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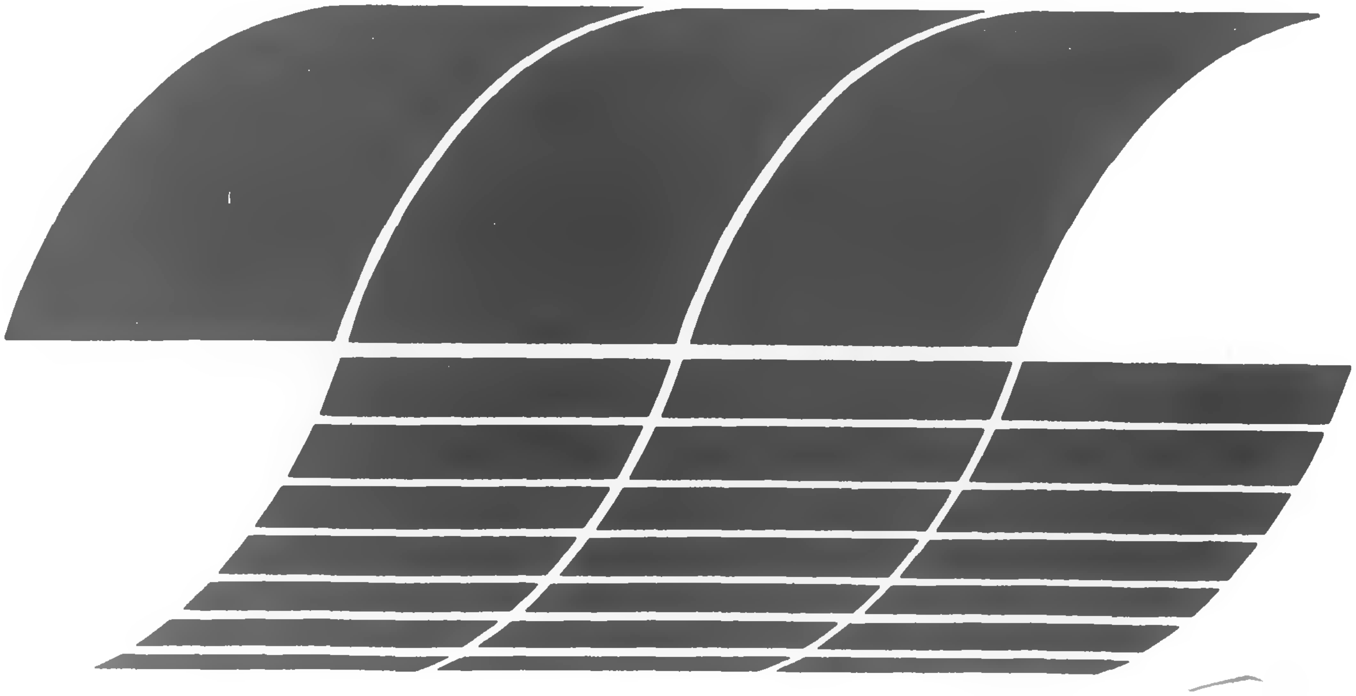
Trace Element Characterization of Coal Wastes - Third Annual Progress Report

Interagency Energy/Environment R&D Program Report



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Trace Element Characterization of Coal Wastes -

Third Annual Progress Report

October 1, 1977 to September 30, 1978

by

E. M. Wewerka, J. M. Williams, L. E. Wangen, J. P. Bertino,
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CONTENTS

ABSTRACT 1

EXECUTIVE SUMMARY 1

SUMMARY OF TASK PROGRESS 4

TASK PROGRESS DESCRIPTION 7
  Task 1. Environmental Control Technology for Trace Elements in the Drainage From High-Sulfur Coal Preparation Wastes 7
  Task 2. Identify Trace Elements of Environmental Concern in High-Sulfur Coal Preparation Wastes From the Appalachian Region 40

PERSONNEL 44

APPENDIX A. Column Leaching of Calcined Refuse 46

APPENDIX B. Preliminary Cost Comparisons for Selected Environmental Control Options for Contaminated Coal Refuse Drainage 55

APPENDIX C. Column Leaching Studies of Limestone/Refuse Mixtures 65

APPENDIX D. Column Leaching Studies of Lime/Refuse Mixtures 76

APPENDIX E. Program Code for Determining the Cost of Alkaline Neutralization of Coal Waste Drainages 82

REFERENCES 84

TABLES

I. Effect of Calcining on Trace Element Retention From Plant B Coal Refuse 8

II. Presence of Coal Refuse Minerals at Various Calcining Temperatures 9

III. High-Temperature Mineral Transformations of High-Sulfur Coal Refuse 9

IV. Effect of Calcining Temperature on the Leachability of a High-Sulfur Coal Refuse Material 10

V. Trace Element Leachability of a High-Sulfur Coal Refuse Sample Calcined at 1000°C 11

VI. Conditions of Preleaching Experiments Conducted on a High-Sulfur Coal Refuse Material 12

VII. Elemental Analyses of Leachates From Preleaching of Plant B Coal Refuse 13

VIII. Effectiveness of Leaching Agents at Removal of Trace Elements From Plant B Coal Refuse 14

IX. Effectiveness of Ferric Sulfate Treatment on Iron Removal From Coal and Coal Waste 15

X. Analyses of Jemez Limestone Used in Neutralization Control Technology Experiments 17

XI. Description of Dynamic Leaching Studies of High-Sulfur Refuse/Limestone Mixtures 17

XII. Results of Experiments Using Eleven Sorbents for pH Control and Trace Element Attenuation for an Illinois Basin High-Sulfur Coal Refuse Leachate 23

XIII. Comparison of Capabilities of Eleven Sorbents to Elevate pH and to Attenuate Thirteen Trace Elements in Illinois Basin Coal Refuse Leachates 24

XIV. Trace Element Analyses of Soil/Refuse Leachate Equilibrations 25

XV. Batch Attenuation of Trace Elements in Coal Refuse Leachates by Soils 26

XVI. Results From a Column Leaching Experiment With Calcite-Coated Illinois Basin Coal Refuse 28

XVII. Analyses for the Control of Refuse Drainage by Alkaline Neutralization 31

XVIII. Experiment Identification and Catalog Description of Resins Used in Bio-Rad's Ion-Exchange Experiments on High-Sulfur Coal Refuse Leachates .. 32

XIX. Summary of pH, TDS, and Trace Element Compositions Resulting From Ion-Exchange Treatment of a High-Sulfur Coal Refuse Leachate 33

XX. Trace Element Analyses on Reverse Osmosis Experiments 35

XXI. Alkaline Neutralization/Permanganate Oxidation of Contaminated Coal Refuse Drainage 37

XXII. Matrix Grid Summary of Environmental Control Options for Contaminated Coal Refuse Drainage 38

XXIII. Matrix Grid Summary of Environmental Control Options for Contaminated Coal Refuse Drainage 39

XXIV. Mineral Compositions of Coal Refuse Samples 40

XXV. Trace Element and Mineral Content of Coal Waste from Appalachian Plant G 42

XXVI. Trace Element Compositions of Coal Refuse Samples 43

XXVII. Static Leaching of Low-Sulfur Appalachian Plant G Waste 43

XXVIII. MEG/MATE Analyses of Plant G Coal Refuse Leachate 46

A-I. Experiment Identification for Dynamic Leaching Studies of Calcined Refuse 46

A-II. Analyses for Dynamic Leaching Studies of Calcined Refuse, Experiment No. GL-12 47

A-III. Analyses for Dynamic Leaching Studies of Calcined Refuse, Experiment No. GL-18 48

B-I. Landfill Growth and Mass Flow of Water Through Waste Pile During 20-Yr Active Life of Pile 60

B-II. Lime Requirements for Various Treatments of Coal Waste 61

B-III. Fly Ash Demand Requirements for Codisposal Treatment of Coal Waste 61

B-IV. Costs of Various Drainage Treatment/Prevention Processes 62

B-V. Annual Costs of Various Drainage Treatment/Prevention Processes 63

B-VI. Net Present Value of Costs for Various Drainage Treatment/Prevention Processes 64

C-I. Experiment Identification for Dynamic Leaching Studies of Limestone/Refuse Mixtures 65

C-II. Analyses for Dynamic Leaching Studies of Limestone/Refuse Mixtures, Experiment No. GL-12 66

C-III. Analyses for Dynamic Leaching Studies of Limestone/Refuse Mixtures, Experiment No. GL-14 67

C-IV. Analyses for Dynamic Leaching Studies of Limestone/Refuse Mixtures, Experiment No. GL-15 68

C-V. Analyses for Dynamic Leaching Studies of Limestone/Refuse Mixtures, Experiment No. GL-16 69

C-VI. Analyses for Dynamic Leaching Studies of Limestone/Refuse Mixtures, Experiment No. GL-17 70

D-I. Experiment Identification for Codisposal of Lime and Coal Wastes 76

D-II. Analyses for Dynamic Leaching Studies of Lime/Refuse Mixtures, Experiment No. CTWT-11-1 (Control) 76

D-III. Analyses for Dynamic Leaching Studies of Lime/Refuse Mixtures, Experiment No. CTWT-11-2 (0.5 wt% Lime) 77

D-IV. Analyses for Dynamic Leaching Studies of Lime/Refuse Mixtures, Experiment No. CTWT-11-3 (1.5 wt% Lime) 77

D-V. Analyses for Dynamic Leaching Studies of Lime/Refuse Mixtures, Experiment No. CTWT-11-4 (3 wt% Lime) 78

D-VI. Analyses for Dynamic Leaching Studies of Lime/Refuse Mixtures, Experiment No. CTWT-11-5 (10 wt% Lime) 78

FIGURES

1. Leachate pH and TDS versus leachate volume for column leaching study of limestone/refuse mixtures 18

2. Leachate pH and TDS versus leachate volume for column leaching study of lime/refuse mixtures 20

3. Iron levels in aqueous streams of a reverse osmosis system 36

4. Leachate pH and TDS versus leachate volume for column leaching study of Plant G refuse 45

A-1. The pH, TDS, and trace element concentrations for dynamic leaching experiments with calcined refuse 49

C-1. The pH, TDS, and trace element concentrations for dynamic leaching experiments with limestone/refuse mixtures 71

D-1. The pH and trace element concentrations for dynamic leaching experiments with lime/refuse mixtures 79

TRACE ELEMENT CHARACTERIZATION OF COAL WASTES
THIRD ANNUAL PROGRESS REPORT

October 1, 1977 to September 30, 1978

by

E. M. Wewerka, J. M. Williams, L. E. Wangen, J. P. Bertino,
P. L. Wanek, J. D. Olsen, E. F. Thode, and P. Wagner

ABSTRACT

In 1978, we performed laboratory experiments to investigate the efficacy of several control options for treating coal wastes at the preparation plant or during disposal. Our research revealed that calcining is one of the more effective and permanent means of treating high-sulfur coal wastes before disposal to decrease, quite dramatically, the release of environmentally undesirable pollutants into the drainages from disposal sites. Another promising control method is codisposal of the coal wastes with lime or limestone to neutralize the acid drainage and retain soluble aqueous contaminants within the waste site. Other experiments have examined the feasibility of using natural sealants, such as clays, soils, calcite, and cements, to isolate the disposal site from its immediate environment. The various tradeoffs for these control options are discussed in terms of contaminant reduction, complexity, permanency, and cost.

We have begun an assessment of coal preparation wastes from the Appalachian region. Based on the work we have done on refuse from a single plant, it is clear that coal wastes containing a low percentage of pyrite (<1%) generate worrisome amounts of acid drainage. Our experimental results show that the trace elements of environmental concern in the leachates from these low-sulfur wastes are aluminum, manganese, iron, nickel, and copper when their concentrations are in excess of the Environmental Protection Agency's recommended Minimum Acute Toxicity Effluent (MATE) values.

EXECUTIVE SUMMARY

The mineral wastes from coal preparation and coal mine development constitute a major environmental problem in the United States.[1] More than 3 billion tons of these materials have accumulated thus far and are increasing yearly at the rate of more than 100 million tons. In an effort to produce cleaner coals and also to upgrade their environmental acceptability, the level of waste production is expected to increase markedly with increased use of coal, possibly doubling in the next decade. In addition to being large volume wastes, these coal preparation discards present problems of serious environmental concern. Of the nearly 5000 coal waste dumps, half pose some type of health, environmental, or safety problem. One of the growing environmental concerns is the effect that trace metals in the waste dump drainages will have as they collect in the surrounding streams and soils. In acknowledging this latter concern, the Department of Energy (DOE) and the Environmental Protection Agency (EPA) have jointly sponsored, since 1975, research at the Los Alamos Scientific Laboratory (LASL) to evaluate the trace element problem and to determine and recommend corrective measures.

