GEOCHEMICAL STUDIES OF COMMERCIAL LOW-LEVEL RADIOACTIVE WASTE DISPOSAL SITES

TOPICAL REPORT

R. Dayal, R.F. Pietrzak, and J.H. Clinton

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NUCLEAR WASTE MANAGEMENT DIVISION
DEPARTMENT OF NUCLEAR ENERGY, BROOKHAVEN NATIONAL LABORATORY
UPTON, LONG ISLAND, NEW YORK 11973

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R. Dayal,* R.F. Pietrzak, and J.H. Clinton

*Present Address: Rock Sciences Section, Civil Research Department,
Ontario Hydro, Toronto, Ontario, Canada

ABSTRACT

The results of source term characterization studies for the commercially operated low-level waste (LLW) disposal sites located in the eastern United States provide an understanding of the importance of hydrological and geochemical factors in controlling the mechanics of leachate formation, evolution of leachate compositions, microbial degradation of organic waste and development of anoxia in the trenches, and the nature and extent of leaching of waste materials. The varying degrees of the intensity of these processes, as determined by the different site characteristics, are clearly reflected in the contrasting leachate geochemistries of Maxey Flats and West Valley trenches, as compared to those of Barnwell and Sheffield trenches. These are important geochemical considerations which not only define LLW source terms but also shed light on the nature and extent of geochemical changes that are likely to occur along a redox gradient outside of the trench environment.

Unsegregated, poorly packaged, and unstabilized wastes, which were buried at the sites, are readily leachable and biodegradable. Especially at the Maxey Flats and West Valley sites, where the infiltrated water accumulates in the trenches leading to a bathtub effect, the relatively long residence time of accumulated water results in continually intense leaching and microbial degradation of waste materials for extended time periods. These leachates exhibit strongly anoxic, reducing conditions, as reflected by negative redox potentials, depletion of dissolved oxygen and sulphate, high alkalinity and ammonia concentrations, as well as high concentrations of dissolved iron and manganese, present primarily as Fe^{2+} and Mn^{2+}. Although the Barnwell and Sheffield leachates exhibit a fair degree of anoxia, as reflected in depletion of dissolved oxygen and some alkalinity and ammonia enrichments, no significant sulphate depletion relative to ambient groundwaters is observed, indicating that the anaerobic degradation processes are not as advanced as those reflected by the leachate characteristics of Maxey Flats and West Valley trenches. This is also evident from the relatively higher redox potentials measured for the Barnwell and Sheffield leachates.

The enrichments, to varying degrees, of inorganic organic, and radionuclide constituents associated with fuel cycle and non-fuel cycle LLW reflect not only the nature of the leaching process and reactivity and amount of waste materials but also the differences in the site characteristics. Relative to ambient groundwaters, the trench leachates are generally enriched in Na^+, K^+, Fe_T, Mn_T, Cl^-, dissolved organic and inorganic carbon and several organic compounds, as well as radionuclides such as H-3, Co-60, Cs-134, Cs-137, Sr-90, Pu-238, Pu-239,-240, Am-241, attributable to leaching of waste materials. The Maxey Flats and West Valley leachates exhibit modifications which are generally considerably more pronounced than those observed in the Barnwell and Sheffield leachates. This is also reflected in generally higher values for specific conductance, ionic strength, and total dissolved solids in Maxey Flats and West Valley leachates.

The observations made at the existing sites have important implications for site selection and disposal of low-level radioactive wastes at future shallow land burial sites in that waste package requirements will include

waste segregation, improved stabilization, and proper packaging. In accordance with current disposal procedures, stabilized and/or packaged waste not only ensures trench stability but also decreases the rate and extent of leaching and microbial degradation of waste materials. In addition, the uncertainties in the source term are considerably reduced. Furthermore, to avoid water accumulations in the trenches, future sites will most likely be located in relatively well-drained systems, such as that represented by the Barnwell site, where the residence of accumulated water in the trenches is relatively short. Many of the current waste package requirements and site selection criteria outlined in 10CFR61 for the shallow land disposal of low level radioactive wastes are based on lessons learned from experiences at existing shallow land burial sites.

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EXECUTIVE SUMMARY

As part of the U.S. Nuclear Regulatory Commission's (NRC's) efforts to develop a data base on LLW source term characteristics, Brookhaven National Laboratory (BNL) during the past several years has been involved in geochemical studies of commercially operated shallow land burial sites located in the eastern United States (Barnwell, South Carolina; Maxey Flats, Kentucky, Sheffield, Illinois, and West Valley, New York). The BNL research effort has generated a considerable amount of data on source term characteristics and geochemical controls on the compositions of trench leachates from existing shallow land burial sites. Although the current disposal procedures are much improved from those practiced in the past, it is likely that several of the factors, conditions, and processes observed by BNL at these sites can be expected to occur at future shallow land burial sites.

The study has provided an understanding of the importance of hydrological and geochemical factors in controlling the mechanics of leachate formation, evolution of leachate compositions, microbial degradation of organic waste and development of anoxia in the trenches, and the nature and extent of leaching of waste materials. A knowledge of these processes and controls affecting the source term geochemistry as well as an understanding of the important factors that are likely to contribute to variability, modifications, and uncertainties in the source term is essential for evaluating the performance of the waste package and the site, making valid predictions of release for dose calculations, and for planning site performance monitoring as well as remedial actions.

In this report, we present the results of our investigation involving geochemical characterizations of trench leachates from the Maxey Flats, West Valley, Barnwell, and Sheffield LLW disposal sites. Data on trench chemistry are reviewed and discussed in terms of mechanisms and processes controlling the compositions of trench solutes, emphasizing the contrasting leachate geochemistries observed at Maxey Flats and West Valley and Barnwell and Sheffield sites.

The Maxey Flats and West Valley trenches are excavated in non-porous shale and glacial till of relatively low hydraulic conductivity which results in accumulation of rainwater that infiltrates through the trench caps. Because of the relatively long residence time of accumulated water in the trenches at these sites, prolonged leaching and microbial degradation of waste materials occurs continually, leading to leachate formation. As a result of such interactions for extended time periods, the resultant trench leachates acquire geochemical properties which are unique and considerably modified compared to ambient groundwaters.

The Barnwell and Sheffield trenches, on the other hand, represent generally well-drained systems, where the infiltrated water has a relatively short residence time and is continually not present in the trenches, thereby

resulting in leachate geochemistries which are not modified, compared to ambient groundwaters, to such an extent as the Maxey Flats and West Valley leachates are.

The development of anoxia observed in practically all the trenches at the sites can be attributed to redox conditions in the trench leachates largely controlled by microbial degradation of organic matter present in waste materials. As a result of aerobic degradation processes, the dissolved oxygen present in the trenches is consumed rapidly. Following depletion of all molecular oxygen and concurrent development of anoxia in the trenches, further degradation of organic material occurs by anaerobic processes which involve denitrification, followed successively by sulphate reduction and methane generation. During these degradation processes, there is a continual production and buildup of decomposition products such as carbon dioxide and ammonia, thereby resulting in elevated levels of alkalinity, total aqueous CO_2, and ammonia in trenches. Concurrently, dissolved oxygen, nitrate, and sulphate are consumed, depleting the leachates in these constituents.

Given the relatively stagnant water accumulations in the trenches at Maxey Flats and West Valley, the leachates exhibit strongly anoxic, chemically reducing conditions, as reflected by negative redox potentials, depletion of dissolved oxygen and sulphate, high alkalinity and ammonia concentrations, as well as high concentrations of dissolved iron and manganese, present primarily as Fe^{2+} and Mn^{2+}.

Although the Barnwell and Sheffield leachates exhibit a fair degree of anoxia, as reflected in depletion of dissolved oxygen and some alkalinity and ammonia enrichments, no significant sulphate depletion relative to ambient groundwaters is observed, indicating that the anaerobic degradation processes are not as advanced as those reflected by the leachate characteristics of Maxey Flats and West Valley trenches. This is also evident from the relatively higher redox potentials measured for the Barnwell and Sheffield leachates.

The modifications in ambient groundwaters, as a result of waste leaching processes, are also reflected in contrasting character of leachate geochemistries. The enrichments, to varying degrees, of inorganic, organic, and radionuclide constituents associated with fuel cycle and non-fuel cycle LLW reflect not only the nature of the leaching process and reactivity and amount of waste materials but also the differences in the site characteristics.

Relative to ambient groundwaters, the trench leachates are generally enriched in Na^+, K^+, Fe_T, Mn_T, Cl^-, dissolved organic and inorganic carbon and several organic compounds, as well as radionuclides such as H-3, Co-60, Cs-134, Cs-137, Sr-90, Pu-238, Pu-239, Pu-240, Am-241, attributable to leaching of waste materials. Because of the relatively long residence of accumulated water in the trenches at Maxey Flats and West Valley leading to continually prolonged leaching of waste materials and subsequent buildup of dissolution products, the Maxey Flats and West Valley leachates exhibit modifications which are generally considerably more pronounced than those

observed in the Barnwell and Sheffield leachates. This is also reflected in generally higher values for specific conductance, ionic strength, and total dissolved solids in Maxey Flats and West Valley leachates.

The solubility calculations indicate that because of strongly reducing conditions in the trenches at Maxey Flats and West Valley calcite, dolomite, and rhodochrosite mineral phases are likely to exert control on the concentrations of Ca^{2+}, Mg^{2+}, Mn^{2+}, and CO_3^{2-}. In case of iron equilibria, gross supersaturations are found in these leachates with respect to siderite, mackinawite, amorphous FeS, and pyrite. In contrast, because of relatively mildly reducing conditions and correspondingly lower alkalinities, the Barnwell and Sheffield leachates exhibit undersaturation with respect to the carbonate phases and supersaturation with respect to the iron oxide minerals. The calculations show the importance of contrasting leachate geochemistries, especially redox potential and alkalinity, in carbonate and iron equilibria.

The acid-generating potential and buffering capacity of a leachate are important geochemical controls that maintain a balance between acidity and alkalinity which, in turn, determines to a large extent the reactivity, partitioning, cycling, and mobility of radionuclides along a redox gradient. In this respect, the Maxey Flats and West Valley leachates generally represent well-buffered systems and exhibit high acid-generating potentials, whereas the Barnwell and Sheffield leachates are characterized by relatively low buffering capacities and acid-generating potentials.

The problems associated with disposal of unsegregated, poorly packaged, and unstabilized wastes, especially at the Maxey Flats and West Valley sites, have helped establish the consequences of past disposal practices and site characteristics on waste package performance, the trench environment and stability, source term characteristics and variability, leaching and microbial degradation of waste materials, and radionuclide behavior and mobility.

The observations made at the existing sites have important implications for site selection and disposal of low-level radioactive wastes at future shallow land burial sites in that waste package requirements will include waste segregation, improved stabilization, and proper packaging. In accordance with current disposal procedures, stabilized, packaged waste not only ensures trench stability but also decreases the rate and extent of leaching and microbial degradation of waste materials. In addition, the uncertainties in the source term are considerably reduced. Furthermore, to avoid water accumulations in the trenches, future sites will most likely be located in relatively well-drained systems, such as that represented by the Barnwell site, where the residence of accumulated water in the trenches is relatively short.

Many of the current waste package requirements and site selection criteria outlined in 10CFR61 for the shallow land disposal of low level radioactive wastes are based on lessons learned from experiences at existing shallow land burial sites.

1. INTRODUCTION

Studies of existing LLW disposal sites provide an unique opportunity to investigate the behavior and fate of radioactive waste nuclides in a natural setting and to assess the suitability of similar sites for future disposal of low-level radioactive wastes. In addition, such investigations provide valuable information on LLW source terms under a range of hydrological and geochemical conditions, as well as on site behavior and waste package performance. The effects of the disposal of unsegregated, unstabilized, and poorly packaged waste on the trench environment can also be estimated.

As part of the NRC's efforts to develop a data base on LLW source term characteristics, BNL during the past several years has been involved in geochemical studies of commercially operated shallow land burial sites located in the eastern United States (Barnwell, South Carolina; Maxey Flats, Kentucky; Sheffield, Illinois; and West Valley, New York). The BNL research effort has generated a considerable amount of data on source term characteristics and geochemical controls on trench leachates from existing shallow land burial sites. Although the current disposal procedures are much improved from those practiced in the past, it is likely that several of the factors, conditions, and processes observed by BNL at these sites can be expected to occur at future shallow land burial sites. A knowledge of such factors, controls, and processes, which characterize the trench leachate chemistry, is essential for obtaining a reliable estimate of the source term, an important consideration in evaluating the capability of future sites to conform with 10CFR61 of the federal regulations code.

In this report, we present the results of our investigations involving characterization of trench leachates from the Maxey Flats, West Valley, Barnwell and Sheffield disposal sites. Data on trench leachate chemistry are reviewed and discussed in terms of mechanisms and processes controlling the compositions of trench solutes, emphasizing the contrasting leachate geochemistries observed for Maxey Flats and West Valley and for Barnwell and Sheffield sites. A preliminary evaluation of the contrasting nature of Maxey Flats and Barnwell leachate compositions was presented in an earlier report (Dayal et al., 1984a).

The results of laboratory experiments investigating extra-trench processes affecting the geochemistry of trench leachates are not discussed here. Specifically, extra-trench processes relating to oxidation-induced geochemical changes in iron-rich, anoxic waters and leachate-soil interactions have been discussed in earlier reports (Dayal et al., 1984b; 1986a; 1986b).

The main areas addressed in this report include:

- The processes which control leachate formation and development of anoxia in the trenches,

- The important solute constituents of trench leachates,

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- The processes and redox couples which may determine redox equilibria and buffering in trench leachates,

- The important biogeochemical processes controlling leachate compositions,

- The effects of disposal of unsegregated, unstabilized, and poorly packaged waste on the trench environment,

- Those solute constituents which may serve as tracers for in situ leaching and microbial degradation of buried wastes,

- The contrasting leachate geochemistries of the Maxey Flats and West Valley and the Barnwell and Sheffield sites,

- The factors which contribute to uncertainties in the source term, and

- The relevance of these research findings to NRC needs.

1.1. Commercial Disposal of Low-Level Radioactive Waste

Disposal of low-level radioactive wastes at commercially operated disposal sites began in 1962 at Beatty, Nevada. Since that time additional disposal sites were established in Maxey Flats, Kentucky; West Valley, New York; Richland, Washington; Sheffield, Illinois; and Barnwell, South Carolina, as shown in Figure 1.1.

Figure 1.1. Commercial low-level radioactive waste disposal sites in the United States.

The disposal sites at West Valley, Maxey Flats, and Sheffield have discontinued operations leaving Barnwell as the only disposal site accepting commercial radioactive wastes in the Eastern United States. That site plus the Beatty and Richland sites now handle all of the nations commercially generated low-level wastes, which amount to 75,000 m^3/yr (Robertson, 1984). Table 1.1 lists the existing disposal sites, the year licensed, and their current status.

Table 1.1. Commercial low-level radioactive waste disposal sites.

Site	Year Licensed	Current Status
Beatty, Nevada	1962	open
Maxey Flats, Kentucky	1963	closed
West Valley, New York	1963	closed
Richland, Washington	1965	open
Sheffield, Illinois	1967	closed
Barnwell, South Carolina	1971	open

Low-level waste consists of a variety of laboratory, hospital, and reactor equipment, residues, and trash which are contaminated with radioactive materials. For the most part, the types of materials buried include clothing, plastics, paper, ion exchange resins, scintillation vials, animal carcasses, solidification agents, decontamination agents, and other materials in small quantities.

Transuranic elements, e.g., plutonium, americium, neptunium, etc., in concentrations greater than 10 nCi/g of waste are classified as TRU waste and are not accepted at commercial disposal sites. However, in the past, solid wastes contaminated with plutonium and other transuranic radionuclides were mixed with LLW and buried at the commercial sites.

Solid low-level radioactive wastes, such as those generated at commercial power plants, hospitals, research facilities, and universities, are usually buried in the containers in which they are shipped. The purpose of the package is to minimize personnel exposure and to prevent the loss of radioactive materials during shipment to the disposal site.

Burial practices at the commercial disposal sites are similar in that, typically, solid wastes are buried in shallow rectangular trenches with varying dimensions, ranging from 60-150 m long, 8-15 m wide, and 6-12 m deep. The trench floor is sometimes sloped toward one or more sumps, and riser pipes are provided for routine observation of water levels and for removal of water. Solid wastes are buried in an assortment of containers, such as cardboard and wooden boxes, plastic bags, cement casks, steel drums, and steel bins.

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When filled with waste, the trench is backfilled with previously exca-
vated earth, compacted, and covered with capping material. Generally, the
compacted cap contains clay material that expands when moist, to retard infil-
tration of rain water. The finished trench is covered with soil and planted
with shallow rooted ground cover to prevent soil erosion. Rain or groundwater
that enters a trench may leach the contents of the buried containers that have
failed either during burial operations or through corrosion and decomposition
processes. A prime concern of burial operation is that the leached material
may not be carried to the environment around the trenches by the infiltrated
water.

In this report, the terms "trench water" and "trench leachate" are used
interchangeably to identify water that resides in a trench, and that may be
removed via sumps or test wells in the trench.

1.2. Site Characteristics and Operations

The disposal sites are located in sparsely settled areas that were con-
sidered suitable for shallow land burial of low-level radioactive wastes. A
summary of selected geohydrologic characteristics of four commercial LLW dis-
posal sites studied by BNL, as reported by Clancy et al. (1981), is presented
in Table 1.2.

Table 1.2. Characteristics of commercial low-level
radioactive waste sites.[1]

Disposal Site	Mean Annual Precipitation (mm)	Surface Material	Interstitial Permeability	Bedrock Material	Depth to Groundwater (m)	Depth to Regional Aquifier (m)	Sorption Capacity of Surface Material
Maxey Flats	1,050	Clay, Weathered-Siltstone and Stone	Low	Clay-Shale, Siltstone Sandstone	Unknown	85	Moderate to High
West Valley	1,040	Weathered Till-silty clay and gravel	Low	Shale Siltstone	31–38	>60	High
Barnwell	1,200	Sand-Clay	Low	Sedimentary-Sand	10–20	200	Moderate
Sheffield	900	Silt Sand	Low	Shale, Limestone	6–15	>50	Moderate

[1]Adapted from Clancy et al. (1981).

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1.2.1. Maxey Flats Site

Detailed descriptions of site characteristics and hydrogeology have been reported by Zehner (1983) and Meyer (1976) and summarized by Dayal et al. (1984b). Briefly, the burial site is located on Maxey Flats, an eroded plateau in the knobs region of northeastern Kentucky. The plateau rises approximately 90 to 120 m above the surrounding alluvium-filled valley. The Maxey Flats region has a humid continental climate with sharp contrasts between winter and summer months. Based on the reported mean annual precipitation of 1,050 mm, the Maxey Flats site can be characterized as a humid site.

The burial trenches are excavated entirely in the Nancy Member of the Borden Formation. The upper 0.3 to 7.5 m at the site is yellow brown regolith composed of weathered shale. McDowell (1971) reported the presence of two sandstone beds of variable thickness located at about the middle and at the base of the regolith. In some places, the shale between the two sandstone beds is partly weathered, imparting a mottled yellow brown and gray color to the rock unit. The unweathered part of the Nancy Member, which averages about 4.5 m in thickness, extends from the base of the lower sandstone bed in the weathered part of Nancy shale and is underlain by a 0.5 m-thick sandstone bed, at a depth of approximately 7.5 m. About 80% of the rocks underlying the disposal site are shale. Because of the extremely low hydraulic conductivity of the bedrock, essentially all groundwater movement occurs through fractures and joints.

The Maxey Flats site was operational from May 1963 until the end of 1977. By 1972, some of the completed trenches were filled or partially filled with infiltrating water. A water management program was initiated to remove the accumulated water in the trenches and to control further entry of water. Efforts towards water removal included pumping the water out of trenches, storing the leachates in a surface storage tank, reducing the leachate volume by evaporation, and solidification of evaporated residues for eventual disposal on site. Efforts to minimize water ingress included grading and improving surface drainage, recapping older trenches to reduce cap permeability, and establishing a vegetation cover over completed trenches (Mills and Page, 1972). A temporary remedial measure, taken in 1981 to minimize entry of infiltrated water into the trenches, involved installation of a PVC plastic cover over the surface of trenches located in the middle and western sections of the burial area. As a short-term solution, the trench cover appears to be effective in preventing surface runoff from infiltrating the trenches.

1.2.2. West Valley Site

The site is located on a ridge of glacial till ranging in thickness from 0 to 170 m, which is underlain by shale of Paleozoic age. The unweathered till into which the trenches are excavated has a very low hydraulic conductivity ($\approx 10^{-7}$ cm/s). The depth of the water table ranges from 31 to 38 m. The climate is cool and moist with the average annual precipitation being 1040 mm, much of which falls in the form of snow.

The site accepted waste for burial between November 1963 and March 1975, when disposal site operations were suspended following detection of leachate seeping through the cover of one of the trenches. Some of the waste trenches have been reported to accumulate enough infiltrated water, resulting in a bathtub effect. This was reported to be a major problem affecting the site performance (Clancy et al., 1981).

1.2.3. Barnwell Site

The site is located in Barnwell county in southwestern South Carolina. As reported by Cahill (1982), the Barnwell disposal site is underlain by sediments of the Atlantic Coastal Plain, which are relatively unconsolidated and are composed of stratified gravel, sand, silt, clay and limestone. These unconsolidated sediments, in turn, are underlain by Triassic rocks. The unsaturated zone at Barnwell extends from the land surface to just above the water table and is comprised of the aeolian sands and part of the Hawthorne Formation. The sediments in the unsaturated zone are generally fine grained sands admixed with minor amounts of clay and coarse sand. The depth of the water table varies from 10 to 20 m. The climate of the area is characterized by warm, humid summers and mild winters. The mean annual precipitation for the area is reported to be 1,200 mm.

The burial site is owned by the State of South Carolina and has been operated by Chem-Nuclear Systems, Inc. since it was licensed in 1971. About 75% by volume of the radioactive waste buried at Barnwell comes from the nuclear power industry. The remaining material is derived from non-fuel cycle activities, such as pharmaceutical industry, hospitals, universities and medical research institutions. There have been no significant problems encountered at the site, since it first opened in 1971.

1.2.4. Sheffield Site

The site is located in northwestern Illinois, about 5 km west to southwest of the town of Sheffield. Burial trenches are excavated primarily in Pleistocene loess and glacial deposits of clay, silt, sand, and gravel, 3 to 15 m thick. These are underlain by consolidated Paleozoic shales. The water table depth ranges from 6 to 15 m. The permeability of the upper sediments is reported to be sufficiently high to prevent accumulation of water in the trenches, except during periods of heavy rainfall and rapid snow melt.

The climate at the site is humid continental with cold winters and warm to hot summers. The average annual precipitation is 900 mm with the majority of the rainfall occurring between April and September, while annual snowfall averages about 750 mm. It is estimated that of the 900 mm of precipitation occurring annually, a small fraction ($\approx 6.5\%$) makes its way to the water table.

The disposal of LLW was initiated at the Sheffield site in 1967. Since 1978, the facility has been closed and no waste has been accepted for disposal. The principal problems encountered at the site include erosion, trench subsidence, and detection of radioactivity in on-site wells.

1.3. Waste Character, Inventory and Burial

1.3.1. Maxey Flats Site

As summarized by Dayal et al. (1984b), the following information has been gathered on the waste inventory of the site and on the character and volumes of wastes buried at the site. A schematic of the Maxey Flats disposal site, showing the location and identification of the various waste trenches, is presented in Figure 1.2.

During 1962-1977, the period of commercial operation, approximately 140,000 m^3 of waste were buried at the site. This waste was comprised of 2,400,000 Ci of by-product material, 431 kg of special nuclear material, and 242,000 kg of source material. By-product material represents materials that become radioactive by neutron activation in reactors; special nuclear material consists of plutonium, U-233, and enriched U-235; and source material corresponds to uranium and thorium, but does not include special nuclear material.

Most of the wastes buried at Maxey Flats were in solid form and consisted of low activity wastes such as paper, trash, clothing, protective apparel, laboratory glassware, obsolete equipment, radiopharmaceuticals, and miscellaneous rubble. Higher activity wastes included solidified liquids, shielding accessories, filter cartridges, ion-exchange resins, and activated metals. Transuranic wastes were generally associated with glove boxes, rubber tubing, gaskets, plastics, paper and rags.

The more hazardous solid wastes were usually contained in 210 L steel drums. The less hazardous wastes were generally packaged in fiberboard boxes or wooden crates. During the early years of operation, burial of container-enclosed liquids and solidified liquid wastes also occurred. The packaged solid wastes were dumped at random in large rectangular trenches separated by 1.5 to 3 m of shale and covered by 1 to 3 m of compacted clay admixed with crushed shale. Many packages were crushed from the impact of dumping or during compaction of the trench cap.

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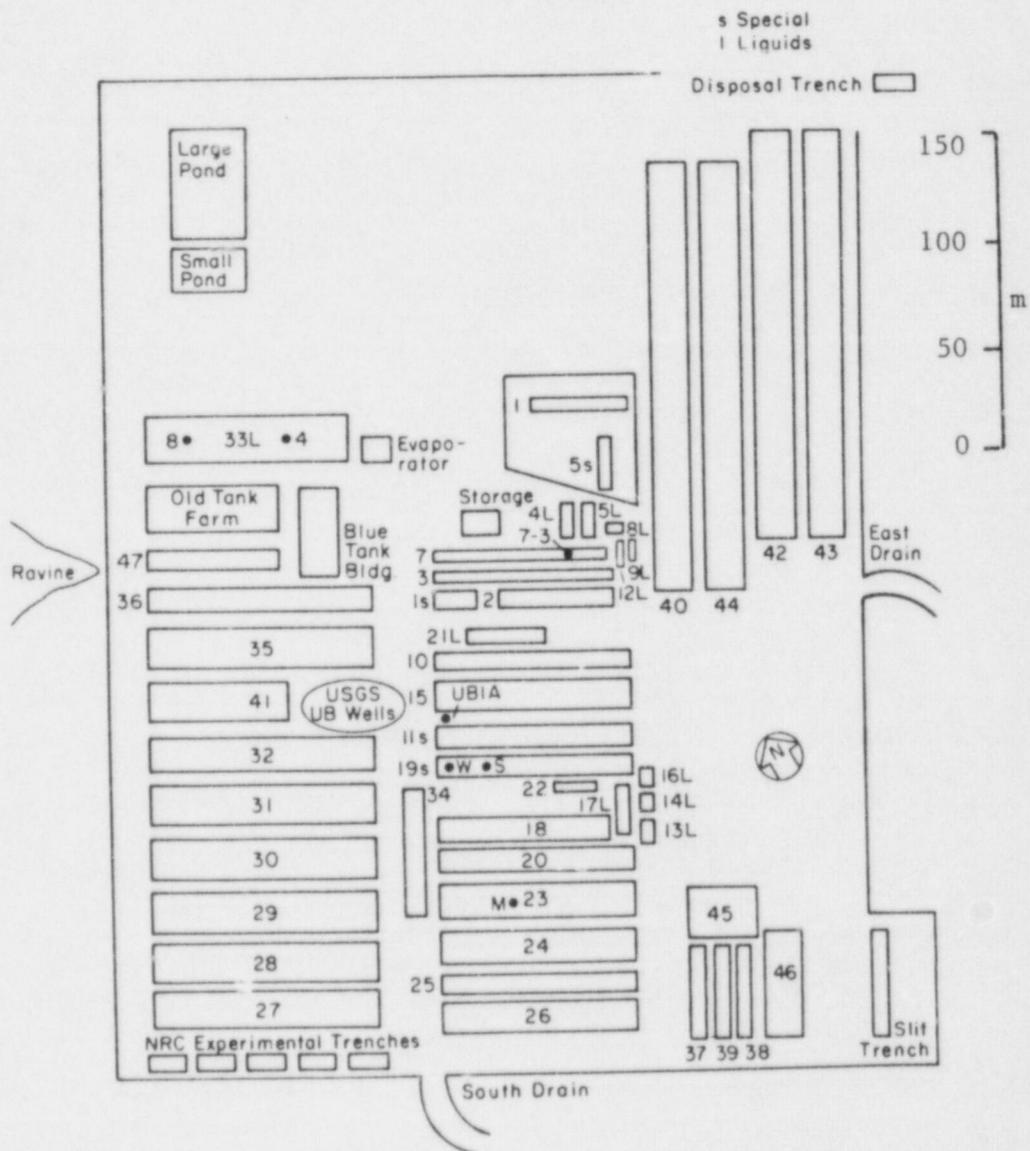
Figure 1.2. Schematic of the Maxey Flats site, showing the location and identification of the disposal trenches. The exact location of the sump standpipes in the trenches is also shown as M in trench 23, W and S in trench 19s, 7-3 in trench 7, and 4 and 8 in trench 33L.

