DISTRIBUTION AND TRENDS OF NATURAL RADIONUCLIDE CONCENTRATIONS AT CTBT RADIONUCLIDE MONITORING STATIONS

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ABSTRACT

The Atmospheric Radionuclide Monitoring Program at the prototype International Data Center (pIDC) began developing technical support for the Comprehensive Nuclear-Test-Ban-Treaty (CTBT) in October 1995. Since then, over 20 particulate monitoring stations have transmitted data to the pIDC. The data include gamma-ray spectra of aerosol samples collected with high-volume air samplers. Sampling methodology (e.g., sampling time, volume of air sampled, decay time, gamma-ray acquisition time) currently varies from station to station, but will soon be standardized.

Successful environmental monitoring in support of the CTBT requires a detailed knowledge of the natural radionuclide concentrations that dominate the background. Natural radionuclides can both help and hinder monitoring efforts. On the positive side, they may be utilized for quality assurance/quality control (QA/QC) measures. However, significantly high natural radionuclide background levels will adversely affect a station's detection capabilities.

Natural radionuclides may originate from interactions of cosmic-rays with matter or from the singular event at the beginning of the universe (primordial). This study characterizes the natural radionuclide concentrations and seasonal trends of four naturally measured radionuclides: ^{7}Be, ^{210}Pb, ^{212}Pb, and ^{40}K. This information is necessary to model natural radionuclide concentration levels for station characterization.

Key Words: radionuclide monitoring, aerosol, pIDC, natural radionuclides, ^{7}Be, ^{210}Pb, ^{212}Pb, and ^{40}K

OBJECTIVE

While the focus of the radionuclide monitoring effort at the pIDC is to monitor the environment for radioactive aerosols and gas representative of a nuclear weapon test, the overwhelming majority of radionuclide measurements are those of natural origins. The presence of these natural radionuclides can both help and hinder the pIDC's monitoring efforts. On the positive side, the natural radionuclides may be utilized in system performance monitoring, station QA/AC measures, and provide information about the local environment of the station. On the negative side, the signal from the natural radionuclides increase the Compton continuum background of the spectrum and interfere with the detection of anthropogenic radionuclides expected to be produced in a nuclear weapon test.

To optimize our benefits from the presence of natural radionuclides within the gamma-ray spectrum, an increased understanding of their occurrence rates, distributions, and trends needs to be achieved. This information may then be utilized to enhance detector calibrations, monitor the performance of the detection equipment, and provide QA/QC guidance on the sampling metrics. The information may be included in models that calculate prediction intervals for the expected natural radionuclide concentrations. If a natural radionuclide is measured outside the predicted range, then a flag is raised indicating that the sample should be reviewed with increased scrutiny.

The modeling of expected radionuclide concentrations at the pIDC is referred to as sample characterization. Samples with natural radionuclides present at normal levels and have no anthropogenic radionuclides indicative of a nuclear test are characterized as Level 1. Natural radionuclides present at abnormal levels and have no anthropogenic radionuclides indicative of a nuclear test are characterized as Level 2. Detailed descriptions of the sample characterization methods may be found in Evans (1996) and Bohner (1998). Within the current software, radionuclide concentrations may be modeled as having normal, log-normal, or square-root distributions. This research will demonstrate which distributions best represent the radionuclides at the different stations.

The natural radionuclides considered in this research include ^{7}Be, ^{210}Pb, ^{212}Pb, and ^{40}K. These are the most common natural radionuclides that may be quantitatively determined. Many radionuclides are detected within the radon and thoron decay series that can only be qualitatively identified. The pIDC is currently evaluating methods to accurately determine activity concentrations of radionuclides within a decay series, but no such method is in place at the current time. Numerous other natural radionuclides are observed less predictably; many generated through interactions with cosmic radiation.

