (0303) exercise samples were analyzed and consequently no results were reported to EML. QAP exercise 50 (9903) results initially submitted to EML are given in Table-5.a. These results did not compare well with the EML results. Therefore the results were re-examined and it was discovered that the wrong reference date (01/03/98 instead of 01/03/99) was used to calculate

these activities. The correct results are presented in Table-5.b and show better agreement with the EML data.

#### Performance Evaluation

Control limits for performance evaluation have been presented for each analyte/ matrix pair in

Tables 3 to 14. These were assigned by EML and were initially established from percentiles of historic QAP data distributions for the last 10 years (1982-1992) [15]. The limits are re-evaluated on an annual basis using the most recent 10 years of historic data. Therefore a participant's analytical performance is evaluated based on the historical analytical capabilities for individual analyte/matrix pairs. A frequency distribution is calculated from all reported measurements relative to the EML value over the previous ten years of the programme for each analyte/matrix pair. The criteria for acceptable performance, "A", has been chosen to be between the 15<sup>th</sup> and the 85<sup>th</sup> percentile of the cumulative normalized distribution, which can be viewed as the middle 70% of all historic measurements. The acceptable with warning criteria, "W", is between the 5<sup>th</sup> and 15<sup>th</sup> percentile and the 85<sup>th</sup> and 95<sup>th</sup> percentile. In other words, the middle 90% of all reported values are acceptable, while the outer 5<sup>th</sup> - 15<sup>th</sup> (10%) and 85<sup>th</sup>-95<sup>th</sup> percentiles (10%) are in the warning area. The not acceptable criteria, "N", is established at less than the 5<sup>th</sup> percentile and greater than the 95<sup>th</sup> percentile, that is, the outer 10% of the historical data.

#### *Pitfalls in Measurements*

Looking at the results in Tables 3 to 14, [4-14] it can be seen that, generally, fewer radionuclides were reported in vegetation by our laboratory while the greatest numbers of radionuclide were reported in soil samples. In soil samples our reported values were in close agreement with the EML values. This was due to the fact that higher count rates were obtained from soil samples as compared to the air filter and vegetation samples. The results for the glass fiber filter samples generally fulfilled the acceptable criteria but our laboratory did not report many radionuclides for this matrix. This point will be taken up later when the performance of the NAA MNSR laboratory throughout the QAP Programme will be discussed.

Scrutiny of Table-14 reveals that after submission of the preliminary results the data for the soil sample were examined again and data for two more radionuclides, <sup>214</sup>Bi and <sup>214</sup>Pb, have been included in this Table. The activities of these radionuclides were not reported earlier as in the past QAP exercises our results for these radionuclides

generally did not compare well with the EML results. Initially two sets of values were obtained for <sup>234</sup>Th and the results obtained with 2 out of the 3 reference samples (SO0109 and SO0203) were submitted to EML. However re-examination of the results shows better comparison of the EML results with the 3<sup>rd</sup> reference sample (SO0209), as this RM has a reference date closest to the actual QAP 60 soil sample. This shows that using earlier intercomparison samples with consensus values may provide incorrect results even when up to three different such samples are used as RMs, as is the case here. Thus, it is recommended that, wherever possible, certified reference materials (CRMs) and standards should be used. To overcome this problem, these RMs should have been analysed in each analysis run and the results should have been followed over the entire study period to demonstrate that no statistically deviating results or trends are present. As can be seen from Table-1, these RMs have been infrequently analysed throughout the QAP programme. These results were compiled for quality assurance purposes and show that with the passage of time greater confidence was obtained in our results. Consequently the numbers of radionuclides reported at the end of the QAP Programme were greater than at its beginning. Moreover, as the study progressed, better agreement with EML data was obtained for all matrices.

