The Nucleus **47**, No.1 (2010) 47-53

The Nucleus
A Quarterly Scientific Journal of Pakistan
Atomic Energy Commission
NCLEAM, ISSN 0029-5698

US CONTINENTAL NUCLEAR TESTS CONDUCTED DURING 1961–1992 : STATISTICAL ANALYSIS

*M. WASIM, M. ARIF and J. H. ZAIDI

Chemistry Division, Directorate of Science, PINSTECH, P.O. Nilore, Islamabad, Pakistan

(Received December 14, 2009 and accepted in revised form January 29, 2010)

The paper presents statistical analysis of the data obtained from US continental nuclear tests conducted during 1961 - 1992. The analysis summarizes the types of tests, their yield, radioactivity released and the radionuclides emitted during the tests. The application of principal component analysis shows that some radionuclides are emitted in correlation with other radionuclides, which need not originate from the same decay chain. The overall analysis puts emphasis on the detection of radioiodines and radioxenons both for event screening and for event backtracking.

Keywords : Fission products, Event screening, Event backtracking, US continental nuclear tests

1. Introduction

Natural and anthropogenic radionuclides are measured in order to get information about their origin, amount, distribution and behaviour in the environment. There are around twenty singly occurring primordial radionuclides, among them some have very long half lives and some are beta or alpha emitters [1]. ^{40}K is the only suitable singly occurring primordial radionuclide, which is readily detectable by γ-ray spectrometry. Other radionuclides of terrestrial origin are characterized in term of radioactive series. Most of the natural radionuclides identified in a γ-ray spectrum are from two series, one starting with ^{238}U and the other with ^{232}Th. Since ^{238}U and ^{232}Th both emit very weak γ lines, they are estimated by the presence of their progenies in their decay series. The most abundant anthropogenic radionuclides identified on the earth surface or in the atmosphere include ^{90}Sr and ^{137}Cs, which are from nuclear weapon tests and nuclear accidents. The activity ratio of ^{137}Cs:^{90}Sr is approximately 1.6 in depositions found at many sites. ^{90}Sr is a pure beta emitter and is not detectable by γ-ray spectrometry. ^{137}Cs with a long half life (~ 30 y) and high fission yield was released in the atmosphere in large quantities by nuclear weapon tests conducted during the 1950s and 1960s in the atmosphere. The Chernobyl accident in 1986 also deposited ^{137}Cs over a large part of Europe. Now ^{137}Cs has become a permanent component of the background γ-spectrum collected in the Northern Hemisphere. Its concentration varies due to the re-suspension of soil [2]. The deposition in both hemispheres is highest at midlatitude and decreases on moving towards the poles or the equator. With the exception of local contamination due to reactor accidents ^{137}Cs is virtually undetectable in the atmosphere of the Southern Hemisphere [3]. Sometimes ^{137}Cs is detected around a nuclear facility, due to leakage from the primary cooling system [4, 5] or from nuclear waste storage [5].

Airborne radionuclide monitoring is a major surveillance technique employed in the Comprehensive Test Ban Treaty (CTBT). Unlike seismometry, radionuclide monitoring is not a real-time detection system; i.e. an explosion is detected after a few days or weeks. However, radionuclide monitoring complements other methods of detection due to its high sensitivity, backtracking capability, and most importantly, its event discrimination capability. Experts [6] consider particulate and noble gas monitoring as an essential component of the radionuclide monitoring system. The present paper discusses selection of suitable radionuclides primarily those emitting γ-rays as useful marker for the identification of a nuclear explosion. These radionuclides will become part of the NUCDATA [7] library. NUCDATA was primarily developed by us for neutron activation analysis (NAA). The current

* Corresponding author : mwasim@pinstech.org.pk

version of NUCDATA has been modified for k_0-NAA; with this update it now contains data of 1034 nuclides with 22,450 gamma lines. The database is used by our in-house written programs "GAMMACAL" and "GAMMALAB" for radionuclide identification and quantification. The data analysed in this paper has been taken from US DOE reports [8, 9].

