Conf-9205174--1

PNL-SA--20486

DE92 015094

MULTIVARIATE METHODS IN NUCLEAR WASTE REMEDIATION: NEEDS AND APPLICATIONS

B. A. Pulsipher

May 1992

Presented at the
Seventh International Conference on
Multivariate Statistics
May 5-9, 1992
University Park, Pennsylvania

Work supported by
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

MULTIVARIATE METHODS IN NUCLEAR WASTE REMEDIATION: NEEDS AND APPLICATIONS

Brent A. Pulsipher
Pacific Northwest Laboratory [1]
Richland, Washington 99352

ABSTRACT

The United States Department of Energy (DOE) has developed a strategy for nuclear waste remediation and environmental restoration at several major sites across the country. Nuclear and hazardous wastes are found in underground storage tanks, containment drums, soils, and facilities. Due to the many possible contaminants and complexities of sampling and analysis, multivariate methods are directly applicable. However, effective application of multivariate methods will require greater ability to communicate methods and results to a non-statistician community. Moreover, more flexible multivariate methods may be required to accommodate inherent sampling and analysis limitations. This paper outlines multivariate applications in the context of select DOE environmental restoration activities and identifies several perceived needs.

INTRODUCTION

The U.S. Department of Energy (DOE) is commencing major environmental cleanup at its principal facilities throughout the nation. During the preceding 50 years, DOE and its predecessor agencies focused on nuclear materials production while maintaining much of the nuclear waste in temporary storage awaiting future permanent disposal. Some waste management practices resulted in environmental contamination. Today, DOE is actively formulating waste management, environmental restoration, and decommissioning strategies. However, due to the inherent complexities and risks involved with the cleanup of radioactive and hazardous waste, the challenges are great.

---

[1] Operated for the U.S. Department of Energy by Battelle Memorial Institute under contract DE-AC06-76RLO 1830.

Multivariate statistical methods are directly applicable to many aspects of the DOE environmental restoration activities. Although it is readily apparent that complex correlative relationships exist for many of the radioactive and hazardous components, some issues must be addressed before practical implementation of multivariate statistical methods is fully realized. Before outlining these issues, the range of DOE cleanup activities is discussed to give an appreciation for the magnitude of DOE's environmental restoration requirements. Several proposed treatment technologies are briefly presented and multivariate issues are discussed. Finally, several needs are identified for practical implementation of multivariate statistical methods.

BACKGROUND: DOE'S CHALLENGE

A variety of storage and disposal practices for nuclear and hazardous wastes from previous DOE production activities have been employed depending on waste form and risk level. Waste disposal and environmental restoration efforts can be categorized into several general classifications that correspond to specific waste forms and disposal practices as follows:

- Waste Contained in Underground Storage Tanks
- Solid and Liquid Waste in Drums, Containers, and Trenches
- Soil and Groundwater Contamination
- Contaminated Facilities

Waste Contained in Underground Storage Tanks

DOE is managing 100 million gallons of radioactive waste in 327 tanks across the DOE complex (DOE 1991). This waste resulted from the production of strategic nuclear materials for national defense. Waste forms within the tanks range from liquids to hard salt cake and sludge. Characterization of the tank contents is the first step in the remediation process. Although records were kept on transfer of materials in and out of the tanks, tank contents are only qualitatively known due to the unknown chemical and radiochemical reactions that have occurred and the lack of detailed records available.

Current characterization of tank contents is somewhat limited because of the safety and risks involved in obtaining and handling samples, cost of acquiring and analyzing samples, and limited sampling access to the tanks. At DOE's Hanford site, core samples are obtained by driving a small pipe into the waste through existing tank risers. The material in the pipe is extracted for characterization purposes. The process of obtaining a single core sample and conducting thorough chemical, radiochemical, and physical analyses costs approximately $1 million, thereby significantly limiting the number of core samples available. Also little is known about the spatial distribution of chemical and radiochemical constituents within the tanks. This creates significant uncertainty about the representativeness of a few core samples obtained through a few existing risers.

Near-term stabilization of tank wastes is also a major objective. Some of the material contained in single shell tanks at Hanford has leaked into the environment. Also conditions exist in several of the tanks that could potentially result in fires or explosions (DOE 1991). Characterization is crucial for development of proper interim stabilization measures.

