CESIUM-137 DISTRIBUTIONS AND TRENDS AT CTBT RADIONUCLIDE MONITORING STATIONS AND THEIR IMPLICATIONS ON CTBT MONITORING

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ABSTRACT

As part of the development support for the Comprehensive Nuclear-Test-Ban Treaty (CTBT), the prototype International Data Center (pIDC) has been processing radionuclide data since 1995. Radionuclide data received from field stations includes gamma-ray spectra, meteorological data, and state of health (SOH) information. To date over 20 radionuclide monitoring stations have transmitted data to the pIDC. The radionuclide monitoring system collects both aerosol and gas samples. Gamma-ray spectral analyses are performed on the samples to determine if they contain anthropogenic radionuclides indicative of nuclear debris.

A key radionuclide monitored by this system is ^{137}Cs [WP.224]. This radionuclide has a large fission yield and is produced in large quantities during a nuclear explosion [Vladimirski et al., 1998]. Due to the half-life of ^{137}Cs (30.17 years), amounts of this nuclide from past nuclear tests and reactor releases are still present in the soil and atmosphere. Cesium-137 from these sources is routinely detected in the prototype CTBT radionuclide monitoring system.

The pIDC has analyzed and reviewed gamma-ray spectra from over 7500 samples collected by the radionuclide monitoring network. Eight per cent of these samples contain the fission product ^{137}Cs. Samples from northern Europe have the highest percentage of ^{137}Cs detections due to the re-suspension of fall-out from the Chernobyl accident [Hötzl et al., 1992]. At most stations, ^{137}Cs concentrations follow a seasonal trend with high levels being detected in the spring and fall. This trend is similar to what is expected for aerosols from crustal origin. It is important to understand the behavior of this fission product in the monitoring environment for accurate characterization in the context of nuclear test ban verification.

Key Words: radionuclide monitoring, ^{137}Cs, aerosol, and pIDC

OBJECTIVE

The objective of this research is to examine the distribution, trends, and sources of airborne ^{137}Cs measured at radionuclide stations contributing data to the PDIC. Multiple sources contribute to atmospheric activity concentrations of ^{137}Cs measured at different sampling stations. The primary source of ^{137}Cs is the resuspension of fallout from past nuclear weapons tests and the Chernobyl event in 1986. Other airborne ^{137}Cs sources include reactor releases, industrial accidents, and laboratory contamination.

The spectra received at the pIDC are characterized with respect to their content [Bohner, 1998; Evans, 1996]. The characterization algorithm assumes either a normal, log-normal, or square-root distribution for the radionuclide atmospheric activity concentrations. Initial studies of radionuclide data indicated that a square-root distribution was a good assumption. Now that the network has acquired more data, ^{137}Cs distributions are re-examined to determine the best distribution for characterization modeling.

Seasonal trends are also observed in this data set. These trends are the result of meteorological influences on crustal resuspension and atmospheric transport. Understanding these seasonal influences is important for determining the sources of ^{137}Cs.

RESEARCH ACCOMPLISHED

Table 1 lists the stations that have contributed radionuclide data to the pIDC, their locations, date at which the pIDC began processing station data in operations, and the station's sampling methodology. Most stations have continued to send data to the pIDC on a regular basis with some data gaps due to routine maintenance and equipment failures. Most 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 the aerosol sample collection to the end of the gamma-ray spectrum acquisition is longer than a week. For compliance with the CTBT International Monitoring System standards, stations must adhere to a sampling methodology where the time between the beginning of the aerosol sample collection and the end of the gamma-ray spectrum acquisition is less than or equal to 72 hours.

Measurements of airborne ^{137}Cs have been made at 9 of the 20 stations listed in Table 1. These stations are shown in Figure 1 along with their respective percent of samples containing ^{137}Cs at detectable levels. Stations in Europe have the highest percentage of ^{137}Cs detections due to the re-suspension of fallout from the Chernobyl event. The DE002 (Schauinsland, Germany) station has the highest ^{137}Cs detection percentage. This is largely due to the fact that the long sampling methodology at the DE002 station equates to lower ^{137}Cs detection limits. It is very likely that ^{137}Cs is present in all European aerosol samples at some level. Detection of ^{137}Cs is dependent on the station's sampling methodology, gamma-ray spectroscopy system, and other factors that affect the detection limit. ^{137}Cs detections in the current Southern Hemisphere stations are virtually non-existent. An exception to this are the ^{137}Cs detections made at the AR001 (Buenos Aires, Argentina) station. The source of the AR001 ^{137}Cs is yet to be completely determined, but it is likely due to local detector/laboratory contamination or local medical industry sources.