^{7}Be is a cosmogenically produced radionuclide that decays with a 53.28 day half-life. It is produced as a result of cosmic-ray interactions with nitrogen and oxygen atoms in the stratosphere and upper troposphere. The magnitude of the ^{7}Be production is affected by the energy level of the bombarding cosmic radiation and the atmospheric density of nitrogen and oxygen targets. Maximum production of ^{7}Be occurs in the stratosphere [Koch *et al*., 1996]. This production is also a function of longitude and the 11-year solar cycle. To be detected on the ground by the radionuclide monitoring system contributing data to the pIDC, vertical atmospheric mixing must occur to transport the ^{7}Be to lower altitudes. While the production of ^{7}Be does not vary with seasons, the meteorological influences on vertical atmospheric transport does. Thus, seasonal trends are often observed with ground-based ^{7}Be atmospheric activity concentration measurements. ^{7}Be may be quantified through its 477.6 keV gamma-ray with 10.3% abundance.

^{210}Pb is a terrestrial radionuclide in the radon decay series. Many other radon daughters are also found within the spectra evaluated at the pIDC including ^{214}Pb and ^{214}Bi. Radon concentrations vary with respect to geographical regions and are a function of local crustal uranium concentrations and soil permeability. Following production, radon daughters attach indiscriminately to available aerosol and therefore spread evenly with respect to area over the ambient aerosol size distribution [Koch *et al*., 1996]. The 22.6 year half-life of ^{210}Pb allows for substantial geographical distribution. Therefore, ^{210}Pb atmospheric activity concentrations are less regionally dependent than radon concentrations. This nuclide may be quantified

through its 46.50 keV gamma-ray with 4.1 % abundance. However, within International Monitoring System (IMS) specifications, there is no requirement to analyze spectra at energies as low as this. ^{212}Pb is another terrestrial radionuclide but it is in the thoron decay series. Other radionuclides in the thoron decay series measured at the pIDC are ^{212}Bi and ^{208}Tl. Unlike ^{210}Pb, the ^{212}Pb atmospheric activity concentrations are more closely tied to the concentrations of its parent, thoron. This is largely a function of this radionuclide's half-life of 10.6 hours. ^{212}Pb may be quantified through its primary decay line at 238.63 keV with 43.60 % abundance.

^{40}K is a radionuclide of primordial origin. It has a long half-life of 1.3 x 10^9 years and is widely distributed throughout the environment. Its natural abundance is 0.0119 %. This radionuclide is present not only in atmospheric aerosols, but also in the aerosol filters used to collect samples as well as in the surrounding building materials. ^{40}K may be quantified through its 1460.75 keV line with 10.67 % abundance.

RESEARCH ACCOMPLISHED

Table 1 lists the stations that have contributed radionuclide data to the pIDC, their locations, date at which the pIDC began processing the station data in operations, and the station's sampling methodology. Most stations have continued to send data to the pIDC on a regular basis through the present time with some data gaps due to routine maintenance and equipment failures. Most of the stations contributing to the pIDC operate on a long sampling methodology. A long sampling methodology is one in which the time from the beginning of aerosol sample collection to the end of gamma-ray spectrum acquisition is longer than a week. For compliance with CTBT International Monitoring System standards, stations must adhere to a sampling methodology in which the time between the beginning of aerosol sample collection and end of the gamma-ray spectrum acquisition is less than or equal to 72 hours. Stations with long sampling methodologies are not able to detect certain short-lived radionuclides including ^{212}Pb.

Table 1
Radionuclide Stations Contributing Data to the pIDC

Station Code	Location	Start of Data Processing in pIDC Operations	Sampling Methodology
AR001	Buenos Aires, Argentina	January, 1996	Short
AU001	Melbourne, Australia	September, 1995	Short
AU002	Perth, Australia	January, 1997	Long
AU003	Darwin, Australia	February, 1997	Long
AU004	Townsville, Australia	February, 1997	Long
CA001	Ottawa, Canada	December, 1995	Long
CA002	Vancouver, Canada	April, 1996	Short
CA003	Resolute, Canada	May, 1996	Long
CA004	Yellowknife, Canada	May, 1996	Long
CA005	St. John's, Canada	June, 1996	Long
DE002	Schauinsland, Germany	January, 1996	Long
FI001	Helsinki, Finland	January, 1996	Long/Short*
KW001	Kuwait City, Kuwait	September, 1995	Short
NZ001	Kaitaia, New Zealand	May, 1995	Long
NZ002	Rarotonga, New Zealand	May, 1996	Long
NZ003	Hokitika, New Zealand	May, 1996	Long
RU001	Ussuriysk, Russia	November, 1995	Short
SE001	Stockholm, Sweden	September, 1995	Short
UK001	Chilton, England	June, 1996	Long
US001	Charlottesville, USA	December, 1995	Short