The overall objectives of the LASL research program are to assess the problem of trace element contamination in coal waste drainages and to identify suitable control technologies. More specifically these are to

- assess the nature and magnitude of trace elements in the effluents from coal preparation wastes,
- identify the chemistry of the trace constituents of environmental concern,
- identify and experimentally verify effective environmental strategies to control the release of hazardous constituents, and
- analyze the tradeoffs associated with the different control technologies and recommend control measures or necessary research development and demonstration (RD&D) programs.

To understand why coals and coal preparation wastes release elements in the amounts they do, we have studied the levels and releases of elements from a low-sulfur Appalachian coal area where the mineral drainage is not highly visible and from high-sulfur Illinois Basin areas where mineral drainage has long been recognized as a severe problem. Both types of wastes are composed primarily of clay minerals, quartz, iron sulfides, and calcite. Low-sulfur Appalachian wastes differ from the high-sulfur Illinois wastes by having <1% iron sulfide minerals (pyrite and marcasite) as compared to 20-30% for the latter. Some 55 elements have been identified and undoubtedly there are more. The most abundant of these (aluminum, iron, and silicon) form the major minerals. The trace constituents are probably present as minor minerals (even as microparticles), components of residual coal, or substituents in the major minerals. A number of elements that are generally considered to be environmentally sensitive are present in significant quantities (>30 μg/g of waste). These elements of concern include aluminum, arsenic, cobalt, copper, fluorine, iron, lead, manganese, nickel, and zinc. Although the relative amounts of some of the trace elements are small, the absolute quantities available in a large waste dump have grave consequences when they are released by natural processes into the surrounding environment.

The high-sulfur (high-pyrite) wastes from the Illinois Basin, when exposed to air and water, produce highly acidic drainages (pH ~ 2 to 4). Even low-sulfur (low-pyrite) Appalachian wastes produce acidic drainages (pH ~ 4) though the total amount of acid is much less. Our experiments to simulate the intermittent rainfall and weathering to which coal waste dumps are subjected have revealed that alternate oxidation and leaching of the pyrite in the waste is a most effective way to regenerate acid leachates continuously. We have demonstrated experimentally that intermittently leached coal wastes pose a far greater pollution threat, in both quantity and time, than do those wastes that are always submerged in water or are isolated from air, water, or both in some manner.

Acidic leachates in coal waste dumps are very efficient in dissolving or degrading many of the minerals present and releasing the elements associated with them. Last year, we reported that aqueous leachates from high-sulfur Illinois Basin coal wastes contained nine elements (aluminum, cadmium, cobalt, copper, fluorine, iron, manganese, nickel, and zinc) in environmentally hazardous concentrations. More recently, we have analyzed leachates from a low-sulfur Appalachian coal waste by the EPA's Multimedia Environmental Goal/Minimum Acute

Toxicity Effluent (MEG/MATE) system for classifying potentially hazardous contaminants and have found that six elements (aluminum, copper, iron, manganese, nickel, and zinc) were present in levels of possible environmental concern. The severity of contamination caused by the low-sulfur waste, however, is much less pronounced than that caused by the high-sulfur waste.

From our studies and from information about actual dump drainages, it is evident that similar and suitable control technologies to prevent environmental degradation by the release of acid and trace elements are needed for both high-sulfur and low-sulfur coal preparation wastes, as the difference in waste types is more a matter of degree than of kind.

Control strategies for expedient and environmentally acceptable disposal of coal preparation wastes fall into three logical categories.

I. Alter the waste structure to produce an environmentally acceptable waste.

II. Dispose the waste in a manner that will produce an environmentally acceptable drainage.

III. Collect and treat the contaminated drainage from disposed, but untreated, waste.

Category I eliminates the source of the problem by immobilizing or removing the potential pollutants from the waste. The second category could be implemented by retaining the pollutants in the dump, providing back-up safeguards (if necessary), and monitoring the dump to insure environmentally acceptable containment. A properly devised and monitored disposal scheme in Category II could provide for an orderly release of environmentally acceptable levels of pollutants. This category also recognizes that an acceptable, environmental control can be found without the need to completely destroy or alter the waste itself. In the third alternative, one could isolate the dump, let it generate pollutants as it would, collect the contaminated drainage, and remove the pollutants from the drainage before releasing the water into the local waterways. This strategy is the least desirable from an environmental viewpoint because it is nearly impossible, with such large volumes of wastes and extensive drainage areas, to insure that the pollutants will not inadvertently escape into the environment in unacceptable quantities. Our studies show that the release of pollutants is certainly extensive enough that monitoring and treatment of such drainages will be required for many years and perhaps for centuries. All three control strategies, when viewed in terms of the tradeoffs among their economic impact, technical complexity, and overall effectiveness, have good and bad features and no single strategy is obviously more promising than the other two.

Two methods that immobilize or remove metal pollutants from coal wastes (Category I) are calcining and preleaching, respectively. Our laboratory results on calcining show that heating the waste to 800-1000°C releases the acid-producing sulfur and causes mineralogical transformations that seal the remaining trace pollutants in the residue. Leachates from such calcined residues have no trace element concentrations of concern. Unlike calcining, preleaching with oxidizing agents has not been very fruitful.

Building environmentally acceptable, controlled pollutant-release coal waste dumps (Category II) has many possibilities. Codisposing the waste with neutralizing agents or sorbents and sealing the surfaces of the waste particles have been investigated experimentally. Small particle limestone, lime, certain types of clays and soils, and industrial wastes, such as fly ash and alkaline sludges, have all shown promise as neutralizing agents when codisposed with highly acidic coal wastes in our laboratory testing. Encapsulating the wastes in cement is also effective in preventing trace element releases. This method of contaminant release control has the potential of rendering coal wastes nonhazardous under the provisions of the Resource Conservation and Recovery Act (RCRA). Such a possibility plus the effective attenuation of trace elements already demonstrated in laboratory-scale codisposal experiments make this method look especially promising.

The third environmental control strategy, collection and treatment of contaminated coal waste discharge, is largely the application of well-known commercial water pollution control methods.

Of the various controls we have considered for this purpose, alkaline neutralization of these acid drainages appears to have the best potential as an effective and economical method. Higher technological methods that we have investigated, such as ion exchange and reverse osmosis, have better potential for polishing partially purified water than as a primary treatment.

Making a selection from among the various control options is by no means a straightforward matter. The applicability of any of the control strategies must be evaluated in terms of the pertinent tradeoffs. For example, coal refuse calcining has excellent potential for preventing the release of trace elements from coal refuse piles and is a one-time treatment, but it is expensive. Alkaline neutralization, on the other hand, is low in cost. However, all the effluent from a given dump will have to be treated, and when the acid-regeneration capability of the high-sulfur waste is considered, the acid effluent from a given refuse pile might have to be treated for upwards of a hundred years. Taking several of the more important factors into consideration, we have generated a comparison of several of the control options in the form of the following grid. (More comparisons are made in the main body of the report.) The options seem to be choices based primarily on economics (favoring drainage treatment) and commitment to responsibility (favoring immobilization). A blend of the factors may make Category II (waste pile construction) a favorable compromise.

COMPARISON OF ENVIRONMENTAL CONTROL OPTIONS

Parameter	Calcining	Codisposal	Alkaline neutralization
Cost	High	Moderate	Low
Effectiveness	Good	Good	Good
Complexity	High	Moderate	Moderate
Treatment duration	Short	Short	Long
Permanency	Good	Uncertain	Poor

In summary, our laboratory studies that have examined in detail the sources of trace elements in coal waste drainages, the mechanisms of their release, and their fate upon weathering and leaching have allowed us to understand the problem sufficiently well to address the key environmental control technology issues effectively. A substantial portion of our future efforts will be directed at identifying the most promising control options, demonstrating more clearly their utility, and analyzing the balances between their advantages and disadvantages. To broaden the scope of our work, we will also include further studies on high-sulfur coal wastes from the Appalachian region. These studies will define the potential of trace elements in these wastes to lead to undesirable environmental impacts and will identify the control technologies appropriate to mitigate those of environmental concern.

SUMMARY OF TASK PROGRESS

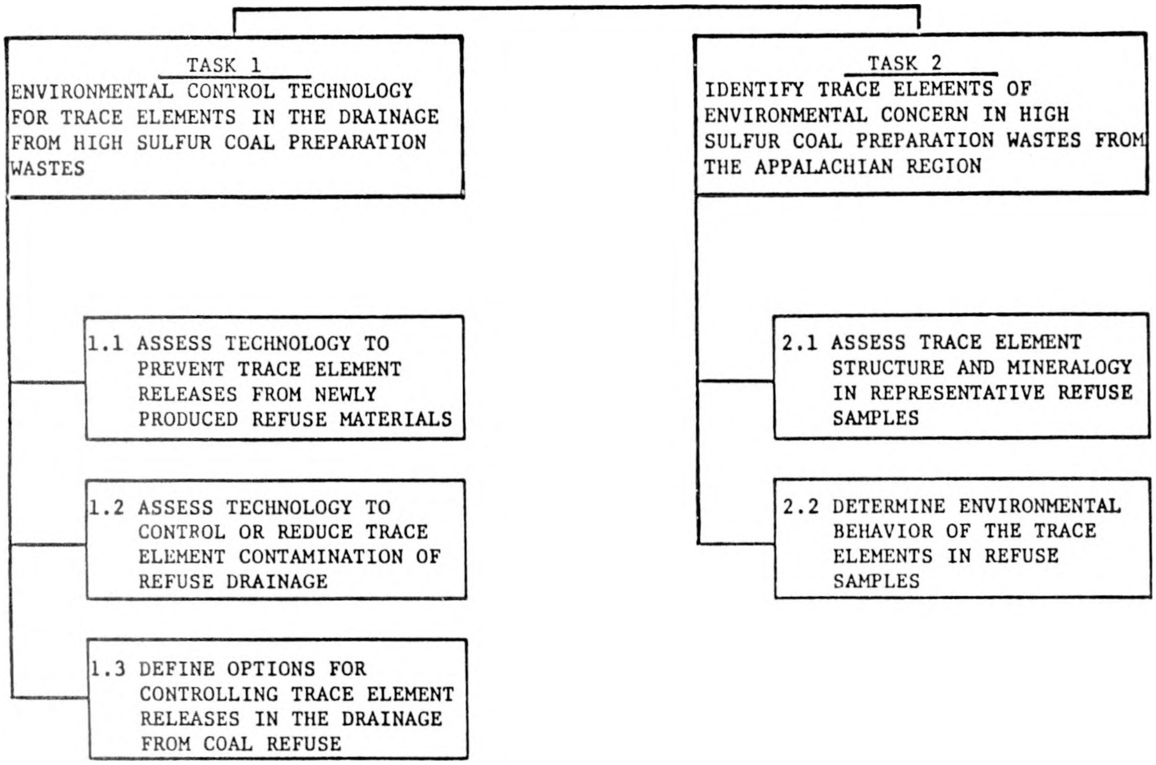
The objectives of this ongoing research program are to assess the potential for environmental pollution from trace elements released by the drainages from coal cleaning wastes and to identify necessary environmental control technologies for this type of contamination. This report describes the technical accomplishments in each of the main research areas of the program for the period October 1, 1977 to September 30, 1978.

The research activities in this program are broken down into major tasks and subtasks as listed in the Task Breakdown chart.

Task 1—Environmental Control Technology For Trace Elements in the Drainages From High-Sulfur Coal Preparation Wastes—is divided into three subtasks. The first of these (Subtask 1.1)

TASK BREAKDOWN

TRACE ELEMENTS CHARACTERIZATION AND REMOVAL/RECOVERY FROM COAL AND COAL WASTES



is a laboratory investigation of various options for treating coal refuse materials either at the preparation plant or during disposal to reduce subsequent releases of harmful trace elements during waste dump weathering and leaching. Among the potential methods that we investigated this year are calcining and preleaching of the refuse before disposal and applying neutralizing agents, adsorbents, and sealants to the dump itself to prevent contaminated drainage.

One of the more promising control strategies under investigation in the program is calcining. High-temperature heat treating of the refuse materials is used to remove volatile, acid-forming constituents and to chemically immobilize potentially toxic trace elements in the fused matrix of the refuse. In these researches, we have studied the changes in both the chemical and physical characteristics of the calcined refuse and the leachabilities of the trace elements that remain in the calcined mass. Our research has revealed that refuse calcining is technically one of the more effective and permanent means of treating high-sulfur coal wastes to eliminate subsequent releases of acidic or contaminated drainage from refuse disposal sites.