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Most trenches are about 90 m long, 7.5 m wide, and 7.5 m deep. The bottoms are slightly sloping to facilitate collection of infiltrating water accumulated at the bottom of the steel riser pipes. Some trenches have one or more riser pipes for water removal. The riser pipes are identified by a number or letter following the trench number. Trench 34 has four riser pipes, trench 31 has risers in its east and west ends, and trenches 40, 43, and 44 have risers in the north and south ends. The 33L slit tenches have one or more riser pipes.

Zehner (1983) reported that "s" trenches, such as 5s and 19s, were used for burial of "special" types or amounts of wastes having relatively long biological or radioactive half-lives (Sr-90, Pu). Solidified liquid wastes, or liquid holding tanks, were buried in "L" trenches. Most "L" trenches are much smaller than the common trenches. Trench 33L consists of several adjacent slit trenches, ranging in width from 0.6 to 2.5 m. The slit trenches, about 75 m long and 3 m deep, were lined with plastic and filled with liquid waste slurries, which were subsequently stabilized. Cement and urea formaldehyde were used as solidification agents in slit trenches 33L4, 33L11, 33L16 and 33L17, and 33L3 and 33L8, respectively, to stabilize the liquid waste slurries.

Approximate trench dimensions and their closure dates, waste volumes and the amount of radioactivity at the time of burial in each disposal trench are summarized by Dayal et al. (1984b) in an earlier report. The ratio of waste volume to trench volume gives an indication of the magnitude of void space in each trench. In most cases, more than 50% of the trench volume represented void spaces at time of burial. Zehner (1983) reported further that the overall void space in a trench has probably decreased since the time of burial due to compaction of the waste by heavy equipment during burial and trench capping, settling of the waste by itself, microbial degradation, and leaching of waste with time. In addition, container failure at the time of burial probably also contributed to compaction of packaged waste, thus resulting in a net increase in void space. The observed subsidence of trench caps in recent years is a result of trench instability attributable primarily to an increase in void space with time. The occurrence of subsidence holes observed on the trench caps indicates a net decrease in void space in the trenches.

1.3.2. West Valley Site

Clancy et al. (1981) reported that between November 1963 and March 1975, approximately 66,837 m^3 of radioactive waste containing 704,500 Ci of by-product material, 465,394 kg of source material, and 56 kg of special nuclear material (including 4 kg of plutonium) had been disposed at West Valley. These wastes came from offsite medical, educational, research, industrial, pharmaceutical, federal installations, nuclear power plants and (about 20%) from the onsite nuclear fuel reprocessing facility.

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The majority of waste buried at West Valley consisted of paper trash, animal carcasses, evaporator bottoms, filters, filter sludges, protective apparel, residues, plastic, glass and packing material. These materials were packaged in drums, liners, crates, bags and boxes. Through 1972, the predominant radionuclides reported as disposed at West Valley were tritium and C-14 (over 158,000 Ci), Co-60 (about 76,000 Ci), mixed fission products (about 20,843 Ci; presumably dominated by Cs-137 and Sr-90 mixtures), miscellaneous wastes or waste not specifically identified (totaling over 85,000 Ci), and Pu-238 (34,982 Ci).

As reported by Clancy et al. (1981), the disposal site consists of 14 shallow land burial trenches (Fig. 1.3). Seven trenches were constructed in the northern area and the other seven in the southern area of the site. The trenches on the average are relatively long and narrow, measuring about 180 to 240 m in length, about 10 m in width, and about 6 m in depth. Trench 6 is not a true trench but rather a series of individual bore holes for waste with high external exposure rates. Trench 7 is actually a narrow and shallow concrete vault.

Drains were installed in the trench floors leading to sumps into which standpipes had been emplaced. From 1963 until 1968, leachate was periodically pumped out of the trenches and discharged after considerable dilution, into an adjacent stream. A lagoon was constructed adjacent to the southern area trenches to hold rainwater which was pumped from open trenches. This lagoon and the two lagoons in the northern area were connected by pipeline to the LLW treatment facility located in the nearby fuel reprocessing plant (Clancy et al., 1981).

Most of the waste delivered to the disposal facility and disposed was packaged in 55-gallon steel drums. Wastes having a radiation level at the container surface in excess of 200 mR/hr were required to be solidified in concrete. Many of the waste containers were placed (rolled or dropped) manually into the trenches. Heavy containers and packages with high external radiation levels were placed into the trench with the aid of a large capacity crane. Many of the 55-gallon drums were stacked in place.

1.3.3. Barnwell Site

Waste disposal was initiated at the Barnwell site in 1971. Clancy et al. (1981) reported that, during the first year of operation, approximately 12,405 m^3 of waste containing 4200 Ci of byproduct material was accepted and disposed. The disposal rate rose steadily over the next nine years to the point where the annual rate reached 63,862 m^3 in 1979. Due to restrictions in the annual volume of waste received, however, the disposal rate in 1980 was reduced to about 53,800 m^3. Through 1980, over 323,560 m^3 of waste containing 1,665,100 Ci of byproduct material radioactivity has been disposed. In addition, 2,567,000 kg of source material and 1,120 kg of special nuclear
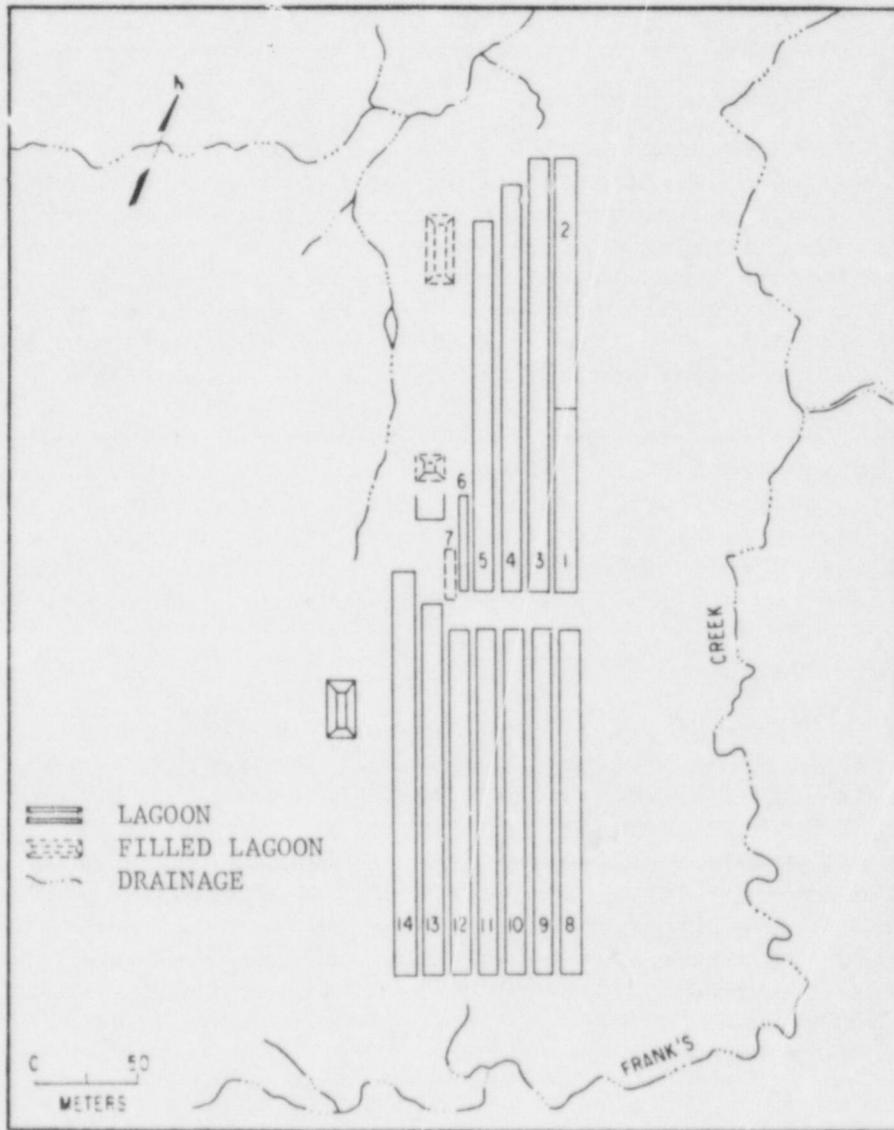
Figure 1.3. Schematic of the West Valley site, showing the location and identification of the disposal trenches.

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material have been disposed through 1980. Transuranic-contaminated waste in concentrations exceeding 10 nCi/gm have never been accepted for disposal at the site.

Two types of disposal trenches have been employed at the Barnwell facility: slit trenches and "standard" shallow land burial trenches. Each of the two slit trenches that have been constructed at the site measure about 75 to 150 m long, 1 m wide, and 6 m deep. A map of the disposal site is shown in Figure 1.4.

According to Clancy et al. (1981), the slit trenches have been used in the past for disposal of waste material having high surface radiation levels such as non-fuel bearing reactor core components (poison curtains, control rods, and other miscellaneous core hardware), with the intention of reducing occupational exposures. Most of the waste received at the site has been disposed in the "standard" trenches. These trenches were initially relatively small but more recent disposal trenches are larger, typically measuring 300 m long by 30 m wide. Somewhat smaller trenches are also occasionally used.

In practice, waste emplacement is a combination of stacked and random disposal. Boxes and ion exchange liners are typically stacked while drums and other small waste packages are typically dumped into the spaces alongside the stacked waste. After waste emplacement, the trench is backfilled with a sandy soil and compacted. The trench covers are graded to promote drainage, top soil is added, and the surface is seeded with grass (Clancy et al., 1981).

1.3.4. Sheffield Site

According to Clancy et al. (1981), between 1967 and April 1978, approximately 90,500 m^3 of low-level solid waste containing over 60,200 Ci of byproduct material was disposed at the site. The quantities of source material, special nuclear material, and plutonium disposed at the site were 271,790 kg, 60 kg, and 13.4 kg, respectively. The State license at the Sheffield site generally limited the concentration of disposed radioactive material to 35 Ci/m^3, although some exceptions were made on a case by case basis. Liquids were occasionally received on site and were solidified in urea-formaldehyde or cement. Disposal of plutonium waste at the site was discontinued in 1975.
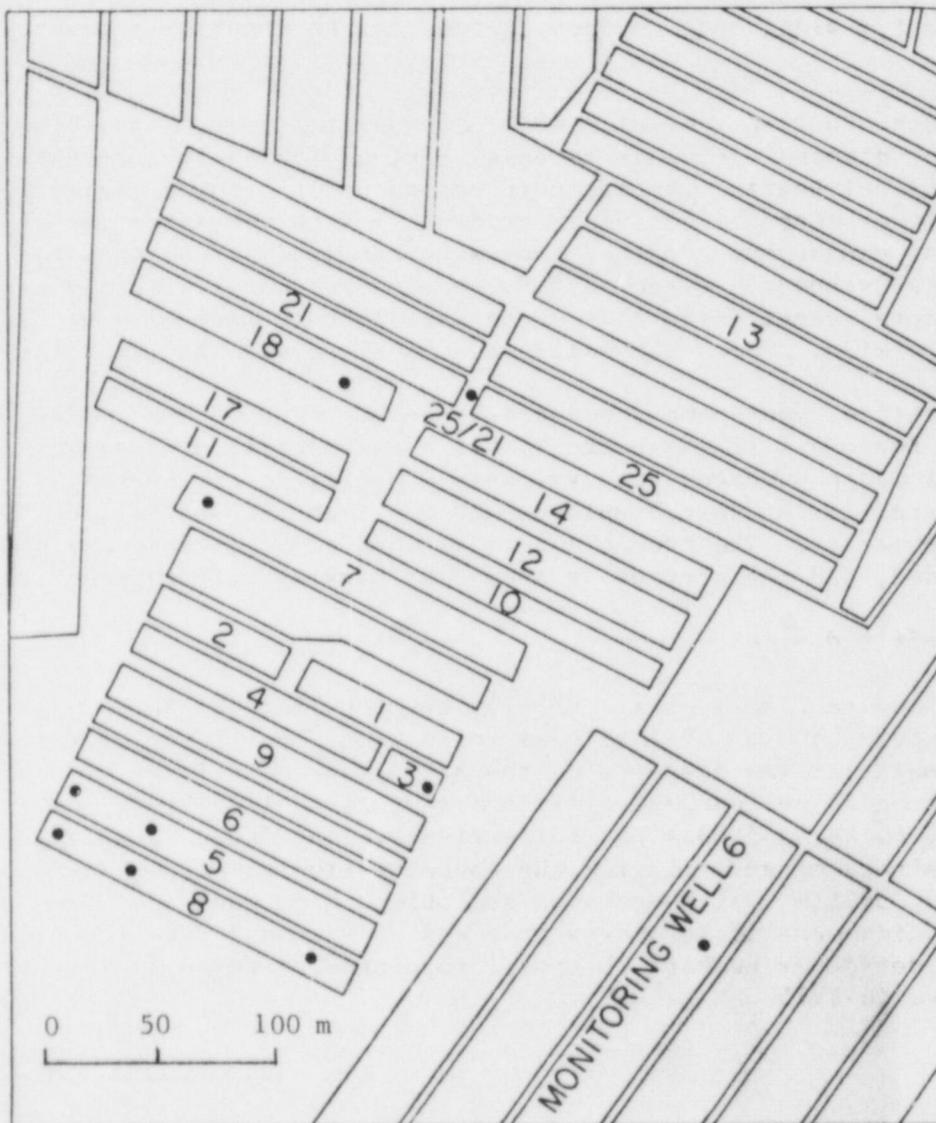
Figure 1.4. Schematic of a portion of the Barnwell site, showing the location and identification of the disposal trenches.

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The disposal trenches at the Sheffield site have dimensions generally ranging from 60 to 150 m in length, 10 to 25 m in width, and 6 to 12 m in depth (Fig. 1.5). With the exception of several slit trenches, the trenches have been excavated roughly parallel to one another with about 3 m spacing separating the trench side walls. All trench tops are above the probable maximum flood elevation and the trench bottoms (with the exception of trench 18) are above the maximum groundwater elevation. The bottoms of the trenches were sloped toward one end and are equipped with French drains that lead to sumps and riser pipes for sampling. Each trench has been capped and mounded for surface drainage (Clancy et al., 1981).
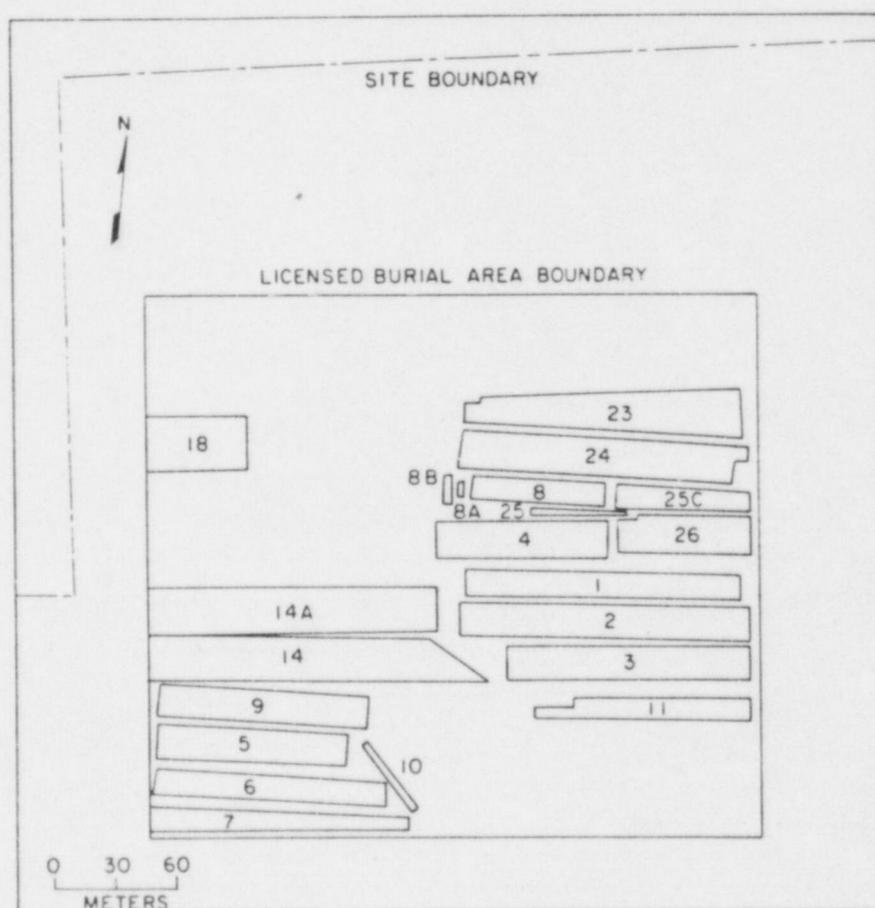
Figure 1.5. Schematic of the Sheffield site, showing the location and identification of the disposal trenches.

2. WATER SAMPLING AND ANALYSIS

The following is a summary of the field sampling and laboratory procedures that were developed at BNL and employed for the collection, storage and analysis of trench and well water samples from the LLW disposal sites. Further details of these procedures can be found in Pietrzak et al. (1982), Czyscinski and Weiss (1981), and Weiss and Colombo (1980).

2.1. Sampling Methods and Analytical Procedures

Procedures were developed to maintain the anoxic character of the trench waters during sampling. A schematic diagram of the anoxic water sampling system is shown in Fig. 2.1. After collection, the trench water samples were stored in 4-liter borosilicate glass bottles designed to maintain the trench redox conditions. The anoxic collection procedure allows samples to be stored for months without oxidation, which results in a reddish-brown ferric hydroxide precipitate. A scheme for sequential processing of trench water samples collected in the field and subsequent analysis for the various dissolved and suspended constituents is shown in Fig. 2.2.
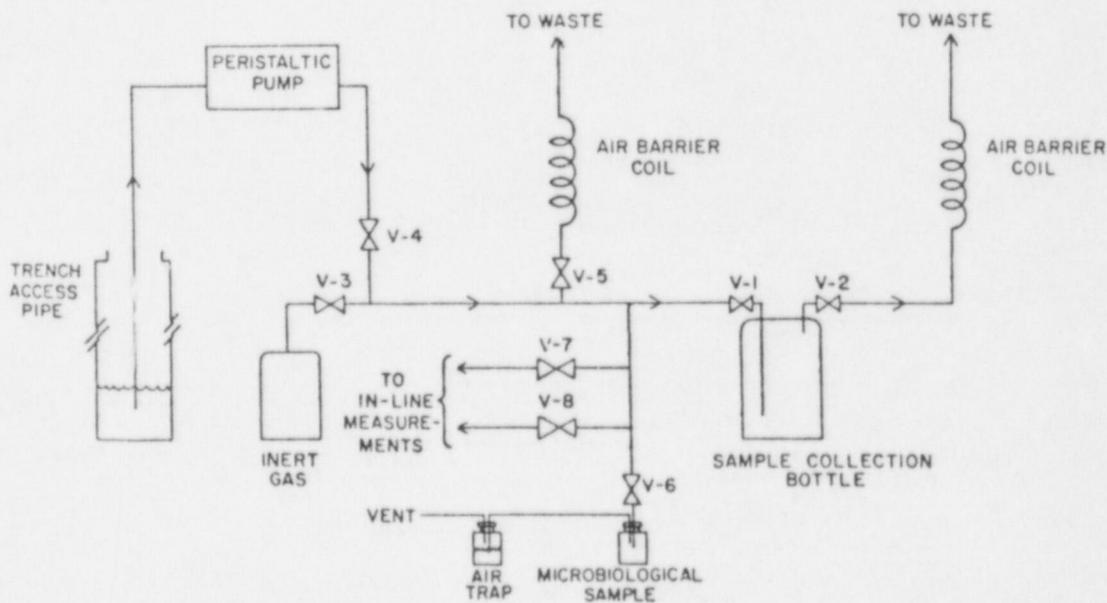
Figure 2.1.   Schematic of the anoxic water sampling system.

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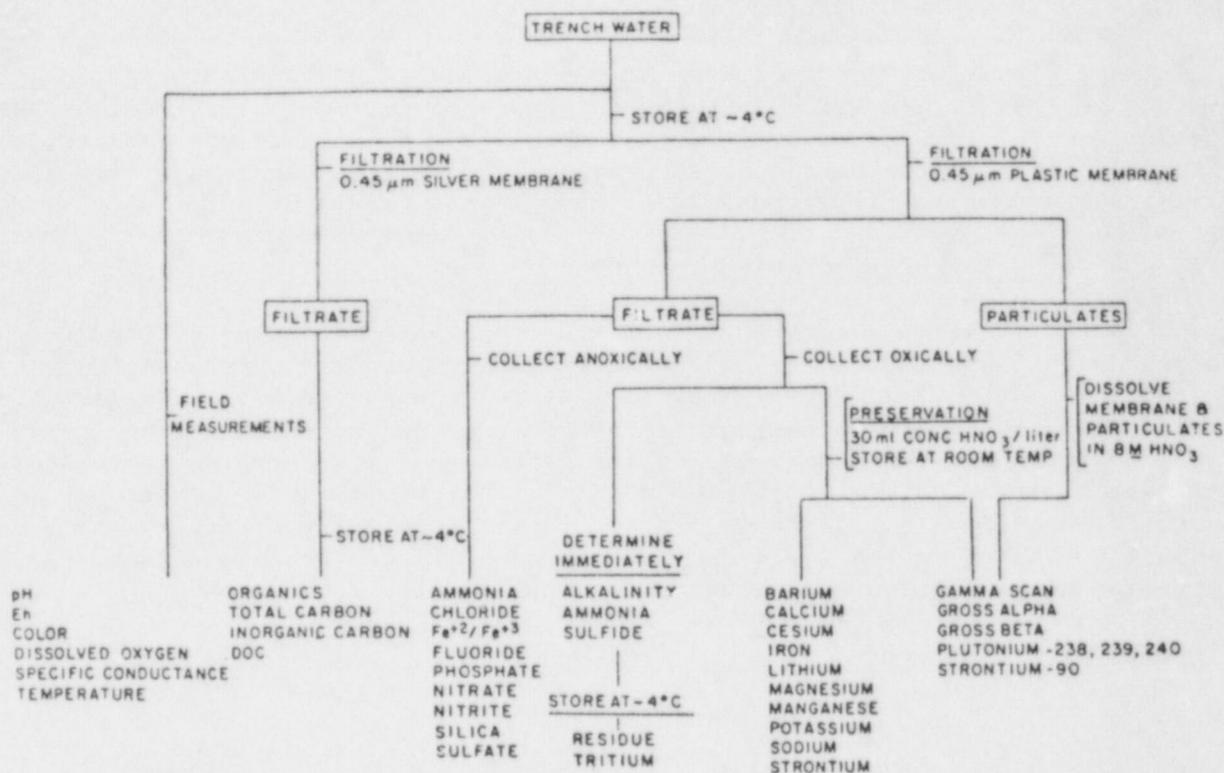
Figure 2.2. Scheme for sequential processing of water samples for analysis.

2.1.1. In-Line Field Measurements

Physico-chemical characteristics of trench water which are subject to change during storage were measured at the time of sample collection. The temperature, pH, redox potential (Eh), dissolved oxygen and sulphide, and specific conductance of the trench water samples were measured in-line while the water sample was being collected. A schematic diagram of the in-line field measurement system is shown in Fig. 2.3.

2.1.2. Anoxic Filtration

Upon arrival at BNL, the trench water samples were filtered through 0.45 μm membrane filters to remove suspended particulate matter. The filtration was also conducted under an inert atmosphere to maintain the anoxic character of the waters. The filtrate was divided into several fractions and processed according to the scheme shown in Fig. 2.2. The dissolved fraction of each sample was analyzed for inorganic, organic, and radiochemical constituents. The particulate fraction was analyzed for radiochemical constituents only. A schematic diagram of the anoxic filtration system is shown in Fig. 2.4.

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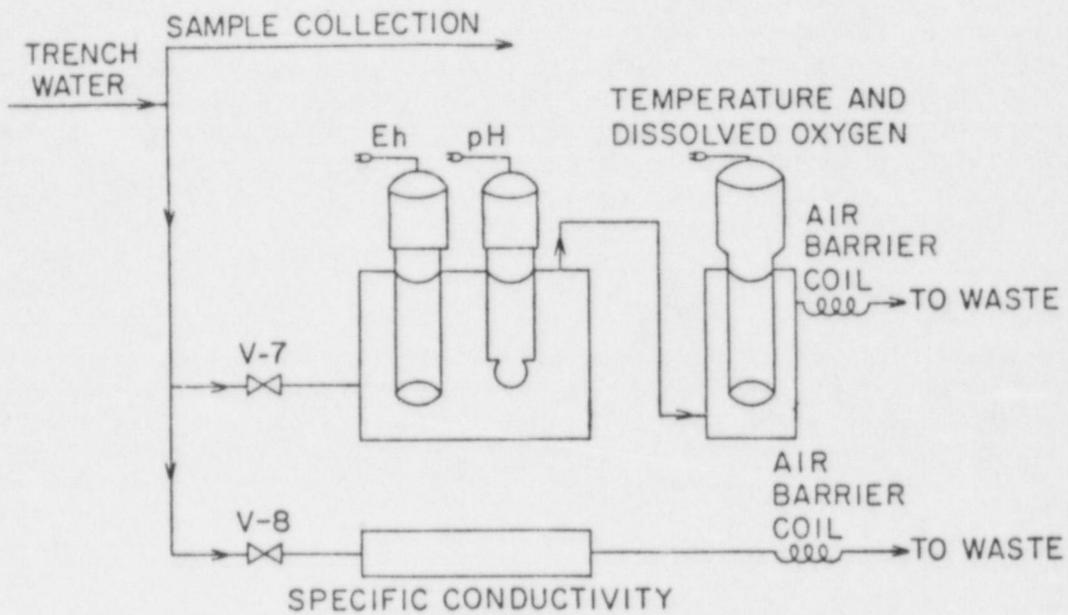
Figure 2.3. Schematic of the in-line field measurement system.
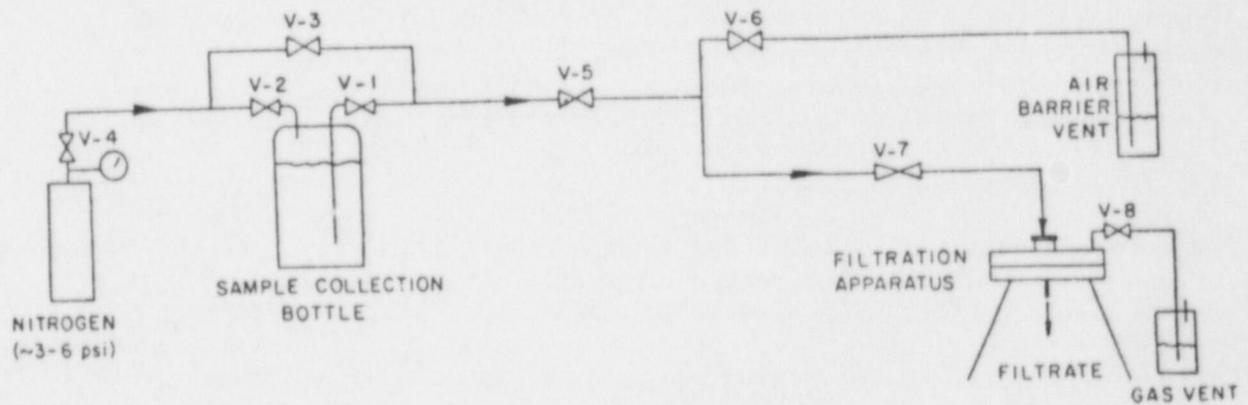
Figure 2.4. Schematic of the anoxic filtration system.

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2.1.3. Inorganic Constituents

The following is a brief summary of methods used to measure the dissolved chemical constituents in trench waters sampled prior to 1981. Trench waters collected during the 1981 sampling trip and subsequent trips were analyzed for inorganic constituents at BNL using ion chromatographic methods. Specifically, the anions chloride, sulphate, nitrate, phosphate, fluoride, and bromide were analyzed on a Dionex Model 10 ion chromatograph using an anion column with a 0.002 M sodium bicarbonate, 0.002 M sodium carbonate buffering solution. Ions were detected by changes in the electrical conductivity of the buffering solution.