#### *Performance of NAA MNSR Laboratory Throughout the QAP Programme*

The performance of our laboratory, as evaluated by EML, has been compiled from Tables 3 to 14 into Table-15 for the entire QAP Programme for the three matrices studied. The total numbers of radionuclide reported in each exercise are also listed in Table-15. It can be seen from this table that 2 to 4 radionuclides were quantified in air filter samples and apart from the results of the initial two QAP exercises all data were considered acceptable by EML. The results for the soil matrix show that as many as 9 radionuclides were reported for QAP 56. The vegetation matrix data ends at QAP 55, after which no results for this matrix were submitted to EML. In each QAP exercise 2 to 3 radionuclides were reported for this matrix. Comparing the results of all three matrices it was observed that due to the lower count rates obtained for the vegetation samples, the results cited for this matrix differed the most from the EML

Table-6: Results of QAP 51 (9909) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	Evaluation	QAP 49 Evaluation
Matrix: AI Air Filter Bq / filter								
1	<sup>57</sup> Co	6.420	0.430	7.730	0.033	0.831	A	N
1	<sup>60</sup> Co	7.470	1.410	6.350	0.410	1.176	W	W
1	<sup>137</sup> Cs	7.220	0.550	6.430	0.420	1.123	A	A
1	<sup>54</sup> Mn	8.720	0.470	7.910	0.450	1.102	A	
Matrix: SO Soil Bq / kg								
1	<sup>228</sup> Ac	116.300	32.500	124.000	4.800	0.938	A	
1	<sup>212</sup> Bi	145.000	23.200	140.000	14.000	1.036	A	
1	<sup>214</sup> Bi	54.990	3.750	69.500	1.800	0.791	W	
1	<sup>137</sup> Cs	237.670	9.700	204.000	5.000	1.165	A	A
1	<sup>40</sup> K	993.100	73.100	780.000	27.000	1.273	W	A
1	<sup>212</sup> Pb	109.300	31.700	127.000	4.800	0.861	W	
1	<sup>214</sup> Pb	61.360	17.200	72.000	0.420	0.852	W	
Matrix: VE Vegetation Bq / kg								
1	<sup>137</sup> Cs	394.000	4.800	440.000	20.000	0.895	W	A
1	<sup>40</sup> K	275.500	38.800	513.000	20.000	0.537	N	A

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

Table-7: Results of QAP 52 (2003) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	Evaluation	QAP 51 Evaluation
Matrix: AI Air Filter Bq / filter								
1	<sup>57</sup> Co	4.760	0.490	5.310	0.220	0.896	A	A
1	<sup>60</sup> Co	6.360	0.530	5.320	0.260	1.195	W	W
1	<sup>137</sup> Cs	6.730	0.610	6.100	0.300	1.103	A	A
1	<sup>54</sup> Mn	28.000	1.790	27.200	0.800	1.029	A	A
Matrix: SO Soil Bq / kg								
1	<sup>137</sup> Cs	311.800	11.100	339.000	9.300	0.920	A	A
1	<sup>40</sup> K	837.300	66.400	811.000	29.000	1.032	A	W
1	<sup>212</sup> Pb	90.290	6.060	97.300	4.600	0.928	A	W
1	<sup>214</sup> Pb	52.100	1.900	86.500	6.800	0.602	N	W
Matrix: VE Vegetation Bq / kg								
1	<sup>60</sup> Co	19.570	2.700	52.800	1.000	0.371	N	
1	<sup>137</sup> Cs	1319.000	48.000	1380.000	20.000	0.956	A	W
1	<sup>40</sup> K	516.600	85.300	521.000	20.000	0.992	A	N

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

Table-8: Results of QAP 53 (2009) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	Evaluation	QAP 52 Evaluation
Matrix: AI Air Filter Bq / filter								
1	<sup>57</sup> Co	15.600	1.200	14.550	0.460	1.072	A	A
1	<sup>60</sup> Co	7.720	0.230	8.430	0.480	0.916	A	W
1	<sup>137</sup> Cs	8.040	0.120	7.410	0.360	1.085	A	A
1	<sup>54</sup> Mn	45.700	2.200	43.200	1.300	1.058	A	A
Matrix: SO Soil Bq / kg								
1	<sup>228</sup> Ac	77.300	4.200	80.200	3.600	0.964	A	
1	<sup>214</sup> Bi	103.300	10.000	83.300	4.200	1.240	A	
1	<sup>137</sup> Cs	1055.000	26.000	1020.000	51.000	1.034	A	A
1	<sup>212</sup> Pb	67.600	3.200	79.300	4.300	0.852	W	A
1	<sup>214</sup> Pb	9.300	2.300	86.300	4.300	0.108	N	N
1	<sup>234</sup> Th	223.000	54.000	148.000	10.000	1.507	A	
Matrix: VE Vegetation Bq / kg								
1	<sup>137</sup> Cs	838.100	15.500	867.000	44.000	0.967	A	A
1	<sup>40</sup> K	533.200	40.800	639.000	34.000	0.834	W	A

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

results. These results show that as the exercise progressed the number of radionuclides reported increased. In addition, the NAA MNSR laboratory performance improved as the numbers of unacceptable results were reduced to zero by the middle of the Programme [QAP 54 (0103)].