Figure 2 is a plot of the ^{137}Cs atmospheric activity concentrations. Multiple years of the aerosol data are plotted in a "box and whisker" format. A line in the box represents the mode of the 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 highest ^{137}Cs airborne activity concentrations were measured at the KW001 (Kuwait City, Kuwait) station at levels above 100 _Bq m $^{-3}$. The detections of ^{137}Cs at KW001 are strongly correlated to sandstorm activity and consequently to significant sand deposits on the filters. An investigation is currently being performed coorelating the ^{137}Cs concentrations in the Kuwait soil with those found in the sand collected on the filters. Figure 2 shows that a good portion of all the ^{137}Cs concentrations are in the 1 to 10 _Bq m $^{-3}$ range. Detections of ^{137}Cs at lower levels are observed at the DE002 (Schauinsland, Germany) and UK001 (Chilton, England) stations since their long sampling methodology equates to lower detection limits. No special treatment was given to this data for values below the limit of detection. All ^{137}Cs atmospheric activity concentrations measured at the pIDC are well below levels those that would adversely affect human health.

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.

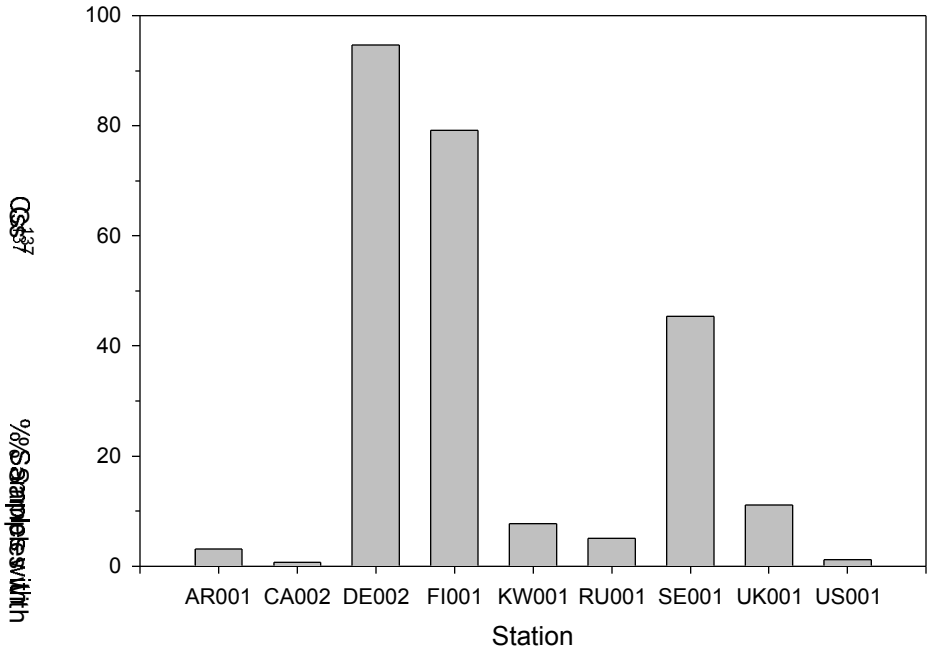


Figure 1. Percent of samples with detectable levels of ^{137}Cs.