Refuse preleaching has been studied to explore the feasibility of pretreating high-sulfur coal refuse materials with aqueous leaching agents before disposal to remove some or all of the environmentally active trace elements and acid-forming constituents. Among the leaching agents that we investigated for this purpose were (1) water, (2) mixtures of water and oxidizing agents, such as ferric salts and hydrogen peroxide, and (3) an oxidizing acid, nitric acid. Only the strong oxidizing acid appears to hold promise for removing a substantial proportion of the contaminants of concern through preleaching of the refuse samples that we studied.

Several methods were considered to treat coal refuse during disposal to prevent the release of trace contaminants during subsequent waste dump weathering or leaching by surface or ground water. These included codisposal of the refuse material with neutralizing agents or trace element adsorbents and the application of watertight sealants to all or parts of the waste dump mass. Especially promising among these techniques is the codisposal of high-sulfur refuse with lime or limestone to neutralize acid drainage *in situ* and retain aqueous contaminants within the refuse disposal site.

Our research has shown that the codisposal of attenuating agents or sorbents, other than lime or limestone, with acidic coal refuse materials also has good potential for reducing or abating trace element contamination of disposal site drainage. Many natural materials, such as certain types of clays and soils, and many industrial wastes, such as fly ash or alkaline sludges, have considerable capacity to attenuate contaminated refuse drainage, and often they are available in large and accessible quantities near refuse disposal sites. Some of our research during the year was directed at assessing the effectiveness of various attenuating agents for reducing the trace element and acid compositions of coal refuse leachates.

Another area of control technology that we are addressing is to seal the refuse pile, dump, or burial site to prevent the intrusion of air or water. The concept of sealing has overtones in all aspects of coal waste (and other waste)disposal. Sealants can be used for existing refuse piles and dumps and for near surface and underground burial of wastes. A variety of sealant scenarios, with an emphasis on clays, soils, calcite, and various cementing agents (Portland and silicate cements and polymers) as sealing agents, are being considered, and we have begun laboratory experiments to test some of these materials.

The second portion of this task (Subtask 1.2) was the assessment of environmental controls to reduce or attenuate undesirable trace elements in the drainages from coal refuse dumps. Our attention in this area was given to pollution abatement techniques that have proved effective for treating acid mine drainage. These techniques include alkaline neutralization, ion exchange, and reverse osmosis. We have also initiated studies on the effectiveness of using a variety of sorbents, such as clays, soils, and solid coal combustion by-products, on high-sulfur coal refuse leachates.

Alkaline neutralization was shown to be the most effective and least costly of the refuse drainage treatment options that we studied. Ion exchange and reverse osmosis both proved to be technically feasible methods for reducing the contaminants in refuse drainage to acceptable levels; however, the necessity to further treat the solutions for acidity sharply reduces the acceptability of these methods. This and the known tendency of ion exchange and reverse osmosis to overload or plug when the contaminant or suspended solid contents are high lead us to believe that these techniques might be most applicable as secondary treatment methods to clean up the effluents from some other primary control process.

The last portion of this task (Subtask 1.3) involves a discussion of the results and major implications from the research that we have conducted thus far on control technologies for trace element contamination of coal refuse drainage. Included in this section is a consideration of the relative costs, effectiveness, treatment duration, likely RCRA classification, and premanency of the most promising control method being studied.

Our research this year has begun to classify the nature of the tradeoffs to be made among the various control options for the disposal of acidic coal refuse materials. The methods that potentially provide the most effective and permanent means of abating trace element contamination of refuse drainage (notably calcining) are also the most costly and complex methods to use. The control techniques that are designed to retain contaminants within the refuse disposal site, such as codisposal with various agents, are effective for attenuating the trace element compositions of refuse drainages for at least short durations, but some of these may lack permanence. Acceptability for nonhazardous RCRA disposal requirements is another questionable aspect. Finally, the methods to treat refuse drainage (alkaline neutralization and reverse osmosis) appear to be quite attractive because of their relatively low costs and effective trace element reduction. These methods, however, are fraught with potential problems, such as indefinite treatment duration, possible contamination escape, and cost to meet RCRA permit and performance requirements of a hazardous waste.

Task 2—Identify Trace Elements of Environmental Concern in High-Sulfur Coal Preparation Wastes From the Appalachian Region—was split into two subtasks. One (Subtask 2.1) involved studies of the mineralogy and elemental composition of low-sulfur coal refuse samples collected from the Appalachian area; the other (Subtask 2.2) concerned the aqueous leaching behavior of these materials.

The mineralogy of the low-sulfur refuse material is notably different from that of the Illinois Basin that we studied earlier. There is very little detectable pyrite or marcasite in the Appalachian samples, whereas, these acid-forming minerals composed 20-30% of the Illinois Basin materials. Clay minerals and quartz represent about 60% of the detectable mineral composition of the Appalachian samples.

The elemental composition of the Appalachian refuse is a reflection of its mineral matrix. Aluminum and silicon are by far the most abundant elements present in the refuse. From an environmental viewpoint, this low-sulfur Appalachian refuse was found to contain potentially troublesome quantities ($>40\ \mu g/g$ of refuse) of aluminum, copper, iron, manganese, nickel, and zinc.

Static and dynamic leaching tests were conducted on the Appalachian refuse material. These studies were designed to simulate the weathering and leaching behavior of the refuse materials and to yield data on those potentially troublesome trace elements that may be released into the environment. An analysis of the data from the leaching experiments was made using Multimedia Environmental Goals established by the EPA. This analysis identified aluminum, copper, iron, manganese, nickel, and zinc as the elements of most environmental concern in the Appalachian refuse samples that we studied.

TASK PROGRESS DESCRIPTION

TASK 1—ENVIRONMENTAL CONTROL TECHNOLOGY FOR TRACE ELEMENTS IN THE DRAINAGE FROM HIGH-SULFUR COAL PREPARATION WASTES

Subtask 1.1—Assess Technology to Prevent Trace Element Releases From Newly Produced Refuse Materials

The purpose of this subtask is to investigate in the laboratory the options available for treating high-sulfur coal refuse materials, either at the preparation plant or during disposal, to prevent or reduce subsequent releases of environmentally harmful trace elements during waste dump weathering and leaching. Such control technology could include chemical or physical processing to remove the undesirable elements from the refuse; treating the refuse materials to immobilize these elements; applying neutralizing agents, adsorbents, or sealants at the refuse dump site; and burying, grading, and compacting the waste materials to control the flow of water and air through refuse piles.

Calcining to Immobilize Refuse Constituents

The possibility that the release of toxic trace elements into the environment can be controlled by pretreatment of coal preparation wastes has been investigated experimentally. One approach that we are examining is calcining (high-temperature heat treating) of these materials to remove volatile, acid-forming constituents and to chemically immobilize potentially toxic trace elements in the refuse matrix. In these researches, we have studied the changes in both the chemical and physical characteristics of the refuse and trace element mobilities that result from the heat treatment. Our research has revealed that refuse calcining is technically one of the more effective and permanent means of treating high-sulfur coal wastes to eventually completely eliminate subsequent releases of acidic or contaminated drainage from refuse disposal sites.

Our initial set of calcining experiments was performed using high-sulfur coal preparation wastes from Illinois Basin Plant B to determine the effects of heat treatment on the elemental composition of this type of refuse material. The sample was prepared by crushing the refuse to −3/8 in. and calcining it in a quartz tube at 800 to 850°C in air for 6 h. The calcined material, which had partially agglomerated, was ground to -20 mesh for subsequent studies. The analysis of the chemical and trace element composition of the calcined refuse sample showed a marked decrease in the concentration of the volatile components in the refuse. Of particular interest was the loss of sulfur that occurred as a result of the calcining. From an original concentration of 13.4 wt% sulfur in the noncalcined sample, the described sample treatment yielded a product that contained only 0.7 wt% sulfur. Other volatile components whose concentrations were decreased by the calcining were bromine, lead, and cadmium. The complete elemental analyses for the calcined refuse sample appear in Table I.

TABLE I

EFFECT OF CALCINING ON TRACE ELEMENT RETENTION FROM PLANT B COAL REFUSE

Element	Level[a]	Retention (%)[b]	Element	Level[a]	Retention (%)[b]
Na	1 140	90	Zn	300	120
Mg	4 900	120	Ga	29	-
Al	115 000	120	As	110	70
SiO_2	168 000	80	Br	<0.1	**<5**
P	560	-	Mo	35	**40**
S	11 400	**5**	Cd	0.29	**40**
Cl	<100	-	Cs	9	80
K	24 500	140	La	60	100
Ca	1 900	110	Ce	130	110
Sc	20	100	Eu	2	100
Ti	5 490	100	Yb	5	100
V	120	90	Lu	0.6	90
Cr	100	100	Hf	5	100
Mn	190	80	Ta	1.7	120
Fe	190 000	110	Pb	12	**20**
Co	70	140	Th	15	100
Ni	110	100	U	6	140
Cu	73	130			

[a] Experimentally determined concentration before calcining, in parts per million.
[b] Error approximately ±30% of value.

Several sets of calcining and leaching experiments were conducted to determine optimal heat-treatment conditions necessary to immobilize the potentially toxic trace elements in the refuse matrix. These experiments were performed using high-sulfur coal preparation waste from Illinois Basin Plant C. The waste was ground to -20 mesh and calcined in air at 600, 800, 1000, and 1200°C for 2 h.

The effect of refuse calcining treatments on the mineral composition of the refuse is illustrated by Table II, which delineates the changes in refuse mineralogy that occurred at various temperatures. The two most environmentally active species, pyrite (marcasite) and calcite, have been transformed to high-temperature phases by 600°C. By 1000°C, even the clay minerals have been converted to structurally indefinable aluminosilicates, and the samples have become fused or sintered at particle surfaces. The x-ray diffraction analyses reported in Table II support the concept that the mineralogical transformations thought to occur in coal waste burning (Table III) have been effected by heat treatment in the range of 800 to 1000°C.

TABLE II

PRESENCE OF COAL REFUSE MINERALS AT VARIOUS CALCINING TEMPERATURES

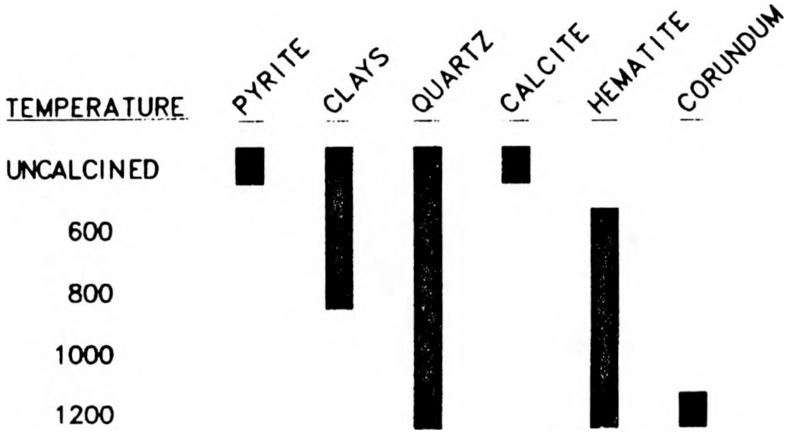


TABLE III

HIGH-TEMPERATURE MINERAL TRANSFORMATIONS OF HIGH-SULFUR COAL REFUSE

Clays	Complex Aluminosilicates
Calcite	$CaO + CO_2$ (gas)
Pyrite or Marcasite	$Fe_2O_3 + SO_2$ (gas)
Quartz	Quartz

Extensive studies of the trace element leachabilities of the calcined refuse samples have been conducted.* The effects of calcining temperature on leachate pH and total dissolved solids (TDS) content are seen in Table IV. Calcining to 600°C and higher results in leachates with elevated pH values. This is a consequence of the conversion of the acid-forming mineral species, pyrite and marcasite, to more stable oxide forms and the driving off of the sulfur. Calcining of the refuse also significantly reduced the general leachability of the material as evidenced by the reduced TDS values of the leachates.

The success of calcining at reducing trace element releases during refuse leaching is illustrated by Table V, which lists trace element data from a comparison leaching test of calcined and uncalcined refuse samples. (More complete data are presented in Appendix A.) The refuse samples referred to in the table had been subjected to static leaching for 48 h. It is seen that the concentrations of the group of toxic elements listed are reduced in the leachates from the calcined refuse by as much as two orders of magnitude over the concentration in the uncalcined refuse leachates.