The NAA MNSR laboratory's overall performance is shown in Figure 1 and Table-15. This

figure confirms the above-mentioned observations. However it can be seen that from the QAP exercise 51 to 57, the total number of radionuclides reported was the highest (11-16). In these exercises generally all three matrices were studied with the maximum number of radionuclides identified in all matrices. Study of Table-15 also reveals that as experience was gained in quantifying radionuclides, majority of the reported results fit the acceptable criteria and none of

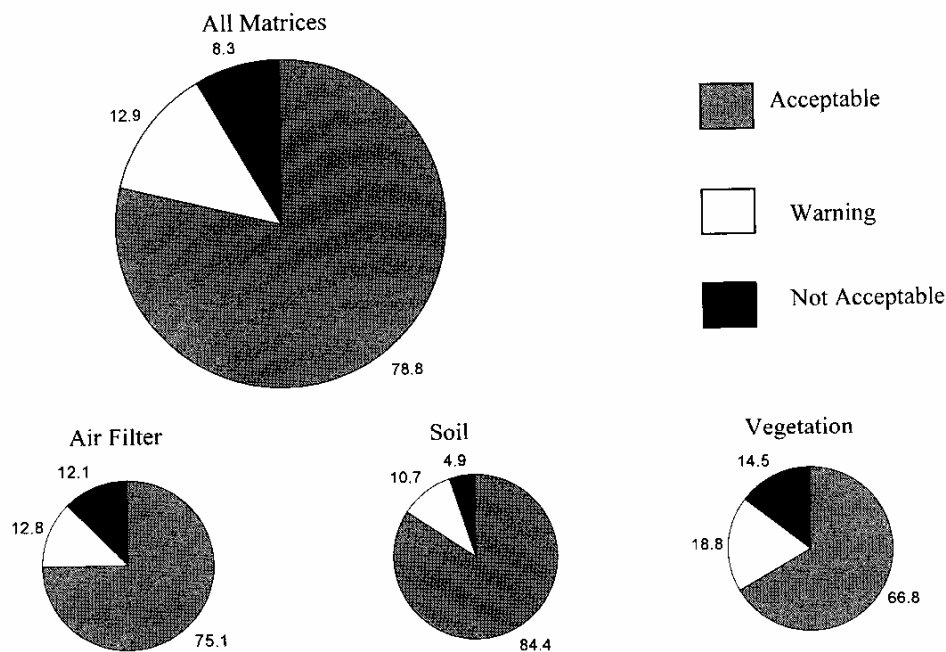


Fig. 1: Overall average performance of NAA MNSR laboratory, PINSTECH throughout the QAP programme (1998-2004).

Table-9: Results of QAP 54 (0103) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	Error/ EML	Evaluation	QAP 52 Evaluation
Matrix: AI Air Filter Bq / filter									
1	<sup>60</sup> Co	19.160	2.040	19.440	0.500	0.986		A	A
1	<sup>134</sup> Cs	2.920	0.020	2.830	0.160	1.032		A	A
1	<sup>137</sup> Cs	10.230	0.490	8.760	0.340	1.168		W	A
1	<sup>54</sup> Mn	6.340	0.370	6.520	0.280	0.972		A	A
Matrix: SO Soil Bq / kg									
1	<sup>228</sup> Ac	49.220	1.760	42.700	1.700	1.153		A	A
1	<sup>212</sup> Bi	56.020	10.920	42.000	4.100	1.334		N	
1	<sup>214</sup> Pb	36.240	2.360	32.600	1.400	1.112		A	A
1	<sup>137</sup> Cs	1756.000	29.000	1740.000	90.000	1.009		A	A
1	<sup>40</sup> K	522.900	18.400	468.000	25.000	1.117		A	A
1	<sup>214</sup> Pb	39.000	2.400	34.300	1.600	1.137		A	N
Matrix: VE Vegetation Bq / kg									
1	<sup>137</sup> Cs	831.300	4.300	842.000	42.000	0.987		A	A
1	<sup>40</sup> K	514.100	66.200	603.000	32.000	0.853		W	W