2.1.3.1. Alkalinity

The alkalinity titration was performed on filtered trench water, immediately after filtration. Generally carbonate and bicarbonate concentrations in groundwater are determined by titrating the water sample with a standard acid to pH 8.3 and pH 4.5, respectively. The equations that govern the reactions are

$$CO_3^{2-} + H^+ \rightleftarrows HCO_3^- \tag{2.1}$$

$$HCO_3^- + H^+ \rightleftarrows H_2O + CO_2 \tag{2.2}$$

The end points of these titrations are usually sharp in the absence of other ions that are in competition for the hydrogen ion. In the presence of salts of weak organic and inorganic acids the titration curve does not give a sharp endpoint due to buffering of the system. The titration alkalinity in this case is not a simple function of the carbonate-bicarbonate concentrations. However, for the sake of simplicity, we have assumed the titration alkalinity to be equal to carbonate alkalinity. The shape of an acid-base titration curve can yield useful information in addition to carbonate and bicarbonate relationships.

2.1.3.2. Chloride

Thiocyanate ion is liberated from mercuric thiocyanate by the formation of non-ionic but soluble mercuric chloride. In the presence of ferric ion, the liberated thiocyanate forms a highly colored ferric thiocyanate whose concentration is proportional to the original chloride concentration. The intensity of the ferric thiocyanate was measured spectrophotometrically.

2.1.3.3. Dissolved Metals

Dissolved metals in the acidified aliquots of filtered trench waters were determined by atomic absorption spectroscopy.

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2.1.3.4. Ferrous-Ferric Iron

Iron in the ferrous state was reacted with 1,10-phenanthroline between pH 3.2 and 3.3. The orange-red ferrous-phenanthroline complex was measured spectrophotometrically. Ferric iron was determined by subtracting the ferrous iron concentration from the total iron concentration. Total iron was determined colorimetrically by reduction with hydroxylamine hydrochloride and subsequent reaction with 1,10-phenanthroline. Atomic absorption determination of total iron was a convenient check on this method.

2.1.3.5. Fluoride

Fluoride was determined with a specific ion electrode consisting of a single-crystal lanthanum fluoride membrane and an internal reference. The crystal is an ionic conductor in which only fluoride ions are mobile. The potential developed across the membrane in the Orion 94-09 probe is proportional to the fluoride ions in solution and was measured against an external reference with the Orion 407A specific ion meter.

2.1.3.6. Phosphate

The determination of orthophosphate involved the formation of molybdophosphoric acid with the addition of ammonium molybdate in acid medium. This was reduced to molybdenum blue by ascorbic acid and measured spectrophotometrically.

2.1.3.7. Nitrogen-Ammonia

A gas sensing electrode, containing a hydrophobic gas permeable membrane, allows dissolved ammonia generated in the sample to diffuse through the membrane until the partial pressure of ammonia is the same on both sides. The partial pressure of ammonia is proportional to its concentration according to Henry's law. When sodium peroxide and sodium hypochlorite are added to a solution containing an ammonium salt, the Berthelot reaction takes place with the formation of a green compound related to endophenol. The color intensity was measured spectrophotometrically which is proportional to the ammonia concentration. Methods have been developed at BNL to perform this analysis in an inert nitrogen or argon environment.

2.1.3.8. Nitrogen-Nitrate

Nitrite reacts with sulphanilamide under acidic conditions to yield a diazo compound which couples with N-1-napthylethylene-diamine dihydrochloride to form a red colored compound. The color intensity measured spectrophotometrically was related to the concentration of nitrite.

2.1.3.9. Nitrogen-Nitrite plus Nitrate

Nitrate was reduced to nitrite when passed through a cadmium-copper column. The nitrite was then treated with sulphanilamide and N-1-naphthylenediamine dihydrochloride to form the red azo dye as above. When both nitrite and nitrate were present in a sample, the nitrate content was obtained by subtracting the nitrite value from the combined nitrite plus nitrate value.

2.1.3.10. Silica

Ammonium molybdate reacts with silicic acid (H_4SiO_4) in acid medium to form molybdosilicic acid. This was reduced to molybdenum blue by ascorbic acid and measured spectrophotometrically. Oxalic acid was added before the ascorbic acid to eliminate interference from phosphates.

2.1.3.11. Sulphate

When equimolar barium chloride and methylthymol blue are added to a sulphate-containing sample at pH 2.5-3.0, barium sulphate precipitates. After adjusting the pH to 12.5-13.0, the barium remaining in solution reacts with the methylthymol blue to form a chelate. The uncombined methylthymol blue remaining in solution was measured spectrophotometrically and was proportional to the sulphate initially present in the sample.

2.1.3.12. Sulphide

A silver-silver sulphide electrode used in conjunction with a reference electrode develops a potential which is a direct function of the logarithm of the activity of the sulphide ion. The sulphide electrode was calibrated using the method developed by Berner (1963).

2.1.4. Organic Constituents

2.1.4.1. Dissolved Carbon

The total dissolved carbon and inorganic carbon contents were determined using a Beckman Model 915 Total Carbon Analyzer. The difference between the total dissolved carbon and the inorganic carbon measurements is the dissolved organic carbon (DOC) content of the sample.

2.1.4.2. Organic Carbon Compounds

A liquid extraction technique using methylene chloride was employed to isolate acidic, neutral, and basic hydrophobic organic compounds from the trench waters. These fractions were analyzed by gas chromatography and identified by mass spectroscopy (GC/MS) methods.

Hydrophilic compounds were isolated at PNL using BF_3 in methanol to prepare methylated derivatives which were subsequently analyzed by gas chromatography and identified by mass spectroscopy methods (GC/MS).

2.1.5. Radiochemical Constituents

Radiochemical measurements were performed on the filtrate and particulates collected following filtration of each trench water sample. Except for tritium, all of the radiochemical measurements were made on an acidified aliquot of the filtrate. Tritium was measured on a non-acidified fraction of filtrate. The following is a brief summary of the procedures used to measure radionuclides in trench and well waters.

2.1.5.1. Gross Alpha/Gross Beta

An aliquot of acidified filtrate was heated to dryness in a 50 mm planchette and counted with a Canberra model 2200 low-level alpha/beta gas flow proportional counter. Each sample was counted at two instrument settings corresponding to the plutonium-239 alpha plateau and the strontium-90/yttrium-90 beta plateau. This measurement was not made for the particulate fraction. Gross counting should be regarded as a rapid, semi-quantitative measure of sample activity.

2.1.5.2. Tritium

An aliquot of tritiated water, distilled by conventional methods at atmospheric pressure, was emulsified with Packard Insta-Gel liquid scintillation reagent and counted in a Searle Analytic-92 liquid scintillation counter.

2.1.5.3. Strontium-90

Strontium-90 was determined by radiochemically separating strontium from the trench water sample and counting the in-growth of yttrium-90 with a low-level beta counter. These determinations were made by LFE Laboratories, Richmond, California.

2.1.5.4. Plutonium-238,239,240

Plutonium isotopes were radiochemically separated from other alpha emitting radionuclides by anion exchange chromatography and electroplated onto a counting disc. Plutonium isotopes on the disc were measured by alpha spectroscopy with a surface barrier silicon detector. The Pu isotopes were analysed by LFE Laboratories, Richmond, California.

2.1.5.5. Gamma-Ray Emitters

Gamma-ray emitting radionuclides, such as Am-241, Cs-137, Cs-134, Co-60, Na-22, and Mn-54, were measured by counting aliquots of the filtered trench water with a 2 keV resolution Ge(Li) detector. Radionuclide identifications were made on the basis of measured gamma ray energies and relative peak heights of nuclides emitting more than one gamma.

3. RESULTS

3.1. Selection of Leachate Data

As reported earlier, several sampling trips were undertaken by BNL to the waste disposal sites during the period 1976-1982 to collect trench and well water samples. In many instances, the same trench was sampled multiply during subsequent sampling trips.

During preliminary sampling trips to the study areas, adequate precautions were not taken to prevent air from coming in contact with water samples during collection. Consequently, the water samples do not represent the actual leachates in the trenches. We consider these data suspect and have not discussed them further in this report.

Subsequent leachate samplings involved use of anoxic sampling procedures. In addition, the leachate analyses were relatively complete in that most of the samples were analyzed for major ion constituents, radionuclides, and in many cases, for organic constituents.

An ion balance of the major cations and anions present in each leachate sample was conducted to determine completeness of the trench water analyses. Equivalents of cation and anion sums were compared to calculate the ion balance error. In several cases, the leachate samples exhibited appreciable error in ion balance. As a result, these data have been excluded from further consideration. Based on a relatively small ion balance error, we have compiled in Table 3.1 a list of trench water samples that will be considered further in the interpretation of trench leachate compositions. It should be noted, however, that for the interpretation of radionuclide and organic data all leachate samples have been considered (including the 1977 survey samplings), since these constituents are present in trace amounts and do not contribute significantly to bulk composition of the trench leachates.

3.2. Selection of Well Water Data

Both on- and off-site wells were sampled several times at the disposal sites by BNL to obtain baseline information on ambient groundwater characteristics.

At the Maxey Flats site, an on-site well UB1A was sampled multiply during the study period. Analyses of the well water samples revealed the presence of contaminants such as radionuclides and dissolved organic compounds, presumably derived from neighboring trenches. Consequently, the composition of an off-site well UA3, which is the shallowest of UA series wells (Zehner, 1983), was selected for baseline information. The water in this well is derived from rock units corresponding to lower part of Nancy Member and upper part of Farmers Member. According to Zehner (pers. comm.), well water UA3 may be used as a substitute for UB1A to represent ambient groundwater composition. The average major ion composition of UA3 well water sampled in 10/78, 6/79, and

Table 3.1. Selected trench leachates considered in this study.

Trench	Sampling Date	Leachate Designation
Maxey Flats Site		
2	September 1976	2(76)
7	September 1976	7(76)
7	October 1981	7(81)
18	September 1976	18(76)
19s	September 1976	19s(76)
19s	October 1981	19s(81)
23	October 1981	23(81)
26	September 1976	26(76)
27	September 1976	27(76)
27	May 1978	27(78)
27	October 1979	27(79)
27	October 1981	27(81)
30	October 1979	30(79)
32	September 1976	32(76)
33L4	May 1978	33L4(78)
33L4	October 1981	33L4(81)
33L8	October 1981	33L8(81)
35	October 1981	35(81)
37	September 1976	37(76)
West Valley Site		
2	November 1977	2(77)
3	October 1978	3(78)
4	November 1977	4(77)
5	October 1978	5(78)
9	October 1978	9(78)
Barnwell Site		
6	May 1980	6(80)
8	March 1979	8(79)
8	May 1980	8(80)
25/21	March 1979	25/21(79)
25/21	May 1980	25/21(80)
Sheffield Site		
14A	April 1979	14A(79)
18	April 1979	18(79)
18	June 1982	18(82)

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10/79 by USGS personnel are reported by Zehner (1983). In our interpretation of Maxey Flats leachate data, we have used the average composition of UA3 to represent the ambient groundwater composition.

At the West Valley site, three on-site wells and seven off-site streams, located in the vicinity of the burial site, were sampled by BNL during the survey study. However, most of these water samples were found to be contaminated with tritium, presumably derived from the neighboring trenches. As a result, no further samplings were conducted at these locations during subsequent field trips. For interpretation of West Valley trench leachate data, we have considered West Valley spring water composition, as reported by the USGS (Cartwright and Ziarno, 1980), to be representative of the ambient groundwater.

Although several on-site wells were sampled at the Barnwell site (Weiss and Colombo, 1980), monitoring well #6 was considered to be the most typical in terms of ambient groundwater characteristics.

At the Sheffield site, a series of on-site wells were sampled by BNL during the survey study (Weiss and Colombo, 1980). However, more than half of the well waters sampled were found to be contaminated with radionuclides and organic carbon. Well water #525, which was sampled multiply and contained no significant levels of contaminants, was selected for the interpretation of Sheffield trench leachate data, relative to ambient groundwater composition.

For the purposes of this study, the well waters selected to represent the ambient groundwaters at the Maxey Flats, West Valley, Barnwell, and Sheffield waste disposal sites are summarized in Table 3.2.

Table 3.2. Selected well waters considered in this study.

Disposal Site	Well Water	Groundwater Designation
Maxey Flats	UA3	GW(MF)
West Valley	West Valley Spring	GW(WV)
Barnwell	#6	GW(BW)
Sheffield	#525	GW(SH)

3.3 Characteristics of Trench Leachates and Well Waters

Field data on specific conductance, dissolved oxygen, Eh, pH, sulphide and temperature for selected trench leachates and ambient groundwaters are presented in Table 3.3. Also included in the Table are the values for dissolved organic carbon and inorganic carbon, total dissolved solids and ionic strength for each water sample. The values for total dissolved solids and ionic strength represent calculated values based on WATEQF geochemical code.

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Table 3.3. Characteristics of trench leachates and groundwaters from LLW disposal sites.

Sample	Temperature (°C)	pH	Eh (mV)[a]	Dissolved Oxygen (ppm)	Specific Conductance (μMho/cm)	Ionic Strength ($\times 10^{-2}$)[b]	Total Dissolved Solids (ppm)[b]	DOC (ppm)[c]	DIC (ppm)[c]
Maxey Flats Site									
2(76)	20.0	6.7	d	d	3400	6.0	4460	210	270
7(76)	22.5	6.9	d	d	2530	3.5	2220	250	210
7(81)	16.5	7.4	−44	0.10	12000	14	8200	730	220
18(76)	21.5	7.0	d	d	3450	5.1	3660	500	440
19s(76)	21.0	6.6	d	d	2340	3.3	1850	620	80
19s(81)	15.0	6.5	−28	0.10	2100	3.5	2080	430	170
23(81)	17.0	7.5	−39	0.05	4800	7.2	5000	780	520
26(76)	21.0	6.8	d	d	2910	3.6	2430	950	150
27(Av)[e]	18.5	6.2	79	0.10	13120	20	8420	920	25
27(81)	16.0	6.8	17	<0.05	6000	10	4540	490	57
30(79)	16.0	6.5	140	0.10	6900	17	10580	260	1500
32(76)	20.0	7.3	d	d	5750	6.9	4950	790	510
33L4(78)	12.0	12.1	−7	4.1	5580	3.2	3070	1100	10
33L4(81)	17.0	12.0	−54	0.05	6400	4.2	4190	1500	<2
33L8(81)	18.0	6.0	−135	0.25	2000	2.9	1800	160	330
35(81)	17.0	8.2	−14	0.10	3400	6.4	4180	540	390
37(76)	20.0	5.1	d	d	6900	19	11250	3280	20
GW(MF)	d	6.9	d	d	d	7.2	3670	6[f]	11[f]
West Valley Site									
2(77)	11.7	7.7	47	d	6700	8.7	6120	200	670
3(78)	10.5	7.3	−3	0.1	7600	9.5	5590	1700	95
4(77)	13.5	7.2	210	d	8100	1.1	6830	350	130
5(78)	10.5	6.7	40	0.2	6750	10.7	5970	2900	10
9(78)	13.5	6.7	18	0.2	3400	4.9	3030	1700	120
GW(WV)	d	7.8	d	d	d	0.43	240	d	d
Barnwell Site									
6(80)	15.3	6.1	350	4	260	0.37	240	7	8
8(79)	19.0	6.6	308	2	1400	1.5	1120	170	130
8(80)	16.0	7.4	130	0.3	2600	3.11	2220	203	258
25/21(79)	18.5	5.9	538	1	550	0.62	390	12	38
25/21(80)	13.3	6.2	100	0.2	190	0.37	230	d	18
GW(BW)	18.0	6.8	d	0.1	35	0.18	13	6	6
Sheffield Site									
14A(79)	8.5	5.0	143	0.3	600	0.93	500	100	40
18(79)	10.0	6.8	181	0.1	1600	3.0	1700	50	190
18(82)	10.5	7.1	256	<0.1	2000	3.4	1900	57	275
GW(SH)	10.0	7.5	2	2.2	510	1.1	620	3	70

[a]Field measurements of Eh are reported relative to the Standard Hydrogen Electrode (SHE).
[b]Ionic strength and total dissolved solids values are based on WATEQF calculations.
[c]Represent dissolved organic carbon and dissolved inorganic carbon, respectively.
[d]Not determined.
[e]Average values based on leachate samplings conducted in September 1976, May 1978, and October 1979 (Dayal et al., 1984b).
[f]These species were not measured in well UA3 water. The listed values are means for the species in well UB1A water samples.

The data show that, relative to ambient groundwaters, most trench leach-ates exhibit high specific conductance, ionic strength, total dissolved solids and dissolved organic and inorganic carbon. In addition, the leachates are generally depleted in dissolved oxygen, with most Maxey Flats and West Valley leachates exhibiting strong chemically reducing conditions as indicated by negative redox potentials. In contrast, the Barnwell and Sheffield leachates are relatively mildly reducing.

3.4. Major Ion Composition

The major ion compositions of the trench leachates and ambient ground-waters are presented in Table 3.4. The data show that in a majority of the trench leachates, Na^+, NH_4^+, dissolved iron and manganese, Ca^{2+}, Mg^{2+}, Cl^-, and alkalinity are generally enriched, relative to ambient groundwaters. How-ever, the degree of enrichment is highly variable from one trench to another at a given site or from one site to another. In addition, the Maxey Flats and West Valley leachates exhibit significant depletion in SO_4^{2-} relative to ambient groundwater. Nitrate and nitrite concentrations are generally very low to below detection in both the trench leachates and ambient groundwaters. However, some Maxey Flats leachates show elevated concentrations of these con-stituents.

3.5. Major Water Types

In Figures 3.1-3.4, the cation and anion equivalents are displayed on Piper trilinear diagrams to graphically represent the dissolved constituents in trench leachates and to help classify distinct water types. For the sake of comparison, the composition of ambient groundwaters are also projected on the diamond shaped field to show their distinct chemical identity.

In terms of both anion and cation contents, the leachate compositions can be classified, as shown in Table 3.5, into several groupings of specific water types.

3.6. Dissolved Radionuclides

Table 3.6 presents radionuclide data (data corrected to October, 1981), representing average radionuclide concentrations based on BNL samplings at the Maxey Flats, West Valley, Barnwell, and Sheffield disposal sites during the period 1976-1982 including the 1977 survey samplings. These radionuclide data are displayed in Figures 3.5-3.8, where the error bars reflect the extent of variation observed due to multiple leachate samplings in a given trench.

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Table 3.4. Major ion compositions of trench leachates and groundwaters from LLW disposal sites.

Sample	Na^+	K^+	NH_4^+ (as N)	Fe_T [a]	Mn_T [a]	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	NO_3+No_2 (as N)	Alkalinity (as $CaCO_3$)
Maxey Flats Site											
2(76)	700	66	b	40	0.8	29	79	310	11	<0.1	1560
7(76)	240	140	b	61	0.5	130	73	220	<5	<0.1	1090
7(81)	2140	329	75	17	1.9	107	193	2500	1320	9	1150
18(76)	540	50	b	33	<0.1	14	160	310	18	<0.1	2050
19s(76)	100	25	b	150	0.8	58	130	150	<5	<0.1	980
19s(81)	231	27	45	65	0.5	49	128	231	<2	0.5	1040
23(81)	825	77	100	7	<0.1	11	230	575	57	28	2420
26(76)	240	39	b	65	0.7	31	130	290	<5	<0.1	1320
27(Av)[c]	630	85	70	1250	116	530	468	4733	24	0.8	354
27(81)	554	87	116	165	1.7	220	350	2340	<2	17	312
30(79)	1000	43	50	10	0.3	32	1300	200	85	0.1	6400
32(76)	700	210	b	16	1.2	75	230	370	11	<0.1	2720
33L4(78)	180	30	18	0.3	<0.1	650	<0.1	168	<5	11	1600
33L4(81)	180	102	26	0.2	<0.1	864	<0.2	361	<2	10	2120
33L8(81)	50	11	50	43	1.8	190	49	37	15	2	1080
35(81)	614	51	37	0.9	0.3	26	330	235	<2	0.7	2310
37(76)	680	20	b	1100	42	250	730	180	8000	13	125
GW(MF)	300	24	<1	<0.1	<0.1	220	380	70	2100	0.2	459
West Valley Site											
2(77)	900	330	230	13	<0.1	72	220	470	<5	<0.1	3120
3(78)	1000	320	300	56	0.3	150	180	1300	26	2	1730
4(77)	970	330	68	82	0.5	180	160	2100	<5	<0.1	1800
5(78)	690	270	180	540	2.3	300	200	820	<5	1	2300
9(78)	430	91	84	57	0.2	130	150	82	<5	<0.1	1000
GW(WV)	11	1	<1	10	<0.01	42	5	12	17	0.2	167
Barnwell Site											
6(80)	28	3	4	<1	0.7	14	1	13	45	4	86
8(79)	87	12	59	1.2	0-7	34	18	85	34	8	600
8(80)	120	18	205	24	0.9	82	40	47	7	<0.1	1340
25/21(79)	37	4	25	0.2	0.3	21	3	42	56	15	80
25/21(80)	11	1	35	6	0.6	10	3	12	<5	<0.1	104
GW(BW)	15	12	2	<1	<0.1	4	0.2	3	17	<0.1	61
Sheffield Site											
14A(79)	50	13	5	11	1.6	52	17	20	78	0.1	200
18(79)	67	72	9	0.4	1.1	190	94	28	190	0.4	850
18(82)	76	73	9	<0.1	b	219	126	27	199	b	1060
GW(SH)	35	1	<0.1	10	0.2	74	34	13	47	<0.1	320

[a]Represents total dissolved iron and manganese.
[b]Not determined.
[c]Average values based on leachate samplings conducted in September 1976, May 1978, and October 1979 (Dayal et al., 1984b).
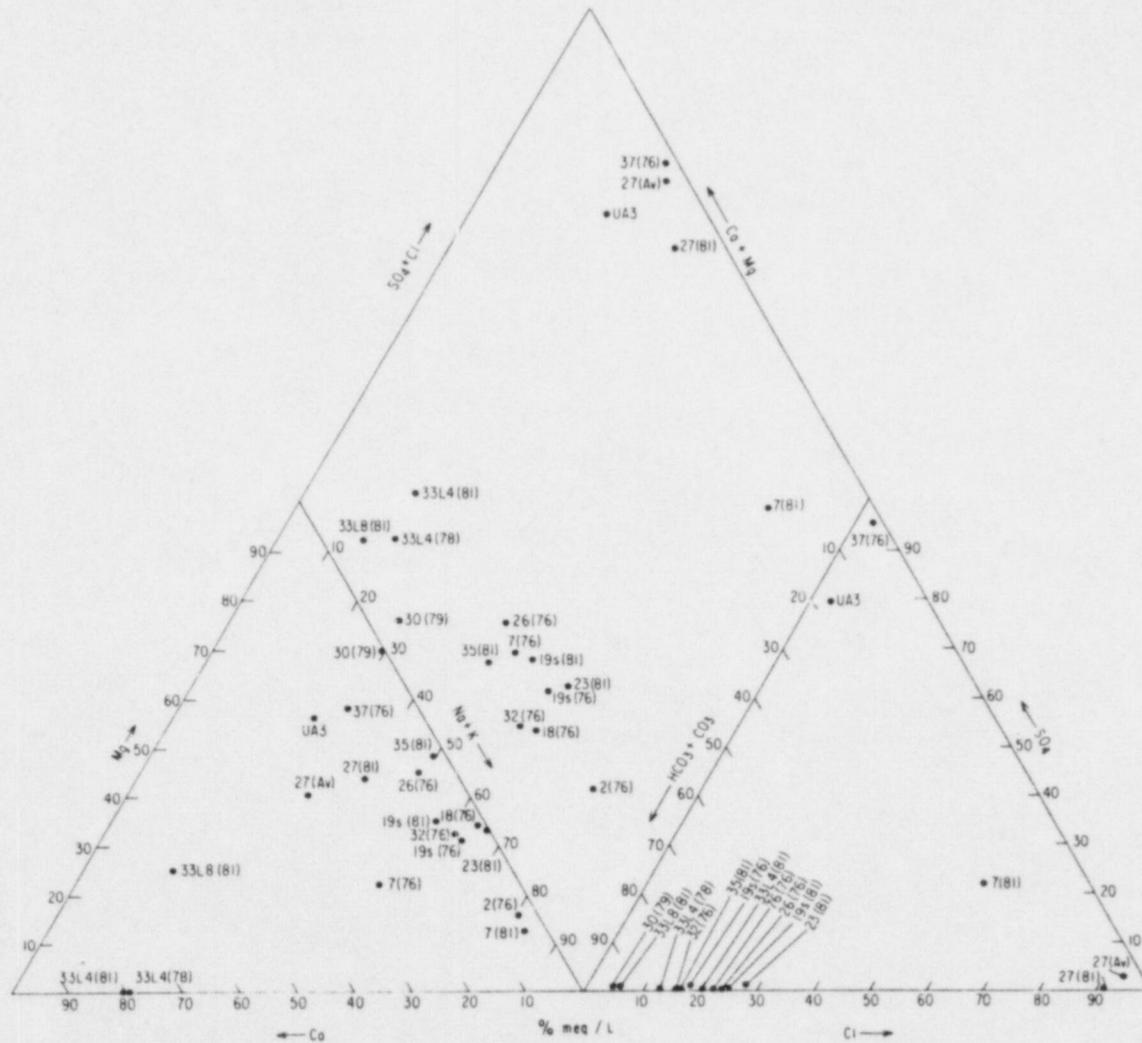
Figure 3.1. Piper trilinear diagram showing the different water types representing trench leachates and ambient groundwater (UA3) from Maxey Flats site.
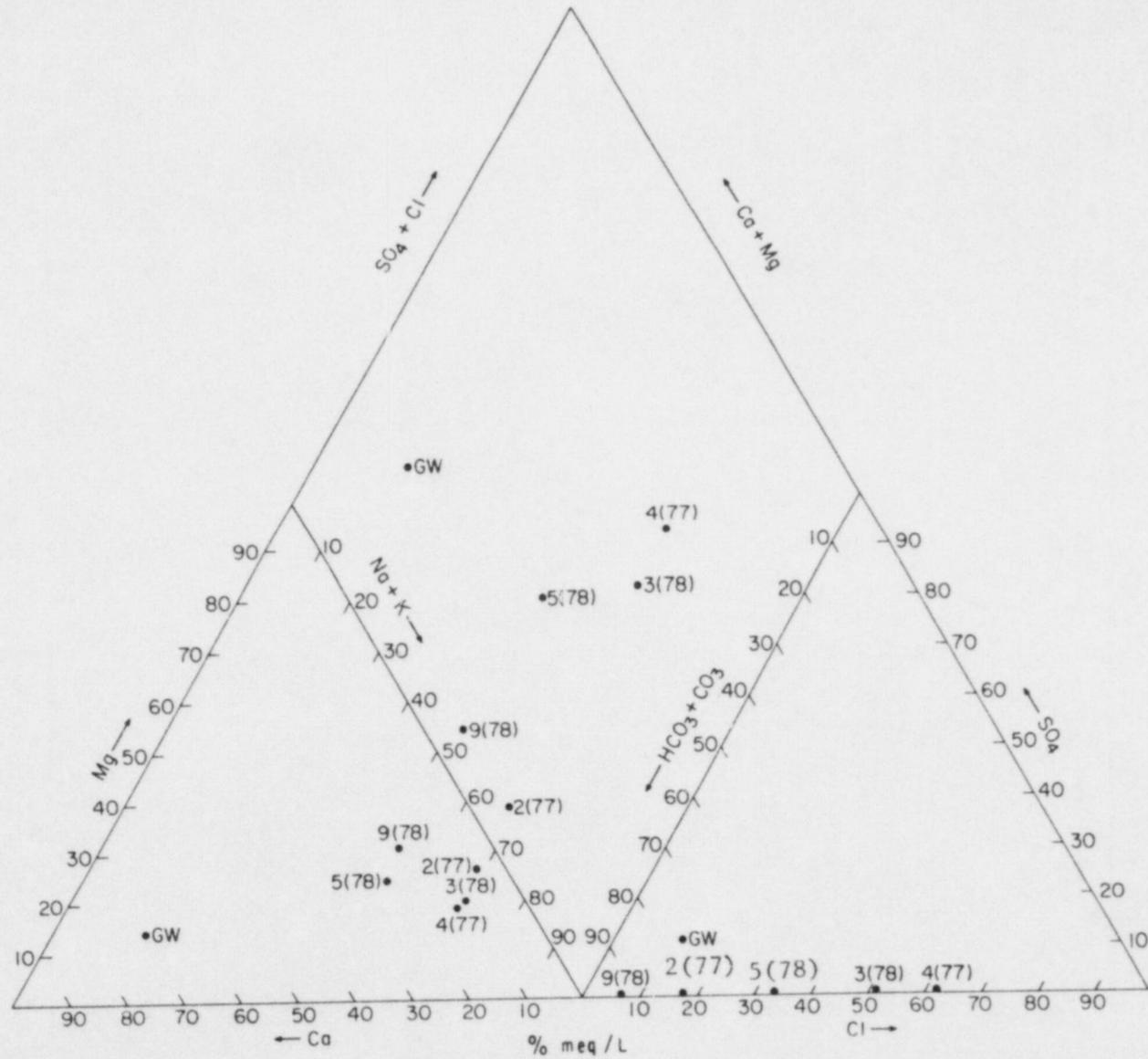
Figure 3.2. Piper trilinear diagram showing the different water types representing trench leachates and ambient groundwater (GW) from West Valley site.