Calcining acid coal refuse materials before disposal could produce several beneficial effects. Foremost among these is the conversion of an active, highly polluting waste material into a chemically and geologically inert mass that can be easily and safely disposed almost anywhere with ordinary landfill practices. Thus, calcining presents a highly effective and permanent solution to a most difficult waste control problem. As a corollary, the calcined refuse materials might be classified as nonhazardous under the criteria developed in conjunction with the Resource Conservation and Recovery Act (RCRA). This circumvents the need to meet the cumbersome and costly permit and performance requirements that RCRA dictates for the disposal of hazardous wastes, a category into which most if not all untreated high-sulfur refuse materials will undoubtedly fall. Lastly, there is high potential for the recovery of by-products in connection with refuse calcining that does not exist for many other control technology schemes. Potentially recoverable products include sulfur, iron, and aggregate materials.

The major drawback for refuse calcining is the cost of constructing and operating a calcining plant. Interestingly, most of this expense is for operation of the scrubber system that is necessary to remove sulfur oxide emissions from the calcining plant's gaseous effluents (see Table III). With a scrubber system based on lime or mixtures of lime and limestone, calcining of high-sulfur coal

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*The calcined residues were statically leached (stirred with distilled water) for a period of 48 h using a ratio of 4 mℓ water to 1 g of calcined refuse, and pH and total dissolved solids values were determined.

TABLE IV

EFFECT OF CALCINING TEMPERATURE ON THE LEACHABILITY OF A HIGH-SULFUR COAL REFUSE MATERIAL[a]

Calcining Temp (°C)	Sample Wt Loss (%)	Leachate pH	Leachate TDS (Wt %)
Uncalcined	-	2.9	1.4
600	23	5.9	0.2
800	23	6.2	0.3
1000	23	8.0	0.2
1200	23	8.0	-

[a] 20-g samples of crushed refuse/100 mℓ H_2O/48 h.

TABLE V

TRACE ELEMENT LEACHABILITY OF A HIGH-SULFUR COAL REFUSE SAMPLE CALCINED AT 1000°C[a]

Element	Uncalcined Refuse (ppm)	Calcined Refuse (ppm)
Al	100	0.4
Fe	600	<0.03
Mn	5.8	0.03
Co	2.8	<0.01
Ni	4.8	0.01
Cu	0.10	0.01
Zn	2.8	0.05
Cd[b]	68[b]	0.3[b]
pH	2.9	8.0
TDS (%)	1.4	0.2

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[a] 50-g samples of crushed refuse/200 mℓ H_2O/48 h.
[b] In parts per billion.

refuse materials may cost in the range of $3 to $5 or more per ton of cleaned coal (see Appendix B, Table B-IV). Studies are now under way to seek methods to reduce the costs. Perhaps the most straightforward way to reduce the cost of refuse calcining is to decrease the need to scrub sulfur oxide from the process effluents. One way to accomplish this is through sulfur retention during calcining by adding limestone to the refuse materials, as is practiced in the fluidized-bed combustion of coal. The results from current research in this area suggest that the cost of calcining can be reduced by one-half to two-thirds by using this sulfur retention technique.

Other ways that the effective cost of calcining high-sulfur coal refuse can be reduced have already been touched upon. These include offsetting values for recovered by-products, such as sulfur and iron, and what might best be termed as value added through the substantial savings by not having to comply with the RCRA hazardous waste requirements. Both of these possibilities could aid substantially in decreasing the total cost of waste disposal based on a refuse calcining concept as compared to less effective or less desirable control methods.

Refuse Preleaching to Remove Mobile Trace Elements or Acid-Forming Constituents Before Disposal

The purpose of our research in this area was to explore the feasibility of pretreating high-sulfur coal refuse materials with aqueous leaching agents before disposal to remove some or all of the environmentally active trace elements and acid-forming constituents. Among the leaching agents that we investigated for this purpose were (1) water, (2) mixtures of water and oxidizing agents, such as ferric salts and hydrogen peroxide, and (3) an oxidizing acid, nitric acid. Only the strong oxidizing acid appears to hold promise for removing a substantial proportion of the contaminants of concern.

Two basic types of experiments were conducted in this series. The first involved agitation of various aqueous agents (usually about 250 mℓ) with 50 g of crushed Plant B coal refuse (-20 mesh) at room temperature. The apparatus for this type of study normally consisted of a 500-mℓ Erlenmeyer flask equipped with an open 15-cm glass chimney or extension at the top of the flask to allow air to enter yet retain the contents. The second type of preleaching experiment used similar ratios and amounts of refuse and leaching agents but was conducted at elevated temperature. The apparatus used here was a 500-mℓ, three-necked, round-bottomed flask equipped with a reflux condenser, a heating mantle, and a gas delivery tube. Agitation was provided by a magnetic stirrer. At the completion of the experiments, the solid and liquid contents were separated by successive filtrations through Whatman Nos. 541 and 50 filter papers.

A description of the experimental parameters maintained during the preleaching experiments appears in Table VI. Note that Samples 1, 4, 6, and 8 are analytic controls used to evaluate the initial compositions of various leaching agents.

The elemental analyses of the various leachates resulting from the refuse leaching treatments appear in Table VII. In Table VIII, the experimental data are reported as the percent of each element that was removed from the solid refuse by the leaching treatments. (This latter representation makes the interpretation of the experimental data somewhat easier.) In addition, the information in Table VIII has been arranged to reflect the effectiveness of the various leaching agents at removing iron (mainly as the acid-forming minerals, pyrite and marcasite) from the refuse samples. The interpretation of the data in Tables VII and VIII is complicated by the many

TABLE VI

CONDITIONS OF PRELEACHING EXPERIMENTS CONDUCTED ON A HIGH-SULFUR COAL REFUSE MATERIAL

CTWT-9-	ml WATER [a]	ml Fe^{+++} [b]	MISCELLANEOUS	TIME(Days)	TEMP(°C)
1[c]	250	–	–	–	–
2	250	–	–	1	20
3	250	–	–	10	20
4[c]	–	250 A	–	–	–
5	–	250 A	–	10	20
6[c]	–	250 B	–	–	–
7	–	250 B	–	10	20
8[c]	–	250 C	–	–	–
9	–	250 C	–	1	20
10	–	250 C	–	10	20
11	250	–	–	1	92
12	–	250 C	–	1	92
13	240	–	10 ml 30% H_2O_2	1	92
14	227.5	12.5 C	10 ml 30% H_2O_2	1	92
15	212.5	12.5 C	25 ml tetrahydrofuran	1	92
16	212.5	12.5 C	25 ml absolute ethanol	1	92
17	162.5	62.5 C	25 ml 30% H_2O_2	1	92
18	250	–	250 ml 0.1N NaOH + 1 g Na_2CO_3	1	92
19	225	–	250 ml 0.1N NaOH + 1 g Na_2CO_3 + 25 ml 30% H_2O_2	1	92
20	237.5	12.5 C	–	1	92
21	240	–	10 ml 30% H_2O_2	1	20
22	250	–	255 ml 16*N* HNO_3[d]	2	92

[a] Millipore >10 megohms-cm water.

[b] A is 0.005 molar $Fe_2(SO_4)_3$; B is 0.025 molar; C is 0.125 molar

[c] Samples 1, 4, 6, and 8 are controls.

[d] Added at a rate of 15 ml/h; 16-h break after 8th addition

TABLE VII

ELEMENTAL ANALYSES OF LEACHATES FROM PRELEACHING OF PLANT B COAL REFUSE

						Solution Parameters[b]		Element Levels Removed From Waste[c]											
Sample No.	Reaction Time (Days)	Reaction Temp (°C)	Fe^{+++} (Mole/ℓ)[a]	Misc. Added	Waste Present	pH	TDS (%)	F PPM	Na PPM	Al PPM	K PPM	Ca PPM	Cr PPB	Mn PPM	Fe[e] PPM	Co PPM	Ni PPM	Cu PPM	Zn PPM
1	1	20	–	–	–	5.3		<0.2	0.7	<2	0.2	0.1	<7	<0.2	<1	<0.2	<0.25	<0.5	<0.05
2	1	20	–	–	Yes	2.1	1.04	9	13	970	38	580	630	29	10600	16	28	2.7	41
3	10	20	–	–	Yes	1.6	1.40	15	16	1100	10	750	630	32	14600	18	30	1.6	56
4	1	20	0.01	–	–	2.3	0.25	0.6	2	<3	0.4	0.8	<9	<0.25	2760	<0.25	2	<0.5	0.2
5	10	20	0.01	–	Yes	1.6	1.60	14	18	1100	9	680	630	31	16900	17	30	1.9	56
6	1	20	0.05	–	–	1.5	1.26	4	3	4	0.3	4	130	1	14800	1	8	0.6	0.4
7	10	20	0.05	–	Yes	1.3	2.50	15	18	1100	7	680	420	33	27900	16	38	3.2	63
8	1	20	0.25	–	–	1.2	6.58	0.3	5	<2	0.8	1	420	5	74500	5	32	1.5	1
9	1	20	0.25	–	Yes	1.1	6.66	11	93	1000	37	650	590	31	75000	17	61	8.3	50
10	10	20	0.25	–	Yes	1.0	6.96	12	20	1100	10	670	560	33	87500	17	63	6.1	70
11	1	92	–	–	Yes	2.0	1.38	26	68	1000	220	670	100	35	15000	15	30	<0.5	56
12	1	92	0.25	–	Yes	1.0	7.72	27	64	2600	660	760	1300	37	94300	19	63	<0.5	84
13	1	92	–	H_2O_2	Yes	1.8	1.57	29	37	1100	240	690	750	36	17200	12	29	0.4	59
14	1	92	0.05	H_2O_2	Yes	1.6	2.85	27	52	1500	400	690	840	37	31300	18	36	0.6	69
15	1	92	0.05	THF	Yes	1.6	8.28	25	22	1200	160	730	750	34	24900	18	35	0.4	63
16	1	92	0.05	EtOH	Yes	1.6	2.42	27	31	1400	220	670	710	40	27300	17	35	0.4	88
17	1	92	0.25	H_2O_2	Yes	1.1	2.42	27	64	2400	580	710	1400	39	98000	19	66	<0.05	64
18	1	92	–	Na_2CO_3	Yes	4.8	7.58		21300[d]	<5	200	490		15	320	<0.5	1	<1	5
19	1	92	–	$Na_2CO_3 + H_2O_2$		4.1	7.74		20800[d]	10	150	610		21	1700	<0.5	2	22	23
20	1	92	0.05	–	Yes	1.7	2.41	15	28.1	1530	188	730	810	37.5	26200	10.4	31	<0.05	47
21	1	20	–	H_2O_2	Yes	2.0	1.04	6.1	15.6	900	65	560	630	30.6	10000	15.1	25	4.6	33.1
22	2	92	–	HNO_3	Yes	<0	6.48	42	19.8	15500	3480	890	12700	114	94000	14.4	49	24.5	82

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[a] Iron added to solution.

[b] Solution/Solid Ratio = 5/1.

[c] Based on solid, solid assumed to be present in all cases.

[d] Sodium added with original leaching solution.

[e] Iron may have been added with original leaching solution.

TABLE VIII

EFFECTIVENESS OF LEACHING AGENTS AT REMOVAL OF ELEMENTS FROM PLANT B COAL REFUSE

Iron Leach Effectiveness	Time (days)	Temp(°C)	Fe^{+++} added	Misc added	Element Removed by Treatment (wt%)									
					Fe	Ca	Co	Ni	Zn	Mn	Cu	Al	Cr	F
Very Poor	1	92		Na_2CO_3	0.3	44.5	<1	1.1	3.3	10.3	<0.1	<0.1		
	1	92		Na_2CO_3/H_2O_2	1.5	55.4	<1	2.5	15.4	14.4	60.7	<0.1		
Poor	1	20		H_2O_2	9.1	50.9	49.7	33.9	22.2	21.1	11.6	1.8	1.0	1.6
	1	20		Control	9.8	52.7	52.7	39.1	27.5	20.0	6.2	1.9	1.0	2.3
	1	20	0.25N		9.9	59.0	39.3	40.6	32.8	17.9	17.8	2.0	0.3	2.8
	1	92	0.05N	THF[a]	10.5	65.6	56.0	38.7	42.0	24.8	<0.1	2.3	1.0	6.0
	1	92	0.05N		11.8	65.6	30.7	30.3	31.2	25.2	<0.1	3.0	1.1	3.4
	10	20		Control	13.3	68.2	59.3	41.9	37.6	22.1	3.1	2.2	1.0	4.0
	10	20	0.01N		13.2	61.7	55.2	39.1	37.4	21.2	2.5	2.2	1.0	3.5
	10	20	0.05N		13.5	61.9	49.3	40.1	42.0	22.1	6.2	2.1	0.5	3.4
	1	92		Control	13.8	60.9	49.3	41.9	37.6	24.2	<0.1	2.0	0.2	6.9
	1	92	0.05N	EtOH	14.2	60.9	52.7	38.7	58.8	26.9	<0.1	2.7	0.9	6.6
	1	92		H_2O_2	15.9	62.7	39.3	40.5	39.6	24.9	<0.1	2.9	1.1	6.6
	1	92	0.05N	H_2O_2	16.0	62.0	56.0	40.1	46.1	24.9	<0.1	2.2	1.2	7.7
Fair	10	20	0.25N		20.8	60.8	39.3	43.4	46.2	19.3	11.6	2.2	0.2	3.1
	1	92	0.25N		28.0	69.0	46.0	43.4	55.6	22.1	<0.1	5.1	1.4	7.1
	1	92	0.25N	H_2O_2	32.6	64.4	46.0	47.6	42.2	23.5	<0.1	4.7	1.6	7.1
Excellent	2	92		HNO_3	91.4	80.9	47.3	68.7	55.0	79.0	67.8	30.4	20.5	11.2

[a] Tetrahydrofuran.

chemical conditions involved in the experiments. However, it is readily apparent that the most effective preleaching agent is the strong aqueous oxidizing acid, nitric acid.