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

the reported values were rejected by EML. This was due to the use of matrix specific reference samples obtained from earlier QAP exercises. In Table-1 it can be seen that initially standards such as IAEA 326, IAEA 327, IAEA 375 and IAEA 152 were used, the first three of which are soil samples whereas the last standard is a milk powder sample. Using these non-matrix specific standards, the activities cited for <sup>40</sup>K and <sup>137</sup>Cs for the QAP 48 soil samples were all acceptable whereas the activities reported for <sup>134</sup>Cs and <sup>137</sup>Cs in the air filter matrix were not acceptable.[4]

An overview of all QAP results reported by the NAA MNSR laboratory shows that in the air filter matrix the radionuclides which are generally incorrectly determined are <sup>134</sup>Cs, <sup>137</sup>Cs and <sup>57</sup>Co. It was observed that our results for <sup>214</sup>Pb and <sup>212</sup>Bi are more likely to differ from EML values for the soil samples whereas for the vegetation samples <sup>60</sup>Co and <sup>40</sup>K activities are generally declared as not acceptable by EML.[4-14] Of these radionuclides <sup>214</sup>Pb and <sup>212</sup>Bi are short-lived intermediates whereas <sup>57</sup>Co has a half-life of 274.74 days and the remaining, <sup>60</sup>Co, <sup>134</sup>Cs, <sup>137</sup>Cs and <sup>40</sup>K are all long lived and have half-lives of

Table-10: Results of QAP 55 (0109) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	Evaluation	QAP 54 Evaluation
Matrix: AI Air Filter Bq / filter								
1	<sup>60</sup> Co	19.130	0.260	17.500	0.470	1.093	A	A
1	<sup>137</sup> Cs	13.080	0.190	12.950	0.362	1.010	A	A
1	<sup>137</sup> Cs	17.930	0.700	17.100	0.580	1.049	A	W
1	<sup>54</sup> Mn	86.700	5.000	81.150	4.760	1.068	A	A
Matrix: SO Soil Bq / kg								
1	<sup>228</sup> Ac	57.700	4.300	59.570	2.090	0.969	A	A
1	<sup>241</sup> Am	4.700	0.450	4.432	0.312	1.060	A	A
1	<sup>212</sup> Bi	59.800	12.400	62.067	5.152	0.963	A	N
1	<sup>214</sup> Bi	41.200	4.500	36.900	1.530	1.117	A	A
1	<sup>137</sup> Cs	596.000	30.000	612.330	30.620	0.973	A	A
1	<sup>40</sup> K	573.000	26.000	623.330	33.040	0.919	A	A
1	<sup>212</sup> Pb	59.300	4.700	58.330	3.130	1.017	A	A
1	<sup>214</sup> Pb	41.500	3.900	39.670	1.720	1.046	A	A
1	<sup>234</sup> Th	119.000	10.000	100.067	6.204	1.189	A	A
Matrix: VE Vegetation Bq / kg								
1	<sup>60</sup> Co	35.110	1.810	35.300	1.436	0.995	A	A
1	<sup>137</sup> Cs	1115.500	30.000	1030.000	51.800	1.083	A	A
1	<sup>40</sup> K	914.700	40.800	898.670	48.230	1.018	A	W

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

†

Table-11: Results of QAP 56 (0203) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	Evaluation	QAP 55 Evaluation
Matrix: SO Soil Bq / kg								
1	<sup>228</sup> Ac	42.260	7.020	51.167	1.941	0.826	W	A
1	<sup>241</sup> Am	12.080	1.750	10.927	0.373	1.106	A	A
1	<sup>212</sup> Bi	42.200	4.500	53.430	5.215	0.790	A	A
1	<sup>214</sup> Bi	46.600	1.900	53.933	2.249	0.864	W	A
1	<sup>137</sup> Cs	1286.000	19.000	1326.670	66.510	0.969	A	A
1	<sup>40</sup> K	614.800	23.000	621.670	33.860	0.989	A	A
1	<sup>212</sup> Pb	46.700	2.500	51.100	2.753	0.914	A	A
1	<sup>214</sup> Pb	52.000	4.600	54.367	2.249	0.956	A	A

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

Table-12: Results of QAP 57 (0209) reported by NAA MNSR laboratory, PINSTECH.