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Figure 3.3. Piper trilinear diagram showing the different water types represent trench leachates and ambient groundwater (GW) from Barnwell site.
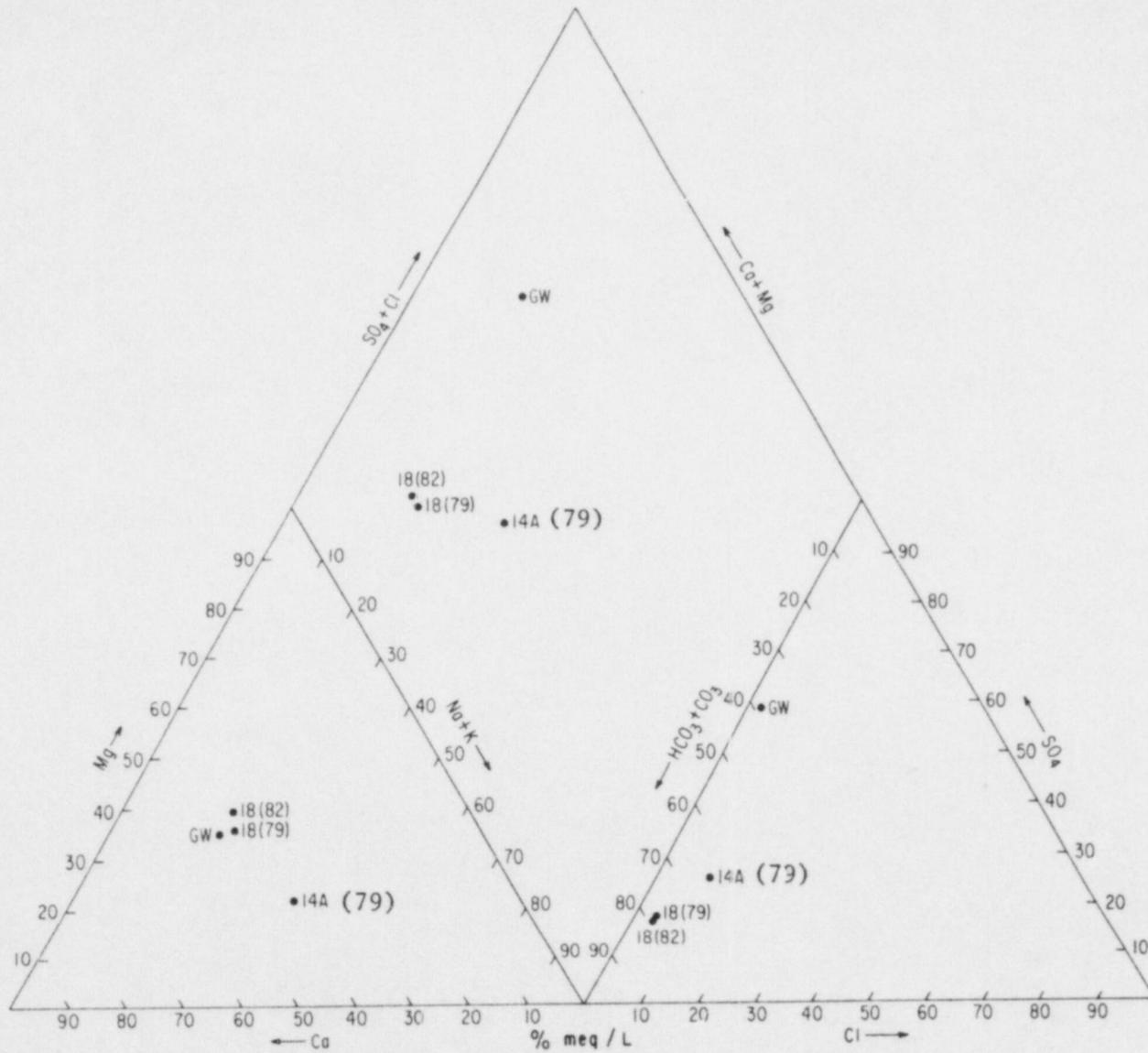
Figure 3.4. Piper trilinear diagram showing the different water types representing trench leachates and ambient groundwater (GW) from Sheffield site.

Table 3.5. Various water types showing nature and extent of chemical variability in trench leachates and ambient groundwaters.

Water Sample	Water Type
Maxey Flats Site	
2(76), 7(76), 18(76), 19s(76,81), 23(81), 26(76), 30(79), 32(76), 35(81)	$(Na + K) - Mg - HCO_3$
37(76), GW(MF)	$Mg - (Na + K) - SO_4$
7(81)	$(Na + K) - Cl$
27(Av), 27(81)	$(Na + K) - Mg - Cl$
33L4(78,81), 33L8(81)	$Ca - HCO_3$
West Valley Site	
GW(WV)	$Ca - HCO_3$
5(78), 9(78), 2(77)	$(Na + K) - HCO_3$
4(77), 3(78)	$(Na + K) - Cl - HCO_3$
Barnwell Site	
25/21(80), 8(80)	$(Ca + Mg) - Na - HCO_3$
8(79), GW(BW)	$(Na + K) - HCO_3$
6(80), 25/21(79)	$(Na + K) - (Cl + SO_4)$
Sheffield Site	
18(82), 18(79), 14A(79)	$(Ca + Mg) - HCO_3 - SO_4$
GW(SH)	$(Ca + Mg) - (Cl + SO_4)$

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Table 3.6. Average radionuclide concentrations in trench leachates sampled during the period 1976–1982.

	Concentration (pCi/L)[a]							
Trench	H-3	Sr-90	Pu-238	Pu-239,240	Am-241	Cs-134	Cs-137	Co-60
Maxey Flats Site								
2	1.7×10^7	4.6×10^3	6.4×10^3	3.5×10^2	4.0×10^3	$<1.0 \times 10^2$	$<1.0 \times 10^2$	7.9×10^3
7	3.0×10^8	1.1×10^6	7.1×10^0	2.6×10^0	3.9×10^2	1.0×10^2	3.6×10^4	1.7×10^4
18	3.4×10^8	4.7×10^4	5.7×10^2	5.1×10^1	$<2.0 \times 10^1$	$<1.0 \times 10^2$	4.8×10^3	1.2×10^4
19s	7.0×10^7	2.4×10^5	2.0×10^5	5.8×10^3	1.4×10^4	$<5.0 \times 10^1$	6.1×10^3	6.9×10^3
23	1.0×10^9	1.9×10^4	2.2×10^4	6.9×10^1	1.4×10^2	$<2.0 \times 10^1$	3.2×10^3	1.5×10^3
26	1.1×10^8	2.9×10^4	8.0×10^4	3.1×10^3	1.0×10^3	3.6×10^1	5.2×10^3	5.7×10^2
27	2.3×10^9	1.4×10^5	1.3×10^4	1.3×10^3	4.8×10^3	7.6×10^1	9.3×10^3	5.3×10^3
30	1.1×10^{10}	1.9×10^4	2.0×10^2	3.3×10^2	1.2×10^2	2.2×10^2	5.6×10^4	2.2×10^4
32	1.3×10^9	3.8×10^5	5.0×10^4	1.3×10^3	6.0×10^3	5.6×10^1	4.1×10^3	2.0×10^3
33L4	4.0×10^7	1.7×10^4	3.5×10^3	5.3×10^3	2.0×10^2	$<4.0 \times 10^1$	1.9×10^2	1.8×10^1
33L8	7.0×10^6	4.4×10^2	7.9×10^0	1.2×10^0	$<3.0 \times 10^1$	$<2.0 \times 10^1$	1.4×10^2	1.6×10^3
33L9	3.3×10^7	9.7×10^3	b	b	$<4.0 \times 10^1$	$<1.0 \times 10^1$	3.4×10^3	1.4×10^4
33L18	3.8×10^7	3.1×10^4	7.7×10^3	2.4×10^3	4.3×10^3	6.4×10^2	2.6×10^4	4.1×10^3
35	3.7×10^9	1.5×10^4	5.1×10^3	7.1×10^1	3.4×10^2	1.2×10^2	5.2×10^3	1.5×10^2
37	7.8×10^6	1.7×10^3	1.7×10^4	3.1×10^2	1.9×10^4	2.5×10^2	6.2×10^3	2.3×10^4
West Valley Site								
2	8.3×10^7	3.1×10^4	3.2×10^1	1.9×10^2	$<2.1 \times 10^2$	$<1.2 \times 10^2$	2.5×10^4	2.0×10^2
3	3.5×10^8	8.4×10^5	6.9×10^1	8.7×10^1	$<5.1 \times 10^2$	$<1.9 \times 10^2$	1.2×10^6	2.3×10^4
4	2.8×10^8	1.5×10^7	1.8×10^1	4.3×10^1	$<1.1 \times 10^3$	2.9×10^1	1.5×10^4	5.0×10^2
5	1.3×10^9	3.5×10^5	2.6×10^2	5.7×10^1	$<2.7 \times 10^2$	6.8×10^2	1.1×10^5	5.3×10^2
8	2.9×10^9	1.4×10^5	1.4×10^5	3.3×10^2	3.6×10^2	3.4×10^2	1.3×10^5	9.8×10^1
9	3.5×10^8	2.0×10^5	2.5×10^2	2.7×10^0	$<2.2 \times 10^2$	5.0×10^2	3.0×10^4	7.3×10^2
Barnwell Site								
3	1.0×10^4	b	b	b	b	$<2.1 \times 10^1$	$<2.3 \times 10^1$	$<2.2 \times 10^1$
5	8.5×10^6	3.9×10^1	1.4×10^6	$<2.8 \times 10^{-1}$	b	$<2.4 \times 10^1$	1.6×10^2	1.3×10^2
6	6.2×10^5	$<4.3 \times 10^0$	3.6×10^1	$<2.8 \times 10^{-1}$	b	$<2.2 \times 10^1$	$<2.2 \times 10^1$	$<2.2 \times 10^1$
8	3.9×10^8	4.1×10^1	1.2×10^0	4.6×10^{-1}	b	$<2.2 \times 10^1$	$<4.9 \times 10^2$	3.1×10^1
13	$<1.4 \times 10^6$	$<4.6 \times 10^0$	7.3×10^{-1}	9.3×10^{-1}	b	$<2.3 \times 10^1$	1.0×10^0	$<2.0 \times 10^1$
18	$<7.1 \times 10^2$	$<4.6 \times 10^0$	$<3.6 \times 10^{-1}$	$<2.8 \times 10^{-1}$	b	$<2.3 \times 10^1$	$<2.1 \times 10^1$	$<2.4 \times 10^1$
25/21	2.6×10^5	$<4.6 \times 10^0$	4.5×10^0	2.8×10^{-1}	b	$<2.3 \times 10^1$	$<2.5 \times 10^1$	$<2.8 \times 10^1$
Sheffield Site								
14A	5.4×10^5	2.1×10^0	1.4×10^0	2.3×10^{-1}	b	6.1×10^3	2.8×10^4	1.0×10^4
18	4.7×10^5	3.1×10^0	4.0×10^0	1.5×10^0	b	$<2.3 \times 10^1$	4.9×10^1	2.9×10^2

[a] Decay corrected to October 1981.
[b] Not determined.

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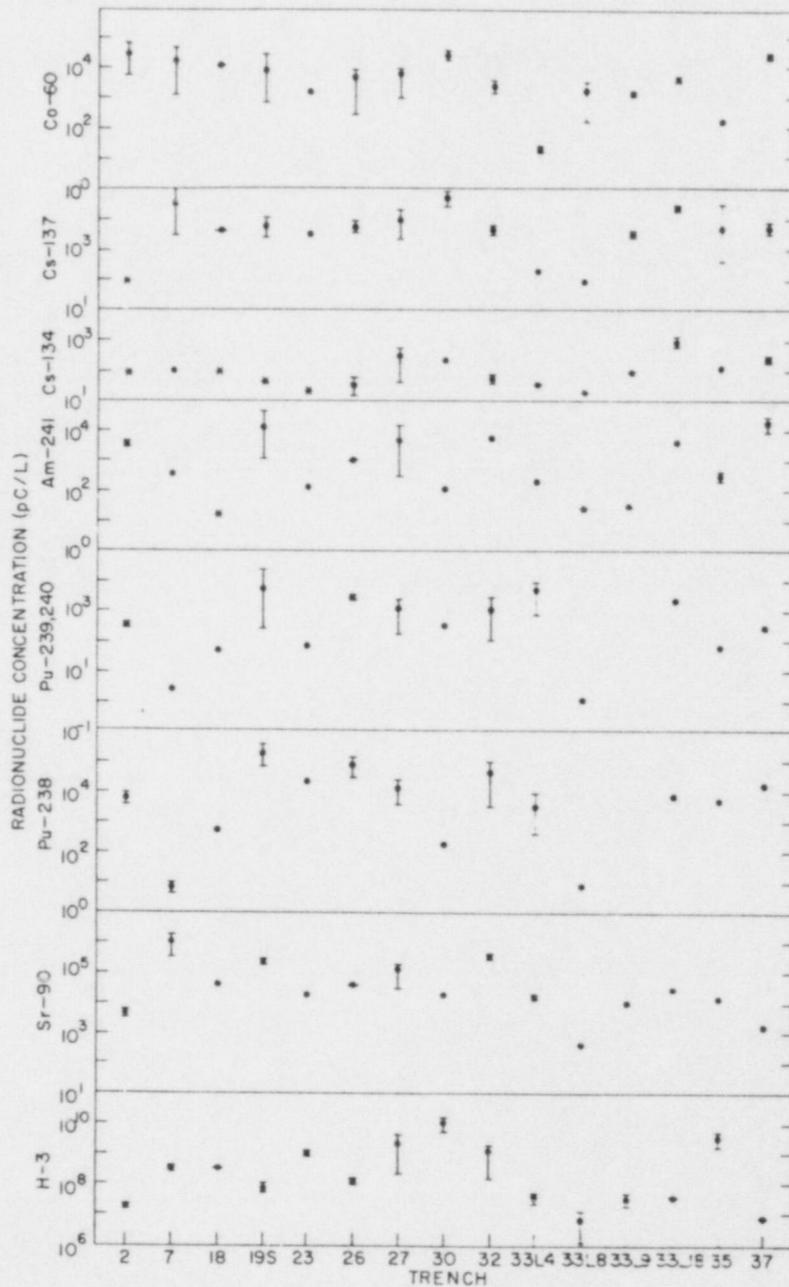
Figure 3.5. Average radionuclide concentrations in Maxey Flats trench leachates (decay corrected to October 1981) based on multiple samplings during the period 1976-1981. The error bars represent the extent of variation observed in radionuclide concentrations in a given trench sampled multiply. Asterisk (*) indicates concentrations below detection limits.
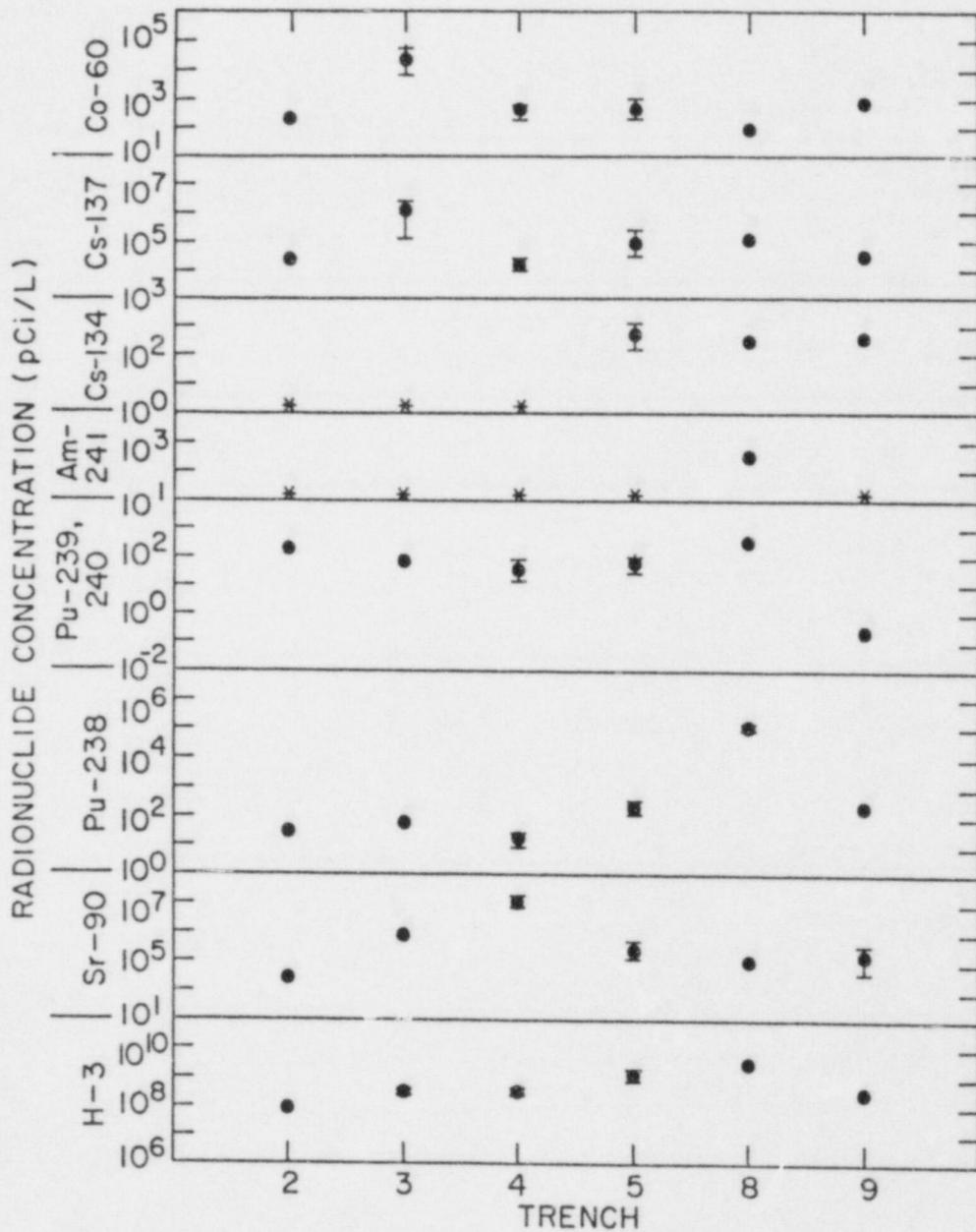
Figure 3.6.   Average radionuclide concentrations in West Valley trench leachates (decay corrected to October 1981) based on multiple samplings during the period 1977-1978. The error bars represent the extent of variation observed in radionuclide concentrations in a given trench sampled multiply. Asterisk (*) indicates concentrations below detection limits.
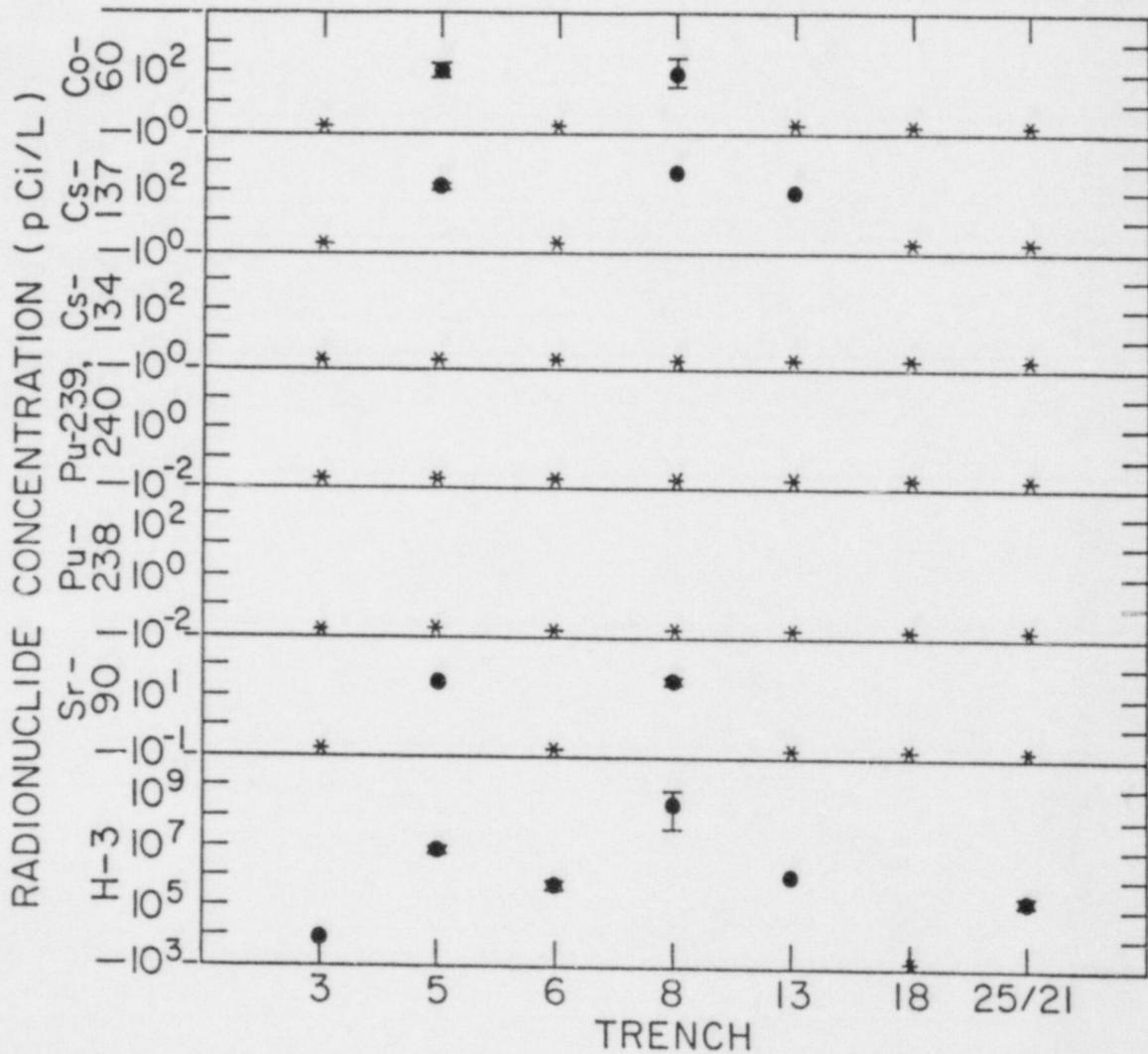
Figure 3.7. Average radionuclide concentrations in Barnwell trench leachates (decay corrected to October 1981) based on multiple samplings during the period 1979-1980. The error bars represent the extent of variation observed in radionuclide concentrations in a given trench sampled multiply. Asterisk (*) indicates concentrations below detection limits.

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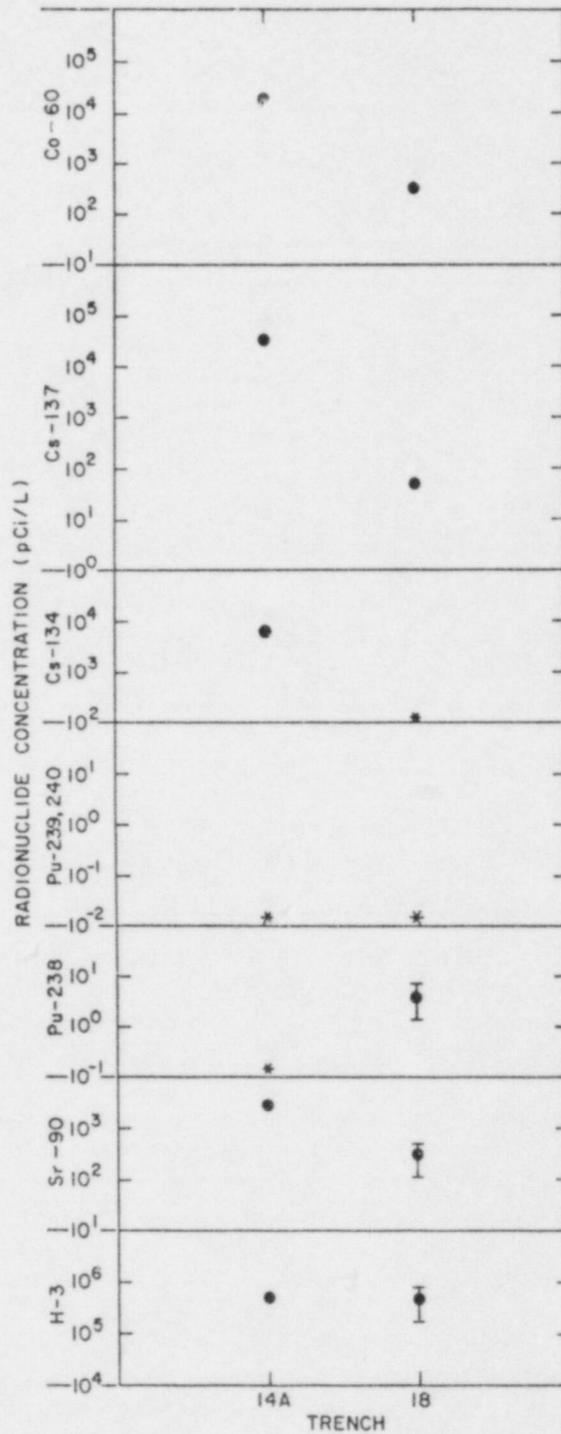
Figure 3.8. Average radionuclide concentrations in Sheffield trench leachates (decay corrected to October 1981) based on multiple samplings during the period 1979-1982. The error bars represent the extent of variation observed in radionuclide concentrations in a given trench sampled multiply. Asterisk (*) indicates concentrations below detection limits.

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4. DISCUSSION

4.1. Mechanics of Leachate Formation and Development of Anoxia in Trenches

A combination of hydrological and geochemical factors determines to a large extent the nature and amount of leachate produced in a trench at a given site. In particular, at the Maxey Flats and West Valley sites, the waste trenches are located in non-porous shale and glacial till of relatively low hydraulic conductivity. Consequently, this leads to accumulation of rainwater that infiltrates through the trench caps. Such stagnant accumulations of water and eventual overflow at the Maxey Flats and West Valley sites have been termed the "Bathtub Effect." Because of the relatively long residence time of accumulated water in the trenches, prolonged leaching and microbial degradation of buried waste materials occurs continually, leading to leachate formation. As a result of such interactions for extended time periods, the resultant trench leachates acquire geochemical properties which are unique compared to ambient groundwaters. In contrast, Barnwell and Sheffield trenches represent generally well-drained systems, where the infiltrated water has a relatively short residence time and is not continually present in the trenches, thus resulting in a different leachate geochemistry. The important biogeochemical processes controlling formation and composition of trench leachates are discussed in the following sections.

The development of anoxia observed in the waste trenches can be attributed to redox conditions in the trench leachates largely controlled by microbial degradation of organic matter present in buried wastes. A large fraction of waste materials consists of unconsolidated organic wastes. Many components of these materials are subject to both aerobic and anaerobic microbial degradation processes. The presence of both aerobic and anaerobic (sulphate reducing, denitrifying, and methanogenic) bacteria has been reported in the trench leachates at the Maxey Flats and West Valley sites (Francis, 1982; 1980).