Neither a weak base (sodium carbonate) nor water alone proved to be very effective at removing iron or most other trace elements from the refuse samples. Ferric ion, added as a leaching agent in the form of ferric sulfate, proved to be a more effective agent for preleaching the refuse samples. Refluxing of ferric (0.25*N*) sulfate solution in the presence of crushed refuse material for 1 day (Sample No. 12) resulted in the removal of close to 30% of the total iron from the sample, as well as considerable amounts of several other key trace elements. A review of the literature on coal desulfurization with ferric ion[2] suggests that much greater pyrite (iron) removal efficiencies can be obtained from the refuse samples by increasing the ferric sulfate concentrations in the leaching solutions to about 1*N* (Table IX). Finally, by far the most effective preleaching agent for removing iron and the several other environmentally important trace elements considered in this investigation was nitric acid solution (Sample No. 22). More than 90% of the total iron and about 50% or more of the total cobalt, copper, manganese, nickel, and zinc were removed by a 2-day treatment of the refuse material with 8*N* nitric acid.

Our research on preleaching of coal refuse materials to remove labile trace elements, the acid-forming mineral constituents, or both is still at a rather early stage, and we do not yet possess sufficient technical information on this potential control technology option to conduct a solid economic assessment of it. In a properly designed control technology scheme in which the leaching agent is recycled, the use of nitric acid, for example, to preleach coal refuse could prove to be economically viable, especially considering the strong possibility for resource recovery offered by this technique. Although more experiments are needed before a final assessment can be made, the usefulness of ferric ion solutions to preleach coal refuse materials appears marginal because of the relatively low extractabilities achieved for many of the elements of greatest environmental concern.

Addition of Neutralizing Agents to Discarded Refuse Materials

Several methods are being considered to treat coal refuse during disposal to prevent the release of trace contaminants during subsequent waste dump weathering or leaching by surface or

TABLE IX

EFFECTIVENESS OF FERRIC SULFATE TREATMENT ON IRON REMOVAL FROM COAL AND COAL WASTE

	Percentage of Iron (Pyrite) Removed					
	Coal Waste			Coal		
Fe^{+++} Level (*N*)	0	0.05	0.25	0	0.4	0.9
92°C/24 h[a]	4[b]	2[b]	18[b]	---	---	---
100°C/6 h	---	---	---	~1[c]	33-43[d]	50-64[d]

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[a] See text for experiment description.

[b] Observed value is 10% higher, but 10% is also soluble in water at 20°C!

[c] Ref. 1, p. 176.

[d] Ref. 1, p. 67.

ground water. These include codisposal of the refuse material with neutralizing agents or trace element adsorbents and the application of watertight sealants to all or parts of the waste dump mass. Especially promising among these techniques is the codisposal of high-sulfur refuse with lime or limestone to neutralize acid drainage *in situ* and retain aqueous contaminants within the refuse disposal site.

One of the major conclusions from our earlier studies of the environmental behavior of coal refuse materials concerned the importance of pH in controlling trace element releases during refuse leaching. In all instances when leachate pH was maintained at or near the neutral point, only minimal amounts of trace elements were solubilized by the leachates. Conversely, when oxidative degradation of the pyritic materials in the refuse caused leachate acidities to build up, substantial quantities of such environmentally troublesome elements as aluminum, cobalt, copper, iron, manganese, and nickel were lixiviated by the acid leachates.[3,4] This marked dependence of trace element contamination on leachate pH suggested that a potentially fruitful means of preventing trace element releases from discarded refuse materials might be the addition of neutralizing agents to the refuse before disposal to negate leachate acidity as soon as it is formed.

Column leaching experiments that used mixtures of crushed limestone and high-sulfur refuse were conducted to test the effectiveness of this *in situ* neutralization concept and also to examine what effect the location of limestone application had on the results. The refuse was from Illinois Basin Plant B. This refuse contains relatively large amounts of pyrite and marcasite but no detectable calcite. This combination represents a worst-case example of acid-forming potential, and in fact, our earlier studies showed that the leachates formed by passing water through a packed column of this material were not only highly acidic but were also highly contaminated with trace elements. Interestingly, the limestone itself contains troublesome amounts of several environmentally sensitive elements including copper, iron, lead, manganese, and zinc. The elemental analysis of the limestone used is given in Table X.

The combinations of refuse and limestone incorporated into the leaching studies are listed in Table XI. Crushed or powdered limestone was combined with the refuse or placed in the column in three different geometric arrangements: at the inlet (simulating a limestone layer placed on top of a refuse pile), at the outlet (simulating refuse disposed on top of a limestone layer), and limestone and refuse intermixed. These column leaching experiments were conducted by passing distilled water through the column packed with the refuse/limestone mixtures at a rate of 0.5 mℓ/min. Periodically, samples of leachate were collected at the column outlet, and pH, total dissolved solids, and trace element compositions were determined. Leachate flow was interrupted once during several of the experiments (after a little more than 10 ℓ had been eluted), and dry air was passed through the packed columns for three weeks before recommencing leachate flow. This was done to explore contaminant regeneration in the refuse/limestone mixtures.

The overall effect of the various limestone additions to the refuse columns is illustrated by the behavior of the leachate pH shown in Fig. 1a. In general, it is seen that adding limestone to the acid refuse material was only partially successful in controlling leachate acidity. The pH values of the refuse/limestone leachates for experiments GL-14, 15, and 17 (where the limestone was intermixed with the refuse or placed at the column outlet) are higher throughout than for refuse alone (GL-12). However, even in the best instance it took about 5 ℓ of water for 1300 g of refuse to reach neutrality. Placing the limestone layer on the inlet side of the refuse column (GL-16) resulted in no decrease in leachate acidity over the control system. This undoubtedly is due to the slow rate of dissolution of limestone in neutral solution (water).

The effects of the various limestone additions on the TDS composition of the refuse leachates are depicted in Fig. 1b. There is very little difference among the TDS values for any of the leachates. This most likely results from the fortuitous balancing of the constituents removed (by elevating the leachate pH) with those added by limestone dissolution (see Fig. 1a).

TABLE X

ANALYSES OF JEMEZ LIMESTONE USED IN NEUTRALIZATION CONTROL TECHNOLOGY EXPERIMENTS

Element	Level (μg/g)	Element	Level (μg/g)
Na	120	Rb	<20
Mg (%)	0.34	Ag	0.3
Al (%)	0.41	Cd	0.6
SiO_2 (%)	3.4	Sb	<0.5
P	220	La	0.4
S (%)	<0.1	Ce	<0.8
K	790	Sm	<0.2
Ca (%)	42	Eu	<0.1
Sc	0.1	Yb	<0.3
Cr	18	Lu	<0.1
Mn	560	Hf	<0.2
Fe (%)	0.26	Ta	<0.5
Cu	19	W	<0.1
Zn	19	Hg	0.1
Ga	<0.5	Pb	82
As	0.6	Th	<0.2
Br	0.3	U	1.2

TABLE XI

DESCRIPTION OF DYNAMIC LEACHING STUDIES OF HIGH-SULFUR REFUSE/LIMESTONE MIXTURES

Experiment No.	Limestone Location	Sample[a]
GL-12	(None - control)	1500 g refuse (-3/8 in.)
GL-14	Intermixed	1300 g refuse (-3/8 in.) 220 g limestone (-3/8 in.)
GL-15	Layered at outlet	1300 g refuse (-3/8 in.) 229 g limestone (-3/8 in.)
GL-16	Layered at inlet	1300 g refuse (-3/8 in.) 221 g limestone (-3/8 in.)
GL-17	Layered at outlet	1300 g refuse (-3/8 in.) 220 g limestone (-20 mesh)

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[a] Illinois Basin Plant B refuse used throughout.

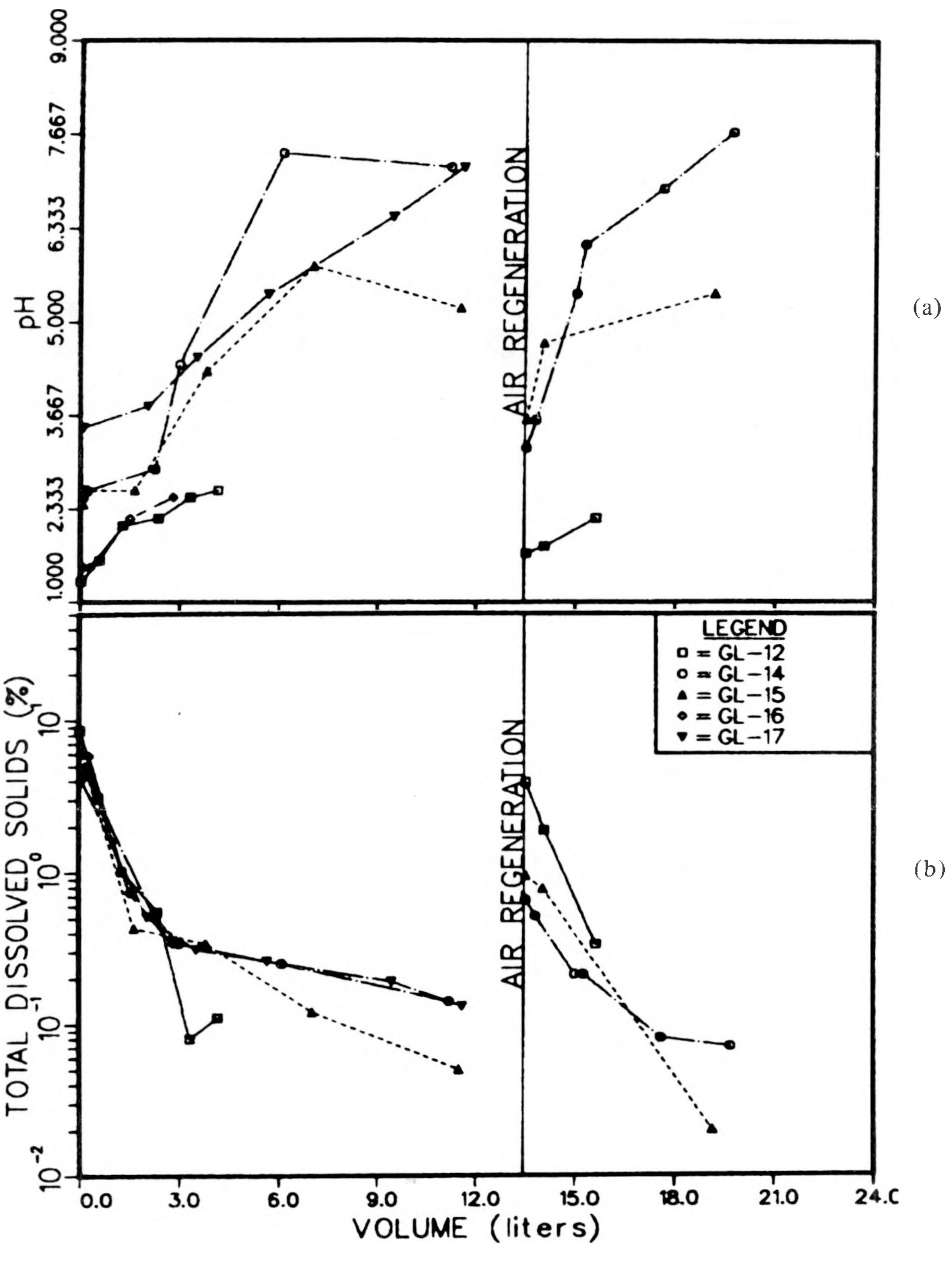

Fig. 1.