No. Test	Radionuclide	Reported Value	Reported Error	EML Value	EML Error	Reported/ EML	Evaluation	QAP 56 Evaluation
Matrix: AI Air Filter Bq / filter								
1	<sup>60</sup> Co	21.410	0.820	23.000	0.059	0.931	A	A
1	<sup>137</sup> Cs	29.790	1.320	32.500	0.777	0.917	A	A
1	<sup>54</sup> Mn	44.130	2.940	52.200	1.170	0.845	A	W
Matrix: SO Soil Bq / kg								
1	<sup>228</sup> Ac	43.125	3.791	42.300	1.560	1.020	A	W
1	<sup>241</sup> Am	7.789	1.121	6.767	0.301	1.151	A	A
1	<sup>212</sup> Bi	47.503	6.046	45.930	4.510	1.034	A	A
1	<sup>214</sup> Bi	45.718	4.422	33.630	1.560	1.359	W	W
1	<sup>137</sup> Cs	808.330	11.370	829.330	41.580	0.975	A	A
1	<sup>40</sup> K	683.900	28.800	637.670	34.260	1.072	A	A
1	<sup>212</sup> Pb	45.645	0.840	43.430	2.710	1.051	A	A
1	<sup>214</sup> Pb	43.886	1.183	35.200	1.510	1.247	A	A

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

Table-13: QAP 59 (0309) preliminary results submitted to EML by NAA MNSR laboratory, PINSTECH.

Sample ID	Radio nuclide	Repetition	Reported Value	Reported Error	EML Value	EML Error	Reported /EML	Evaluation
Air (Bq/filter)								
0309AIPK	<sup>54</sup> Mn	1	58.59	2.32	58.0	1.3	1.010	A
0309AIPK	<sup>60</sup> Co	1	55.43	1.95	55.1	1.1	1.006	A
0309AIPK	<sup>137</sup> Cs	1	54.74	1.78	54.8	1.1	0.999	A
0309AIPK	<sup>241</sup> Am	1	0.4136	0.0356	0.435	0.043	0.951	A
Soil (Bq/Kg)								
0309SOPK	<sup>40</sup> K	1	538.5	19.3	488.0	26.0	1.103	A
0309SOPK	<sup>137</sup> Cs	1	2014.25	55.78	1973.0	99.0	1.021	A
0309SOPK	<sup>241</sup> Am	1	22.49	2.69	18.4	1.8	1.222	A

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

Table-14: QAP 60 (0403) preliminary results submitted to EML by NAA MNSR laboratory, PINSTECH.

Sample ID	Radio nuclide	Repetition	Reported Value	Reported Error	EML Value	EML Error	Reported /EML	Evaluation
Air (Bq/filter)								
0403AIPK	<sup>60</sup> Co	1	35.45	0.57	35.4	0.85	1.001	A
0403AIPK	<sup>134</sup> Cs	1	18.62	0.3	18.2	0.402	1.023	A
0403AIPK	<sup>137</sup> Cs	1	26.64	0.18	26.4	0.86	1.009	A
0403AIPK	<sup>241</sup> Am	1	0.131	0.006	0.1045	0.0025	1.254	A
Soil (Bq/Kg)								
0403SOPK	<sup>40</sup> K	1	621.9	13.5	539.0	29.11	1.154	A
0403SOPK	<sup>137</sup> Cs	1	1484.8	24.2	1323.0	66.17	1.122	A
0403SOPK	<sup>212</sup> Pb	1	54.94	2.22	47.73	2.53	1.151	A
0403SOPK	<sup>228</sup> Ac	1	55.84	3.24	49.0	1.96	1.140	A
0403SOPK	<sup>234</sup> Th	1	146.13	15.95	84.0	5.96	1.740	W (obtained with 2 standards)
0403SOPK	<sup>234</sup> Th	1	73.83	13.13	84.0	5.96	0.879	A (One standard Only)
0403SOPK	<sup>241</sup> Am	1	14.87	2.08	13.0	0.43	1.144	A
0403SOPK	<sup>214</sup> Bi	1	60.9	6.6	58.4	2.2	1.043	A (Not reported to EML)
0403SOPK	<sup>214</sup> Pb	1	56.07	2.2	61.0	2.38	0.919	A (Not reported to EML)
Vegetation (Bq/Kg)								
0403VPK	<sup>40</sup> K	1	715.3	34.1	720.0	37.92	0.993	A (Not reported to EML)
0403VPK	<sup>137</sup> Cs	1	590.9	37.5	584.67	29.23	1.011	A (Not reported to EML)
0403VPK	<sup>60</sup> Co	1	14.15	3.6	14.47	0.63	0.978	A (Not reported to EML)