Treatment technologies for the waste contained in tanks are being developed and demonstrated. Solidification is the primary technology being pursued for this waste (DOE 1991). The low-level liquid wastes will be removed and treated to extract any radioactive components. This low-level waste will be combined with cement-forming materials and made into a grout. The resulting cement-form will be placed in vaults specificly designed for permanent storage. The high-activity sludge and recycle from the low-level treatment will be combined with glass-forming chemicals or crushed glasses, then continuously fed into a ceramic melter to be vitrified. The molten material will be poured into steel canisters where the radioactive components will become encapsulated as a glass waste form. The canisters will then be shipped for permanent disposal in a deep-geological repository.

These treatment strategies involve many steps such as characterization, pre-treatment, mobilization of sludges, retrieval, grout formulation and production, glass formulation and production, etc. Large programs that deal with each aspect of the remediation of waste stored in tanks are progressing.

Solid and Liquid Wastes

More than 14 million 55-gallon drums of varying waste forms have been stored or buried across the DOE complex (DOE 1991). Early disposal practices allowed the commingling of various types of wastes (Kostelnik 1991). As a result, much of the buried waste contains both hazardous and radioactive constituents. Solid wastes have been buried since the 1940s, and practices have changed considerably over time. Records for the older solid wastes are essentially non-existent, but there is indication that burial trenches may contain low-level wastes, transuranic materials, explosives, hazardous chemicals, mixed waste and large pieces of contaminated equipment. Many of the original containers have degraded significantly, resulting in contamination of the immediately surrounding soil. Some of the materials were placed in less durable containers (i.e., cardboard boxes) instead of 55-gallon drums.

Characterization of buried waste is a necessary first step in the remediation process. Classification of waste types is required before appropriate remediation technologies can be applied. The heterogeneous properties of buried waste make the sampling process difficult. For example, contaminated clothing could be included in the same drum as a hazardous substance from a chemical laboratory. Moreover, the drum may have degraded such that material from one drum could be exposed to material from another drum. Mixtures of radioactive and hazardous wastes present a unique problem uncommon to normal cleanup of hazardous waste sites.

Technologies are being developed and tested to handle these difficulties. Some waste may be treated in-situ or retrieved, sorted, and repackaged or treated depending on waste type. If the waste is deemed to be TRU waste [2], it will be packaged for permanent disposal in the Waste Isolation Pilot Plant site in New Mexico (DOE 1991). For low-level waste removal several treatment technologies being investigated include

[2] TRU waste is defined as containing more than 100 nanocuries per gram of alpha-emitting transuranium radionulides with half-lives greater than 20 years.

incineration, grout, saltstone and repackaging in drums for burial in shallow land trenches. Other technologies being applied to hazardous EPA Superfund sites are also being considered.

For drums that are still intact, retrieval and sorting strategies would appear to be fairly straight-forward, although sorting operations must minimize the exposure to personnel and if possible maintain the integrity of the containers. These goals make the task much more difficult. For the wastes in decomposed drums or buried in less durable containers, the retrieval and sorting process becomes a significant challenge.

Soils and Groundwater

The history of operations dating back to the 1940s shows that spills of hazardous substances occurred at several sites. Waste management and disposal practices were also implemented that are unacceptable under today's regulations and current knowledge of the effects of chemicals on the environment (Keller 1991). DOE estimates that there are more than 3,700 hazardous substance release sites under its jurisdiction (DOE 1991). Numerous types of waste exist at these release sites generated from past production and testing of nuclear devices. Contaminants include unstabilized mill tailings, petroleum products, VOCs, PCBs, carbon tetrachloride, heavy metals, radionuclides, acids, and bases.

Groundwater at some sites has been contaminated by constituents that have been carried into the soil by rainwater percolating through contaminated soil sites. Liquid wastes that have leaked from high-level waste tanks have also contaminated nearby soils. Less radioactively contaminated liquid wastes were discharged to the environment via cribs, trenches, or ponds.