Leachate pH and TDS versus leachate volume for column leaching study of limestone/refuse mixtures.

The trace element compositions for the leachates from the refuse/limestone mixtures were followed throughout the experiment (Appendix C). Some elements, such as aluminum, chromium, potassium, scandium, and vanadium, were apparently sensitive to leachate pH and, hence, tended to precipitate from the refuse/limestone mixtures. (The exception was GL-16 where the pH remained low.) Other elements, including cobalt, copper, iron (probably in ferrous state), manganese, and zinc were not apparently so highly pH-dependent in these mixtures; therefore, there was little effect of the limestone addition on the leachate concentrations of these elements. Unfortunately, most of the elements that we have identified as being of greatest environmental concern (listed in Ref. 3) fall into the latter category.

In summary, these experiments revealed that crushed limestone (-3/8 in.) is only moderately effective in controlling the acidity of refuse leachates, largely, we believe, because of the slow rate of dissolution of the limestone under the conditions of the experiments. During the year, we have extended these studies to include, as additives, more finely powdered limestone and limestone that has been slurry-mixed with the refuse. Both should be more effective (in a kinetic sense) than the coarser dry-mixed limestone at controlling leachate pH and, indirectly, trace element composition. Preliminary data from these later experiments, which will be tabulated and discussed in future reports, reveal that both the fineness of the limestone and the manner in which it is mixed with the refuse are indeed very important variables in determining the effectiveness of limestone at controlling refuse leachate pH and trace element composition.

Our efforts involving the additions of powdered lime to high-sulfur refuse materials to control leachate pH and trace element content proved to be very fruitful. For these experiments, powdered lime in varying amounts (3 to 50 g) was slurried in 150 mℓ of distilled water with 530 g of -3/8-in., high-sulfur coal refuse from Illinois Basin Plant B. The resultant mixture was subsequently dried in air at 50°C and recrushed to -3/8-in. particles. Four different lime concentrations were used. The experiments are identified as follows.

Experiment No.	**Lime Level (wt%)**
CTWT-11-1 (control)	0
CTWT-11-2	0.5
CTWT-11-3	1.5
CTWT-11-4	3
CTWT-11-5	10

Column leaching experiments were conducted with about 500 g of each of the above samples to determine the effects of the lime additions. The refuse mixtures were packed into pyrex columns 40 cm long by 5 cm in diameter and subsequently were leached with distilled water at a flow rate of 0.5 mℓ/min until more than 4 ℓ of water had been passed through the refuse beds. Leachate flow was interrupted once during the experiment at the 4.2-ℓ point, and dry air was passed through the column for 2 wk to test the acid-regeneration potential of the refuse/lime mixtures. Tables and plots of leaching data, pH, and trace element analyses for these experiments are compiled in Appendix D.

Figure 2 shows pH and total dissolved solids behavior as a function of lime addition. A consistent pattern of the effects of the lime additions emerges from these data. The additions of 0.5 and 1.5 wt% lime to the acid refuse had only a small influence on leachate pH and trace element concentration because the acid neutralization provided by these amounts of lime was overwhelmed by the acid present in the refuse. The additions of 3 and 10 wt% of lime, on the other hand, did indeed effectively counteract the acid properties of the refuse. The pH values of the leachates for these mixtures were higher, TDS values were relatively low, and the trace element concentrations were depressed.

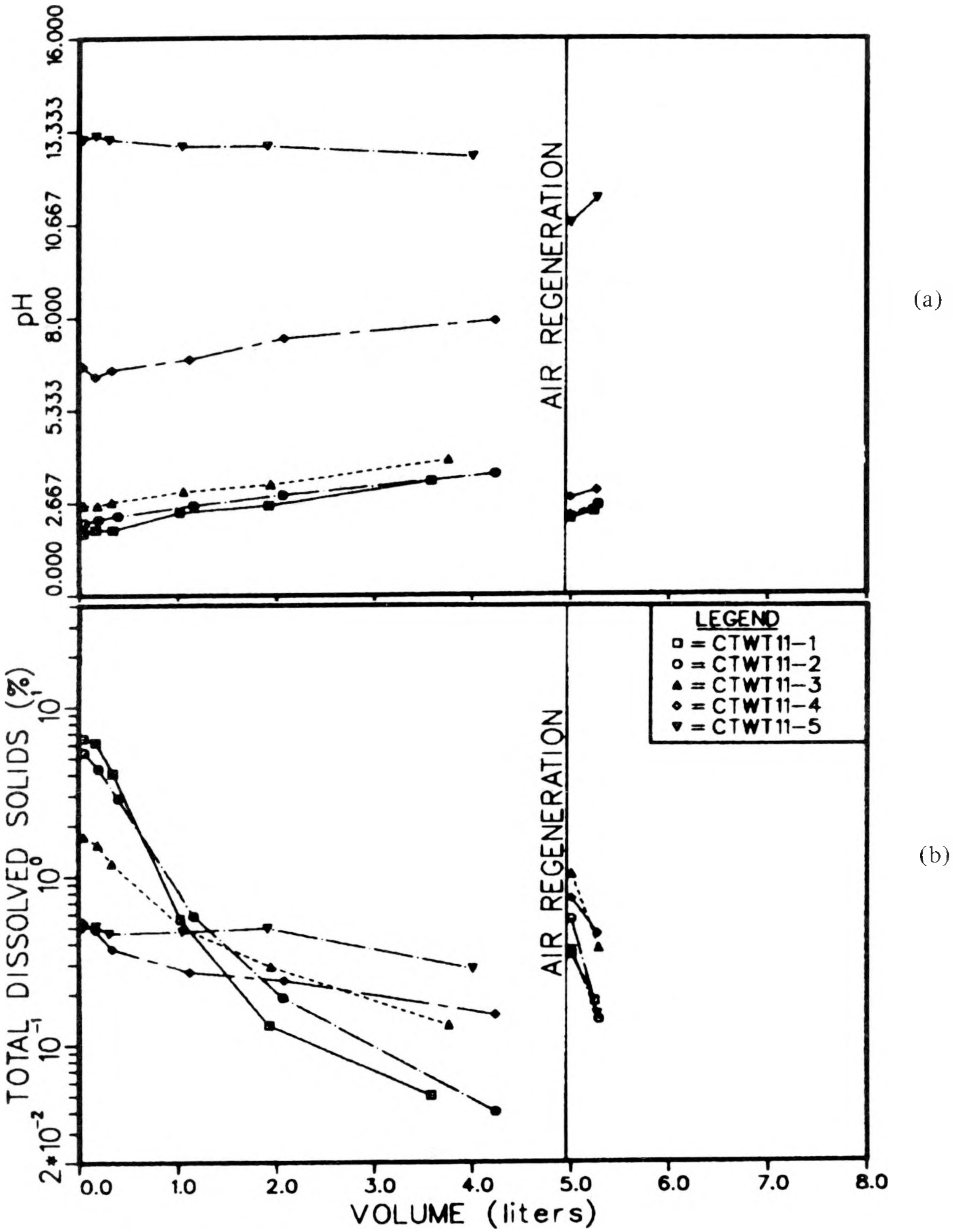

Fig. 2.
Leachate pH and TDS versus leachate volume for column leaching study of lime/refuse mixtures.

The mixture containing 3 wt% lime was especially interesting because a leachate pH of 7 was maintained for nearly the entire duration of the continuous part of the leaching experiment (until 4.2 ℓ had been passed through the column). The TDS values for this refuse/lime combination were also very respectable (ranging downward from about 0.6 wt%), especially considering that the dissolution of the lime itself adds substantially to the dissolved solids content of the solution. By the end of the continuous part of the leaching experiment, concentrations of troublesome trace elements, especially iron and manganese, had been reduced to environmentally acceptable levels. Regeneration of this refuse/lime mixture with air did tend to lower the leachate pH and to elevate the trace element concentrations. However, we did not continue the study long enough after the regeneration point to determine subsequent behavior.

The codisposal of alkaline agents, such as lime, with acidic coal refuse materials does appear to be an attractive option for controlling trace element contamination of disposal area drainages. The technique is only moderately costly ($0.50 to $1.00 per ton of cleaned coal, see Appendix B) and appears to be a highly effective means of preventing the release of a contaminated drainage from coal refuse dumps. The technology for mixing alkaline agents with coal refuse materials should be relatively simple and is immediately effective.

There are also a few questionable aspects connected with the use of alkaline additives for coal refuse materials. One uncertainty involves the long-term effectiveness or permanency of the method. Also, the durability and immobility of the alkaline additives over long geologic periods must be demonstrated. Another potential drawback of codisposing alkaline additives with high-sulfur coal refuse materials concerns the RCRA classification of the resulting refuse/additive mixtures. It is not at all clear whether such a mixture would be classified as hazardous or nonhazardous. As pointed out earlier, a hazardous RCRA designation could be quite costly for the disposal site operator. Another somewhat negative aspect of refuse codisposal with alkaline agents (as compared to refuse calcining, for example) is its low potential for by-product recovery. The lack of such potential, of course, negates the possibility of offsetting environmental costs with recovered product value.

Addition of Sorbents or Attenuating Agents to Discarded Refuse Materials

The codisposal of attenuating agents or sorbents, other than lime or limestone, with acidic coal refuse materials also has great potential for reducing or abating trace element contamination of disposal site drainage. Many natural materials, such as certain types of clays and soils, and many industrial wastes, such as fly ash or alkaline sludges, may have considerable capacity to attenuate contaminated refuse drainage, and often these materials are available in large and accessible quantities near refuse disposal sites. Some of our research during the year was directed at assessing the potential of various attenuating agents for reducing the trace element and acid compositions of coal refuse leachates and thus at revealing the possible effectiveness of this class of agents as refuse dump additives.

Our initial investigation into this area included a series of natural and man-made materials collected from various parts of the country. In one set of experiments, acidic coal refuse leachates were equilibrated with eleven solid sorbent materials to evaluate their trace element attenuation capacities. The solids used were

- $CaCO_3$ (standard)
- Acid mine drainage treatment sludge
- Bottom ash from a western power plant
- Precipitator ash (fly ash)
- Bottom slag from a midwestern plant burning western coal
- SO_2 scrubber sludge from a midwestern plant burning western coal

- Alabama soil
- Illite clay
- Montmorillonite clay
- Kaolinite clay
- Sea sand (two replicates).

The experimental procedure consisted of shaking the solid with the coal refuse leachate* for 15 h, measuring the pH, and analyzing the filtrate for trace elements. Companion experiments in which the solids were shaken with distilled water were carried out to evaluate the alkalinity of the sorbent and to determine its water soluble components. The pH values of the filtrates from the solid attenuating materials previously mixed with distilled water ranged from 5.8 (sea sand) to 11.2 (precipitator ash). The pH values of the filtrates from the refuse leachate/solid mixture ranged from 2.8 (sea sand) to 9.6 (precipitator ash). As a general rule, the higher the pH, the lower the trace element concentrations. Total dissolved solids are not included in these discussions because after equilibration, the soluble matter of some sorbent materials artificially elevated the TDS values in the leachates.

The results of these experiments are discussed with reference to Tables XII and XIII. Table XII has essentially all the data pertinent to the experiments, including the liquid/solid ratios, the measured pH values, and the trace element concentrations. In general, Table XII is self explanatory and points out the potential benefits of using some of the coal combustion by-products and naturally occurring clays as a treatment for coal refuse drainages, even where the acid content of the drainage is quite high. Table XIII lists in a more qualitative manner, the performance of the various sorbents with regard to leachate pH elevation and attenuation of the 13 trace elements that we have identified as being of greatest environmental concern in the Illinois Basin coal refuse effluents. We have isolated different sections in Table XIII to draw attention to some of the salient features. For example, because the solubilities of Fe^{+3} and Al^{+3} are highly dependent on pH in acidic solutions, those sorbents that are most effective in elevating the pH are also most effective in decreasing the concentrations of Fe^{+3} and Al^{+3}. The results listed in Table XIII are quite striking and demonstrate clearly that 7 of the 11 sorbents tested are effective in controlling the key leachate parameters. Cost analyses of various codisposal options, including lime and fly ash with and without limestone modification, are included in Appendix B (Table B-IV) and are favorable for several options.

In another series of investigations, 14 subsurface soils from the Illinois Basin were tested to determine the ability of these materials to reduce the trace element and acid concentrations in contaminated coal refuse leachates. These soils represent a cross section of the types found in the coal producing regions of the Basin. Soil properties ranged from noncalcareous to calcareous, unweathered to weathered, low to high clay content, and low to high cation-exchange capacities. Only one soil had appreciable organic content.