Note: The letters A, W and N denote acceptable, warning and not acceptable result status, respectively.

Table-15: Radionuclides determined in the environmental matrices throughout the QAP programme (1998-2004).

QAP No.	Acceptable	Warning	Un-acceptable	No. of Radionuclides	
				Reported	% Acceptable Results
Air Filter (AI)					
48				<sup>134</sup> Cs, <sup>137</sup> Cs	0
49	<sup>60</sup> Co, <sup>137</sup> Cs, <sup>54</sup> Mn, <sup>125</sup> Sb				4
50*	<sup>60</sup> Co, <sup>137</sup> Cs	<sup>57</sup> Co			3
51	<sup>57</sup> Co, <sup>137</sup> Cs, <sup>54</sup> Mn	<sup>60</sup> Co			4
52	<sup>57</sup> Co, <sup>137</sup> Cs, <sup>54</sup> Mn	<sup>60</sup> Co			4
53	<sup>57</sup> Co, <sup>60</sup> Co, <sup>137</sup> Cs, <sup>54</sup> Mn				4
54	<sup>134</sup> Cs, <sup>60</sup> Co, <sup>54</sup> Mn	<sup>137</sup> Cs			4
55	<sup>134</sup> Cs, <sup>137</sup> Cs, <sup>54</sup> Mn, <sup>60</sup> Co				4
57	<sup>60</sup> Co, <sup>137</sup> Cs	<sup>54</sup> Mn			3
59	<sup>60</sup> Co, <sup>137</sup> Cs, <sup>54</sup> Mn, <sup>241</sup> Am				4
60	<sup>60</sup> Co, <sup>137</sup> Cs, <sup>134</sup> Cs, <sup>241</sup> Am				4
Overall					2 - 4
Soil (SO)					
48	<sup>40</sup> K, <sup>137</sup> Cs				2
49	<sup>137</sup> Cs				1
50*	<sup>40</sup> K, <sup>137</sup> Cs				2
51	<sup>228</sup> Ac, <sup>214</sup> Bi, <sup>137</sup> Cs	<sup>214</sup> Bi, <sup>40</sup> K, <sup>212</sup> Pb, <sup>214</sup> Pb			7
52	<sup>137</sup> Cs, <sup>40</sup> K, <sup>212</sup> Pb		<sup>214</sup> Pb		4
53	<sup>228</sup> Ac, <sup>214</sup> Bi, <sup>137</sup> Cs, <sup>234</sup> Th	<sup>212</sup> Pb	<sup>214</sup> Pb		6
54	<sup>228</sup> Ac, <sup>214</sup> Bi, <sup>137</sup> Cs, <sup>40</sup> K, <sup>214</sup> Pb		<sup>212</sup> Bi		6
55	<sup>228</sup> Ac, <sup>212</sup> Bi, <sup>214</sup> Bi, <sup>137</sup> Cs, <sup>40</sup> K, <sup>212</sup> Pb, <sup>214</sup> Pb, <sup>241</sup> Am, <sup>234</sup> Th				9
56	<sup>241</sup> Am, <sup>212</sup> Bi, <sup>212</sup> Pb, <sup>214</sup> Pb, <sup>137</sup> Cs, <sup>40</sup> K	<sup>228</sup> Ac, <sup>214</sup> Bi			8
57	<sup>228</sup> Ac, <sup>212</sup> Bi, <sup>137</sup> Cs, <sup>40</sup> K, <sup>212</sup> Pb, <sup>214</sup> Pb, <sup>241</sup> Am	<sup>214</sup> Bi,			8
59	<sup>137</sup> Cs, <sup>40</sup> K, <sup>241</sup> Am				3
60	<sup>228</sup> Ac, <sup>137</sup> Cs, <sup>40</sup> K, <sup>212</sup> Pb, <sup>241</sup> Am	<sup>234</sup> Th			6
Overall					1 - 9
Vegetation (VE)					
48	<sup>40</sup> K, <sup>137</sup> Cs				2
49	<sup>40</sup> K, <sup>137</sup> Cs				2
50*	<sup>40</sup> K, <sup>137</sup> Cs	<sup>60</sup> Co			3
51		<sup>137</sup> Cs	<sup>40</sup> K		2
52	<sup>40</sup> K, <sup>137</sup> Cs	<sup>60</sup> Co			3
53	<sup>137</sup> Cs	<sup>40</sup> K			2
54	<sup>137</sup> Cs	<sup>40</sup> K			2
55	<sup>40</sup> K, <sup>137</sup> Cs, <sup>60</sup> Co				3
Overall					2 - 3