2. Data Analysis

All data in this study has been taken from reports published by US Department of Energy [8, 9]. The data analysis was performed using MS EXCEL and MATLAB. The data in reference [8] provides only univariate data while the data in reference [9] is suitable for multivariate analysis techniques. One such technique is principal component analysis (PCA), which is further explained in the next section.

2.1. Principal component analysis

PCA [10, 11] decomposes multivariate data into a set of abstract eigenvectors and an associated set of abstract eigenvalues. Each eigenvector is a linear combination of the original variables and each eigen value represents a portion of the total variation in the data and, as more and more variation in the data is explained, the later eigenvalues become smaller in magnitude. PCA when performed on a data matrix $\boldsymbol{X}$ *(M* $\times$ *N),* with suitable *K* number of components selected, decomposes the data matrix into a scores matrix $\boldsymbol{T}$, a loadings matrix $\boldsymbol{P}$ and an error matrix $\boldsymbol{E}$ of dimensions M $\times$ K, K $\times$ N and M $\times$ N respectively. Mathematically it can be defined as

$$X = TP + E \qquad (1)$$

If M represents the number of tests and N the number of variables, then the scores matrix relates to the tests and the loading matrix to the variables. If plotted one scores vector against the other, suitable information about the relation of tests can be obtained. This is also true for the loading matrix.

3. Results and Discussion

The data studied in this paper has been divided into two parts: whole dataset taken from reference [8] and the reduced dataset taken from reference [9]. The whole dataset provides variables such as "*type of test*" and its "*yield*". The reduced dataset contains information about the *"radionuclides emitted", "radioactivity released", "depth of the detonation"* and the *"yield".* Both reports do not throw any light regarding the type of detonation device i.e. whether it was uranium or plutonium based.

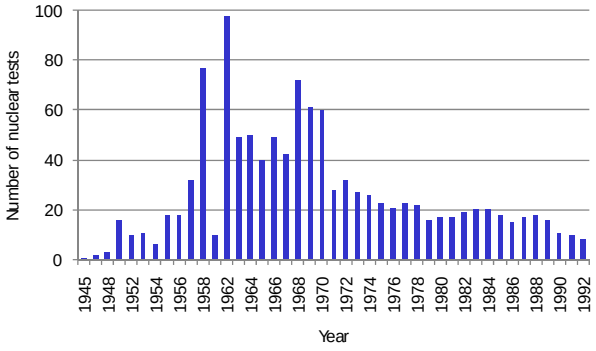

Figure 1. Frequency of nuclear tests conducted during the specified period.

3.2. Whole dataset

Starting from 1945 and ending in 1992, a total of 1149 nuclear devices were detonated in 48 years. During this time, five years went silent including 1947, 1949, 1950, 1959 and 1960. Figure 1 provides year-wise distribution of the number of nuclear detonations. The United States Government entered into a unilateral testing moratorium on October 1, 1958 and on September 15, 1961 the US resumed testing. Figure 1 shows that maximum number of tests (98) was conducted during 1962. The overall average is 24 tests per year. Among these detonations 934 were underground, 210 in the atmosphere and only 5 were underwater. The distribution of the tests in different media is presented in Figure 2, which shows that the types of atmospheric tests include *airburst, airdrop, balloon, barge, rocket, surface* and *tower*. The types of underground tests were *crater, shaft* and *tunnel* with the maximum number of 853 tests as *shaft type.* In these tests the yield varied from zero to 15 Mt. Figure 3 shows the frequency distribution of the yield, where the maximum lies at an average value of 30 kt with 795 events, which is almost 70% of the total tests. On October 2, 1992, the United States entered into another unilateral moratorium on nuclear weapons testing, which is still implemented. There have not been any nuclear tests conducted by the US Government since September 1992.

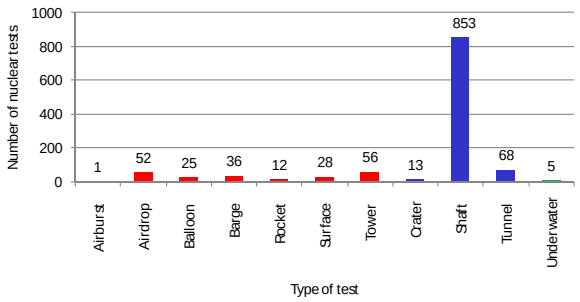

Figure 2. Types of nuclear tests during the specified period.