Many remediation technologies have already been developed and demonstrated for cleanup of soils and groundwater at EPA Superfund sites. Studies have been conducted to determine the applicability of existing technologies to DOE cleanup efforts for soils and groundwater. Because pertinent experience, regulations, and technologies exist, numerous options are available. In one study on the Hanford site, a total of 86 technologies currently available or holding potential for remediation of contaminated soils and groundwater were identified (Keller 1991). However, because of the large volumes of contaminated soils and groundwater, treatment by conventional methods may be ineffective and costly. Contaminants can often be present as mixed waste forms that alter their behavior as they are transported and transformed in subsurface environments. Thus, although many technologies exist, advanced methods will be required to characterize and predict behavior of contaminants, remediate soils and groundwater, and assess the success of remedial actions.

Facilities

Approximately 500 contaminated DOE facilities are currently slated for decommissioning and decontamination (DOE 1991). The decommissioning process has been ongoing for over 20 years at DOE sites. The basic technologies needed to perform decommissioning safely are well understood and routinely applied (Keller 1991). However, improvements are required that provide faster, better, and cheaper results.

The common strategies for decommissioning are characterization, assessment, in-situ disposal, in-situ remediation, removal, treatment, and closure. Some areas requiring testing and demonstration include decontamination and release of valuable materials, volume-reduction of contaminated metals and equipment before disposal, and disintegration of contaminated concrete to permit removal of the contamination from the constituent materials.

WASTE REMEDIATION CONDITIONS AMENABLE TO MULTIVARIATE APPROACHES

The circumstances surrounding environmental restoration across the DOE complex provide ample opportunity for contributions from multivariate statistical approaches. Some areas where multivariate methods seem especially suited are discussed below. The intent is not to develop a comprehensive list herein, but the areas discussed do represent potential opportunities for significant contributions.

Estimation and Decision Objectives

Many environmental cleanup units involve multiple waste constituents. Mixed waste contains both hazardous and radioactive components. Besides the hazardous elemental components, numerous other elemental concentrations and physical properties are determined from samples. Correlative patterns exist between many of these constituents due to the original processes that produced the wastes. Moreover, covariances are often introduced in the measurement process. Multivariate methods should be considered when these correlative patterns exist.

For example, waste contained in underground storage tanks are sampled and analyzed to determine chemical and radiological composition as well as physical properties. Numerous chemical and radiochemical concentrations are reported (Morgan 1988). Certain constituents are correlated due to the production processes, mixing patterns with the tanks, chemical reactions, radioactive decay, and sampling constraints. Certain analytical biases or uncertainties also affect classes of constituents differently. Estimation and modeling methods that account for these interrelationships are most appropriate when multiple decision rules are applied to multiple estimates. In addition, exploration of correlative patterns can provide valuable insight that may be pertinent to modeling, stabilization, and treatment technologies.

One of the limiting factors affecting the use of multivariate methods is the small number of samples that are available for some waste types. Although numerous analyses can be performed on subsamples, very few samples can be obtained from, for example, underground storage tanks. Estimation of variance components is often used to determine how possible sources of error (variance) contribute to the overall uncertainty. Usually the covariance estimates associated with each source of uncertainty are non-estimable (or non-unique) due to the limited number of samples relative to the number of analyzed constituents. Dimension reduction techniques can be applied, but the interpretation of the results becomes difficult.

As DOE begins to address the mixed waste issues, the correlative patterns inherent in the data could provide valuable information. Significant reductions in sampling and analysis may be achievable through the proper use of information on covariance structures. At a minimum, adequate presentations of such information would help decision-makers gain greater insight into the underlying characteristics of waste.

Combining Data from Various Sources

Because of the safety and economic issues inherent in characterization of hazardous and nuclear waste, it is important to utilize relevant information available from all sources. Minimizing uncertainties through maximum use of ancillary information is becoming recognized within DOE as an important concept. Data fusion methods are gaining greater sponsorship in the waste remediation arena. At least two classes of data fusion methods are being explored. The first is quantitatively combining estimates from similar measurement systems. The second is the quantitative use of ancillary information such as historical data, model results, laboratory studies, or related measurements. Because of the multiple constituent characteristics of DOE wastes, multivariate methods for data fusion are applicable.