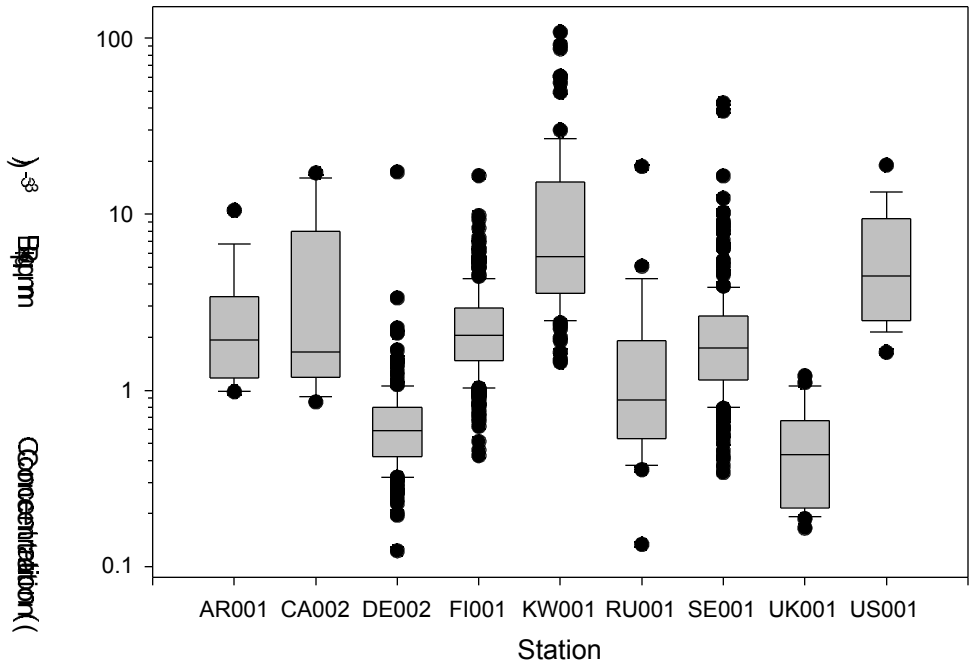


Figure 2. Box plot of all airborne ^{137}Cs measurements made at the pIDC.

Distribution of ^{137}Cs Atmospheric Activity Concentrations

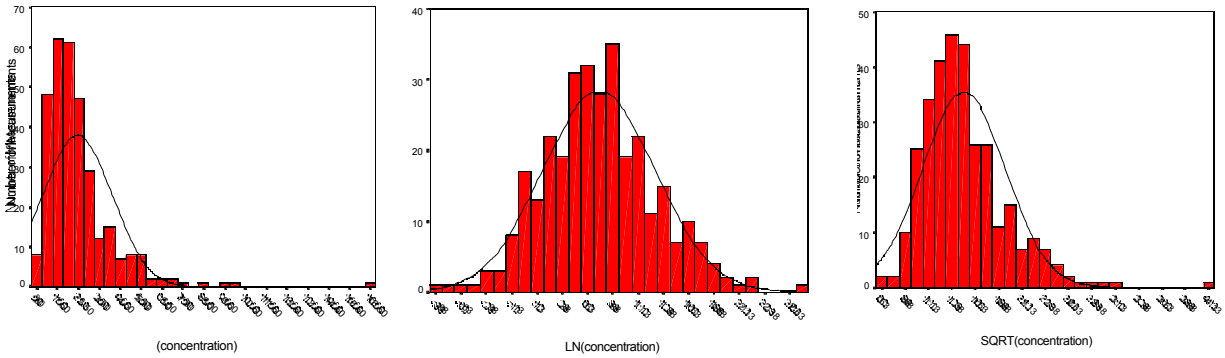
The radionuclide characterization algorithm utilized at the pIDC assumes the atmospheric activity concentrations follow a normal, log-normal, or square-root distribution [Evans, 1996]. Thus, the distribution type affects the prediction limits of the characterization code. Three methods were utilized in this research to examine how well the three different distribution types fit the ^{137}Cs data: histograms, cumulative probability plots, and the Kolmogorov-Smirnov test. Table 2 summarizes the results of the three methods utilized to determine the best distribution fit to the ^{137}Cs data. The results shown in this table indicate that the three methods produce very similar conclusions.

Table 2
^{137}Cs Distribution Fits

Station	Histogram	Cumulative Probability Plot	Kolmogorov-Smirnov Test	Best Distribution Fit
AR001	(Log-Normal)	(Log-Normal)	(Log-Normal)	(Log-Normal)
CA002	(Log-Normal)	(Log-Normal)	(Log-Normal)	(Log-Normal)
DE002	Log-Normal	Log-Normal	Log-Normal	Log-Normal
FI001	Log-Normal	Log-Normal	Log-Normal	Log-Normal
KW001	Log-Normal	Log-Normal	Log-Normal	Log-Normal
RU001	Log-Normal	Log-Normal	Log-Normal	Log-Normal
SE001	Log-Normal	Log-Normal	Log-Normal	Log-Normal
UK001	(Log-Normal)	(Log-Normal)	(Log-Normal)	(Log-Normal)
US001	(Log-Normal or Square-Root)	(Log-Normal or Square-Root)	(Square-Root)	(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.