\* Re-calculated results of QAP 5

the order of years.  $^{214}\text{Pb}$  and  $^{212}\text{Bi}$  are daughter products of the gas  $^{222}\text{Rn}$  (3.6 day half life).  $^{222}\text{Rn}$  may easily escape as noble gas from the sample. In order to establish secular equilibrium, the samples have to be kept in gas-tight counting vessels for 2-3 weeks. The discrepancy in the results for these radionuclides shows that a lack of equilibrium may be the source of this deviation, which may be due to the fact that the sample and/or standard was counted before secular equilibrium was established or the container was not sealed properly. Therefore, hermetically sealed samples should ideally be used in such studies.  $^{40}\text{K}$  is naturally occurring so is observed in the background spectrum as well. Therefore, the discrepancy in the reported results may also be attributed to the low activities of these radionuclides in the samples being analyzed and or greater uncertainty in the certified value of the standards used in their quantification.

After QAP 54 (0103) the amount of soil and vegetation sample counted was increased as the sample depth was increased from ~1 to ~2.5 cm. This more than doubled the volume of the sample and hence had a corresponding effect on the count rate. Nevertheless it may also mean that substantial attenuation may occur especially for low energy  $\gamma$ -rays, such as the 59.5 keV  $\gamma$ -ray in  $^{241}\text{Am}$ . However, due to the fact that soil, vegetation and air filter samples were quantified using soil, vegetation and air filter RMs, and identical counting procedures and parameter were used, the attenuation differences will cancel out to a large extent. In fact it can be seen from Tables 10 to 14 that  $^{241}\text{Am}$  has been reported in QAP 55 to 60 in soil samples. Further scrutiny of Tables 13 and 14 shows that  $^{241}\text{Am}$  has also been reported in the air filter samples for QAP 59 and 60. Additionally, the results show that no "not acceptable" activities were reported after this change occurred. Thus the higher count rate and the resultant improvement in counting statistics provided better agreement between our and EML results and also enabled the identification and quantification of a greater number of radionuclides in all samples, especially soil samples.

The pie charts in Figure 1 and the data in Table-15 show the NAA MNSR laboratory's overall average performance from 1998 to 2004. As stated earlier the best results were obtained for the soil matrix and the worst for the vegetation matrix, whereas the overall performance somewhat

resembles the results obtained for the air filter samples. Apart from the variations in the count rate associated with individual matrices, during the QAP Programme, not all vegetation samples were analyzed whereas all soil samples were investigated; therefore more data is available for the soil matrix thereby giving better overall performance and implying greater confidence in our results. Generally 2-4 (mode 4), 2-9 (mode 6) and 2-3 (mode 2) radionuclides were reported in the air filter, soil and vegetation matrices, respectively. Thus 6-12 (mode 12) radionuclides were reported per QAP exercise with 79%, 13% and 8% of the results being acceptable, acceptable with warning and not acceptable, respectively.