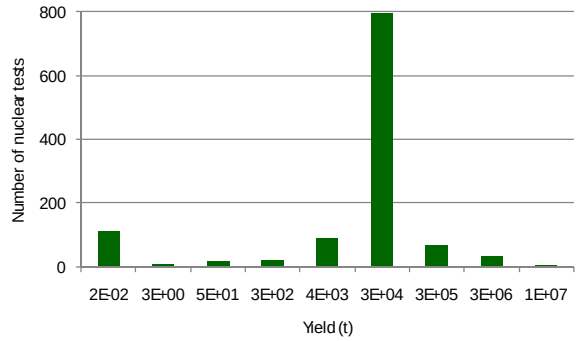

Figure 3. Frequency distribution of yield (tons).

3.3. Reduced dataset

The reduced dataset has been obtained from the U.S. Department of Energy report [9], which contains data of 433 tests. The report catalogues information on *test name, test date, sponsor,* and *time of detonation, depth of burial, location, purpose, type,* and *yield, release detected onsite* or *offsite and type of release*.

To record the radioactivity released, 14 different terminologies were employed, these included *cement back, controlled, drillback, gas sampling (GS), late time seepage* (LTS), *mud point, plutonium dispersal* (PD), *prompt particle sampling* (PPS), *seepage, special packaging operation, surface, test* and *uncontrolled release.* The variation of released activity in different categories is given in Figure 4, which shows that the *"drillback"* release was recorded in 275 tests and *"test"* release in 152 tests. *Drillback* release has been defined as [9] "directional drilling operation, performed after the test activities have ceased, to sample fission product materials in the test cavity", while *test release* is the "activity measured after detonation", the activity is normalised to 12 hours after the detonation. Figure 5 shows frequency distribution of the activity released for *drillback* and *test*. Among the 433 tests the release of only 52 (12%) tests was detected offsite and release of 381 (88%) tests was detected onsite.

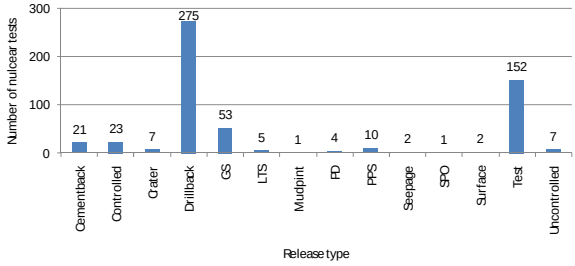

Figure 4. Number of events of release with release type.

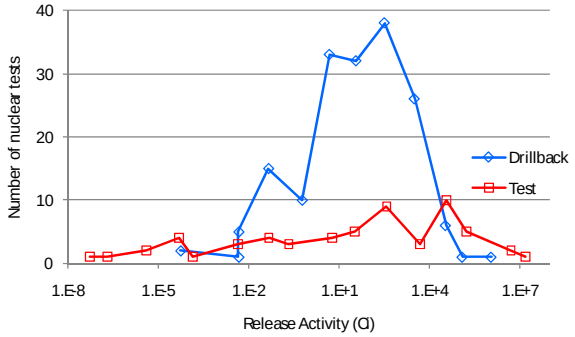

Figure 5. Frequency distribution of release of Drillback and Test.

The radionuclides identified only in the *test* release have been used in further data analysis because in *drillback* release only iodine and xenon were detected. The data in this section belongs to 100 tests; among these 3 are atmospheric tests (*Johnnie Boy*, *Small Boy* and *Little Feller I*). In the data the *released activity* varied from 5×10^{-8} Ci to 1.5×10^{7} Ci and the *yield* varied from zero to 104 kt. In total, 57 different radionuclides were observed. The frequency distribution of radionuclides is given in Figure 6, where it can be observed that ^{135}I was observed in 61 tests, followed by ^{133}I, ^{131}I, ^{135}Xe, ^{133}Xe, ^{138}Xe, ^{138}Cs, ^{88}Kr, ^{87}Kr, ^{140}Ba, ^{140}La, ^{132}Te, ^{88}Rb, ^{103}Ru, ^{132}I and others. The highest number of radionuclides (19) was observed in the *Tee* test. Figure 6 shows that in the radionuclide monitoring of a nuclear test, iodine and xenon are the most important radionuclides.