* Station has maintained both long and short sampling methodologies.

Distribution of Atmospheric Activity Concentrations

As stated, the pIDC utilizes a modeling code to calculate confidence intervals of atmospheric activity concentrations. The confidence intervals are then used to determine if the measured atmospheric activity concentration is within normal bounds. A user may choose one of three distribution types in the initial model set-up procedure. Initial research was conducted in this area [Evans, 1996; Shen, 1998]. However, this work examines all the stations, includes a larger data set, and utilizes a more comprehensive approach in determining the best distribution fits and seasonal trends.

Three methods are utilized to investigate the best distribution fits to the data: the first method includes plotting the data on a histogram, the second method includes plotting the data on a cumulative probability plot, and the third method is the Kolmogorov-Smirnov test. The first two methods are graphical methods that require a visual comparison of the data fit to a given distribution. The third method, the Kolmogorov-Smirnov test, compares the observed cumulative distribution function for a variable with a specified theoretical distribution. The Kolmogorov-Smirnov variable, Z, is computed from the largest difference (in absolute value) between the observed and theoretical distribution function. The goodness-of-fit test determines how well the observations conform to the theoretical distribution. Basically, the lower the Z-value, the better the fit. For this test, the normal distribution is utilized as the theoretical distribution to which the observed data is compared. Natural log and square-root transformations are applied to the data to test the goodness-of-fit with the log-normal and square-root distributions, respectively. However, one must also take the sample set size into account because the Kolmogorov-Smirnov Z-value is a sum of differences and does not normalize to sample set size. The results of the Kolmogorov-Smirnov test provide similar conclusions to the plotting methods, however, the Z value allows for a more quantitative comparison.

Table 2 contains the Kolmogorov-Smirnov test results for ^{7}Be at all the radionuclide monitoring stations. Out of the twenty stations, four were best fit with a normal distribution, seven were best fit by a log-normal distribution, and nine were best fit with a square-root distribution. For many of the stations, large differences were seen between the quality of fit between the distribution types. The data sets were all large enough to provide adequate representation of the atmospheric activity concentration distribution. The results for the KW001 (Kuwait City, Kuwait) station indicate that a square-root distribution best fits the ^{7}Be data. This is commensurate with the results of an initial study reported in Evans (1996). The ^{7}Be data for the DE002 (Schauinsland, Germany) station is best fit by the square-root distribution. This is in contrast to data reported by Winkler *et al*. (1998) for another rural sampling location in southern Germany (about 10 km north of the city of Munich) that is best fit by a log-normal distribution. The Winkler *et al*. data set consists of 46 10-day samples collected from December, 1994 through March, 1996.

Table 2 contains the Kolmogorov-Smirnov test results for ^{210}Pb at all the radionuclide monitoring stations. Out of the twenty stations, four are best fit with a normal distribution, nine are best fit by a log-normal distribution, six are best fit with a square-root distribution, and one station did not have ^{210}Pb measurements available. The 46.50 keV gamma-ray was not always detectable on all gamma-ray spectroscopy systems. The electronic noise was often high in this energy region and some systems had their lower-level energy discriminator set above this energy. As a result, the ^{210}Pb data set is not as complete as the ^{7}Be data set. Also, one should view the results from the NZ001 (Kaitaia, New Zealand), NZ002 (Rarotonga, New Zealand), and NZ003 (Hokitika, New Zealand) stations with caution because the data sets are small. No data is available for the AU001 (Melbourne, Australia) station. ^{210}Pb atmospheric activity concentrations are not normally modeled at the pIDC for sample characterization, so no previous studies have been performed at the pIDC on the ^{210}Pb atmospheric activity concentration distributions. A comparison can be made between the DE002 (Schauinsland, Germany) data and the data reported by Winkler *et al*. (1998). Both data sets show that a log-normal distribution best fits the ^{210}Pb atmospheric activity concentrations within the southern Germany region.