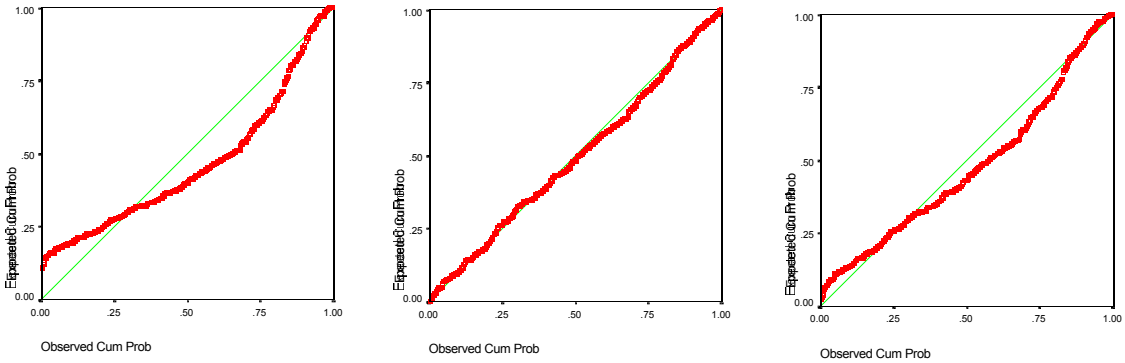
Histograms were made to visually display the ^{137}Cs activity concentration distributions. Figure 3 shows the ^{137}Cs histograms for the FI001 (Helsinki, Finland) Station. A plot of a normal distribution was plotted over each histogram to provide visual guidance. These plots clearly show that the best fit for this data is with the natural log transform. Thus, the log-normal distribution would be best suited for modeling ^{137}Cs at the FI001 (Helsinki, Finland) station. Similar plots were examined for each distribution type for each station where ^{137}Cs was detected. Table 2 contains a summary of the results of the histogram plotting method. The results for stations with small data sets should be interpreted with caution. For the US001 station, it was difficult to judge with the histogram method if the log-normal or square-root distribution best fit the data.



3a 3b 3c

Figure 3. Histograms of FI001 (Helsinki, Finland) ^{137}Cs atmospheric activity concentrations (_Bq m $^{-3}$) with normal distribution overlay. Figure 3a is the data with no transform. Figure 3b is the data with a natural log transform. Figure 3c is the data with a square-root transform.

Cumulative probability plots were also utilized to visually display the fit of the data to a normal distribution. Figure 4 displays the cumulative probability plots for the FI001 ^{137}Cs activity concentrations. A perfect correlation between the expected distribution and the observed data would result in a straight line at 45 degrees. Similar to the histograms shown in Figure 3, the cumulative probability plots show that the data with a natural log transform best fits the FI001 ^{137}Cs data. Table 2 contains a summary of the results of the cumulative probability plot method. Again, the results for stations with small data sets should be interpreted with caution. For the US001 station, it was difficult to judge with the cumulative probability plot method if the log-normal or square-root distribution best fit the data.



4a 4b 4c

Figure 4. Cumulative probability plots of FI001 (Helsinki, Finland) ^{137}Cs atmospheric activity concentrations. Figure 4a is the data with no transform. Figure 4b is the data with a natural log transform. Figure 4c is the data with a square-root transform.

The Kolmogorov-Smirnov test procedure was also applied to the ^{137}Cs data. This test compares the observed cumulative distribution function for a variable with a specified theoretical distribution. The Kolmogorov-Smirnov factor, 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. For this testing, the normal distribution was utilized as the theoretical distribution to which the observed data is compared. Natural log and square-root transformations were applied to the data to test the goodness-of-fit with the log-normal and square-root distributions, respectively. In contrast to the histogram and cumulative probability plot graphical methods, the Kolmogorov-Smirnov test provides a quantitative value by which the comparison may be judged, Z. Basically, the lower the Z, the better the fit. However, one must also take the sample set size into account since 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 3
Kolmogorov-Smirnov Test for ^{137}Cs Distributions

Station	Sample Set Size	Normal Distribution Z	Log-Normal Distribution Z	Square-Root Distribution Z	Best Distribution Fit
AR001	11	(0.863)	(0.528)	(0.709)	(Log-Normal)
CA002	8	(1.136)	(0.759)	(0.992)	(Log-Normal)
DE002	176	4.347	1.020	2.401	Log-Normal
FI001	316	2.963	0.890	1.907	Log-Normal
KW001	77	2.432	1.073	1.846	Log-Normal
RU001	25	1.491	0.569	1.114	Log-Normal
SE001	324	4.989	1.112	2.517	Log-Normal
UK001	18	(0.861)	(0.574)	(0.604)	(Log-Normal)
US001	13	(0.705)	(0.657)	(0.645)	(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.