Given the relatively stagnant leachate accumulations in the trenches at these sites, the dissolved oxygen is consumed rapidly during aerobic decomposition processes. Following depletion of all molecular oxygen, further degradation of organic material occurs by anaerobic processes which represent progressively lower Eh values as shown in Fig. 4.1 (Drever, 1982). The anaerobic degradation or organic matter involves denitrification, followed successively by sulphate reduction and methane generation. During these aerobic and anaerobic degradation processes, there is a continual production and buildup of decomposition products such as carbon dioxide and ammonia which causes the alkalinity, total aqueous CO_2, and ammonia contents of the trench leachates to increase. Concurrently, dissolved oxygen, sulphate, and nitrate are consumed, resulting in negative redox potentials and the presence of sulphide in solution. Iron reduction contributes to high concentrations of dissolved iron, primarily present as Fe^{2+} [Heaton and Dayal (1985); Dayal et al. (1981)]. Methane generation fermentation reaction represents an advanced stage of microbial oxidation of organic matter, following sulphate depletion,

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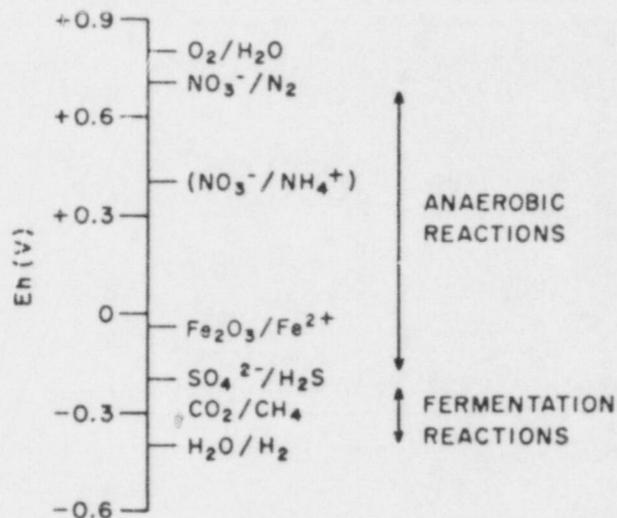
Figure 4.1. Approximate Eh values at which various redox reactions
occur in water at pH 7 and 25°C (after Drever, 1982).

and drives the redox potential to lower Eh values. In fact, methane is reported to be present in several trenches at the Maxey Flats and West Valley sites [Weiss and Colombo (1980); Czyscinski and Weiss (1981)].

4.2 Redox Equilibria and Buffering in Trench Leachates

Although Eh measurements reflect the redox levels of aqueous systems, it is important to know which redox processes control and buffer the system. A redox buffered system is one in which reducible or oxidizable constituents are present that prevent changes in Eh during additions of small amounts of strong oxidizing or reducing agents (Drever, 1982). Figure 4.2 shows the change in Eh upon decomposition of organic matter in fresh water containing sedimentary material. In the absence of solid phases such as Mn and Fe oxides, there is a sharp drop in Eh to a constant value controlled by SO_4^{2-}/H_2S, following complete depletion of all molecular oxygen. The Eh value remains constant until all the dissolved sulphate is consumed by the process of bacterial sulphate reduction, at which point the fermentation reactions take over, as reflected by a steady decrease in Eh with further decomposition of organic matter. If the nitrate content of groundwater is appreciable, reactions involving nitrogenous compounds such as denitrification (NO_3^-/NH_4^+) may provide some buffering between the O_2/H_2O and SO_4^{2-}/H_2S redox levels. In the presence of iron and manganese oxides, reactions involving reduction of the solid oxide phases also provide buffering between the O_2/H_2O and SO_4^{2-}/H_2S pairs in the redox sequence (Fig. 4.2).

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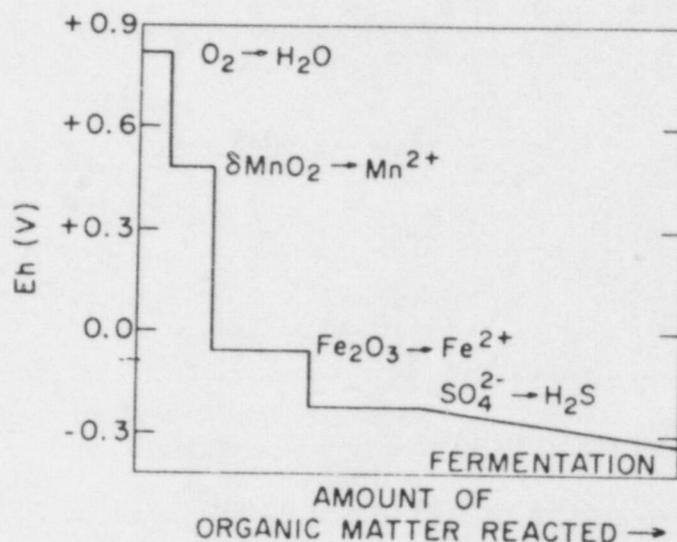
Figure 4.2. Redox sequence based on decomposition of organic matter in fresh water containing sedimentary matter (after Drever, 1982). The lengths of the horizontal segments are arbitrary, depending on the quantity and reactivity of specific solid phases. pH is assumed to remain constant at 7.0. Because of the initially low nitrate concentrations in groundwater, denitrification is assumed not to be an important redox process.

In the discussion above, we have only evaluated redox buffers for relatively uncontaminated systems involving organic matter and solutes in the absence and presence of sedimentary material such as iron and manganese oxides. Considering the complexity of the solid phases and the solute chemistry of the trench leachates, it is difficult to establish unambiguously the dominant redox buffers controlling the redox conditions in the trenches. However, plotting the leachate data on an Eh-pH diagram showing the various solid/solution boundaries representing the important redox buffers in a groundwater system (Fig. 4.3), indicates that the redox levels of most Maxey Flats and West Valley leachates (Region A) correspond to buffering by the Fe_2O_3/Fe^{2+} and SO_4^{2-}/H_2S redox pairs, reflecting the presence of reactive organic matter and relatively long residence times for infiltrated water in the trenches. In contrast, the Barnwell and Sheffield leachates generally lie in Region B which appears to be buffered by iron reduction [$(Fe(OH)_3/Fe^{2+}$ pair], indicating either the absence of metabolizable organic matter in the leachates or that their residence time is relatively short.

It has been reported that in complex aqueous systems, the measured redox potential often represents a mixed potential and does not respond to a particular redox couple (Jackson and Patterson, 1982; Stumm and Morgan, 1981;

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Eh (V)

+0.9 — O_2/H_2O

+0.6 — $\delta MnO_2/Mn^{2+}$

+0.3

0.0 — $Fe(OH)_3/Fe^{2+}$

— Fe_2O_3/FeS_2
— SO_4^{2-}/H_2S
-0.3 — Fe_2O_3/Fe^{2+}
— H_2O/H^+

-0.6

5   6   7   8   9   10

pH

B

A
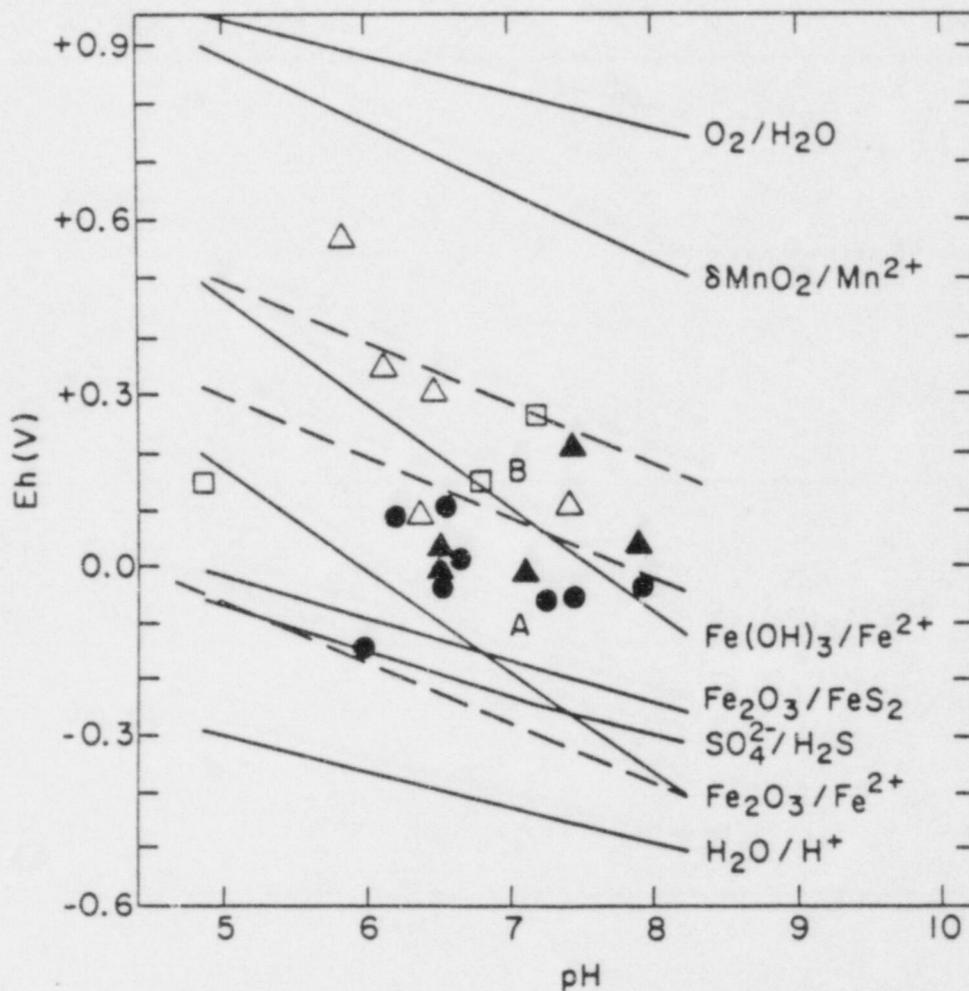
Figure 4.3. Solid/solution boundaries showing the various redox buffers in a groundwater system. Dashed lines delineate Regions A and B showing the redox ranges for trench leachates: (●) Maxey Flats; (▲), West Valley; (△)Barnwell; and (□)Sheffield (adapted from Drever, 1982).

Lindberg and Runnels, 1984). Assuming the SO_4^{2-}/H_2S and Fe_2O_3/Fe^{2+} redox couples to be controlling the redox levels of Maxey Flats and West Valley trench leachates and knowing the sulphate, bisulphide, and ferrous iron activities and the pH, we have obtained two sets of data, representing estimates of leachate redox potentials based on the two redox buffers. Our calculations show that although the estimated redox potentials are consistently lower than the measured values for most trench leachates, the calculated redox potentials still lie in Region A, indicating that the measured redox potentials are fairly representative of the actual redox conditions in the Maxey Flats and

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based on the SO_4^{2-}/H_2S and Fe_2O_3/Fe^{2+} redox couples. This is also consistent with the observation of complete sulphate depletion and methane generation in several trenches at the Maxey Flats and West Valley sites.

4.3. Microbially-Mediated Charges in Major Ion Chemistry

The nature and extent of microbially-mediated changes in major ion chemistry of trench leachates, relative to ambient groundwaters, are presented in Tables 4.1-4.4 and displayed in Figures 4.4-4.7.

Table 4.1. Enrichment and depletion of leachate constituents due to biodegradation in Maxey Flats leachates.

Leachate	Enrichment/Depletion Factors[a]		
	Alkalinity $(\times 10^0)$	SO_4^{2-} (10^{-3})	NH_4^+[b] $(\times 10^1)$
2(76)	3.4	5.2	c
7(76)	2.4	2.4[d]	c
7(81)	2.5	628	7.5
18(76)	4.5	8.6	c
19s(76)	2.1	2.4[d]	c
19s(81)	2.3	1.0[d]	4.5
23(81)	5.3	27	10
26(76)	2.9	2.4[d]	c
27(Av)	0.8	11.	7.0
27(81)	0.7	1.0[d]	12
30(79)	14	40	5.0
32(76)	5.9	5.2	c
33L4(78)	3.5	2.4[d]	1.8
33L4(81)	4.6	1.0[d]	2.6
33L8(81)	2.4	7.1	5.0
35(81)	5.0	1.0[d]	3.7
37(76)	0.3	3809	c

[a]Enrichment/Depletion Factor = C_t/C_w, where C_t represents concentration of species in trench leachate and C_w, the average well water concentration as reported by Zehner (1983) for well UA3.
[b]Detection limit of species in groundwater was used to calculate enrichment factors.
[c]Leachate not analyzed for ammonium.
[d]Detection limits of species in leachate were used to calculate depletion factors.

Table 4.2. Enrichment and depletion of leachate constituents due to biodegradation in West Valley trenches.

Leachate	Enrichment/Depletion Factors[a]		
	Alkalinity $(\times 10^1)$	SO_4^{2-} $(\times 10^{-1})$	NH_4^+ $(\times 10^1)$
2(77)	1.9	2.9b	23
3(78)	1.0	15.	30
4(77)	1.1	2.9b	7
5(78)	1.4	2.9b	18
9(78)	0.6	2.9b	8

[a]Enrichment factor = C_t/C_w, where C_t represents concentration of leachate species and C_w, the groundwater species concentration as represented by West Valley Spring Water.
[b]Detection limits of species in leachate were used to calculate enrichment factors.

Table 4.3. Enrichment and depletion of leachate constituents due to biodegradation in Barnwell trenches.

Leachate	Enrichment/Depletion Factors[a]		
	Alkalinity $(\times 10^0)$	SO_4^{2-} $(\times 10^0)$	NH_4^+ $(\times 10^1)$
6(80)	1.4	2.6	0.2
8(79)	9.8	2.0	3.0
8(80)	22	0.4	10
25/21(79)	1.3	3.3	1.3
25/21(80)	1.7	0.3 b	1.8

[a]Enrichment factor = C_t/C_w represents concentration of leachate species and C_w, the well water species concentration as represented by the well #6.
[b]Detection limits of species in leachate was used to calculate depletion factors.

Table 4.4. Enrichment and depletion of leachate constituents due to biodegradation in Sheffield leachates.

Leachate	Enrichment/Depletion Factors[a]		
	Alkalinity ($\times 10^0$)	SO_4^{2-} ($\times 10^0$)	NH_4^{+}[b] ($\times 10^1$)
14A(79)	0.6	1.7	5.0
18(79)	2.7	4.0	9.0
18(82)	3.3	4.2	9.0

[a]Enrichment factor = C_t/C_w, where C_t represents concentration of leachate species and C_w, the well water species concentration as represented by the well #525.
[b]Detection limit of species in groundwater was used as to calculate enrichment factors.
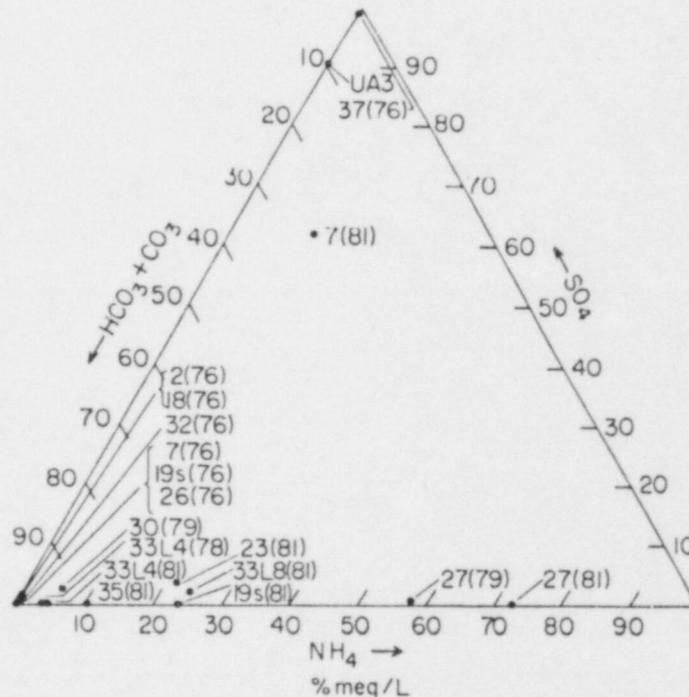
Figure 4.4. Ternary plot showing the nature and extent of modification of infiltrated rainwater as a result of microbial degradation processes in Maxey Flats trench leachates. UA3 represents the composition of ambient groundwater.

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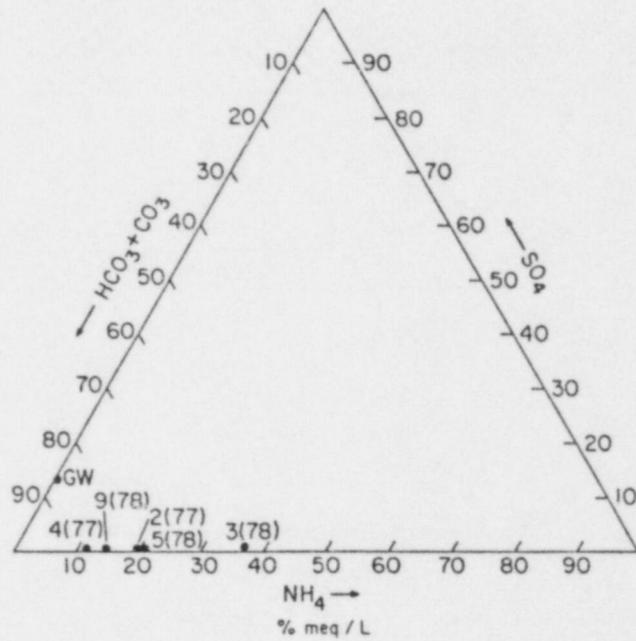
Figure 4.5. Ternary plot showing the nature and extent of modification of infiltrated rainwater as a result of microbial degradation processes in West Valley trench leachates. GW represents the composition of ambient groundwater.
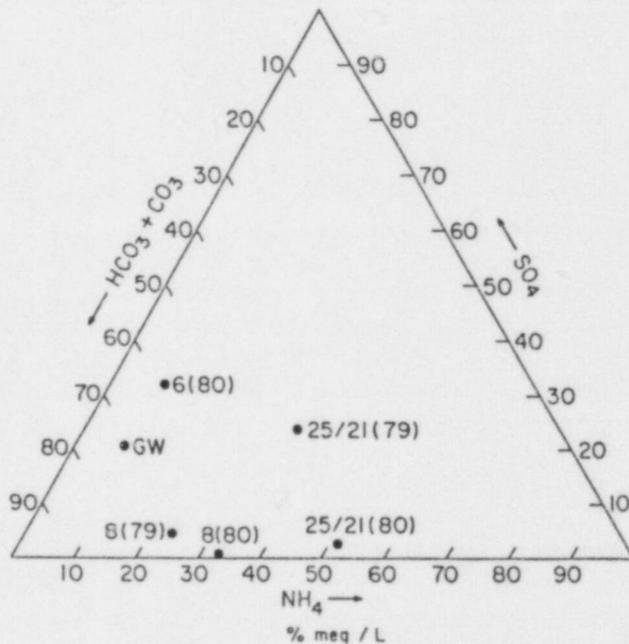
Figure 4.6. Ternary plot showing the nature and extent of modification of infiltrated rainwater as a result of microbial degradation process in Barnwell trench leachates. GW represents the composition of ambient groundwater.
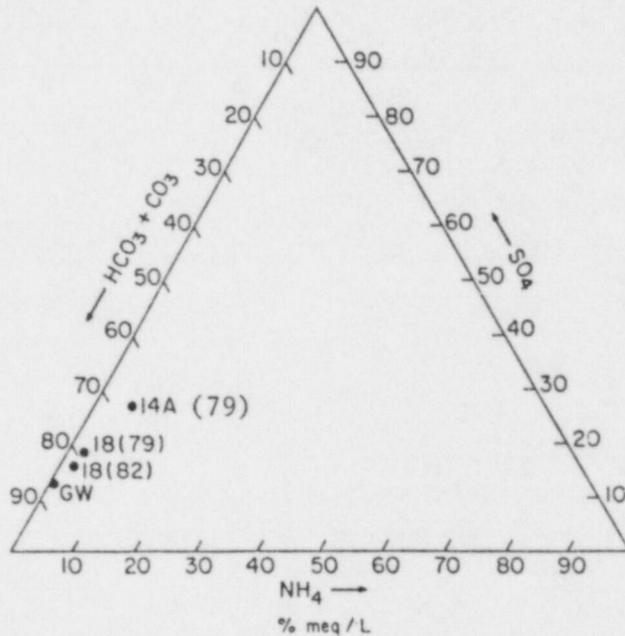
Figure 4.7. Ternary plot showing the nature and extent of modification
            of infiltrated rainwater as a result of microbial
            degradation processes in Sheffield trench leachates.
            GW represents the composition of ambient groundwater.

4.4. Waste Leaching and Resultant Enrichment of Waste-Derived Constituents

Water accumulations in the trenches lead to prolonged leaching of buried
waste materials, thereby resulting in the presence and build up of various
waste-derived constituents in the leachates. The nature and extent of modifi-
cation of infiltrated water due to waste leaching is a function of the quan-
tity and characteristics of the waste and the residence time of accumulated
water. As a consequence, the resulting leachates exhibit enrichments, to
varying degrees, of waste-derived inorganic, organic, and radionuclide consti-
tuents associat. with fuel cycle and non-fuel cycle low-level wastes (Appen-
dix A).

4.4.1. Inorganic Constituents

Relative to ambient groundwaters, most trench leachates are generally
enriched to varying degrees in sodium, potassium, dissolved iron, dissolved
inorganic and organic carbon, and chloride (Tables 4.5-4.8). Unlike Maxey
Flats and West Valley leachates, the Barnwell and Sheffield leachates also
show significant enrichments in calcium and magnesium. The enrichments of
Na^+, K^+, Cl^-, and other inorganic constituents are also reflected in high
specific conductance of the leachates. Relatively low specific conductance is

Table 4.5. Enrichment of leachate constituents, due to waste leaching in Maxey Flats trench leachates.

Leachate	Enrichment Factors[a]						
	$DOC^{b,f}$ $(\times 10^1)$	$DIC^{b,f}$ $(\times 10^1)$	Na^+ $(\times 10^0)$	K^+ $(\times 10^0)$	$Mn_T^{c,d}$ $(\times 10^0)$	$Fe_T^{c,d}$ $(\times 10^2)$	Cl^- $(\times 10^0)$
2(76)	3.4	2.5	2.3	2.8	8.0	4.0	4.4
7(76)	4.0	1.9	0.8	5.8	5.0	6.1	3.1
7(81)	12	2.0	7.1	14	19	1.6	36
18(76)	8.1	4.4	1.8	2.1	1.0	3.3	4.4
19s(76)	10	0.7	0.3	1.0	8.0	15	2.1
19s(81)	6.9	1.5	0.8	1.1	5.0	6.5	3.3
23(81)	13	4.7	2.8	3.4	1.0^e	0.7	8.2
26(76)	15	3.4	0.8	1.6	7.0	6.5	4.1
27(AV)	15	0.2	2.1	3.5	1160	125	67
27(81)	7.9	0.6	1.8	3.6	17	17	33
30(79)	4.2	14	3.3	1.8	3.0	1.0	2.8
32(76)	13	4.6	2.3	8.8	12	1.6	5.3
33L4(78)	18	0.1	0.6	1.2	1.0^e	0.03	2.4
33L4(81)	24	0.02	0.6	4.2	1.0^e	0.02	5.2
33L8	2.6	3.0	0.2	0.5	18	4.3	0.5
35(81)	8.7	3.5	2.0	2.1	3.0	0.09	3.4
37(76)	53	0.2	2.3	0.8	420	110	2.6

[a]Enrichment factor = C_t/C_w, where C_t represents concentration of leachate species and C_w, the well water species concentration as reported by Zehner (1983) for well UA3.
[b]DOC=dissolved organic carbon; DIC=dissolved inorganic carbon.
[c]Mn_T, Fe_T represent total dissolved manganese and iron, respectively.
[d]Detection limits of species in groundwater were used to calculate enrichment factors.
[e]Detection limits of species in leachate were used to calculate enrichment factors.
[f]Since C_w was not available for these species in UA3 well water, UB1A well water concentrations were used for calculating enrichment factors.

Table 4.6. Enrichment of leachate constituents, due to waste leaching in West Valley trench leachates.

Constituents	Enrichment Factors[a]				
	2(77)	3(78)	4(77)	5(78)	9(78)
Sodium $(\times 10^1)$	8.2	9.1	8.8	6.3	3.9
Potassium $(\times 10^2)$	3.3	3.2	3.3	2.7	0.9
$Mn_T^{c,d}$ $(\times 10^1)$	1.0	3.0	5.0	23	2.0
$Fe_T^{c,d}$ $(\times 10^2)$	0.7	2.8	4.1	27	2.9
Chloride $(\times 10^1)$	3.9	11	18	6.8	0.7
Calcium $(\times 10^0)$	1.7	3.6	4.3	7.1^e	3.1^e
Magnesium $(\times 10^1)$	4.4	3.6	3.2	4.0	3.0

[a]Enrichment factor = C_t/C_w, where C_t represents concentration of leachate species and C_w, the groundwater species concentration as represented by the composition of West Valley Spring Water reported by Cartwright and Ziarno (1980).
[b]DOC=dissolved organic carbon; DIC=dissolved inorganic carbon.
[c]Mn_T, Fe_T represent total dissolved manganese and iron, respectively.
[d]Detection limits of species in groundwater were used to calculate enrichment factors.
[e]No values available for the 1978 leachates so the values for the 1977 leachates from the same trench were substituted.

Table 4.7. Enrichment of leachate constituents due to waste leaching in Barnwell trench leachates.

Constituents	Enrichment Factors[a]				
	6(80)	8(79)	8(80)	25/21(79)	25/21(80)
DOC^b (x10^0)	1.2	28	34	2.0	c
DIC^b (x10^0)	1.3	22	43	6.3	3.0
Sodium (x10^0)	1.9	5.8	8.0	2.5	0.7
Chloride (x10^1)	0.4	2.8	1.6	1.4	0.4
Calcium (x10^0)	3.5	8.5	21	5.2	2.5
Magnesium (x10^1)	0.5	9.0	20	1.5	1.5
$Fe_T^{d,e}$ (x10^0)	1.0	1.2	24	0.2	6.0
$Mn_T^{d,e}$ (x10^0)	0.7	0.7	0.9	0.3	0.6

[a]Enrichment factor = C_t/C_w, where C_t represents concentration of the leachate species and C_w, the groundwater species concentration as represented by the well #6.
[b]DOC=dissolved organic carbon; DIC=dissolved inorganic carbon.
[c]Leachate species not determined.
[d]Fe_T represents total dissolved iron.
[e]Detection limit of species in groundwater was used to calculate enrichment factors.

Table 4.8. Enrichment of leachate constituents due to waste leaching in Sheffield trench leachates.

Constituents	Enrichment Factors		
	14A(79)	18(79)	18(82)
DOC^b (x10^1)	3.3	1.7	1.9
DIC^b (x10^0)	0.6	2.7	3.9
Sodium (x10^0)	1.4	1.9	2.2
Potassium (x10^1)	1.3	7.2	7.3
Mn_T^d (x10^0)	8	5.5	c
Fe_T^d (x10^0)	5.5	0.2	0.05^e
Chloride (x10^0)	1.5	2.2	2.1
Calcium (x10^0)	0.7	2.6	3.0
Magnesium (x10^0)	0.5	2.7	3.7

[a]Enrichment factor=C_t/C_w, where C_t represents concentration of leachate species and C_w, the well water species concentration.
[b]DOC=dissolved organic carbon; DIC=dissolved inorganic carbon.
[c]Leachate species not determined.
[d]Fe_T, Mn_T represents total dissolved iron and manganese, respectively.
[e]Detection limit of species in leachate was used to calculate enrichment factors.

observed for some leachates, which are also depleted in the major inorganic constituents, relative to ambient groundwaters. In addition to the principal waste-derived inorganic constituents discussed above, there are several other constituents such as Sr^{2+}, Ba^{2+}, Li^+, PO_4^{3+}, F^-, and silica which are enriched in the trench leachates (Dayal et al., 1984b). It is likely that elevated concentrations of dissolved silica in some leachates are a result of the dissolution of siliceous, diatomaceous earth filter waste, composed mainly of amorphous silica.

The ternary plots in Figs. 4.8 to 4.11 summarize the nature and extent of modification of infiltrated water as a result of leaching of buried waste materials in trench leachates at the disposal sites. The end components in the ternary plot represent the principal suites of inorganic constituents which are believed to be waste-derived, based on the discussion given above.

Relative to the ambient groundwater composition, the compositions of most trench leachates from Maxey Flats reflect enrichment in the constituents $(Na + K + Cl + F + NO_3)$ and depletion of $(Ca + Sr + Ba)$ and Mg. Only leachates 33L4(78,81) and 33L8(81) are enriched in $(Ca + Sr + Ba)$, depleted in Mg and $(Na + K + Cl + F + NO_3)$, reflecting leaching of the cementitious grout matrix, which was used as a solidification agent for stabilizing liquid waste slurries in these trenches. In contrast, 30(79) and 37(76) are the only leachates which exhibit substantial enrichment of Mg and depletion of other waste-derived components, with the overall composition approaching that of ambient groundwater. Therefore, in terms of waste-derived inorganic constituents, leachates 30(79) and 37(76) appear to be the least modified relative to ambient groundwater. The most pronounced modification in the composition of infiltrated rainwater as a result of waste leaching is reflected in the compositions of trench leachates 33L4(78,81) and 7(81).