PCA was performed on data of size (100×61), where 100 represents the tests and 61 represents variables which includes 57 different radionuclides, *released activity* (Ci), *media type* (converted to numbers and designated as *Type Val)*, *yield (t)* and *depth of detonation (ft).* The presence of each radionuclide is denoted as 1 and its absence as 0.

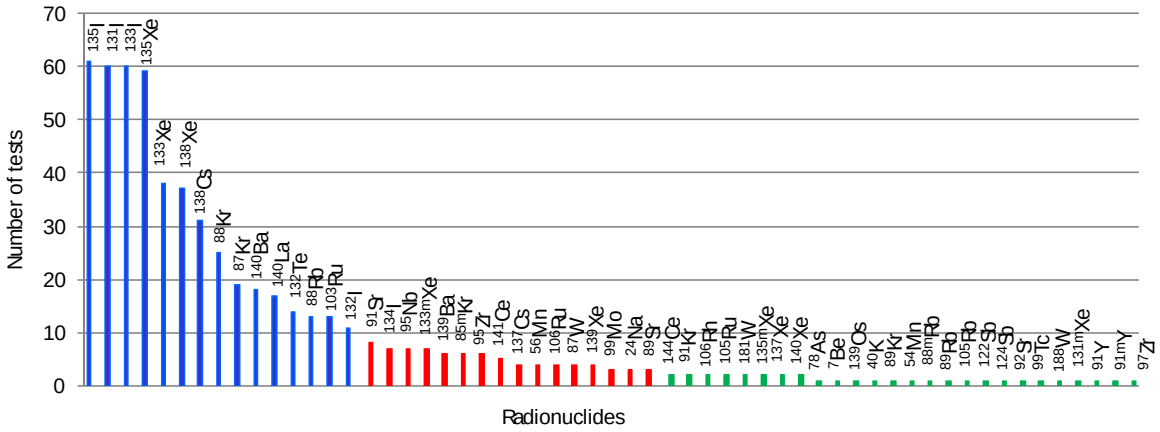

Figure 6. Frequency distribution of different radionuclides emitted in the nuclear tests.

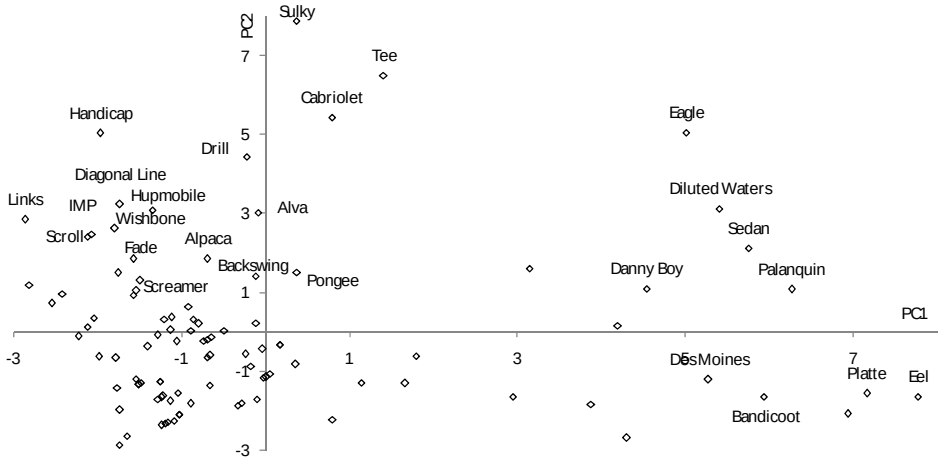

Figure 7: Scores plot of PC2 vs. PC1, showing name of the tests with more than 7 radionuclides in release.