The experimental procedure for this study involved a series of successive dilutions with each soil type. First, a moderately contaminated coal refuse leachate was agitated for about 16 h using a 5:1 leachate:soil ratio (by weight) for each of the noncalcareous soils and a 10:1 leachate:soil ratio for the calcareous soils (see Table XIV). The latter condition was chosen because of the expected higher acid-attenuating capacities of the calcareous soils. (This first set of leachate/soil equilibrations is designated Leach Step 1 in Table XIV.) Fresh soil was then added to the filtrates from the first leach step, and the mixtures were again agitated for a 16-h period (Leach Step 2). This cycle of equilibration followed by fresh soil addition was carried out as many as five times for some of the leachate/soil mixtures. This information, along with data on leachate pH and trace element composition, are shown in Table XIV. A qualitative assessment of the leachate attenuating capacities of each of the soil types appears in Table XV.

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*The coal refuse leachate had a pH of 2.6 and a strong yellow color, indicating that most of the Fe^{+2} had been converted to Fe^{+3}.

TABLE XII

RESULTS OF EXPERIMENTS USING ELEVEN SORBENTS FOR pH CONTROL AND TRACE ELEMENT ATTENUATION FOR AN ILLINOIS BASIN HIGH-SULFUR COAL REFUSE LEACHATE

					Element Levels Removed												
Sample	Sorbent	Liquid	Liquid/Sol Ratio	pH	F PPM	Na PPM	Al PPM	K PPM	Ca PPM	Cr PPB	Mn PPM	Fe PPM	Co PPM	Ni PPM	Cu PPM	Zn PPM	Cd PPB
0		Leachate		2.64	2.0	108	10	12	275	22	4.0	105	1.9	2.6	0.055	1.02	6.0
0		H_2O			<0.1		<0.5			<1	<0.02	<0.05	<0.05	<0.03	<0.02	<0.01	<0.1
1	AMD Treatment Sludge	Leachate	4	7.63	0.2	82	<0.2	11	600	1.0	0.11	0.2	<0.1	<0.05	<0.01	0.04	8
2		H_2O	4	7.67	0.2	5	0.6	2.3	590	5.3	0.05	0.2	<0.1	<0.05	<0.01	0.04	1
3	Bottom Ash	Leachate	3	8.08	1.5	150	<0.2	12.2	520	1.1	3.40	0.2	0.4	0.63	0.05	0.07	1.3
4		H_2O	3	8.59	0.7	35	<0.2	1.8	54	1.5	0.05	0.1	<0.1	<0.05	0.01	0.01	0.2
5	Precipitator Ash	Leachate	3	9.63	1.6	163	0.6	11.2	550	73	0.04	0.2	0.1	<0.05	0.01	0.02	0.1
6		H_2O	3	11.23	1.1	49	2	2.6	65	90	0.04	<0.1	<0.1	<0.05	0.01	<0.005	0.1
7	Illite	Leachate	3	8.08	1.5	145	0.6	50	450	1.6	0.25	0.3	0.1	0.07	<0.01	0.35	0.3
8		H_2O	3	7.84	1.6	22	<0.5	18	34	5.0	0.02	<0.1	<0.1	<0.05	<0.01	0.01	1.5
9	Montmorillonite	Leachate	9	7.95	0.4	770	7	17	110	4.4	0.67	1.4	0.2	0.22	0.03	0.19	2.3
10		H_2O	9	8.46	0.2		21		2	3	<0.1	1	<0.3	<0.3	<0.07	<0.05	<0.4
11	Kaolinite	Leachate	3	4.29	2.6	106	13.5	18	290	3.6	40.1	6.4	1.5	2.47	0.27	18.4	20
12		H_2O	3	6.03	0.3	6	0.4	1.6	<2.5	1.1	0.01	<0.1	<0.1	0.02	0.01	<0.005	0.2
13	Alabama Soil	Leachate	3	4.01	2.3	90	10	4.6	220	1.4	17.1	9.6	1.3	1.02	0.06	0.50	8
14		H_2O	3	6.30	0.2	5	<0.2	0.7	<2.5	<1	0.04	<0.1	<0.1	<0.05	0.01	0.01	1
15	$CaCO_3$	Leachate	3	8.03	0.16	102	<0.2	16	160	29	0.01	<0.1	0.1	0.12	0.02	0.01	0.5
16		H_2O	3	9.14	0.15	11	<0.2	4.9	4	163	<0.01	<0.1	<0.1	<0.05	0.02	<0.005	0.3
17	Bottom Slag	Leachate	3	4.23	0.9	102	2	15	330	<1	3.78	108	1.6	3.34	0.04	1.30	51
18		H_2O	3	7.63	0.2	2.5	0.5	1.5	16	<1	<0.01	0.35	<0.1	<0.05	0.01	0.01	0.4
19	SO_2 Scrubber Sludge	Leachate	3	7.34	4.0	125	<0.2	46	760	2.8	2.20	<0.1	0.7	0.80	0.02	0.60	26
20		H_2O	3	8.35	4.2	25	<0.2	33	400	8.4	0.05	<0.1	<0.1	<0.05	0.01	0.02	3.0
21	Sea Sand	Leachate	3	2.80	1.8	95	11	12	250	29	3.52	120	1.7	2.2	0.13	0.98	9.0
22		H_2O	3	5.88	0.2	<1	<0.2	0.2	<2.5	<1	0.01	<0.1	<0.1	<0.05	0.01	<0.005	0.3
23	Sea Sand	Leachate	3	2.81	1.5	95	11	11	250	21	3.40	113	1.6	2.10	0.13	0.97	7
24		H_2O	3	5.83	0.17	<1	<0.2	0.4	<0.5	2.5	<0.01	<0.1	<0.1	<0.05	<0.01	<0.005	0.8

TABLE XIII

COMPARISON OF CAPABILITIES OF ELEVEN SORBENTS TO ELEVATE pH AND TO ATTENUATE THIRTEEN TRACE ELEMENTS IN ILLINOIS BASIN COAL REFUSE LEACHATES*

Sorbent	pH	Fe	Al	Ni	Mn	Zn	Co	Cr	Cu	F	Cd	Na	K	Ca
$CaCO_3$	EEE	EEE	EEE	GG	EEE	EEE	GG	P	FF	GG	GG	P	P	P
AMD Treatment Sludge	EEE	EEE	EEE	EEE	GG	GG	GG	GG	GG	GG	P	P	P	o
Illite	EEE	EEE	GG	GG	GG	FF	GG	GG	GG	P	GG	P	o	o
Precipitator Ash	EEE	EEE	GG	GG	EEE	GG	GG	o	FF	P	GG	P	P	o
Montmorillonite	EEE	EEE	FF	GG	GG	GG	GG	GG	FF	GG	FF	o	P	FF
Bottom Ash	EEE	EEE	EEE	FF	P	GG	FF	GG	P	P	FF	P	P	o
SO_2 Scrubber Sludge	EEE	EEE	EEE	FF	P	P	P	FF	FF	o	o	P	o	o
Alabama Soil	FF	GG	P	P	o	P	P	GG	P	P	P	P	P	P
Bottom Slag	FF	P	FF	P	P	P	P	GG	P	P	o	P	P	P
Kaolinite	FF	GG	P	P	o	o	P	FF	o	P	o	P	P	P
Sea Sand	P	P	P	P	P	P	P	P	o	P	P	P	P	P

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*EEE = >100x Reduction
GG = 10 to 100x Reduction
FF = 3 to 10x Reduction
P = 0.5 to 3x Reduction
o = >2x Increase

TABLE XIV

TRACE ELEMENT ANALYSES OF SOIL/REFUSE LEACHATE EQUILIBRATIONS

							Element Levels										
ATSB Sample	Soil Sorbent	Comments	Liquid	Leach Step	Liq/Sol Ratio[a]	pH	F PPM	Al PPM	Ca PPM	Cr PPB	Mn PPM	Fe[b] PPM	Co PPM	Ni PPM	Cu PPM	Zn PPM	Cd PPB
0	Initial Leachate	Ferric Leachate				2.65	1.4	8.5	355	4	4.1	130	1.0	2.2	0.04	1.0	6.5
50	Alluvium	Weathered	H_2O		5	8.3	0.8	<1	26	<2	<0.06	0.3	<0.2	<0.2	<0.03	<0.04	<0.2
51		Partly Calcareous	Leachate	1	10	6.7	0.6	<0.5	500	<1	1.5	<0.06	<0.1	0.2	<0.02	<0.02	0.5
52				2	5	7.4	0.5	<0.5	420	<1	0.07	<0.06	<0.1	<0.1	<0.02	<0.02	0.1
90	Alluvium	Weathered	H_2O		5	7.7	0.6	2.4	2	<2	<0.06	2.1	<0.2	<0.2	<0.03	<0.04	<0.2
91		*Leached*	*Leachate*	1	5	4.2	2.4	3	310	3.5	9.5	<0.3	<0.5	0.8	<0.02	9	13
92				2	4	5.7	0.3	<3	310	<5	7.5	<0.3	<0.5	<0.5	<0.02	4	4
93				3	4	5.9	0.1	<0.5	290	<1	5.3	<0.06	<0.1	<0.1	<0.02	2	2
10	Loess	Unweathered	H_2O		5	7.6	0.7	<0.5	13	<1	<0.03	<0.06	<0.1	<0.1	<0.02	<0.02	0.1
11		Calcareous	Leachate	1	5	6.1	16	<2	410	<5	3.1	<0.3	<0.5	<0.5	<0.08	<0.1	2.5
12				2	4.5	7.6	2.3	<2	390	<5	<0.8	<0.4	<0.5	<0.5	<0.08	<0.1	0.4
13				3	4	7.6	0.6	0.5	330	<1	0.1	<0.06	<0.1	<0.1	<0.02	<0.02	0.3
20	Loess	Weathered	H_2O		5	5.6	0.3	<1	2	<2	<0.06	1	<0.2	<0.2	0.1	<0.04	0.4
21		Leached	Leachate	1	5	3.5	3.4	15	260	<5	7.0	<0.3	<0.5	1.5	<0.08	0.7	7
22				2	4.5	4.0	2.4	8	210	<5	5.8	<0.3	<0.5	<0.6	<0.1	0.3	7
23				3	4	4.1	3.5	4	190	<5	4.6	<0.3	<0.5	<0.5	<0.02	0.3	5
24				4	4	4.1	2.5	<6	140	<10	4.9	<0.8	<1	<1	<0.2	<0.3	5
25				5	2	4.1	1.6	<5	120	<10	3.0	<0.6	<1	<1	<0.2	<0.2	3
60	Loess	Unweathered	H_2O		5	8.1	0.5	<1	16	<2	<0.06	<0.1	<0.2	<0.2	<0.03	<0.04	<0.2
61		Calcareous	Leachate	1	10	6.4	0.6	<0.5	410	<1	2.8	<0.06	0.2	0.7	<0.02	0.02	1.1
62				2	5	7.4	0.6	<0.5	410	<1	0.7	<0.06	<0.1	<0.1	<0.02	<0.02	0.2
80	Loess	Unweathered	H_2O		5	8.2	0.4	<0.5	16	<1	<0.03	<0.06	<0.1	<0.1	<0.02	<0.02	<0.1
81		Calcareous	Leachate	1	10	6.4	0.6	<0.5	440	<1	2.7	<0.06	0.35	<0.1	<0.02	0.04	2
82				2	5	7.4	<1	<3	430	<5	0.35	<0.3	<0.5	<0.5	<0.02	<0.1	<0.5
83				3	4	7.6	<2	<6	380	<10	<0.4	<0.8	<1	<1	<0.02	<0.3	<1
84				4	2	7.4	<2	<6	390	<10	<0.4	1.6	<1	<1	<0.02	<0.3	<1
85				5	1	7.3	0.7	<2	390	<10	0.2	<0.2	<0.2	<0.3	<0.02	<0.05	<0.5
130	Loess	Weakly Weathered	H_2O		5	8.0	0.5	<1	11	<3	<0.08	<0.2	<0.3	<0.3	<0.04	<0.05	<0.2
131		Leached	Leachate	1	5	3.8	1.1	5	370	<3	6.9	0.3	0.2	1.2	<0.05	0.6	6
132				2	4.5	6.5	<0.6	<3	360	<5	3.2	<0.3	<0.5	0.5	<0.08	<0.1	0.9
133				3	5	7.2	0.3	<0.5	360	<1	0.9	<0.1	<0.06	<0.1	<0.02	<0.04	0.2
30	Organic	Weakly Weathered	H_2O		5	8.1	0.3	<1	620	<2	2.1	<0.1	<0.2	<0.2	<0.03	<0.04	<0.2
31		Calcareous	Leachate	1	10	6.8	6.8	<0.5	640	<1	2.7	<0.06	<0.1	<0.1	<0.02	<0.02	0.2
32		Organic		2	5	6.9	2.2	<2	660	<4	1.6	<0.2	<0.4	<0.4	<0.06	<0.07	<1
33				3	4	7.0	4.5	<6	650	<1	4.8	<0.8	<1	<1	<0.2	<0.3	<1
34				4	2	6.8	5.0	<6	710	<1	2.0	1.4	<1	<1	<0.2	<0.3	<1
35				5	1	7.0	1.1	<3	750	<6	2.4	0.3	<0.6	<0.6	<0.1	<0.1	<1
40	Till	Unweathered	H_2O		5	8.2	0.3	<0.5	40	<1	<0.03	<0.06	<0.1	<0.1	<0.02	<0.02	<0.1
41		Calcareous	Leachate	1	10	6.9	1.0	<0.5	580	<1	2.1	<0.06	0.1	0.3	<0.02	<0.02	0.8
42				2	5	7.5	1.0	<0.5	570	<1	0.3	<0.06	<0.1	<0.1	<0.02	<0.02	<0.1
70	Till	Strongly Weathered	H_2O		5	7.9	0.8	<1	6	4	<0.06	0.2	<0.2	<0.2	<0.03	<0.04	<0.2
71		Leached	Leachate	1	5	4.6	1.3	<2	290	<5	3.6	<0.3	<0.5	<0.5	<0.08	0.2	0.43
72				2	4.5	7.2	<1	<2	240	<5	<0.2	<0.3	<0.5	<0.5	<0.08	<0.1	<0.5
73				3	4	7.4	<1	<3	240	<5	<0.2	<0.3	<0.6	<0.6	<0.10	<0.1	<0.5
74				4	4	7.4	<1	<6	220	<10	<0.4	<0.8	<1	<1	<0.2	<0.3	<1
75				5	2	7.4	<1	<5	250	<10	<0.3	<0.6	<1	<1	<0.2	<0.2	<1
100	Till	Unweathered	H_2O		5	8.2	0.5	<0.5	20	<1	<0.03	<0.06	<0.1	<0.1	<0.02	<0.02	<0.1
101		Calcareous	Leachate	1	10	6.9	0.8	<0.5	570	<1	2.0	<0.06	0.15	0.4	<0.02	<0.02	2
102				2	5	7.4	0.6	<0.5	490	<1	0.4	<0.06	<0.1	<0.1	<0.02	<0.02	0.1
110	Till	Unweathered	H_2O		5	8.2	0.4	<0.5	50	<1	<0.03	<0.06	<0.1	<0.1	0.04	<0.02	<0.1
111		Calcareous	Leachate	1	10	6.8	0.7	<0.5	520	<1	2.3	<0.06	0.30	0.7	<0.02	<0.02	<0.1
112				2	5	7.6	0.6	<0.5	510	<1	0.9	<0.06	<0.1	<0.1	<0.02	<0.02	<0.1
120	Till	Partly Weathered	H_2O		5	8.2	0.4	<0.5	41	<1	<0.03	<0.06	<0.1	<0.1	0.02	<0.02	<0.1
121		Calcareous	Leachate	1	10	6.8	0.5	<0.5	540	<1	2.0	<0.06	0.2	0.5	<0.02	<0.02	<0.1
122				2	5	7.6	0.5	<0.5	470	<1	0.3	<0.06	<0.1	<0.1	<0.02	<0.02	<0.1
140	Till	Partly Weathered	H_2O		5	8.5	<1.2	<5	12	<10	<0.3	<0.6	<1	<1	<0.2	<0.2	<1
141		Calcareous	Leachate	1	10	6.9	0.7	<0.5	490	<1	0.7	<0.06	<0.1	0.3	<0.02	<0.02	<0.1
142				2	5	7.6	<0.7	<3	430	<5	<0.2	<0.3	<0.5	<0.5	<0.08	<0.1	<0.5
143				3	4	7.6	<1	<6	390	<10	<0.4	<0.8	<1	<1	<0.2	<0.8	<1
144				4	2	7.7	<1	<6	330	<10	<0.4	<0.8	<1	<1	<0.2	<0.3	<1
145				5	1	7.7	0.3	<3	260	<5	<0.2	<0.3	<0.6	<0.6	<0.1	<0.1	<0.5