The conclusions reached from the histogram, cumulative probability plot and Kolmogorov-Smirnov test are similar. The graphical methods require some subjective interpretation of the plots whereas the Kolmogorov-Smirnov test provides a Z value from which a comparison may be made. With the exception of the US001 station, all of the station's ^{137}Cs concentrations are best fit with a log-normal distribution. However, the US001 data set is small and the square-root distribution is only marginally better than the square-root distribution at fitting the data.

^{137}Cs Atmospheric Activity Concentration Trends

Trace element trends were investigated with the aid of both box and whisker plots and bar graphs. The objective of these plots is to reveal the existing annual trends present in ^{137}Cs atmospheric activity concentrations. In general, ^{137}Cs measurements at the stations may be broken down into about five source categories: 1) local resuspension of weapons and accident fallout, 2) remote resuspension of weapons and accident fallout, 3) deposition of stratospheric ^{137}Cs injected at the time of the initial weapon explosion or nuclear accident, 4) local reactor, medical, or other nuclear facilities, and 5) laboratory contamination. The first three of these source categories are dependent on meteorological factors. These meteorological influences vary by season and affect the atmospheric ^{137}Cs activity concentrations accordingly. Understanding the seasonal trends of the measured atmospheric ^{137}Cs activity concentrations aids in the modeling of the data as well as with the determination of the ^{137}Cs sources.

Figure 5 contains a box and whisker plot of ^{137}Cs at the FI001 (Helsinki, Finland) station. The mode values observed as lines within the box show that ^{137}Cs atmospheric activity concentrations are highest in winter and lowest in the summer. This is a similar trend observed at other European stations. The high ^{137}Cs in the winter is suspected to be a result of the increased probability of transport for resuspended Chernobyl

fallout from heavily contaminated regions. The seasonal low found in July is the result of decreased atmospheric pressure differentials occurring during this month and, consequently, the reduced crustal resuspension. The summer lows are commensurate with trends observed in other crustal materials (Biegalski *et al.*, 1998).

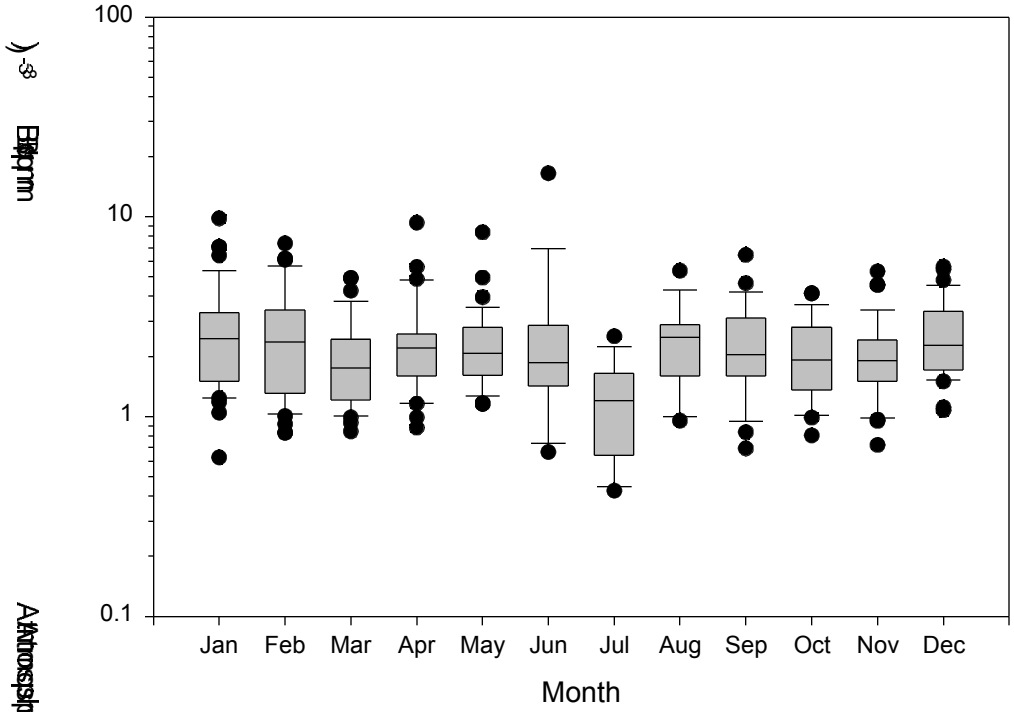


Figure 5. ^{137}Cs atmospheric activity concentrations at FI001 (Helsinki, Finland) as a function of month. Data accumulated from January, 1996 through June, 1999.