The scores plot of PC2 vs. PC1 is given in Figure 7, and loadings plot of PC2 vs. PC1 in Figure 8. Figure 7 shows three different regions of data points, the first region of both negative PC1 & 2 is the region where majority of the data points lie. On comparing Figure 7 with Figure 8, it indicates that this region is not characterized for radionuclides rather it is associated with the *yield, depth* and *Type Val.* Almost all tests with less than 8 radionuclides are clustered in this area. All the tests in Figure 7 having 8 or more radionuclides have been labelled with their names. The tests either on high PC1 or on high PC2 are those tests which are characterized with large number of radionuclides, for instance *Sulky* is associated with 17, *Tee* with 19, *Eagle* with 14 and *Eel* with 13 radionuclides. Once again on comparing Figure 7 with Figure 8, individual tests can be identified with their associative radionuclides such as *Eel* is characterised for the presence of iodine, while *Sulky* is associated with the xenon. The tests which released only iodine and xenon are 38, events released only xenon is 14 and events with iodine only are 9.

On considering the loadings plots of PC2 vs. PC1, implies the following pairs of radionuclides with positive relationships $^{95}Zr/^{95}Nb$, $^{133}I/^{135}I$, $^{140}Ba/^{132}Te$, $^{140}La/^{103}Ru$, $^{133}Xe/^{135}Xe$ and $^{88}Kr/^{87}Kr$. These inter-radionuclide correlations are due to their compatible half-lives. Figure 8 confirms these relations. Moreover, Figure 8 also shows that *yield, depth of burial* and *Type Val* is inversely related with the release. The activity released, however, is highly correlated with the $^{95}Zr/^{95}Nb$ couple. Among the pairs presented for positive or negative correlations, only $^{95}Zr/^{95}Nb$ is in the single decay chain, while the remaining radionuclides are from different decay chains.

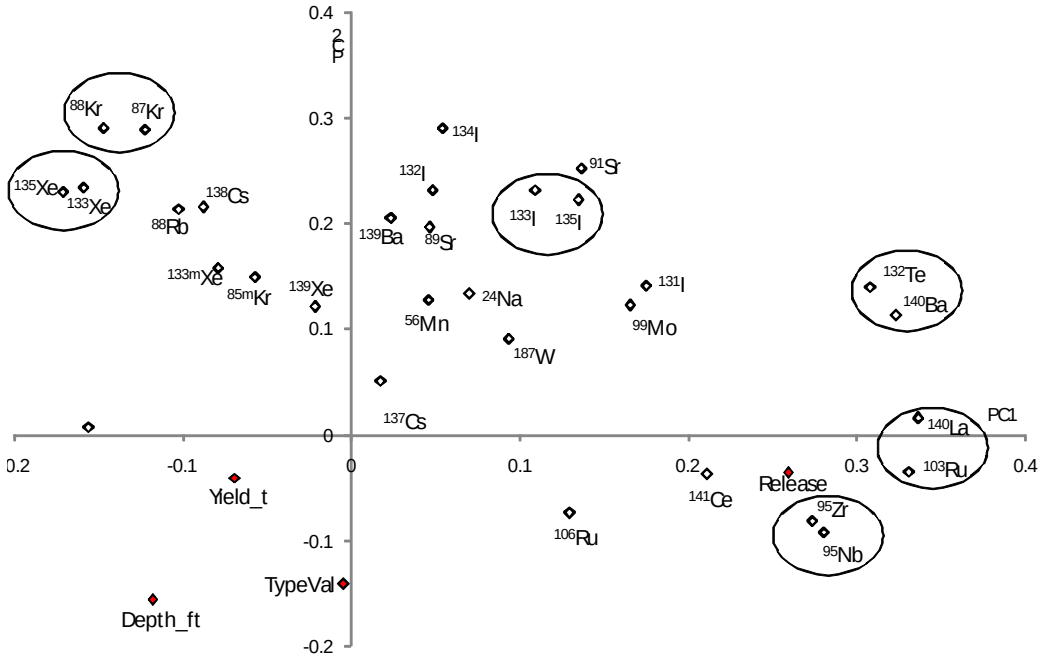

Figure 8. Loadings plot of PC2 vs. PC1, showing name of the variables.