Table 2
^{7}Be Atmospheric Activity Concentration Distributions

Station	Sample Set Size	Normal Distribution K-S Z-Value	Log-Normal Distribution K-S Z-Value	Square-Root Distribution K-S Z-Value	Best Distribution Fit
AR001	363	0.830	1.876	0.881	Normal
AU001	799	1.882	1.732	0.542	Square-Root
AU002	117	1.132	0.603	0.809	Log-Normal
AU003	120	1.160	0.774	0.741	Square-Root
AU004	117	0.690	0.571	0.414	Square-Root
CA001	170	1.242	1.194	1.073	Square-Root
CA002	1104	4.063	1.549	1.945	Log-Normal
CA003	121	1.164	1.145	0.733	Square-Root
CA004	127	1.043	0.606	0.806	Log-Normal
CA005	75	0.641	0.926	0.713	Normal
DE002	187	0.912	0.790	0.336	Square-Root
FI001	409	2.096	0.568	1.286	Log-Normal
KW001	999	1.162	1.929	0.698	Square-Root
NZ001	143	0.964	0.702	0.780	Log-Normal
NZ002	135	0.796	0.974	0.647	Square-Root
NZ003	151	1.105	0.400	0.678	Square-Root
RU001	498	1.030	3.110	1.520	Normal
SE001	718	3.027	1.315	1.515	Log-Normal
UK001	162	0.909	0.486	0.523	Log-Normal
US001	1132	1.234	4.124	1.762	Normal

Table 4 contains the Kolmogorov-Smirnov test results for ^{212}Pb at all the radionuclide monitoring stations. Due to ^{212}Pb's 10.6 hour half-life, only stations that employ short counting methodologies are considered for studying the ^{212}Pb atmospheric concentration distributions. Out of these eight stations, none are best fit with a normal distribution, six are best fit by a log-normal distribution, and two are best fit with a square-root distribution. The ^{212}Pb data set utilized for the FI001 (Helsinki, Finland) station has been abbreviated to include only those samples that adhered to a short sampling methodology. The ^{212}Pb atmospheric activity distribution at KW001 is best fit by a square-root distribution similar to what was shown in an earlier evaluation [Evans, 1996].

Studies on ^{40}K atmospheric activity concentrations indicate that the majority of the counts under the 1460.75 keV peak result from ^{40}K within the filter blank and detector background. While histograms, cumulative probability plots, and the Kolmogorov-Smirnov test are all applied to the ^{40}K data, these results have little meaning because no blank subtraction has been performed on the data. Figure 1 shows the ^{40}K count-rates for the US001USA1 detector at the US001 (Charlottesville, USA) station. The shifts in count-rates are due to changes in the aerosol filter batches. Thus, one can see that for a given batch of aerosol filters, the ^{40}K count-rate remains relatively constant. This phenomenon is often utilized at the pIDC as a QA/QC check on gamma-ray spectrum acquisition time.