Another aspect of data fusion is the ability to bring data from various sources into an integrated data environment. Many scientists desire a data environment that allows visualization, analysis, and modeling of data to facilitate interpretation and data fusion. Visualization of multivariate data is key to the success of multivariate data analysis.

Real-Time Monitoring and Quality Control

Monitoring and control of waste constituents, instrumentation, and processes are required throughout the remediation cycle. Often many constituents or measurements must be monitored simultaneously. In some cases, adequate control can be performed using upper and lower action limits (specification limits). However, multivariate statistical quality control methods are needed to monitor and control many of the processes.

Multivariate methods are being applied to control the process of vitrification of high-activity waste (Kuhn 1989). To ensure an acceptable glass waste form (durable and processable), several of the major constituents must be controlled within acceptable concentration limits. Multivariate methods are employed to simultaneously control multiple constituents before the material is fed into the ceramic melters.

Other processes for retrieval and treatment of soils and groundwater are also candidates for multivariate process control. Many instruments will be gathering data in real-time to monitor waste remediation activities. These instruments and processes would benefit from multivariate statistical quality cortrol.

Experiments and Sampling Strategies Driven By Multivariate Analyses

Application of multivariate exploratory analysis tools can sometimes provide great insights into future experimentation or sampling strategies. Understanding correlative patterns assists scientists develop new theories and models that can be validated through experimental or sampling exercises.

The value of multivariate exploratory analyses in guiding experimental design is illustrated through an application within the high-level vitrification process (Elliott and Pulsipher 1989). Glass-forming chemical constituents can be conveniently added to the waste material in a variety of chemical forms (e.g., hydroxides, nitrates, phosphates, and glass frits). Experience on small-scale, pilot-scale, and full-scale melters at various DOE vitrification facilities provided data with varying glass-forming chemical feed stock. A partial least squares (PLS) analysis was conducted to explore relationships between feed-stock type and production rate. The analysis revealed that the cations that were added in the form of nitrates had a negative effect on the production rate whereas those added as hydroxides or formates had a positive effect on production rate. This result became apparent only after applying multivariate methods. Experimentation followed to validate the observed effects and chemical addition procedures were modified.

Vast amounts of data are being gathered that, if examined using exploratory multivariate data analysis methods, could provide valuable information to the scientific community.

ACTIONS REQUIRED FOR PRACTICAL IMPLEMENTATION OF MULTIVARIATE METHODS

It is apparent that multivariate statistical methods are applicable to the DOE environmental restoration program. Nonetheless, historically, multivariate applications have been somewhat limited. The statistical community must play a significant role in overcoming the roadblocks to practical implementation of multivariate methods.

Several actions should be considered to facilitate overcoming these roadblocks. Some of these have been categorized into four general areas: 1) communication, 2) education, 3) demonstrations, and 4) tools development. Each is briefly discussed below.

- **Communication Through Visualization --** Visualization of multivariate concepts is key to practical implementation. "Black-box" multivariate analyses are not accepted unless the users (scientists and regulators) can visualize the essence of the analyses. Many visualization tools are becoming readily available including geographic information systems and 3-D visualization packages. Downloading visual representations of the multivariate analysis process onto videotape for client viewing is a powerful mechanism for communication. Through this media, manipulation of multi-dimensional data can be presented visually to the clients.

  Although not restricted to multivariate applications, there is a real need for visual display of uncertainty. With visualization tools becoming more available, too often results from models and data are portrayed without consideration for the inherent uncertainties. Methods and tools are needed to adequately communicate uncertainties.

- **Education --** One argument for relying only on univariate approaches is that the regulations do not consider multivariate issues. Most scientists and engineers have little training in statistics and have never been exposed to multivariate statistical methods. What little statistics training they have was derived from elementary statistics courses where little else than formulas and simple univariate methods were taught. Course content for non-statisticians should, however, be focused on concepts rather than how to calculate t-statistics. These students should be taught why and when statistics are applicable, what statistical methods are available for which conditions, and what assumptions are required for their proper use. This would prepare them for recognizing the need for certain types of statistical methodologies.

For the scientists, engineers and policy analysts already in the work environment, statisticians have the responsibility to actively inform them and raise statistical issues to appropriate levels of attention.