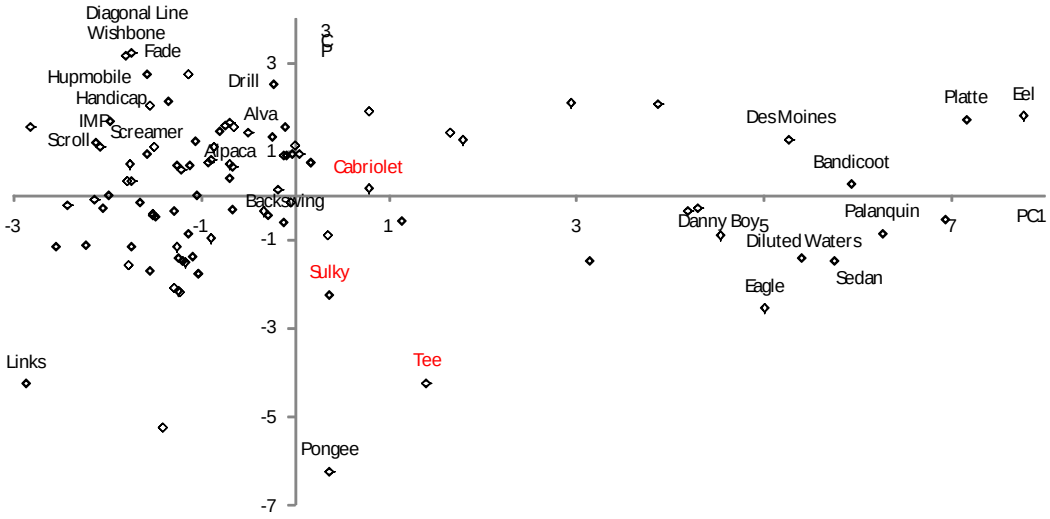

Figure 9. Scores plot of PC3 vs. PC1 showing name of the tests with more than 7 radionuclides in release.

Similar information can be retrieved from scores plot (Figure 9) and loadings plot (Figure 10) of PC3 vs. PC1. On comparing Figure 9 with Figure 10, the tests which appeared on high PC1 scores are still correlated with ^{95}Zr/^{95}Nb, ^{140}Ba/^{132}Te and ^{140}La/ ^{103}Ru couples. The *Sulky* test which has high score on PC2 due to its minimum value of *depth* is somewhere in the middle of PC3 scores. Figure 10, however, makes clear that some points which appear close to each other in Figure 8, such as ^{131}I and ^{99}Mo, are not correlated in the space of PC3 and PC1. Moreover, some pairs of radionuclides such as ^{133}I/^{135}I show some correlation with ^{131}I in both plots. Similarly, ^{133}Xe/^{135}Xe and ^{88}Kr/^{87}Kr are always in the neighbourhood and the same is true for ^{95}Zr/^{95}Nb and ^{140}La/^{103}Ru couples.