Table 3
^{210}Pb Atmospheric Activity Concentration Distributions

Station	Sample Set Size	Normal Distribution K-S Z-Value	Log-Normal Distribution K-S Z-Value	Square-Root Distribution K-S Z-Value	Best Distribution Fit
AR001	72	1.396	2.260	1.517	Normal
AU001	-	-	-	-	
AU002	107	2.040	0.817	1.444	Log-Normal
AU003	112	0.834	0.744	0.546	Square-Root
AU004	109	1.708	0.565	1.203	Log-Normal
CA001	149	1.263	2.979	2.105	Normal
CA002	310	9.145	6.279	9.13	Log-Normal
CA003	106	1.124	0.999	0.728	Square-Root
CA004	109	1.345	0.849	0.878	Log-Normal
CA005	72	1.277	2.200	1.762	Normal
DE002	108	1.107	0.663	0.770	Log-Normal
FI001	288	2.085	1.764	1.698	Square-Root
KW001	199	4.862	1.098	1.785	Log-Normal
NZ001	17	(0.409)	(0.898)	(0.629)	(Normal)
NZ002	5	(0.748)	(0.611)	(0.682)	(Log-Normal)
NZ003	15	(0.778)	(0.986)	(0.690)	(Square-Root)
RU001	227	7.117	2.308	5.412	Log-Normal
SE001	661	3.712	1.346	2.627	Log-Normal
UK001	83	1.181	0.774	0.445	Square-Root
US001	424	7.950	3.010	2.625	Square-Root

Values in parentheses are for sample sets smaller than 20. These values should be viewed with caution since the sample set is small.

Seasonal Trends

Seasonal trends are observed at most of the stations for ^{7}Be, ^{210}Pb, and ^{212}Pb. Since the ^{40}K signals in the gamma-ray spectra are dominated by the blank, no seasonal variations are observed (nor expected) for ^{40}K. The best tool for observing the seasonal trends is the box plot. A box plot of ^{7}Be at the KW001 station (Kuwait City, Kuwait) is shown in Figure 2. Multiple years of the aerosol data are sorted by month and each month's data are plotted in a "box and whisker." A line in the box represents the mode of the monthly air concentrations. The range of the box depicts the bounds of the 25th and 75th percentiles of the data. The whiskers extending from the box represent the bounds of the 10th and 90th percentile of data. Data points that extend beyond the 10th and 90th percentiles are plotted individually. The seasonal trend in this data is obvious with the peak ^{7}Be concentrations occurring in the summer months.

Table 5 shows the months with the highest median atmospheric activity concentration. If no seasonal trend is observed, the space in the table is left empty. This is often the result of insufficient data. ^{212}Pb is not quantified at stations with a long-decay sampling methodology and no seasonal trend is observed for this radionuclide at these stations. While most stations exhibited 12-month cycles in their natural radionuclide concentrations, some stations display trends with two distinct peaks in atmospheric activity concentrations. Figure 3 contains a box plot of the ^{7}Be atmospheric activity concentrations at the RU001 (Ussuriysk, Russia) station. Clear seasonal highs are seen for ^{7}Be during the months of April and September. Since variations in ^{7}Be atmospheric activity concentrations at a given station are largely a function of meteorological influences on vertical atmospheric mixing, it is no surprise that highs are seen both in the spring and fall.

Table 4
^{212}Pb Atmospheric Activity Concentration Distributions

Station	Sample Set Size	Normal Distribution K-S Z-Value	Log-Normal Distribution K-S Z-Value	Square-Root Distribution K-S Z-Value	Best Distribution Fit
AR001	361	1.858	0.869	0.962	Log-Normal
AU001	799	3.540	1.725	1.760	Log-Normal
AU002	-	-	-	-	
AU003	-	-	-	-	
AU004	-	-	-	-	
CA001	-	-	-	-	
CA002	1104	4.199	1.428	2.566	Log-Normal
CA003	-	-	-	-	
CA004	-	-	-	-	
CA005	-	-	-	-	
DE002	-	-	-	-	
FI001	248	4.750	1.214	2.740	Log-Normal
KW001	971	2.934	2.451	1.214	Square-Root
NZ001	-	-	-	-	
NZ002	-	-	-	-	
NZ003	-	-	-	-	
RU001	497	4.322	0.788	2.761	Log-Normal
SE001	713	12.045	1.046	6.146	Log-Normal
UK001	-	-	-	-	
US001	1131	2.654	1.854	0.877	Square-Root

* FI001 sample set size reduced to samples from 01-Sept-1998 to 10-June-1999 that adhere to short-decay sampling methodology.