As evident from Fig. 4.9, the West Valley leachates are generally depleted in $(Ca + Ba + Sr)$ and enriched in $(Na + K + Cl + F + NO_3)$, relative to ambient groundwater. In this respect, the extent of modification appears to be most pronounced in case of leachate 3(78) and least in leachate 9(78).

Figure 4.10 shows that, compared to ambient groundwater, the Barnwell leachates are enriched in $(Ca + Sr + Ba)$ and Mg and slightly depleted in $(Na + K + Cl + F + NO_3)$, with leachate 8(80) exhibiting the most pronounced modification in the composition of infiltrated rainwater as a result of waste leaching. Note that both leachates 8(79) and 25/21(79) exhibit enhanced modifications, relative to ambient groundwater, in subsequent 1980 samplings. This may be a result of extended leaching due to longer period of leaching of wastes.

Compared to the modifications observed in the compositions of infiltrated rainwater as a result of waste leaching at the Maxey Flats, West Valley, and the Barnwell site, the Sheffield leachates exhibit the least modification, relative to ambient groundwater.
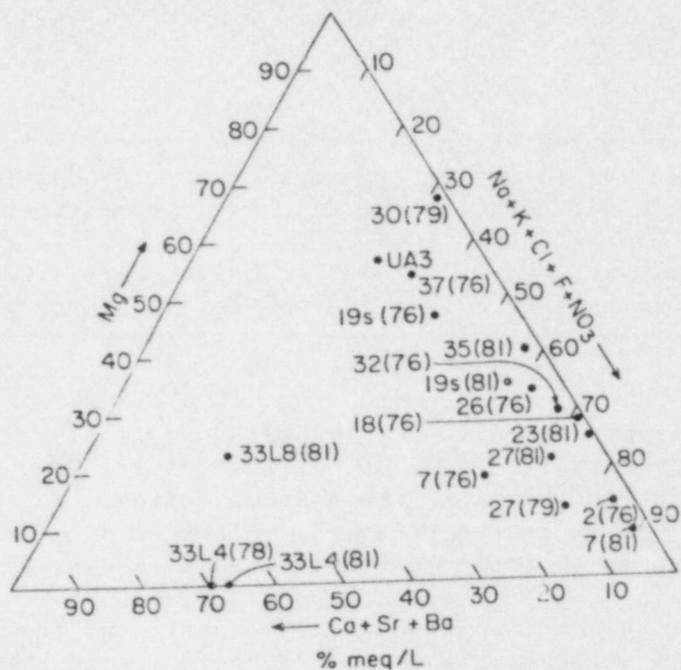
Figure 4.8. Ternary plot showing the nature and extent of modification of infiltrated rainwater as a result of leaching of buried waste materials in Maxey Flats trenches. UA3 represents the composition of ambient groundwater.
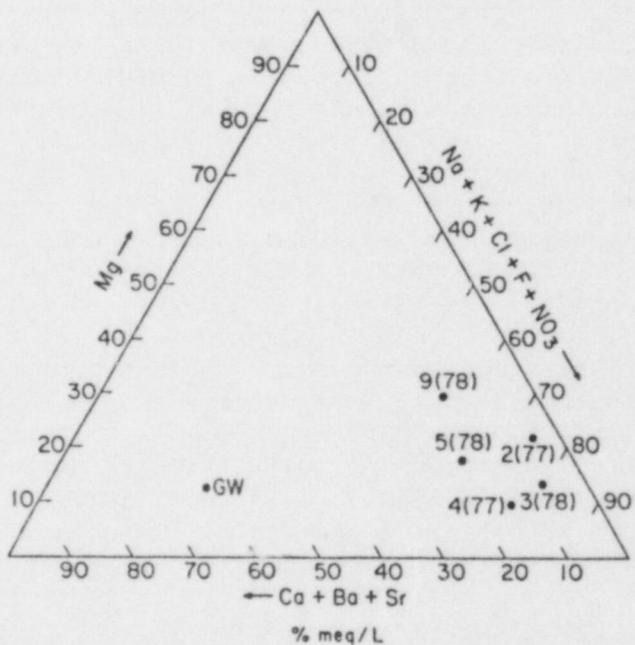
Figure 4.9. Ternary plot showing the nature and extent of modification of infiltrated rainwater as a result of leaching of buried waste material in West Valley trenches. GW represent the composition of ambient groundwater.
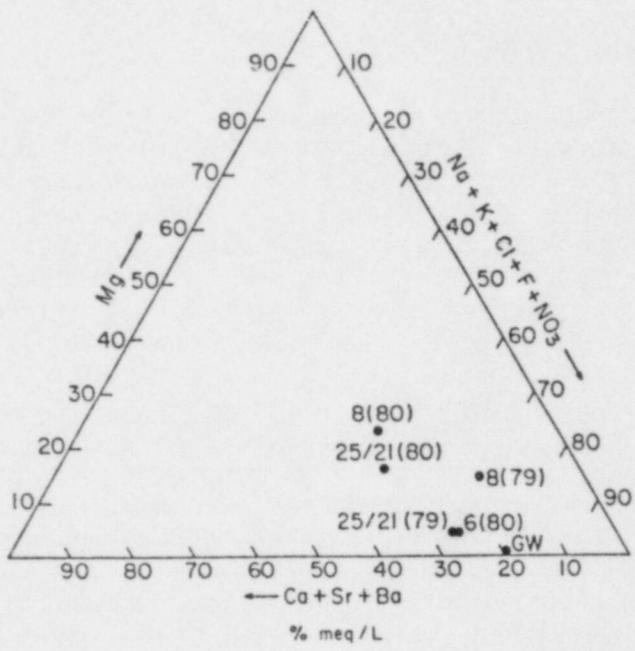
Figure 4.10. Ternary plot showing the nature and extent of modification of infiltrated rainwater as a result of leaching of buried waste materials in Barnwell trenches. GW represents the composition of ambient groundwater.
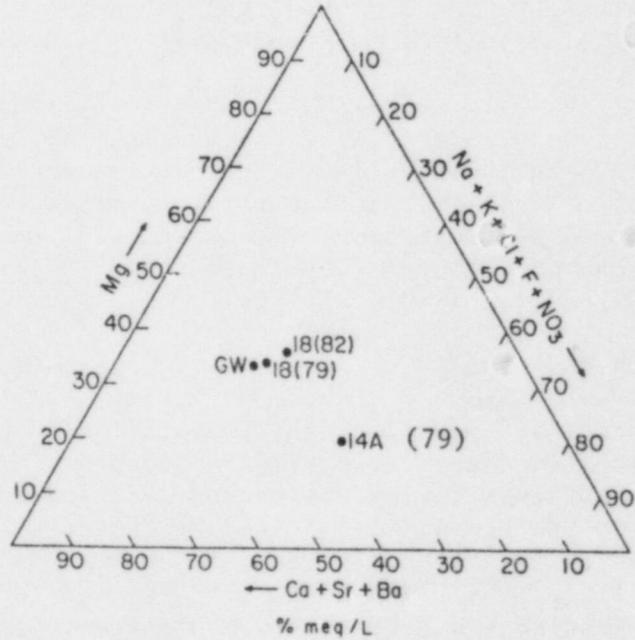
Figure 4.11. Ternary plot showing the nature and extent of modification of infiltrated rainwater as a result of leaching of buried waste materials in Sheffield trenches. GW represents the composition of ambient groundwater.

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4.4.2. Organic Constituents

In most leachates, the dissolved organic carbon represents a major fraction of the total dissolved carbon. As shown in Tables 3.3 and 4.5 to 4.8 the concentrations of dissolved organic carbon are significantly elevated relative to ambient groundwaters. In particular, the Maxey Flats and West Valley leachates are considerably enriched compared to Barnwell and Sheffield leachates. Several of these leachates are enriched in dissolved organic carbon by more than two orders of magnitude, compared to the groundwater concentrations.

A wide variety of hydrophobic and hydrophilic organic compounds have been identified in the trench leachates from the Maxey Flats and West Valley sites (Tables 4.9, 4.10, 4.11). The types of compounds identified reflect to a large extent the nature of the organic waste materials. Solvents or scintillation fluids, such as p-dioxane, toluene, and xylene are present. Tributyl phosphate is presumably derived from the process of solvent extraction of metal ions from solution of reactor products. Chelating agents such as EDTA, NTA, and DTPA are commonly used as chemical decontamination agents in power plants and represent a major fraction of the total hydrophilic compounds in most leachates. Some of these compounds as well as tributyl phosphate, methyl isobutyl ketone, phthalates, aniline, and cyclohexylamine identified in the trench leachates have the potential to form strong and stable radionuclide complexes and thus enhance radionuclide mobility (Dayal et al., 1984b). Many of the hydrophilic organic compounds identified in the trench leachates, especially the straight- and branched-chain organic acids, could be metabolic products formed as a result of anaerobic degradation of organic wastes.

As shown in Fig. 4.12 and 4.13, the data also indicate that the concentration ranges for total hydrophilic and hydrophobic constituents in the various Maxey Flats and West Valley trench leachates are comparable and that they are significantly lower than the total dissolved carbon concentrations, indicating that a large fraction of the organic carbon in solution remains uncharacterized and most likely constitutes high molecular weight organic compounds such as humic and fulvic acids.

4.4.3. Dissolved Radionuclides

The 1977 survey radionuclide data (Dayal et al., 1984b; Weiss and Colombo, 1980) and the decay corrected average radionuclide data based on multiple trench samplings during the period 1976-1982 show that substantial amounts of H-3, Co-60, Sr-90, Cs-137, and Pu isotopes (Pu-238, Pu-239,240) are found in most trench leachates. In many leachates, Cs-134 and Am-241 are also present. In addition, Na-22 and Mn-54 are observed in several leachate samples. The data also indicate considerable inter- as well as intra-trench variability in radionuclide concentrations. Leachates sampled from the same trench but different sumps invariably exhibit significant differences in radionuclide concentrations. As shown in Figs. 3.5-3.8, the Maxey Flats and West Valley leachates exhibit generally higher concentrations of radionuclides than the leachates sampled at the other two sites.

Table 4.9. Hydrophobic organic compounds in Maxey Flats trench leachates.*

Hydrophobic Organic Compounds	Concentration (mg/L)[a]								
	19s(76)	19s(79)	26(76)	27(76)	27(79)	30(79)	32(76)	32(79)	33L4(78)
Carboxylic acids	25	6.9	42	22	14	5.6	64	7.9	35
Benzoic acid	0.22		1.2	0.22	0.24		1.9		0.59
C6 acids	0.35	0.26		0.66	0.16	1.8			1.1
C8 acids	1.4	2.1		b	0.14	b		0.80	
2-Ethylhexanoic acid	5.6	1.9	3.4	17	9.7	1.1	8.8	3.2	27
Hexanoic acid	1.5		1.9	1.2	0.64		4.7		2.0
Isobutyric acid							2.0		
2-Methylbutanoic acid	4.6	0.52	19	0.98	0.60		13		
3-Methylbutanoic acid	1.8	0.80		0.48	0.76		5.8		1.6
2-Methylhexanoic acid	1.5	0.13	1.2	0.40	0.24		3.2	0.57	0.30
2-Methylpentanoic acid			4.2		0.45		4.0		
3-Methylpentanoic acid	3.1	0.67	3.6				1.4	0.98	
2-Methylpropionic acid	0.40	0.12			0.24				
Nonanoic acid		0.08							
Octanoic acid	0.36			0.64	0.10		1.3	0.20	0.39
Pentanoic acid			4.6				4.7		
Phenylacetic acid	0.56	0.34	1.5	0.08	0.16		3.4		0.25
Phenylhexanoic acid	b	b		b	b		b	b	
Phenylpropionic acid	1.2		1.3	0.56	0.50		9.8		0.70
Toluic acid						2.7		2.1	
Valeric acid	2.0				0.28				1.3
Esters	0.54	0.45			0.29		0.36	0.30	
Triethyl phosphate	0.38	0.18		b	0.29				
Tributyl phosphate	0.16	0.17					0.36	0.30	
Hydrocarbons	3.0	9.6	3.8	3.6	0.62		7.8	2.6	0.39
Alkanes (C10-C3ₓ)									
Dimethyl naphthalene									
Naphthalene	0.12	0.03	0.28	0.15	0.06		0.28	0.07	
Toluene	2.9	9.5	3.5	3.4	0.56		7.0	2.4	0.39
Xylene	b	0.03					0.48	0.13	
Alcohols	2.9	0.10	0.31	0.23	0.19		0.73		1.1
Cyclohexanol	2.9	0.10					0.24		1.1
Diethylene glycol		b			b				
Ethylene glycol		b			b	b	b	b	
2-Ethyl-1-Hexanol				0.23	0.17				b
a-Terpineol			0.31				0.49		
Phenols	2.9	1.2	2.0	1.1	0.79		5.4	0.36	9.1
Cresol isom.	2.9	1.2	2.0	0.70	0.56		4.2	0.28	0.28
Phenol		0.04		0.40	0.23		1.2	0.08	0.20
Vanillan									8.6
Ketones and Aldehyes	0.59	0.20			1.8	0.08			1.1
Camphor	b			b	b				
Cyclohexanone									1.1
Dibutylketone	b	b							
Fenchone	0.03			b	b	0.08			
Methylisobutyl ketone	0.56	0.20			1.8				
Total	34	19	48	27	18	6	78	11	47

*Results of trench leachates 2(76), 7(76), 18(76), 37(76) and well water UB1A(79) are reported elsewhere (Weiss and Colombo, 1980). However, qualitative analysis revealed the presence of similar hydrophobic organic constituents in those leachates.

[a]Pacific Northwestern Laboratories (PNL) identified several heterocyclic hydrophobic compounds in leachates 7(81), 19s(81,82), 23(81), 27(81,82) and 35(81). The following compounds were reported: Barbital, Benzofuran, Benzothiazol, 2(3H) bensothiazalone, caffeine, caprolactam, 1,3-dihydro-2H-indol-2-one, p-dioxane, nicotine, pentabarbital, piperidinone, and tetrahydrofuran. Detailed analysis of leachate 27(4/81) revealed the presence of several hydrocarbon and hydrocarbon halide classes of compounds. Further details are given elsewhere (Kirby et al., 1984).

[b]Compounds were identified but not quantified; blank spaces indicate compounds not detected.

Table 4.10. Hydrophilic organic compounds in Maxey Flats trench leachates.*

Hydrophilic Organic Compounds	Concentration (mg/L)[a]						
	7(81)	19s(81)	23(81)	27(78)	33L4(81)	33L8(81)	35(81)
Chelating agents	6.2	23	29	1.2	0.40	0.08	0.87
Ethylenediaminetetraacetic acid (EDTA)	5.0	12	4.9	0.96	0.40	0.08	0.87
N-Hydroxyethylethylene-diaminetriacetic acid (HEDTA)		8.4	20				
Ethylenediaminetriacetic acid (ED3A)	1.2	2.2	3.9	0.21			
Dicarboxylic acids	2.9	0.56	0.45	0.35	2.6	0.38	2.2
Oxalic acid							
Succinic acid	0.35			0.03		0.04	0.08
Methylsuccinic acid	0.19			0.02		0.02	0.23
Pentanedioic acid	0.25			0.04		0.05	0.41
Hexanedioic acid	1.6	0.22	0.24	0.17	0.45	0.10	0.83
Methylhexanedioic acid							0.31
Octanedioic acid					0.78	0.06	
Nonanedioic acid	0.46	0.34	0.21	0.09	1.4	0.11	0.31
Monocarboxylic acids					0.03	0.17	
Dodecanoic acid							
Hexadecanoic acid						0.06	
Nondnoic acid							
Octadecanoic acid					0.03	0.11	
Oxygenated acids	2.7			0.03		0.04	0.19
Citric acid							
4-Oxo-Pentanoic acid	2.7			0.03		0.04	0.19
Aromatic acids	1.9	0.44	0.90	0.08		0.03	2.0
Benzoic acid							
2-Hydroxybenzoic acid						0.03	
1,3-Benzenedicarboxylic acid	0.38	0.28	0.44	0.08			0.88
1,4-Benzenedicarboxylic acid	1.2	0.16	0.46				1.1
1,2,4-Benzenetricarboxylic acid	0.31						
Pentafluorobenzoic acid							
Phthalate esters	8.8	3.0	1.8	1.2		0.10	15
Dibutyl phthalate							
Dimethyl phthalate	8.8	2.8	1.7	0.39		0.10	14
Methyl phthalate		0.21	0.11	0.01			0.87
Dioctyl phthalate				0.85			
Total	22.5	27	32	3.0	3.0	0.8	20

*Methylated (BF$_3$/Methanol) acids identified as methyl esters; analysis conducted by PNL; samples collected by BNL.
[a]No entry indicates compound is below detection levels.

Table 4.11. Hydrophobic organic classes of compounds in West Valley trench leachates.

Class of Compounds	Concentration (mg/L)[a]					
	2(77)	3(78)	4(77)	5(78)	8(78)	9(78)
Carboxylic acids	0.4	297	28	490	849	264
Esters	0.0	0.6	0.1	0.7	0.8	b
Sulfonamides						
Hydrocarbons		3.4	0.9	22	6.6	2.1
Alcohols		8.4	0	11	5.0	5.0
Phenols	0.1	5.8	1.1	12	5.8	10
Ketones		0.3			0.1	<0.1
Amines						
Ethers						
Total	0.5	316	30	536	867	281

[a]No entry indicates compound is below detection limits.
[b]Identified but not quantified.

Tritium is the most abundant of the radionuclides identified and is invariably present in all trench leachates. Its presence and relatively high concentrations can be attributed to the readily leachable nature of H-3 present in waste materials, including the stabilized wastes in "L" series trenches at the Maxey Flats site. Chemically bound as HTO, tritium is the most mobile radionuclide among those identified in the trenches. Because of its relatively non-reactive nature, tritium behaves conservatively and could serve as an inert tracer for evaluating the nature and extent of waste leaching.

The types of radionuclides and their concentration levels observed in the leachates reflect to a large extent the nature and extent of leaching as determined by the reactivity and quantity of waste materials, the leaching behavior of the radionuclides, and the redox and chemical environment of the trenches. For example, most leachates sampled from "L" series trenches at the Maxey Flats site invariably exhibit immeasurably low concentrations of Cs-137, Cs-134, Co-60, and Am-241. This is undoubtedly a result of stabilization of liquid waste slurries disposed of in these trenches. In case of trenches 33L3 and 33L18, however, the liquid wastes were stabilized with urea formaldehyde (UF). Earlier work at BNL has shown that leaching of UF generates a considerable amount of acidity (Czyscinski and Weiss, 1981). The pH of the leachates was observed to be around 2.0 pH units. As a result of such strongly acidic conditions in these trenches, leaching of waste materials is rather intense, giving rise to high concentrations of waste-derived radionuclide constituents.
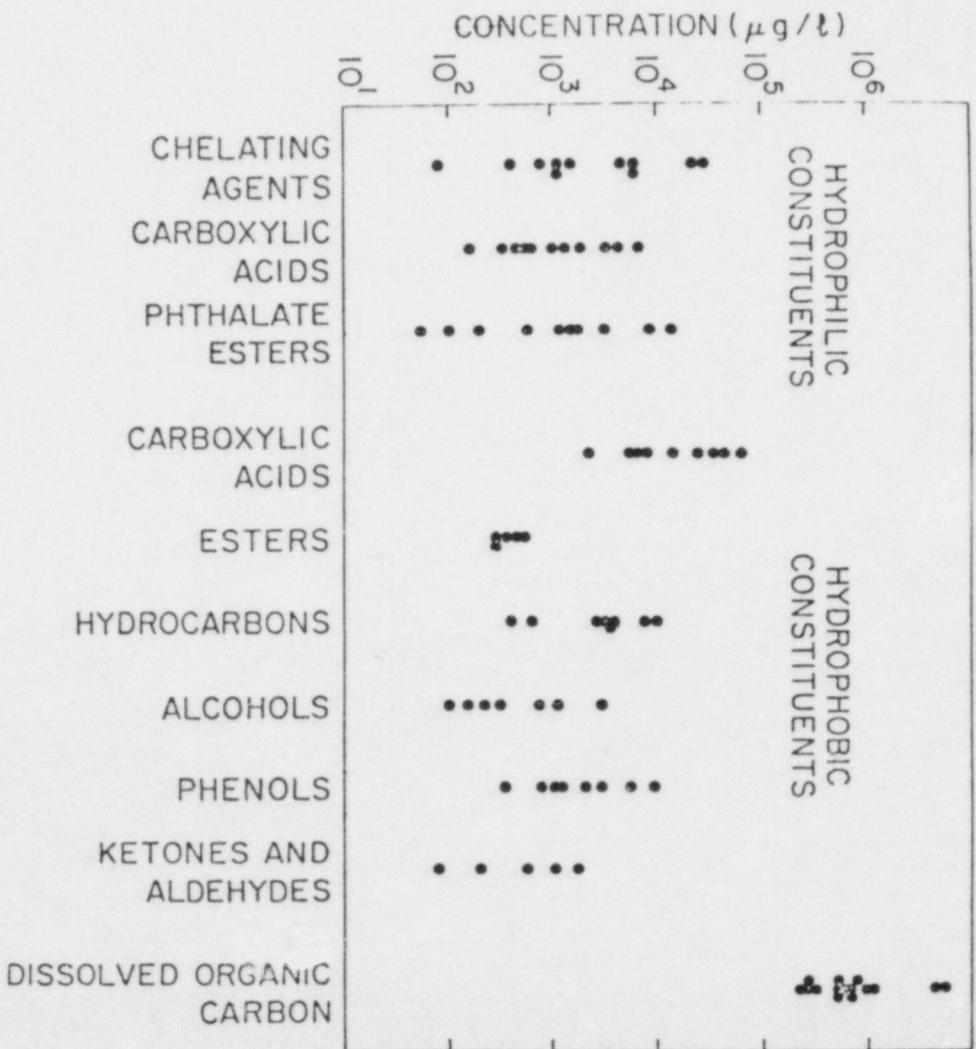
Figure 4.12. Concentration ranges for hydrophilic and hydrophobic constituents observed in the various Maxey Flats trench leachates. The range of dissolved organic carbon concentrations is also presented.

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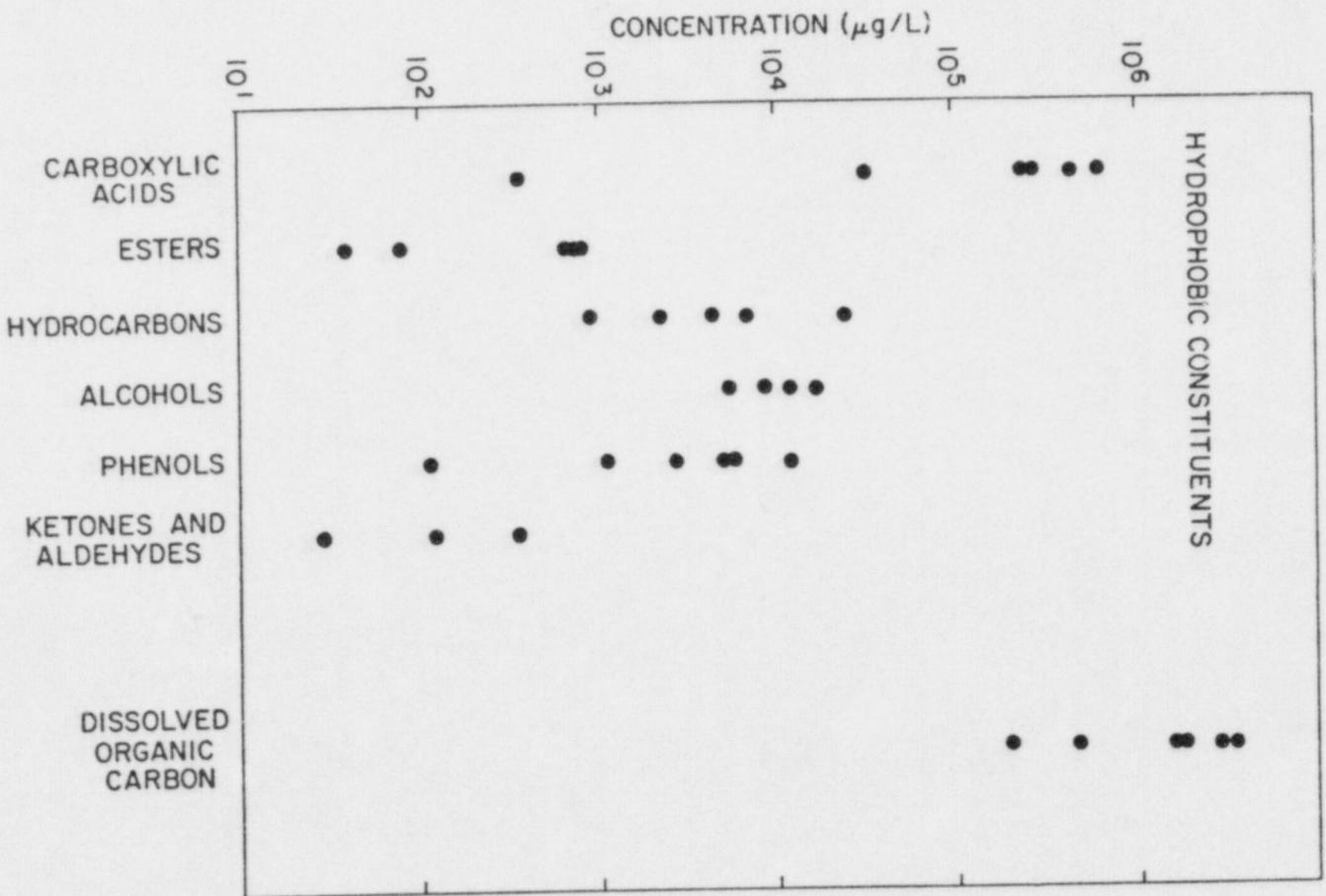
Figure 4.13. Concentration ranges for hydrophobic constituents observed in the various West Valley trench leachates. The range of dissolved organic carbon concentrations is also presented.

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The high concentrations of dissolved organic constituents as discussed above in Section 4.4.2, especially the complexing agents, also tend to keep the radionuclides, such as the Pu isotopes and Co-60, in solution and thus raise their concentrations in solution as well as make them more mobile (Kirby et al., 1984; Cleveland and Rees, 1981; Dayal et al., 1986b; Dayal et al., 1984b; Means et al., 1978; Polzer et al., 1984; Swanson, 1983). In addition, despite the observed anoxia in the trenches, the presence of inorganic complexing agents such as carbonate may keep the actinides in solution and increase their migration potential (Allard, 1982). Because of the heterogeneous and highly reactive nature of the waste materials, it is difficult to establish even qualitatively a direct relationship between radionuclide concentrations, organic and inorganic complexing agents, and varying degrees of anoxia observed in the various trench leachates.

4.5 Saturation States and Precipitation Reactions

In addition to waste leaching and microbial degradation reaction controls on trench leachate compositions, the leachate solutes are also subject to abiogenic precipitation reactions, especially in relatively stagnant systems such as those at Maxey Flats and West Valley trenches where the products of waste leaching and microbial degradation are continually generated and subsequently accumulate. To evaluate equilibrium controls on leachate chemistry, WATEQF geochemical code (Plummer et al., 1983) was used to calculate saturation states of trench leachate solutes with respect to selected mineral phases likely to be present.

In the solubility calculations, total titration alkalinity was assumed to be equal to carbonate alkalinity. Only carbonate, sulphate, phosphate, fluoride, sulphide, and oxide mineral phases have been considered. Ideally, such solubility calculations should be done in conjunction with direct analysis of particulate matter in leachate or sediment for detailed chemical or mineralogical composition, especially for the sulphide, oxide, and carbonate phases. However, in the present study, the main emphasis was placed on solute chemistry. No attempt was made to characterize the particulate solid phases.

In Table 4.12 we have presented the mineral solubility products (K_{sp}) and the reactions used in the WATEQF program code to calculate ion activity products (IAPs). Tables 4.13 to 4.16 and Figs. 4.14 to 4.17 show the calculated saturation indices (Log IAP/KT) for the selected carbonate, sulphate, phosphate, fluoride, sulphide, and oxide minerals plotted for each trench leachate sample. The horizontal bars on either side of the equilibrium line (Log IAP/KT = 0) represent 5% tolerance limits on saturation index values due to uncertainties in the values of mineral solubility products.