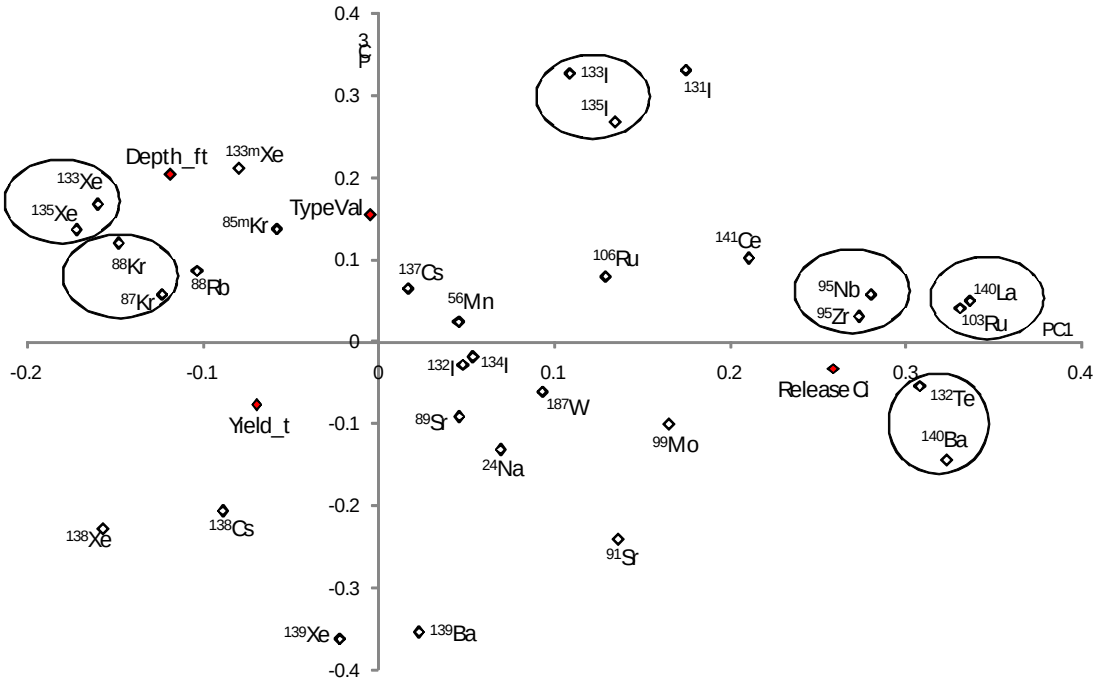

Figure 10. Loadings plot of PC3 vs. PC1, showing name of the variables.

3.4. Event screening

Event screening means to generate sufficient data to discriminate, with high confidence, nuclear device debris from another nuclear event such as a reactor explosion. There are several methods to discriminate an event.

1. In nuclear device debris isotopic ratio of same element generally do not distort. The activity ratios will be within a typical range, which is given only by the degree of fuel burn-up and the physical reactor characteristics before release.
2. Isotopes that have low yield, long half life and are produced as activation products are also good candidate for discrimination. Two such radionuclides are ^{134}Cs and ^{136}Cs. ^{134}Cs is not available in sufficient quantity from a nuclear device to be observed but will be observed in large quantities from a reactor accident. Similarly, ^{136}Cs can be observed in device debris, but in small quantities only, whereas a reactor would release large quantities. Isotopic ratios for some of the radionuclides produced in a nuclear reactor are given in Table 1 [13, 14]. The data of the *Schellbourne* test shows a ratio of ^{133}I/^{131}I as 3.4, which is different from its corresponding ratio for reactor, as found in Table 1. This value indicates a source of nuclear explosion and not a reactor accident.

Table 1. Isotopic ratio at different times after reactor shut down [13, 14].

	Explosion		Reactor	
Isotopic Ratio	1d	7d	30d	720d
^{144}Ce/^{141}Ce	0.08	0.09	0.14	1
^{143}Ce/^{141}Ce	12	0.68	2	1
^{133}I/^{131}I	8	0.11	2.5	2
^{131m}Xe/^{133}Xe	0.08	0.03	-	-
^{97}Zr/^{95}Zr	39	0.11	3	1

3.5. Event time/sample-age in daughter-parent relation

The fission of each parent atom produces a different set of fission product atoms. However, while an individual fission is not predictable, fission products are statistically predictable. The amount of any particular isotope produced per fission is called fission yield, typically expressed as % per parent fission; therefore, yields total to 200% not 100%. For radionuclides present in a decay chain

$$N_p \xrightarrow{\lambda_p} N_d \xrightarrow{\lambda_d} \qquad (2)$$

The event time, t, can be obtained using the following equation.

$$t = \frac{1}{\lambda_p - \lambda_d} \ln \left[\frac{\left(A_d \lambda_p / A_p \lambda_d \right) - k}{\left(N_d / N_p \right) - k} \right] \quad (3)$$

$$k = \frac{\lambda_p}{\lambda_p - \lambda_d} \quad (4)$$

where λ is the decay constant, A is activity, N is the independent fission yield and the subscript p denotes “parent” and d “daughter”.