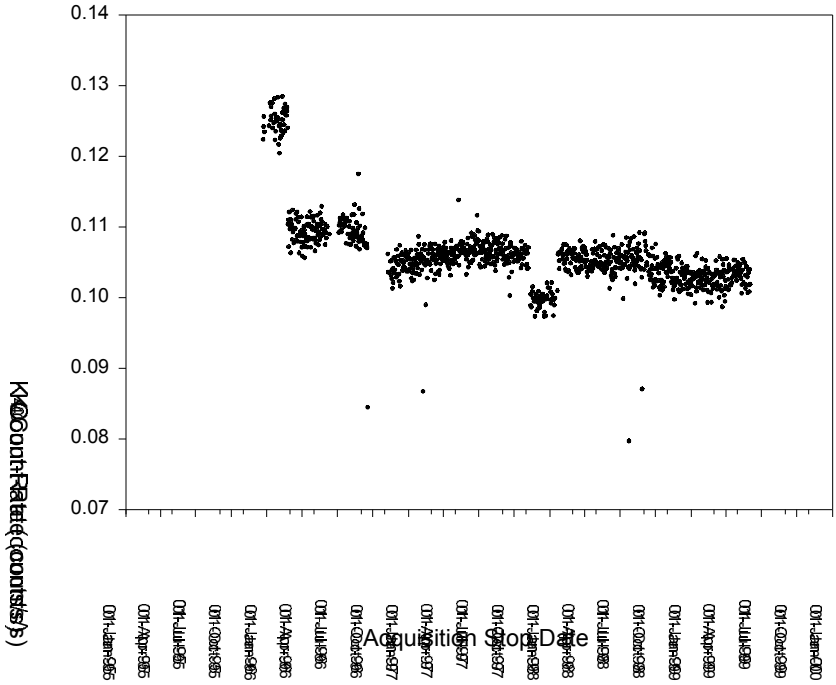

Figure 1. ^{40}K count-rates in sample spectra for US001USA1 detector at US001 (Charlottesville, USA) station. Data from December, 1995 through June, 1999.

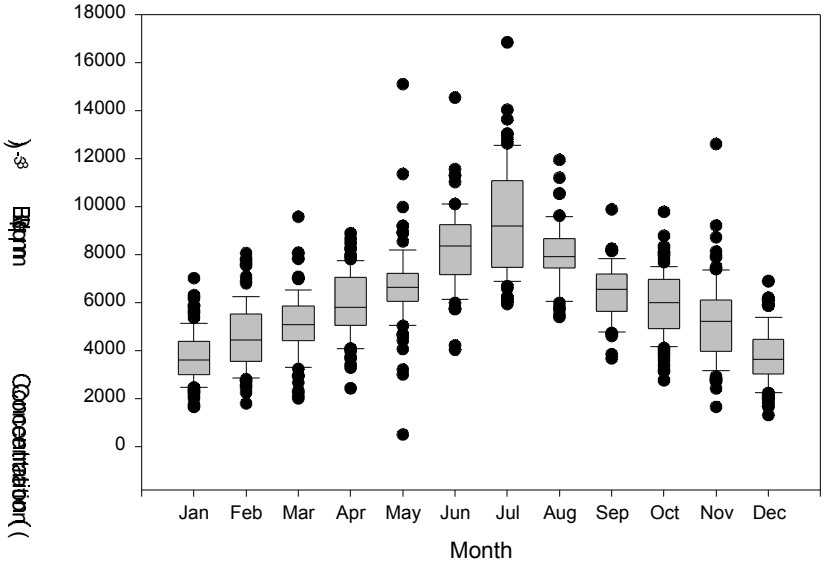

Figure 2. Box plot of ^{7}Be at KW001 (Kuwait City, Kuwait) station. Data from September, 1995 to June, 1999.