The solubility calculations indicate that because of strongly reducing conditions Maxey Flats and West Valley leachates tend to show saturation with calcite, dolomite, and rhodochrosite, implying that the Ca^{2+}, Mg^{2+}, Mn^{2+}, and CO_3^{2-} concentrations appear to be controlled by precipitation of these mineral phases. The saturation with respect to the carbonate phases is caused by the high carbonate concentrations resulting from the generation of CO_2 during

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Table 4.12. Solubility products at 25°C and 1 atm for selected mineral phases.

	Reaction	Solubility Product[a] (log Ksp)
$CaCO_3$ (Calcite)	$\rightleftharpoons Ca^{2+} + CO_3^{2-}$	-8.5
$CaMg(CO_3)_2$ (Dolomite)	$\rightleftharpoons Ca^{2+} + Mg^{2+} + 2CO_3^{2-}$	-16.9
$FeCO_3$ (Siderite)	$\rightleftharpoons Fe^{2+} + CO_3^{2-}$	-10.5
$MnCO_3$ (Rhodochrosite)	$\rightleftharpoons Mn^{2+} + CO_3^{2-}$	-10.5
$CaSO_4 \cdot 2H_2O$ (Gypsum)	$\rightleftharpoons Ca^{2+} + SO_4^{2-} + 2H_2O$	-4.8
$BaSO_4$ (Barite)	$\rightleftharpoons Ba^{2+} + SO_4^{2-}$	-10.1
CaF_2 (Fluorite)	$\rightleftharpoons Ca^{2+} + 2F^-$	-12.3
$Fe_3(PO_4)_2 \cdot 8H_2O$ (Vivianite)	$\rightleftharpoons 3Fe^{2+} + 2PO_4^{3-} + 8H_2O$	-36.0
FeS_2 (Pyrite)	$\rightleftharpoons Fe^{2+} + 2S^{2-}$	-18.6
FeS (Amorphous)	$\rightleftharpoons Fe^{2+} + S^{2-}$	-16.9[b]
FeS (Mackinawite)	$\rightleftharpoons Fe^{2+} + S^{2-}$	-17.5[b]
$Fe(OH)_3$ (Amorphous)	$\rightleftharpoons Fe^{3+} + 3OH^-$	-17.9
$FeOOH$ (Goethite)	$\rightleftharpoons Fe^{3+} + 3OH^-$	-13.5

[a]The K_{sp} values are reported by Plummer et al. (1983).
[b]The K_{sp} values for amorphous FeS and mackinawite are those reported by Aller (1980).

microbial degradation of organic matter. High Mn^{2+} concentrations are presumably a result of the reduction of manganese oxide phases during anaerobic degradation process (Aller, 1980; Dayal et al., 1981; Heaton and Dayal, 1985). The Barnwell and Sheffield leachates, on the other hand, generally show distinct undersaturation, indicating the importance of redox conditions and alkalinity in carbonate equilibria.

As expected, most trench leachates exhibit undersaturation with respect to gypsum. This is not surprising, considering that sulphate concentrations are low due to the bacterial sulphate reduction process. In Maxey Flats leachate 37(76), however, the SO_4^{2-} concentration appears to be controlled by gypsum solubility. It should be recalled that this is the only leachate sample that contains considerably high concentration of sulphate (8000 mg/L).

Table 4.13. Ratios of ion activity products to mineral solubility constants (log IAP/KT) for Maxey Flats leachates.[a]

Leachate	Calcite	Dolomite	Siderite	Rhodochrosite	Gypsum	Barite	Fluorite	Vivianite	Pyrite	FeS (Am)	Mackinawite
2(76)	-0.33	0.004	1.74	-0.19	-3.05		-1.27	2.29			
7(76)	0.44	0.89	2.06	-0.21	-2.46		-0.62	3.39			
7(81)	0.51	1.49	1.57	0.59	-0.83	1.75	2.33	2.18	-10.9	0.80	1.40
18(76)	-0.25	0.81	2.08	-1.22	-3.27		-1.71	2.80			
19s(76)	-0.17	0.27	2.17	-0.27	-2.81		-0.98	3.66			
19s(81)	-0.50	-0.42	1.49	-0.74	-3.68	-0.67	0.25	2.14	0.97	6.87	7.47
23(81)	0.09	1.69	1.81	-0.27	-2.97	0.56	-0.39	1.81	-12.5	-10.2	0.41
26(76)	-0.23	0.41	2.03	-0.11	-3.08		-3.87	3.13			
27(Av)	0.75	-1.26	1.40	0.48	-3.00	0.15	-0.42	2.10	9.92	9.04	9.64
27(81)	-0.21	-0.005	1.51	0.49	-3.34	0.29	-0.69	3.44	-5.01	3.26	3.86
30(79)	-0.24	1.29	1.20	-0.67	-2.68	1.04	-4.46	-0.57	-25.7	-9.90	-9.30
32(76)	0.82	2.34	2.10	0.69	-2.86		-1.05	2.43			
33L8(81)	-0.34	-1.05	0.92	-0.59	-2.08	0.16	0.79	0.09	-25.7	-4.67	-4.07
35(81)	1.11	3.52	1.59	0.89	-4.18	-1.01	1.79	0.43	-1.38	4.51	5.11
37(76)	-2.59	-4.39	-0.02	-1.24	-0.01		-1.03	-1.38			

[a]No entry indicates data not available.

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Table 4.14. Ratios of ion activity products to mineral solubility constants (log IAP/KT) for West Valley
leachates.[a]

Leachate	Calcite	Dolomite	Siderite	Rhodochrosite	Gypsum	Barite	Fluorite	Vivianite	Pyrite	FeS (Am)	Mackinawite
2(77)	-1.11	2.81	2.24	-0.12	-3.26	-0.17	0.27	2.89	10.5	9.8	10.5
3(78)	0.78	1.75	2.17	-0.16	-2.23	2.37	-0.08	4.59	8.33	9.96	10.5
4(77)	0.80	1.70	2.25	-0.05	-2.88	2.60	-0.66	4.64	15.5	9.93	10.5
5(78)	0.58	1.08	2.66	0.13	-2.73	1.73	0.28	5.20	9.00	10.0	10.6
9(78)	0.22	0.66	1.72	-0.88	-2.84	0.06	-0.08	4.55	7.30	9.16	9.76

[a]The concentration of H_2S in the leachates was assumed to be 10^{-6} mg/L, because of strongly reducing conditions in the trenches.

Table 4.15. Ratios of ion activity products to mineral solubility constants (log IAP/KT) for Barnwell leachates.

Leachate	Calcite	Dolomite	Siderite	Rhodochrosite	Gypsum	Barite	Fluorite	Vivianite	$Fe(OH)_3$ (Am)	Goethite
6(80)	-2.29	-5.38	1.70	-1.67	-2.29	1.19	-3.74	-4.04	1.25	7.98
8(79)	-0.63	-1.32	-0.34	-0.49	-2.27	1.00	-3.57	-3.01	1.99	8.86
8(80)	0.75	1.37	2.10	0.51	-2.76	-0.12	-3.28	3.66	2.66	9.42
25/21(79)	-2.22	-5.06	-4.62	-2.12	-2.09	1.39	-3.65	-13.9	1.04	7.89
25/21(80)	-2.29	-4.89	-0.65	-2.37	-3.40	0.19	-3.86	-0.99	-0.35	6.30

Table 4.16. Ratios of ion activity products to mineral solubility constants (log IAP/KT) for Sheffield leachates.[a]

Leachate	Calcite	Dolomite	Siderite	Rhodochrosite	Gypsum	Barite	Fluorite	Vivianite	Fe(OH)$_3$ (Am)	Goethite
14A(79)	-2.63	-5.66	-1.53	2.27	-1.63	-3.18	-1.61	-5.92	-4.68	1.79
18(79)	0.22	0.25	-0.65	-0.22	-0.98	1.70	-2.87	-4.01	-0.13	6.40
18(82)	0.58	1.03	-1.18		-0.94		-1.31	-6.24	1.27	7.81

[a]No entry indicates data not available.

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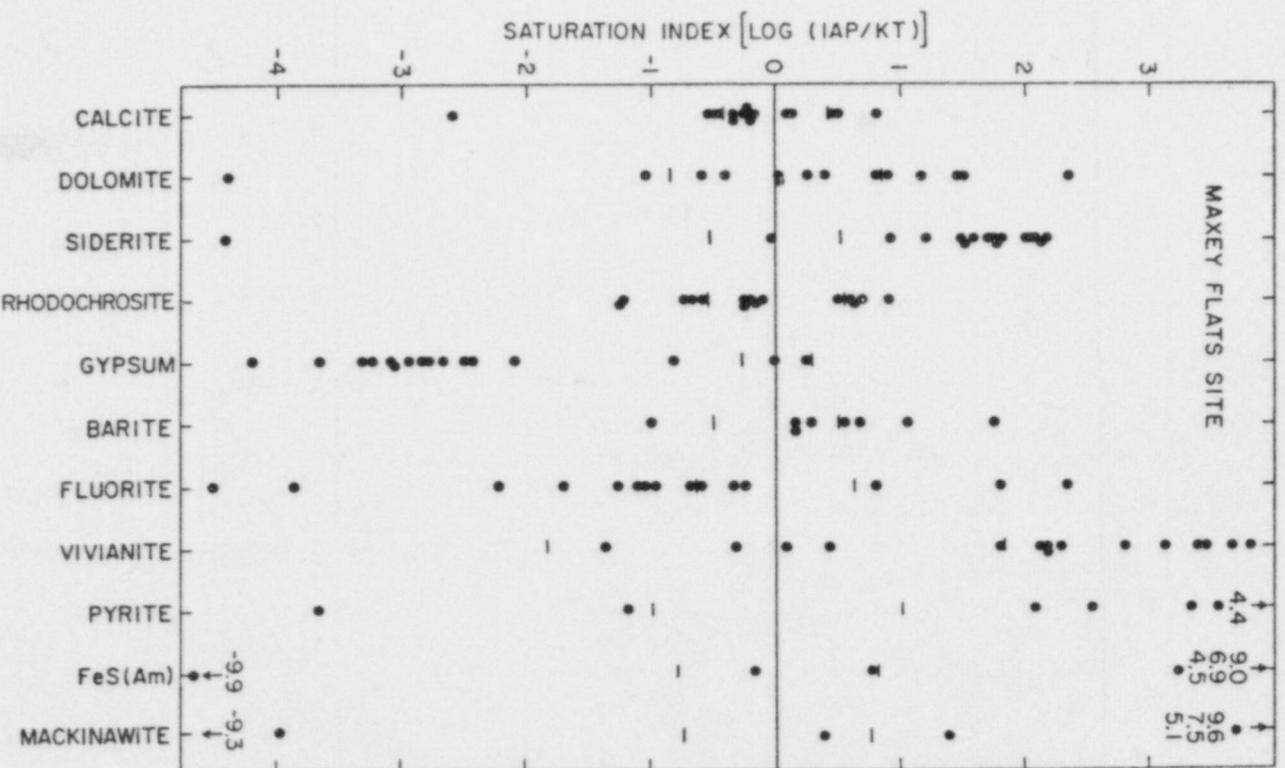
Figure 4.14. Calculated IAPs for the major solutes in Maxey Flats trench trench leachates compared to solubility products (Ksp) of selected minerals likely to form under Maxey Flats trench conditions. The comparisons are made in terms of mineral saturation indices [Log (IAP/KT)]. The data points represent calculated saturation indices based on observed leachate compositions for the various minerals. Horizontal bars represent 5% tolerance limits on saturation indices within which the leachates are believed to be saturated with respect to that mineral.

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SATURATION INDEX $[\log (IAP/KT)]$

-6 -4 -2 0 2 4 6 8

WEST VALLEY SITE

CALCITE

DOLOMITE

SIDERITE

RHODOCHROSITE

GYPSUM

BARITE

FLUORITE

VIVIANITE

PYRITE

10 6
10 5
10 4
10 1

FeS (Am)

MACKINAWITE

Figure 4.15. Calculated IAPs for the major solutes in West Valley trench leachates compared to solubility products (K_{sp}) of selected minerals likely to form under West Valley trench conditions. The comparisons are made in terms of mineral saturation indices [Log (IAP/KT)]. The data points represent calculated saturation indices based on observed leachate compositions for the various minerals. Horizontal bars represent 5% tolerance limits on saturation indices within which the leachates are believed to be saturated with respect to that mineral.

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SATURATION INDEX [log (IAP/KT)]

BARNWELL SITE

-6   -4   -2   0   2   4   6   8

CALCITE

DOLOMITE

SIDERITE

RHODOCHROSITE

GYPSUM

BARITE

FLUORITE

VIVIANITE ← -13.9

Fe(OH)$_3$(Am)

GOETHITE

Figure 4.16. Calculated IAPs for the major solutes in Barnwell trench leachates compared to solubility products (K$_{sp}$) of selected minerals likely to form under Barnwell trench conditions. The comparisons are made in terms of mineral saturation indices. The comparisons are made in terms of mineral saturation indices [Log (IAP/KT)]. The data points represent calculated saturation indices based on observed leachate compositions for the various minerals. Horizontal bars represent 5% tolerance limits on saturation indices within which the leachates are believed to be saturated with respect to that mineral.

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SATURATION INDEX $[\log(IAP/KT)]$

SHEFFIELD SITE

CALCITE

DOLOMITE

SIDERITE

RHODOCHROSITE

GYPSUM

BARITE

FLUORITE

VIVIANITE

Fe(OH)$_3$(Am)

GOETHITE

Figure 4.17. Calculated IAPs for the major solutes in Sheffield trench leachates compared to solubility products (K$_{sp}$) of selected minerals likely to form under Sheffield trench conditions. The comparisons are made in terms of mineral saturation indices [Log (IAP/KT)]. The data points represent calculated saturation indices based on observed leachate compositions for the various minerals. Horizontal bars represent 5% tolerance limits on saturation indices within which the leachates are believed to be saturated with respect to that mineral.

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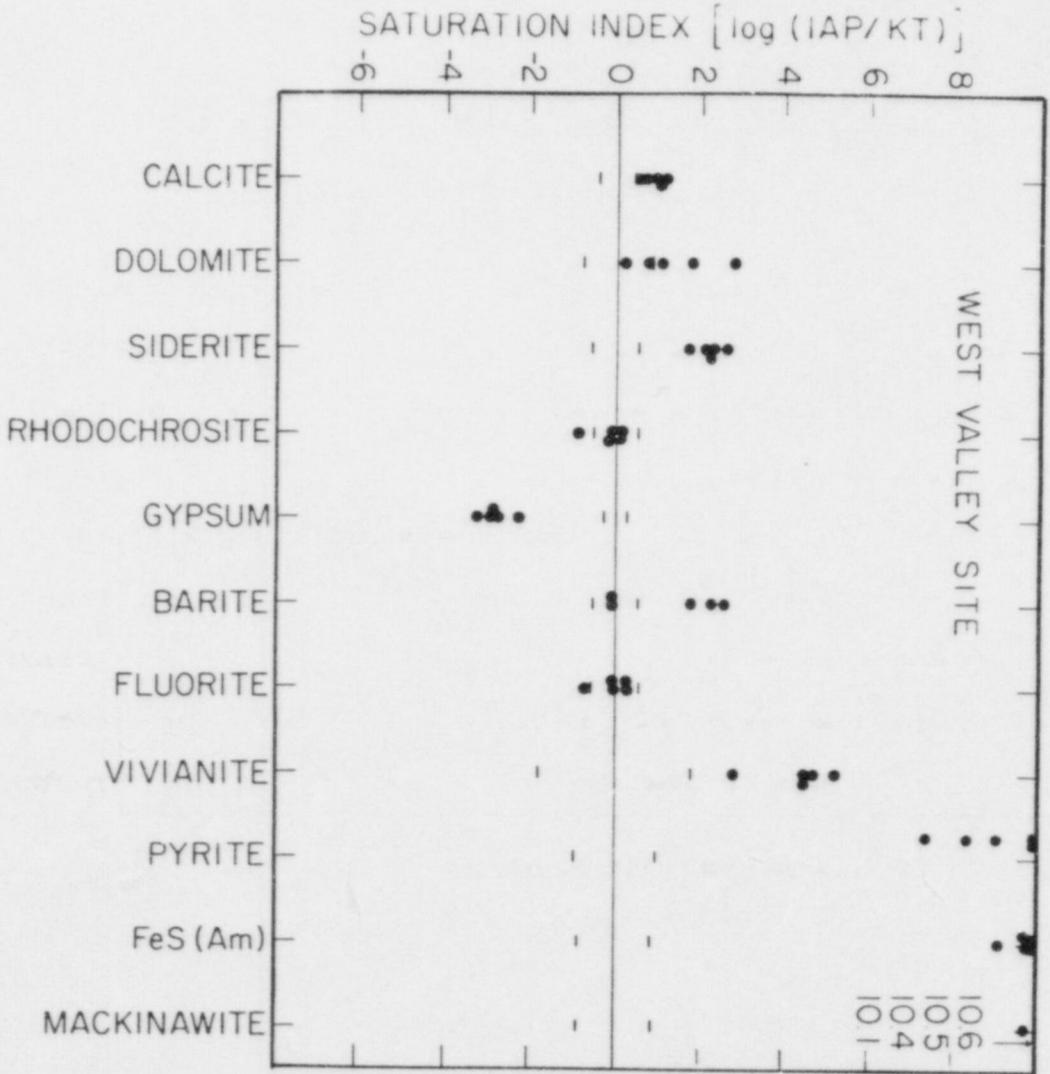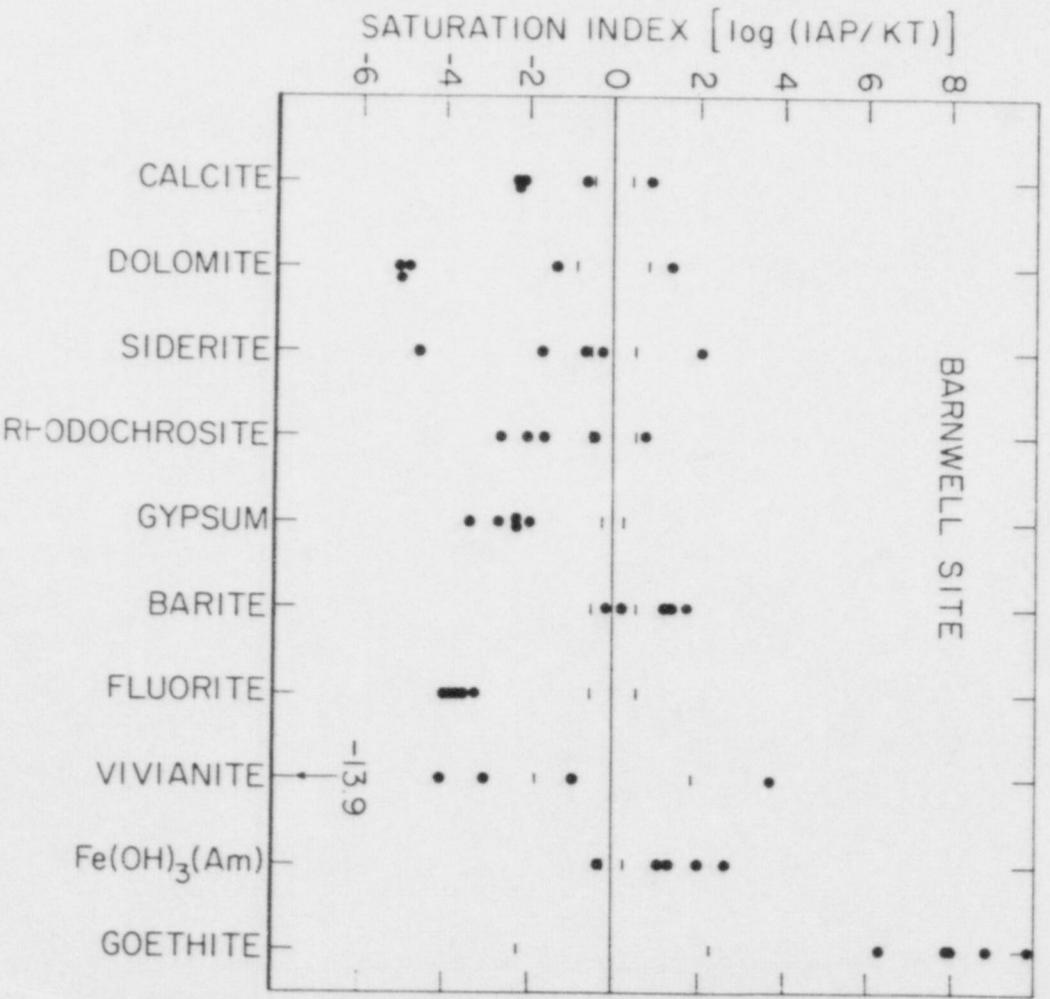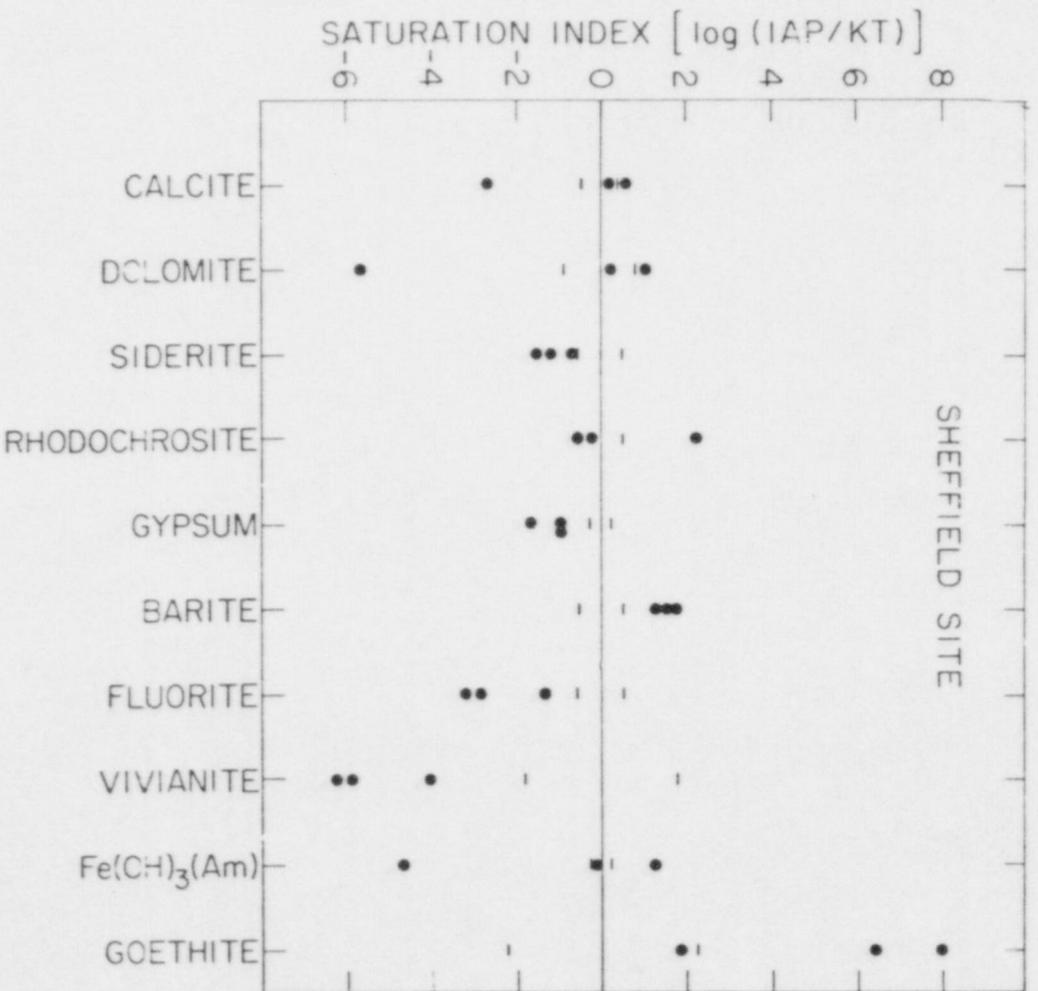In the case of iron equilibria, all Maxey Flats and West Valley leachates are generally distinctly supersaturated with respect to siderite, pyrite, and iron monosulphides (amorphous FeS and mackinawite). Calculated supersaturations can be attributed to several factors: (a) Fe^{2+} is complexed by dissolved organic constituents; (b) the solubility products are incorrect; (c) other phases than carbonate formation are controlling Fe^{2+} concentrations. Consideration of other iron phases indicates that in Maxey Flats leachates [23(81), 30(79), 33L8(81), and 37(76)] and most West Valley leachates Fe^{2+} concentrations may be controlled by vivianite, a ferrous phosphate. It is quite conceivable that Fe^{2+} is complexed with organic chelating agents which have been reported to be present in most Maxey Flats and West Valley leachates. The net effect of iron complexation would be to suppress the Fe^{2+} activity and to increase its solubility. In contrast, since the Barnwell and Sheffield trench leachates are relatively less reducing, these leachates are undersaturated with respect to vivianite and supersaturated relative to the iron hydroxide minerals, goethite and amorphous $Fe(OH)_3$.

In summary, solubility calculations based on trench leachate compositions indicate that calcite, dolomite, and rhodochrosite mineral phases are likely to form in the Maxey Flats and West Valley trench environments and exert control on the concentrations of Ca^{2+}, Mg^{2+}, Mn^{2+}, and CO_3^{2-}. In case of iron equilibria, gross supersaturations are found in these leachates with respect to siderite, mackinawite, amorphous FeS, and pyrite. Because of relatively mildly reducing conditions and correspondingly lower alkalinities, the Barnwell and Sheffield leachates exhibit undersaturation with respect to carbonate phases and supersaturation with respect to the iron oxide minerals.

4.6 Contrasting Geochemistries of Trench Leachates

A combination of hydrological and geochemical factors determines to a large extent the mechanics of leachate formation, evolution of leachate compositions, and development of anoxia in the waste trenches. These, in turn, are reflected in the resultant contrasting leachate geochemistries observed for Maxey Flats and West Valley and for Barnwell and Sheffield sites.

For example, the Maxey Flats and West Valley trenches are excavated in shale and glacial till of relatively low hydraulic conductivity which results in accumulation of rainwater that infiltrates through the trench caps. Because of the relatively long residence time of accumulated water in the trenches at these sites, prolonged leaching and microbial degradation of buried waste material occurs continually, leading to leachate formation. As a result of such interactions for extended time periods, the resultant trench leachates acquire geochemical properties which are unique compared to ambient groundwaters.

In contrast, the Barnwell and Sheffield trenches represent generally well-drained systems, where the infiltrated water has a relatively short residence time and is continually not present in the trenches, thereby

resulting in leachate geochemistries which are not modified, compared to ambient groundwaters, to such an extent as the Maxey Flats and West Valley leachates are.

The development of anoxia observed in practically all the trenches at the sites can be attributed to redox conditions in the trench leachates largely controlled by microbial degradation of organic matter present in waste materials. A large fraction of buried waste in the trenches consists of unconsolidated organic wastes. Many components of these materials are subject to both aerobic and anaerobic microbial degradation processes. As a result of aerobic degradation processes, the dissolved oxygen present in the trenches is consumed rapidly. Following depletion of all molecular oxygen and concurrent development of anoxia in the trenches, further degradation of organic material occurs by anaerobic processes which involve denitrification, followed successively by sulphate reduction and methane generation. During these aerobic and anaerobic degradation processes, there is a continual production and buildup of decomposition products such as carbon dioxide and ammonia, thereby resulting in elevated concentrations of alkalinity, total aqueous CO_2, and ammonia in trench leachates. Concurrently, dissolved oxygen, nitrate, and sulphate are consumed successively, depleting the leachates in these constituents.

Figure 4.18 illustrates the contrasting character of trench leachates in terms of microbial degradation products [$(HCO_3 + CO_3)$, NH_4, and SO_4] from Maxey Flats and West Valley and Barnwell and Sheffield sites, reflecting the nature and intensity of the microbial degradation processes. Given the relatively stagnant water accumulations in the trenches at Maxey Flats and West Valley, the leachates from these sites reflect considerably greater modification of infiltrated water than those from the Barnwell and Sheffield sites. Besides the development of anoxia in the trenches, the Maxey Flats and West Valley leachates are generally characterized by high alkalinity and ammonia concentrations, depletion of sulphate, and negative redox potentials, as well as high concentrations of dissolved iron and manganese, present primarily as Fe^{2+} and Mn^{2+}. In addition, methane generation fermentation reaction, representing an advanced stage of anaerobic degradation processes, is also reported to occur as evidenced from the detection of methane in several trenches at these sites. All these characteristics point to strongly reducing chemical conditions in the Maxey Flats and West Valley trenches.