The NAA MNSR laboratory at PINSTECH participated in the QAP Programme conducted by the DOE from 1998 to May 2004. Experience has been gained by participating in this programme in quantifying radionuclides in contaminated environmental samples such as soil, exposed air filters and vegetation. It has been found that matrix specific RMs provide better results as do larger samples. Furthermore it was also found that establishment of secular equilibrium and use of hermetically sealed samples is essential towards obtaining reliable data. It was discovered that care should be taken to quantify ( $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{57}\text{Co}$ ), ( $^{214}\text{Pb}$  and  $^{212}\text{Bi}$ ) and ( $^{60}\text{Co}$  and  $^{40}\text{K}$ ) in air filter, soil and vegetation matrices respectively. The best results were obtained for the soil matrix due to the higher concentration of radionuclides in this matrix and the worst results were obtained for the vegetation matrix, the results of the air filter matrix being in between. The numbers of radionuclides reported were around 4, 6 and 2 in the air filter, soil and vegetation matrices, respectively.

## Experimental

### Samples

The contaminated environmental samples analyzed consisted of soil, vegetation and exposed air filter samples. The soil and vegetation samples were spiked with radioactivity at a concentration less than 0.002 microcuries per gram. Therefore these materials are not considered radioactive according to Department of Transportation (DOT) USA, regulations (48 CFR 173.403(y)). Glass fiber air-filter samples were received sealed in envelopes. The

activity level of the QAP air filter samples was generally above 0.002 microcuries per gram limit of DOT. Thus the air filter samples are subject to DOT requirements for limited quantities of radioactive material (49 CFR 173.421). Due to time constraints, on occasions, not all samples were studied for each exercise. Details of the samples analyzed throughout the QAP Programme are given in Table-1.

#### *Standards and Reference Materials (RMs)*

At the beginning of the QAP programme suitable reference materials (RMs) were not available therefore radionuclide RMs such as IAEA 326 and 327 (radionuclides in soil samples) were used. Later on air filter, soil and vegetation samples from previous QAP exercises, consensus values for which had been determined and published by EML, were used as RMs. For example, AI0103, AI0109, AI0203, AI0209, S0109, S0203, S0209 etc; here the first two digits represent the matrix, the second two the year and the final two digits the reference month for the calculated results. The consensus values were obtained according to ISO Guide 30: 1992 (E/F)[16] and ISO Guide 43-1:1997 (E)[17] after applying robust statistical methods or by removing outliers prior to calculations.

#### *Gamma Spectroscopic Measurement*

Prior to the study of any of the QAP samples the area surrounding the detector and its shielding were wiped clean and the coverings of all work surfaces were replaced. The QAP samples were counted using a HPGe detector (Canberra Model AL-30) connected to a PC-based Intertechnique Multichannel Analyzer (MCA Model Desk.PRO.286e). The detection system has a resolution of 1.9 keV for the 1332.5 keV peak of  $^{60}\text{Co}$  and a peak-to-Compton ratio of 40:1. The counting time was 16 hours and up to 3 replicate spectra were obtained for each sample. The software "INTERGAMMA, version 5.03" was used for all measurements. The counting system is calibrated daily using calibrated  $\gamma$ -ray sources. The sealed QAP filters were counted directly on detector top, whereas the vegetation and soil samples were counted on the detector top, in plastic screw top containers of diameter ~5cm (approximately the diameter of the detector top). The soil or vegetation sample was loosely packed into the container to a certain depth (i.e. 1cm or 1inch). Depending upon the size of the

samples the soil samples weighed between 20-90g (average weight ~70g) whereas the vegetation samples weighed between 8-50g (average weight ~25g). The screw top was taped to seal the container. The background spectrum was obtained for 16 hours as was the spectrum of the plastic sampling container. The RM samples were also counted under identical conditions as the samples.

The presence of any of the radionuclides listed in Table-2 was investigated in these samples. The most abundant  $\gamma$  peak was used for quantification purposes whereas the less abundant peaks were used to verify the presence of a particular radionuclide. The spectrum data analysis was performed using computer programs developed in our laboratory. All necessary corrections (background subtraction etc) were applied and the final results obtained on dry weight basis for the soil and vegetation samples and per filter for the air filter samples. The dry weights were obtained by drying the soil and vegetation samples at 105 °C for 24 hours. Error propagation rules were applied at each stage of the calculations and accounted for the uncertainties in peak area, the background spectrum, activities of the radionuclides in the RMs etc.

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