- **Demonstrations --** Practical applications of multivariate statistics in environmental restoration programs have not been publicized well. Statisticians are fairly conscientious about presenting results at statistical conferences or in statistics journals. However, more statisticians should present results at conferences and publish in journals outside our discipline. There is a need for more conceptual treatments on uncertainty issues as well as presentations from a statistical point of view.

  Practical demonstrations of applied multivariate statistical methods would provide concrete examples of how these methods can be applied to help solve challenging problems. These demonstrations would help educate scientists and regulators and support related multivariate applications.

- **Tools Development --** Numerous software packages with multivariate analysis and visualization capabilities are emerging. These packages must be amenable to full integration within complete information systems. Tailored integrated information systems that contain modules for data management, statistical analysis, physical modeling, and visualization represent the future wave for information management. Statistics modules that could be portable to other applications or computer platforms may be required. Indeed, integrated information systems would facilitate data fusion and real-time quality control efforts.

Development and demonstration of these integrated information environments requires significant interaction between computer scientists, statisticians, and modelers. Some grassroots efforts are emerging within the DOE arena to develop such integrated information environments (Tzemos 1991). The need for flexible tools for multivariate analysis continues.

SUMMARY

Complexities inherent in DOE environmental restoration programs provide ample opportunities for real contributions through application of multivariate methods. Remediation activities of hazardous and/or nuclear materials found in underground storage tanks, buried drums or trenches, soils and groundwater, and facilities usually involves characterization, treatment, and monitoring of multiple contaminants. Issues where multivariate statistical methods are directly applicable include the following:

- Estimation and Decision Objectives Involving Multiple Constituents
- Combining Data from Various Sources
- Real-Time Monitoring and Quality Control
- Experimental Design and Sampling Strategies

To ensure practical implementation of multivariate methods, some roadblocks must be overcome. Statisticians can help eliminate these roadblocks by actively pursuing improvements in four general categories:

- Communication Through Visualization
- Education
- Demonstrations
- Tool Development

Although isolated applications of multivariate methods can be found within the DOE environmental restoration programs, much more remains to be accomplished. The task of clean-up of DOE sites and facilities would benefit

from multivariate methods implementation. Uncertainty will continue to be an issue throughout the entire remediation process.

REFERENCES

Elliott, M.L., and B.A. Pulsipher. February 1989. "The Effect of Melter Feed on Processing Characteristics." WVSP-89-066, Letter Report, prepared for West Valley Nuclear Services by Pacific Northwest Laboratory, Richland, WA.

Keller, J.F., et.al. September 1991. "The Hanford Model: Environmental Cleanup Problems, Science and Technology Needs, and Facility Requirements." PNL-7805 or WHC-MR-0282, prepared for the U.S. Department of Energy by Pacific Northwest Laboratory and Westinghouse Hanford Company, Richland WA.

Kostelnik, K.M. December 1991. "Buried Waste Integrated Demonstration Plan." EGG-WTD-9870, prepared for U.S. Department of Energy by Idaho National Engineering Laboratory, EG&G Idaho, Inc., Idaho Falls, ID.

Kuhn, W.L., M.R. Toland, and B.A. Pulsipher. May 1989. "Process Simulation and Statistical Approaches for Validating Waste From Qualification Models." PNL-6699, prepared for the U.S. Department of Energy by Pacific Northwest Laboratory, Richland, WA.

Morgan, L.G., W.W. Schulz, M.R. Adams, and K.W. Owens. July 1988. "Summary of Single-Shell Tank Waste Characterization: 1985 to 1987." WHC-EP-0075, prepared for U.S. Department of Energy by Pacific Northwest Laboratory and Westinghouse Hanford Company, Richland, WA.

Tzemos, S., B.J. Evans, and M.E. White. 1991. Developing a GIS to Facilitate Data Analysis for Environmental Restoration of a Large Waste Site. PNL-SA-19158, presented at the 11th Annual Environmental Systems Research Institution User Conference, May 20-24, 1991, Palm Springs, CA. Pacific Northwest Laboratory, Richland, WA.

U.S. Department of Energy. 1991. Environmental Restoration and Waste Management Five-Year Plan, Fiscal Years 1993-1997. DOE/S-0089P, Washington, DC.

END

DATE FILMED

7 / 27 / 92