In contrast, although the Barnwell and Sheffield leachates exhibit a fair degree of anoxia and some alkalinity and ammonia enrichments, no significant sulphate depletion relative to ambient groundwaters, is observed, indicating that the anaerobic degradation processes in these trenches are not as advanced as those reflected by the leachate characteristics of the Maxey Flats and West Valley trenches. This is also evident from the higher redox potentials measured for the Barnwell and Sheffield leachates.

The modifications in ambient groundwaters, as a result of waste leaching, are shown in Fig. 4.19. Generally, the modifications observed in Maxey Flats and West Valley leachates are considerably more pronounced than those observed

Figure 4.18. Ternary plot showing the nature and extent of modification of infiltrated rainwater as a result of microbial degradation processes in Maxey Flats, West Valley, Barnwell, and Sheffield trenches. GFW represents composition of ambient groundwaters.
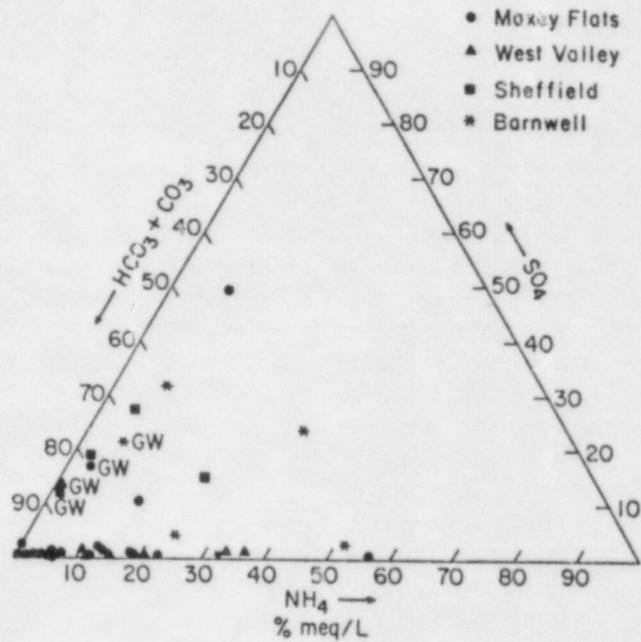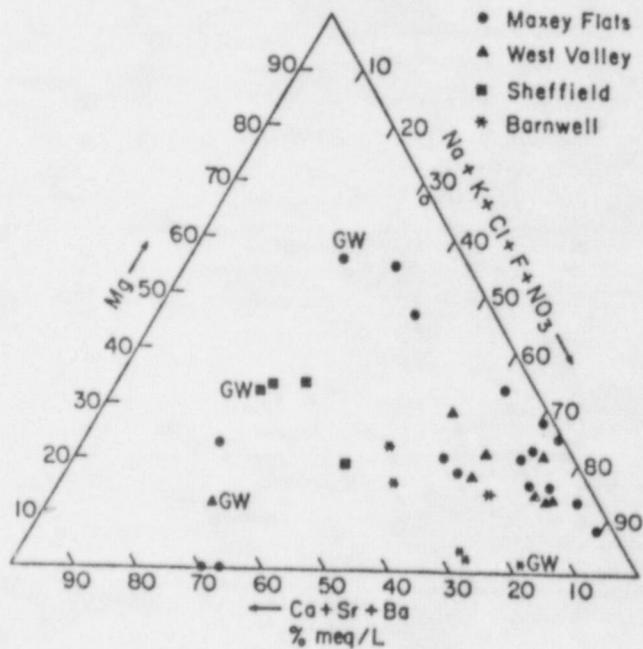

Figure 4.19. Ternary plot showing the nature and extent of modification of infiltrated rainwater as a result of leaching of buried waste materials in Maxey Flats, West Valley, Barnwell, and Sheffield trenches. GW represents composition of ambient groundwaters.

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in Barnwell and Sheffield leachates. This can be attributed to the relatively long residence time of accumulated water in the trenches at the Maxey Flats and West Valley sites, thereby, resulting in continual leaching of waste materials for an extended time period and subsequent buildup of dissolution products. The prolonged leaching of waste materials in the Maxey Flats and West Valley trenches is also reflected in generally higher concentrations of radionuclides in these leachates than those from the Barnwell and Sheffield sites. Several Barnwell leachates exhibit immeasurably low concentrations of Co-60, Sr-90, Cs-134, 137, and the Pu isotopes. Data on specific conductance, ionic strength, total dissolved solids, and dissolved organic carbon, displayed in Fig. 4.20, also show the same general trend. The solubility calculations, based on trench leachate compositions, indicate that because of strongly reducing conditions in the trenches at Maxey Flats and West Valley calcite, dolomite, and rhodocrosite mineral phases are likely to form in these trench environments and exert control on the concentrations of Ca^{2+}, Mg^{2+}, Mn^{2+}, and CO_3^{2-}. In the case of iron equilibria, gross supersaturations are found in these leachates with respect to siderite, mackinawite, amorphous FeS, and pyrite. In contrast, because of relatively mildly reducing conditions and correspondingly lower alkalinities, the Barnwell and Sheffield leachates exhibit undersaturation with respect to the carbonate phases and supersaturation with respect to the iron oxide minerals. These calculations show the importance of contrasting leachate geochemistries, especially redox conditions and alkalinity, in carbonate equilibria.

The acid-generating potential and buffering capacity of a leachate are important geochemical controls that maintain a balance between acidity and alkalinity which, in turn, determines to a large extent the reactivity, partitioning, cycling, and mobility of radionuclides along a redox gradient. In this respect, the Maxey Flats and West Valley leachates generally represent well-buffered systems and exhibit high acid-generating potentials, whereas the Barnwell and Sheffield leachates are characterized by relatively low buffering capacities and acid-generating potentials.

5. RELEVANCE OF RESEARCH FINDINGS TO NRC NEEDS

Modeling of low-level radioactive waste disposal sites is an important consideration in evaluating the capability of future sites to conform with 10CFR61 of federal regulations code. In this respect, the BNL study on characterizations of trench leachates from existing low-level waste disposal sites in the eastern United States deals directly with defining the source term under a range of hydrological and geochemical conditions. In addition to providing information on source term characteristics, the BNL study is unique in that it represents the first detailed geochemical investigation of the sites which attempts to evaluate, based on trench leachate data, not only the fate and behavior of waste materials in a natural setting but also the effects of disposal of unsegregated, unstabilized, and poorly packaged waste on trench environments.

This study has provided an understanding of the importance of hydrological and geochemical factors in controlling the mechanics of leachate formation, evolution of leachate compositions, microbial degradation of organic
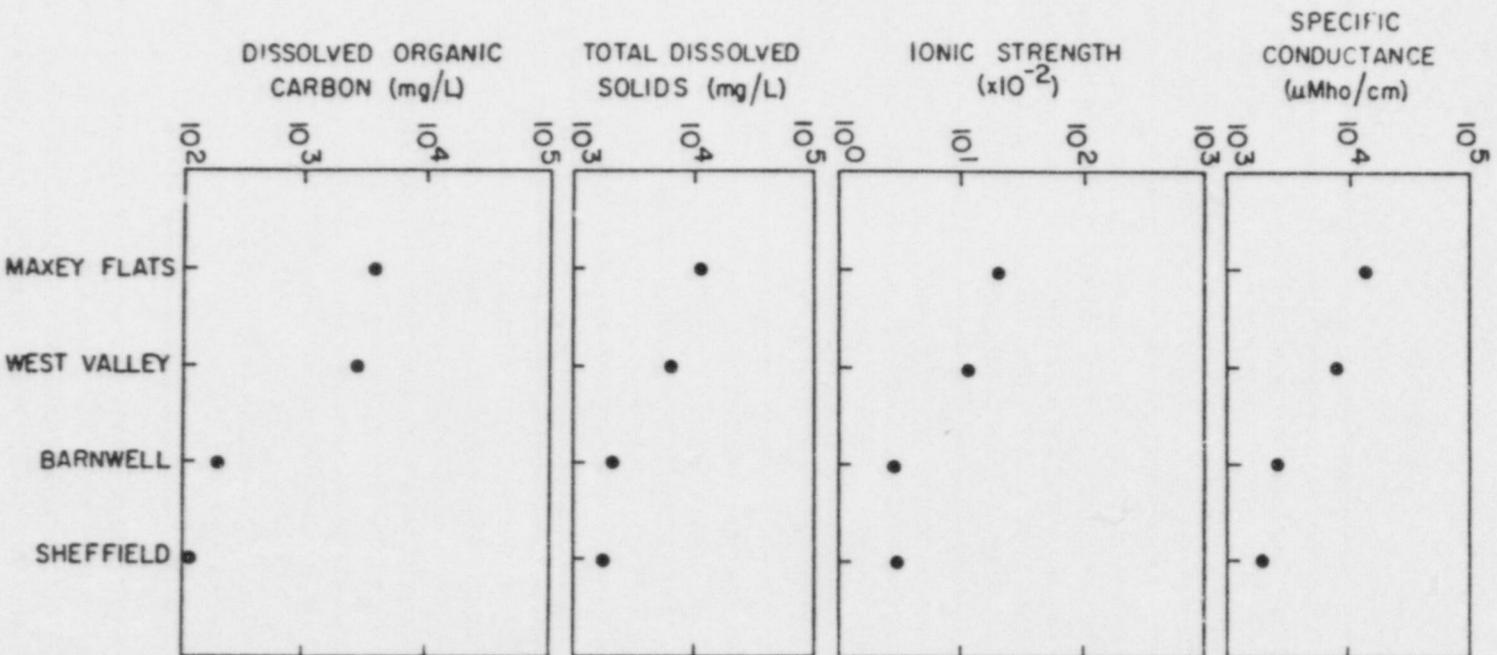
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Figure 4.20. Maximum values of dissolved organic carbon, total dissolved solids, ionic strength, and specific conductance observed in leachates from Maxey Flats, West Valley, Barnwell, and Sheffield site'.

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waste and development of anoxia in the trenches, and nature and extent of leaching of waste materials. The varying degrees of the intensity of these processes in the trenches, as determined by the different site characteristics, are clearly reflected in the contrasting leachate geochemistries of Maxey Flats and West Valley trenches, as compared to those of Barnwell and Sheffield trenches. These are important geochemical considerations which not only define the source term but also shed light on the nature and extent of oxidation-induced changes that are likely to occur along a redox gradient outside of the trench environment. For example, the acid-generating potential and buffering capacity of strongly anoxic chemically reducing source terms are important geochemical controls that maintain a balance between acidity and alkalinity which, in turn, determine the reactivity, partitioning, cycling, and mobility of radionuclides along a redox gradient. Thus, it follows that the modifications likely to occur in the extra-trench environment could very well result in a drastically different source term geochemistry and that they are direct functions of the initial geochemical character of the source term, which implies that, in addition to radionuclide data, other characteristics such as the redox potential, buffering capacity, acid-generating potential, dissolved iron concentration, and organic constituents should be known to effectively use the source term as an input parameter for modeling.

Unsegregated, poorly packaged, and unstabilized wastes, which were buried at the sites, are readily leachable and biodegradable. Especially at the Maxey Flats and West Valley sites, where the infiltrated water accumulated in the trenches leading to a bathtub effect, the relatively long residence time of accumulated water results in continually intense leaching and microbial degradation of waste materials for extended time periods. The net effect of these degradation processes with time is development of large void spaces within the trenches and subsequent trench cap subsidence, followed by increased infiltration and accumulation of water in the trenches. This, in turn, contributes to a greater potential for leaching, mobilization, and migration of radionuclides. Some of the organic complexing agents present in unsegregated waste have the potential to form stable radionuclide complexes, thereby resulting in enhanced nuclide mobility. Furthermore, because of the extreme heterogeneity of the unsegregated waste, both in character and composition, the source terms are highly variable from one trench to another, or even within the same trench.

The problems associated with disposal of unsegregated, poorly packaged, and unstabilized wastes, especially at the Maxey Flats and West Valley sites, have helped establish the consequences of past disposal practices and site characteristics on waste package performance, the trench environment and stability, source term characteristics and variability, leaching and microbial degradation of waste materials, and radionuclide behavior and mobility. Although the current disposal procedures are much improved from those practiced in the past, it is likely that several of these factors, conditions, and processes observed by BNL at these sites can be expected to occur at future shallow land burial sites.

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The observations made at the existing sites have important implications for site selection and disposal of low-level radioactive wastes at future shallow land burial sites in that waste package requirements will include waste segregation, improved stabilization, and proper packaging. In accordance with current disposal procedures, stabilized, packaged waste not only ensures trench stability but also decreases the rate and extent of leaching and microbial degradation of waste materials. In addition, the uncertainties in the source term are considerably reduced. Furthermore, to avoid water accumulations in the trenches, future sites will most likely be located in relatively well-drained systems, such as that represented by the Barnwell site, where the residence time of accumulated water in the trenches is relatively short.

Many of the current waste package requirements and site selection criteria outlined in 10CFR61 for the shallow land disposal of low-level radioactive wastes are based on lessons learned from experiences at existing shallow land burial sites.

6. SUMMARY AND CONCLUSIONS

The major features of this study involving geochemical characterizations of trench leachates from existing low-level radioactive waste disposal sites are highlighted in this section. The relevance of these research findings to NRC needs is also summarized.

This study has provided an understanding of the importance of hydrological and geochemical factors in controlling the mechanics of leachate formation, evolution of leachate compositions, microbial degradation of organic waste and development of anoxia in the trenches, and nature and extent of leaching of waste materials. The varying degrees of the intensity of these processes in the trenches, as determined by the different site characteristics, are clearly reflected in the contrasting leachate geochemistries of Maxey Flats and West Valley trenches, as compared to those of Barnwell and Sheffield trenches. These are important geochemical considerations which not only define the source term but also shed light on the nature and extent of oxidation-induced changes that are likely to occur along a redox gradient outside of the trench environment.

The Maxey Flats and West Valley trenches are excavated in shale and glacial till of relatively low hydraulic conductivity which results in accumulation of rainwater that infiltrates through the trench caps. Because of the relatively long residence time of accumulated water in the trenches at these sites, prolonged leaching and microbial degradation of waste materials occurs continually, leading to leachate formation. As a result of such interactions for extended time periods, the resultant trench leachates acquire geochemical properties which are unique and considerably modified compared to ambient groundwaters.

The Barnwell and Sheffield trenches, on the other hand, represent generally well-drained systems, where the infiltrated water has a relatively

short residence time and is continually not present in the trenches, thereby resulting in leachate geochemistries which are not modified, compared to ambient groundwaters, to such an extent as the Maxey Flats and West Valley leachates are.

The development of anoxia observed in practically all the trenches at the sites can be attributed to redox conditions in the trench leachates largely controlled by microbial degradation of organic matter present in waste materials. As a result of aerobic degradation processes, the dissolved oxygen present in the trenches is consumed rapidly. Following depletion of all molecular oxygen and concurrent development of anoxia in the trenches, further degradation of organic material occurs by anaerobic processes which involve denitrification, followed successively by sulphate reduction and methane generation. During these degradation processes, there is a continual production and buildup of decomposition products such as carbon dioxide and ammonia, thereby resulting in elevated levels of alkalinity, total aqueous CO_2, and ammonia in trenches. Concurrently, dissolved oxygen, nitrate, and sulphate are consumed, depleting the leachates in these constituents.

Given the relatively stagnant water accumulations in the trenches at Maxey Flats and West Valley, the leachates exhibit strongly anoxic, reducing conditions, as reflected by negative redox potentials, depletion of dissolved oxygen and sulphate, high alkalinity and ammonia concentrations, as well as high concentrations of dissolved iron and manganese, present primarily as Fe^{2+} and Mn^{2+}.

Although the Barnwell and Sheffield leachates exhibit a fair degree of anoxia, as reflected in depletion of dissolved oxygen and some alkalinity and ammonia enrichments, no significant sulphate depletion relative to ambient groundwaters is observed, indicating that the anaerobic degradation processes are not as advanced as those reflected by the leachate characteristics of Maxey Flats and West Valley trenches. This is also evident from the relatively higher redox potentials measured for the Barnwell and Sheffield leachates.

The modifications in ambient groundwaters, as a result of waste leaching processes, are also reflected in contrasting character of leachate geochemistries. The enrichments, to varying degrees, of inorganic, organic, and radionuclide constituents associated with fuel cycle and non-fuel cycle LLW reflect not only the nature of the leaching process and reactivity and amount of waste materials but also the differences in the site characteristics.

Relative to ambient groundwaters, the trench leachates are generally enriched in Na^+, K^+, Fe_T, Mn_T, Cl^-, dissolved organic and inorganic carbon and several organic compounds, as well as radionuclides such as H-3, Co-60, Cs-134, Cs-137, Sr-90, Pu-238, Pu-239, -240, Am-241, attributable to leaching of waste materials. Because of the relatively long residence of accumulated water in the trenches at Maxey Flats and West Valley leading to prolonged leaching of waste materials and subsequent buildup of dissolution products, the Maxey Flats and West Valley leachates exhibit modifications which are

generally considerably more pronounced than those observed in the Barnwell and Sheffield leachates. This is also reflected in generaly higher values for specific conductance, ionic strength, and total dissolved solids in Maxey Flats and West Valley leachates.

The solubility calculations indicate that because of strongly reducing conditions in the trenches at Maxey Flats and West Valley calcite, dolomite, and rhodochrosite mineral phases are likely to exert control on the concentrations of Ca^{2+}, Mg^{2+}, Mn^{2+}, and CO_3^{2-}. In case of iron equilibria, gross supersaturations are found in these leachates with respect to siderite, mackinawite, amorphous FeS, and pyrite. In contrast, because of relatively mildly reducing conditions and correspondingly lower alkalinities, the Barnwell and Sheffield leachates exhibit undersaturation with respect to the carbonate phases and supersaturation with respect to the iron oxide minerals. The calculations show the importance of contrasting leachate geochemistries, especially redox potential and alkalinity, in carbonate and iron equilibria.

The acid-generating potential and buffering capacity of a leachate are important geochemical controls that maintain a balance between acidity and alkalinity which, in turn, determines to a large extent the reactivity, partitioning, cycling, and mobility of radionuclides along a redox gradient. In this respect, the Maxey Flats and West Valley leachates generally represent well-buffered systems and exhibit high acid-generating potentials, whereas the Barnwell and Sheffield leachates are characterized by relatively low buffering capacities and acid-generating potentials.

The problems associated with disposal of unsegregated, poorly packaged, and unstabilized wastes, especially at the Maxey Flats and West Valley sites, have helped establish the consequences of past disposal practices and site characteristics on waste package performance, the trench environment and stability, source term characteristics and variability, leaching and microbial degradation of waste materials, and radionuclide behavior and mobility.

The observations made at the existing sites have important implications for site selection and disposal of low-level radioactive wastes at future shallow land burial sites in that waste package requirements will include waste segregation, improved stabilization, and proper packaging. In accordance with current disposal procedures, stabilized, packaged waste not only ensures trench stability but also decreases the rate and extent of leaching and microbial degradation of waste materials. In addition, the uncertainties in the source term are considerably reduced. Furthermore, to avoid water accumulations in the trenches, future sites will most likely be located in relatively well-drained systems, such as that represented by the Barnwell site, where the residence time of accumulated water in the trenches is relatively short.

Many of the current waste package requirements and site selection criteria outlined in 10CFR61 for the shallow land disposal of low-level radioactive wastes are based on lessons learned from experiences at existing shallow land burial sites.

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APPENDIX A

CHEMICAL CONSTITUENTS IN FUEL AND NON-FUEL CYCLE WASTES
(General Research Corporation, 1980)

Table A-1. Chemical characteristics of fuel and non-fuel
cycle low-level waste constituents.

Descriptive Class	Examples	Fuel Cycle	Non-Fuel Cycle
Agricultural compounds	DDT, malathion, $[^{14}C, ^{32}P, ^{35}S]$		X
Alcohols	methanol, cyclohexanol, diacetone		X
Aldehydes and acetals	paraldehyde, 1, 1-diethoxyethane		X
Aliphatic halogenated hydrocarbons	1,1,1-trichloroethane		X
Alkanes	cyclohexane		X
Alkenes	2,3-dimethyl-2-hexene		X
Alkyl halides	1-isobutyl-4-ethyloctyl bromide		
Amino acids	alanine, tryptophan $[^{3}H, ^{14}C, ^{35}S]$		X
Amino sugars			X
Ammonia and ammonium salts	ammonia, ammonium sulfate	X	
Aromatic hydrocarbons	benzene, toluene, p-xylene anthracenes, dibenzene anthracene $[^{14}C]$		X
Asphalt	natural or petroleum-derived	X	
Carboxylic acids	citric acid, tartaric acid oxalic acid, hydroxy-acetic acid	X	X
Chelating agents	EDTA	X	X
	DTPA	X	X
	NTA		
	TTHA	X	X
Carbohydrates	2-deoxy-d-glucose, $[^{3}H, ^{14}C]$		X
Drugs	acetylsalicylic acid, actino-mycin D, $[^{14}C, ^{35}S]$		X

Table A-1. Chemical characteristics of fuel and non-fuel cycle low-level waste constituents. (continued)

Descriptive Class	Examples	Fuel Cycle	Non-Fuel Cycle
Esters			
Acetates	ethyl acetate		X
Adipates	dioctyl adipate, bis-2-ethyl-hexyl adipate		X
Biocarbons	natural or metabolic products of organisms		X
Borates	sodium tetraborate, metaborate boric acid, borate polymers	X	
Heptonates	stannous glucoheptonate		X
Phosphates	chromic phosphate, tributyl phosphate, sodium phosphate, stannous polyphosphate, orthophosphates, ^{32}P		X
Phthalates	di-2-ethylhexyl, diethyl dibutyl, dioctyl phthalates	X	X
Sulfates	ammonium sulfates, calcium sulfate, sodium sulfate, other sulfate salts, ^{35}S	X	X
Ethers	bis(2-chloroethyl) ether, tetrahydrofuran 1,4-dioxane		X
Fatty acids	linoleic acid, oleic acid, oxalic acid, palmitic acid, [^{3}H, ^{14}C]		X
Hormones	histamine dihydrochloride		X
Inorganic acids	boric acid, phosphoric acid, sulfuric acid, [^{32}P]	X	X
Ketones	methyl isobutyl ketone, d-fenchone		X

Table A-1. Chemical characteristics of fuel and non-fuel
cycle low-level waste constituents. (continued)

Descriptive Class	Examples	Fuel Cycle	Non-Fuel Cycle
Lipid-related products	acetylocholine chloride, sphingo-myelin [^{3}H, ^{14}C]		X
Miscellaneous inorganic	calcium fluoride, chloride, copper, diatomaceous earth, fluoride, portland cement, Mg, Zn, sodium hydroxide, sodium iodide	X	X
Nitrosamines	N,N-dimethylnitrosamine		X
Nucleosides	---		X
Nucleotides	adenosine-3',5'-cyclic phosphate [^{3}H, ^{14}C, ^{32}P, ^{125}I]		X
Nucleotide sugars	guanosine diphospho-1-fucose, [^{14}C]		X
Organic acids	acetic acid, benzoic acid, iodoacetic acid, phenylhexanoic acid		X
Peptides	---		X
Phenol and phenolic compounds	cresols, p-tert-butyl phenol		X
Prostaglandins [^{3}H, ^{19}C]	Prostaglandin E$_2$		X
Proteins	---		X
Protein-labelling reagents	acetic anhydride, 1-fluoro-2,4-dinitrobenzene, phenylisothiocyanate, [^{3}H, ^{14}C, ^{32}P, ^{35}S, ^{125}I, ^{131}I]		X

Descriptive Class	Examples	Fuel Cycle	Non-Fuel Cycle
Purines	adenine, guanine sulfate [^{3}H, ^{14}C]		X
Pyrimidines	uracil [^{3}H, ^{14}C]		X
Radionuclides	Ba-137m, 139, 140	X	
	C-14		X
	Ce-144	X	
	Co-58, 60	X	
	Cs-134, 136, 137, 138	X	
	Cr-51	X	
	H-3		X
	I-125, 131		X
	I-129	X	
	I-130, 132, 133, 134, 135	X	
	Fe-55, 59	X	
	La-140	X	
	Mn-54	X	
	Mo-99	X	
	Na-22		X
	P-32		X
	Pu	X	
	Rb-88		X
	Sr-89, 90, 91, 92	X	
	S-35		X
	Tc-99		X
	Th	X	
	U-naturals and daughters	X	
	Y-90, 91	X	
	Zr-95	X	
Resins	ion-exchange, urea		X
	formaldehyde	X	
Steriods	aldosterone, estradiol,		X
	progesterene		X
Vitamins	nicotinamide		X

Table A-2. Summary of chemical constituents in fuel cycle low-level wastes.

Organic Compounds	Elemental Species
asphalt (solidification agents)	barium-137m, 139, 140
urea-formaldehyde resins	bromine-84
(may contain alkali metal	cerium-144
bisulfates)	cesium-134, 136, 137, 138
carboxylic acids (decontaminating	chloride (Cl)
chemicals)	chromium-51
chelating agents	oxide or hydrated oxide
(NTA, EDTA, DTPA, TTHA)	cobalt-58,60
	oxide or hydrated oxide
ion exchange resins	copper
(sulfonated and aminated organic	fluoride
polymers; modified polystrenes,	F⁻, metallic fluorides
copolymers of divinyl benzene	iodine-129, 131, 132, 133, 134, 135
and styrene are typical substrates)	iron-55, 59
phthalates	oxide or hydrated oxide
(for filter testing)	lanthanum-140
vinyl ester styrene	magnesium
	manganese-54
Inorganic Compounds	oxide or hydrated oxide
	molybdenum-99
ammonia	niobium-95
ammonium sulfate	Pu
boric acid	rubidium-103, 106
calcium fluoride	strontium-89, 90, 91, 92
calcium sulfate	uranium
sodium tetraborate, metaborate	natural U and daughters
portland cement	Tellurium-132, 134
(with or without bentonite or	Th
vermiculite)	Tritium
diatomaceous earth	Yttrium-90, 91
	zinc
	zirconium-95
	oxide or hydrated oxide

Table A-3. Summary of identifiable chemicals in non-fuel cycle wastes.

Scintillation Vials	Other Liquids	Solid Waste	Biological Waste
toluene, xylene, sodium hydroxide; (with small amounts of methanol); aquasols, POP	50% is spent scintillation fluids. Other liquids are ogranic waste, alcohol, benzene, ethyl acetate, and RIA Kits which may contain any of the following compounds: ammonium sulfate, sodium barbital, sodium azide, ammonium salt, sulfonic acids, colloids, stannous polyphosphate, stannous glucoheptonate, amino sugars, amino acids, nucleosides, nucleotides, protein hydrolysates, pyruvic acid, NaHCO$_3$, anthracene compounds, phosphoric acid, iodoacetic acid, Bolton-Hunter reagent, iododeoxyuridine, sodium iodide, organic phosphates, orthophosphates, methionine, tritiated thymidine, ethidium bromide (intercalating dye), paradimethyl amino benzalide (PDAB), ethylenediaminetetraacetic acid (EDTA) additional possible liquid waste could be solutions of any of the labeled compounds found in Appendix A-1	cellulosics, PVC plastics, rubber, glass, metal filings, other trash.	animal carcasses, cultures disinfected with Clorox[R], Beau Coup[R] (a phenol solution), or Wescodyn[R] (iodine base); or contaminated animal wastes.

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R. Kempf   T. Sullivan
H. J. C. Kouts   H. Todosow (2)
S. G. Lane (2)   A. J. Weiss

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ABSTRACT

The results of source term characterization studies for the commercially operated low-level waste (LLW) disposal sites located in the eastern United States are used to provide an understanding of the importance of hydrological and geochemical factors in controlling the mechanics of leachate formation, evolution of leachate compositions, microbial degradation of organic waste and development of anoxia in the trenches, and the nature and extent of leaching of waste materials. The varying degrees of the intensity of these processes, as determined by the different site characteristics, are clearly reflected in the contrasting leachate geochemistries of Maxey Flats and West Valley trenches, as compared to those of Barnwell and Sheffield trenches. These are important geochemical considerations which not only define LLW source terms but also shed light on the nature and extent of geochemical changes that are likely to occur along a redox gradient outside of the trench environment.

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