Table 5
Seasonal Peaks of Natural Radionuclide Activity Concentrations

Station	^{7}Be	^{210}Pb	^{212}Pb	^{40}K
AR001	December	(December)	(September)	-
AU001	February		March	-
AU002	March	May		-
AU003	October	June		-
AU004	October	May/October		-
CA001	July	February		-
CA002	April/September	August	August	-
CA003	March	February		-
CA004	March/July	February		-
CA005	March/August	(January)		-
DE002	June	September		-
FI001	June	March		-
KW001	July	June	November	-
NZ001	January			-
NZ002	November			-
NZ003	February			-
RU001	April/September	January	April/November	-
SE001	June	December	August	-
UK001	April/October	March/October		-
US001	April/July	September	September	-

Months in parentheses were for data with only a marginal seasonal trend.

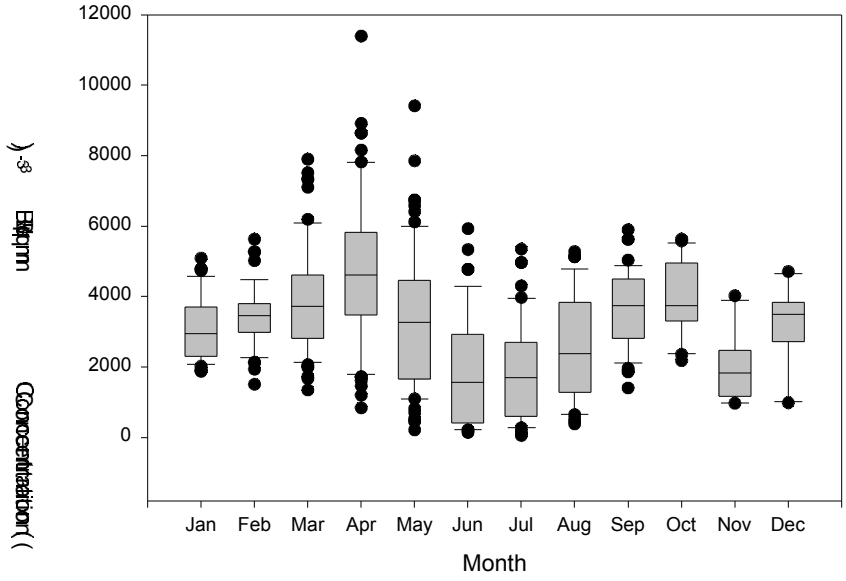

Figure 3. Box plot of ^{7}Be at RU001 (Ussuriysk, Russia) station. Data from November, 1995 to July, 1998.

CONCLUSIONS AND RECOMMENDATIONS

This research shows the distributions and trends of natural radionuclides monitored at twenty stations. These distributions and trends will be utilized to model the expected ranges of the natural radionuclide concentrations at these stations. If an abnormal measurement is detected, a flag will be created to alert radionuclide analysts that the natural radionuclide concentration is out of the expected range. The radionuclide analyst may then pay special attention to sampling metrics like sampling time, decay time, gamma-ray spectrum acquisition time, and sampler flow-rate. Monitoring the levels of these radionuclides in all analyzed samples is an excellent form of performance monitoring.

At many stations the ^{7}Be concentrations fluctuate largely with season. Knowledge of these fluctuations is beneficial to monitoring the atmosphere for fission products from nuclear weapon tests because detection is highly dependent on atmospheric transport. A large degree of vertical mixing in the atmosphere enhances the station's ability to capture fission product aerosols from a nuclear test.

The fluctuations of ^{210}Pb provide some indication of the crustal resuspension at the station. At times of the year when ^{210}Pb concentrations are high, the resuspension of old nuclear weapons fallout and resuspension of fallout from previous nuclear accident releases should also be high. As more data is collected in Europe, correlations should be made between the resuspension of Chernobyl fallout and ^{210}Pb concentrations.

Similar studies should continue to be performed as more stations are brought on-line. This information will help in our understanding of the environment that the station is monitoring. Such information may then be advantageous to the monitoring system as a basis for performance monitoring and other QA/QC measures.

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