For stations with a reduced number of ^{137}Cs measurements, a bar chart was found to be more useful than a box and whisker plot for determining seasonal trends. Figure 6 is a bar chart showing the number ^{137}Cs detections at KW001 (Kuwait City, Kuwait) as a function of month. The majority of detections occurred during the spring. These detections strongly corresponded to the sandstorm activity reported in the local vicinity. These data support the assumption that ^{137}Cs detections at the KW001 station are a result of local resuspension.

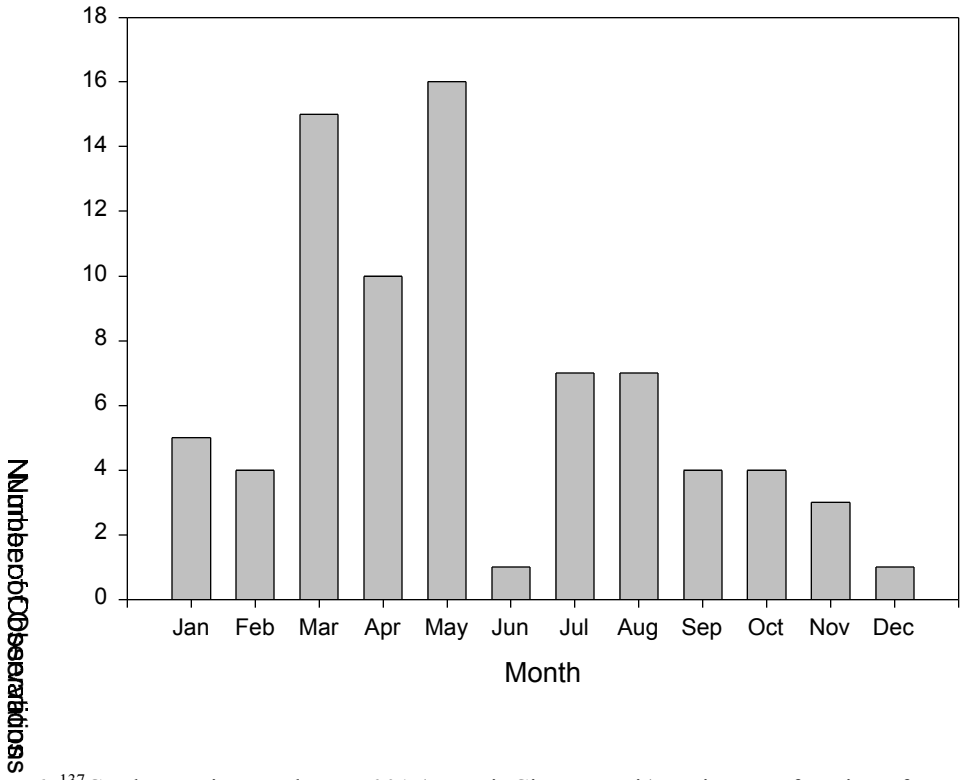


Figure 6. ^{137}Cs observations at the KW001 (Kuwait City, Kuwait) station as a function of month. Data accumulated from September, 1995 to June, 1999.

CONCLUSIONS AND RECOMMENDATIONS

Data presented in this paper show the atmospheric activity concentration distributions and seasonal trends for ^{137}Cs. All of the ^{137}Cs atmospheric activity concentrations follow a log-normal distribution with the exception of the US001 (Charlottesville, USA) station where the sample set is small. Most of the data exhibit seasonal trends with lows in the summer months. The seasonal tends are the result of meteorological influences on the resuspension of crustal material to the atmosphere. This phenomenon is very common with crustal material and the relationship between crustal particle suspension and wind speed is well documented [Gillette and Porch, 1978; Gillette and Passi, 1988].

These data can be used for modeling ^{137}Cs atmospheric activity concentrations at different stations and calculating prediction intervals. The prediction intervals may then be used to characterize radionuclide samples and determine if the atmospheric activity concentrations are at normal levels. Samples with radionuclide atmospheric activity concentrations at abnormally high levels should be examined in more detail than normal analysis, and an effort should be placed to determine the origin of the aerosol.

Similar studies will be performed for new stations that are brought on-line in the radionuclide monitoring system. It is important to understand the specific environment being monitored with special attention given to the distribution, trends, and sources of fission products within that environment. Such studies are vital to differentiating normal measurements from abnormal ones.

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