The important daughter-parent radionuclides found in this study are ^{95}Zr/^{95}Nb, ^{133}Xe/^{133m}Xe, ^{140}La/^{140}Ba and ^{133m}Xe/^{133}I. The analysis of *drillback* data shows that event time for the *Schellbourne* test using ^{133m}Xe/^{133}Xe pair is 23.7 d. Similarly, for the *Metropolis* test data using ^{133m}Xe/^{133}Xe pair is 19.5 d, which shows that the *drillback activity* was measured almost after 20 days of the event.

4. Conclusions

The detection of suitable radionuclide marker of a nuclear test depends on many factors, such as detonation height relative to the earth surface, nature of the device, depth of detonation, nature of the media, type of rock underneath, in case of underground explosion, and other factors. If a device is detonated close to the earth’s surface, the release will contain many activation products originating from soil. However, for a device detonated well below the earth’s surface with good containment, only radioxenons will be released. With an average number of 24 tests per year, 855 tests were conducted underground as shaft type. The frequency distribution shows that radioiodines (^{133}I, ^{131}I and ^{132}I) and radioxenons (^{135}Xe, ^{133}Xe and ^{138}Xe) are important radionuclides from a monitoring point of view. The loadings plot indicates that ^{95}Zr/^{95}Nb, ^{133}I/^{135}I, ^{140}Ba/^{132}Te, ^{140}La/^{103}Ru, ^{133}Xe/^{135}Xe and ^{88}Kr/^{87}Kr are important pairs of radionuclides for aerosol monitoring. For event screening and event backtracking radioiodines and radioxenons are very important radionuclides.

References

[1] R. L. Kathren, Appl. Radiat. Isot. **49** (1998) 149.

[2] S. R. Biegalski, B. Hosticka and L. R. Mason, J. Radioanal. Nucl. Chem. **248** (2001) 643.

[3] M. A. Pires Do Rio, E. C. S. Amaral and H. G. Paretzke, J. Aerosol Sci. **25** (1994) 821.

[4] C. Chung, C. C. Chen and C. N. Hsu, Waste Managements **11** (1991) 241.

[5] C. Chung and C. Y. Chen, J. Radioanal. Nucl. Chem. Lett. **214** (1996) 421.

[6] M. Zahringer, Radionuclides for GARDS Library and Identification of Test Explosions, Informal Radionuclide Monitoring Workshop, New Zealand, 8-10 May (1996).

[7] M. Wasim and J. H. Zaidi, Nucl. Instr. Meth. Phy. A **481** (2002) 760.

[8] U.S. Department of Energy, DOE/NV-209 (Rev. 15) United States Nuclear Tests, July 1945 through September 1992, Nevada Operations Office, Las Vegas, Nevada, December (2000).

[9] C. R. Schoengold, M. E. DeMarre and E. M. Kirkwood, Radiological Effluents Released from U. S. Continental Tests 1961 through 1992, Rept. DOE/NV-317 (Rev. 1), Nevada (1996).

[10] B. G. M. Vandeginste, D. L. Massart, L. M. C. Buydens, S. De. Jong, P. J. Lewi and J. Smeyers-Verbeke: Handbook of Chemometrics and Qualimetrics, Part B, Elsevier, Amsterdam (1998).

[11] S. Wold, K. Esbensen and P. Geladi, Chemom. Intell. Lab. Syst. **2** (1987) 37.

[12] R.W. Perkins and L. A. Casey, Radioxenons: their role in monitoring a comprehensive test ban treaty, DOE/RL-96-51, United States, Department of Energy, Richland, WA 99352, (1996).

[13] M. Zahringer, Radionuclides for GARDS library and identification of test explosions, Informal radionuclide monitoring workshop, New Zealand, May 8-10 (1996).

[14] J. Lucas, Event Discrimination Methods, CTBT Technologies Workshop, LLNL, Beijing, China, Feb. 18-20 (1997).