[a] Grams of liquid per gram of soil

[b] Ferric state

TABLE XV

BATCH ATTENUATION OF TRACE ELEMENTS IN COAL REFUSE LEACHATES BY SOILS

		Soil Parameters				Degree of Attenuation[a]										
Soil Type	Soil No.	pH	CO_3 (%)	CEC (meq/g)[b]	OM (%)[c]	pH	Fe[d]	Al	Zn	Ni	Co	Cr	Cd	Mn	F	Ca
Till	10	8.2	15.1	91	0.4	EEEE	EEEE	EEEE	EEEE	GG	GG	GG	GG	FF	FF	P
Till	11	8.2	13.4	77	0.9	EEEE	EEEE	EEEE	EEEE	GG	GG	GG	GG	FF	FF	P
Till	12	8.2	9.2	96	0.2	EEEE	EEEE	EEEE	EEEE	GG	GG	GG	GG	FF	FF	P
Till	4	8.2	8.6	89	0.9	EEEE	EEEE	EEEE	EEEE	GG	GG	GG	GG	FF	FF	P
Loess	8	8.2	8.3	88	0.3	EEEE	EEEE	EEEE	EEEE	GG	GG	GG	GG	FF	FF	P
Till	14	8.5	7.7	143	0.2	EEEE	EEEE	EEEE	EEEE	GG	GG	GG	GG	GG	FF	P
Organic	3	8.1	6.8	303	7.3	EEEE	EEEE	EEEE	EEEE	GG	GG	GG	GG	FF	P	P
Loess	6	8.1	5.8	116	0.4	EEEE	EEEE	EEEE	EEEE	GG	GG	GG	GG	FF	FF	P
Alluvium	5	8.3	1.6	261	0.7	EEEE	EEEE	EEEE	EEEE	GG	GG	GG	GG	FF	FF	P
Loess	1	7.6	0.7	144	0.3	EEEE	EEEE	GG	GG	GG	FF	-	FF	P	o	P
Loess	13	8.0	0.4	98	0.2	FF	EEEE	FF	FF	FF	FF	FF	P	P	P	P
Till	7	7.9	0.3	280	0.3	FF	EEEE	GG	FF	GG	FF	-	GG	P	P	P
Alluvium	9	7.7	0.2	253	0.6	FF	EEEE	FF	o	FF	FF	P	P	P	P	P
Loess	2	5.6	0.0	279	0.5	P	EEEE	P	P	P	FF	-	P	P	P	P

[a] EEEE = >100x Reduction
GG = 10-100x Reduction
FF = 3-10x Reduction
P = 0.5-3x Reduction
o = >3x Increase

[b] Cation Exchange Capacity.

[c] Organic Material.

[d] Iron primarily in ferric state.

The data in Table XV are ordered according to the percentage of titratable carbonate in each soil. According to the effectiveness scheme used in the table, all of the soils with $>1.6\%$ carbonate content are rated as fair (FF) to excellent **(EEEE)** in attenuating the toxic elements present in the leachate. These results show that many alkaline soils do have a significant capacity to reduce the trace element and acid contents of refuse drainages. This attenuating capacity appears to function mainly on the strengths of these soils in controlling leachate acidity. The additional question concerning whether the cation-exchange capacity (CEC) of each soil type has any major bearing on the capacity of that soil to attenuate leachate contamination is somewhat more difficult to answer based on the data that we have obtained thus far. It is significant, however, that even those soils with essentially no acid-neutralizing capacity (Soils 2, 7, and 9) do attentuate many of the leachate contaminants somewhat. This observation lends credence to the postulate that both the alkalinity and ion-exchange capacity are important in determining the contaminant-attenuating properties of soils.

Even though our work with soils as an environmental control medium for acidic, coal refuse leachates is still in its early stages, it is already apparent that soils as a group (especially alkaline, unweathered soils) have great potential for this purpose because of their abundance and availability near coal refuse dumps. The results of our work this year were sufficiently encouraging to suggest that a cost estimate for this form of environmental control technology be made. Using a locally available soil with a 5 wt% titratable alkalinity, we estimate the cost per ton of cleaned coal to permanently treat highly acidic coal refuse matter (by intermixing with the refuse) to be in the range of \$0.80 to \$1.30, depending on the potential acidity of the refuse (see Appendix B, Table B-IV). These costs would, of course, be lower if some of the more highly alkaline soils listed in Table XV were to be used and if the soils also had significant ion-exchange capacity.

Future studies in this area will be aimed at quantifying both the total capacities and ion specificities of various soils during the attenuation of coal refuse leachates. This will be done by passing contaminated leachates through soil columns and by direct leaching of mixtures of various soils and acid refuse materials. The more promising control options will be scaled up to better duplicate field conditions.

Sealing Refuse From Air and Water

In view of the overwhelming evidence that isolating high-sulfur coal refuse from air and water will prevent the formation of acids and thereby the release of trace elements into the environment, one area of control technology that we are addressing is that of sealing the refuse pile, dump, or burial site to prevent the intrusion of air or water. The concept of sealing has overtones in all aspects of coal waste (and other waste) disposal. Sealants can be used for existing refuse piles and dumps and for near-surface and underground burial of wastes.

Various sealant scenarios, with an emphasis on clays, soils, calcite, and various cementing agents (Portland and silicate cements and polymers) as sealing agents, are being considered, and we have begun laboratory experiments to test some of our ideas. One of the first experiments involved slurrying crushed coal refuse (-3/8 in.) with 5 wt% lime in water and neutralizing the alkaline mixture by bubbling CO_2 through it until the pH was reduced to 7. This had the effect of coating the coal waste particles in the slurry with limestone by the reaction $CaO + CO_2 = CaCO_3$. The effectiveness of the seal was tested by drying the particles, performing a column leaching test on them, and measuring the pH and trace element compositions of the resulting leachates. The results of this experiment are given in Table XVI. These data reveal that this method of coating the refuse particles with a limestone film was very successful in controlling both the acid and trace element compositions of the refuse leachates. The pH values of the emerging leachates were maintained between 7.2 and 7.9 for the entire experiment, and the criteria pollutants (iron and manganese) were controlled within acceptable limits. The other elements reported in Table XVI are greatly reduced in concentration as compared to their levels in leachates from untreated refuse materials. (See control sample in Table D-II.)

Calcium carbonate coating of acid refuse materials is a promising method for controlling contamination of aqueous drainages. In practice a local self-contained unit could be designed to use this principle. The needed lime could be supplied from a small kiln, and the CO_2 and heat for drying the coated particles could also be produced by the kiln. Furthermore, coal fines or middlings from the cleaning plant could conceivably be used as the principal fuel for the kiln. The advantages of this combination in savings of energy and expense could be considerable.

In other work in the area, we have begun to investigate the feasibility of producing a concrete-like aggregate from mixtures of Portland cement and crushed acid coal refuse. The resulting product should be a marked improvement over the untreated refuse aggregate, including reduced permeability, acid-generating potential, and increased structural integrity. Several small cylinders (3.1 cm in diameter and 2.5 cm long) were produced using various proportions of mortar and -20-mesh refuse. Static leaching tests of several of these cylinders with distilled water for periods of up to 34 days revealed that the structure of the cylinders was not appreciably degraded by contact with water. Furthermore, the pH of the leachates ranged from about 9 to 11.5, suggesting that trace element leachability of the refuse would be substantially reduced.

One problem with using commercial cements to produce refuse aggregates is the high cost of structural-grade cement. Therefore, we will begin to explore the possibility of producing cements

TABLE XVI

RESULTS FROM A COLUMN LEACHING EXPERIMENT WITH CALCITE-COATED ILLINOIS BASIN COAL REFUSE

Sample No.[a]	1	2	4	11	17
Vol (ℓ)	0.100	0.201	0.697	2.309	3.326
pH	7.4	7.2	7.9	7.7	7.7
TDS (%)	0.84	0.63	0.34	0.27	0.22
F	0.3	0.4	0.3	0.3	0.4
Na	7	6	2.5	1	1
Al	<0.5	<0.5	<0.5	<0.5	<0.5
K	7	8	4	2	1
Ca	900	870	630	540	480
Cr (μg/ℓ)	<0.5	1	<0.5	<0.5	<0.5
Mn	0.7	0.5	0.2	0.1	0.07
Fe	5	2	0.4	<0.3	<0.3
Co	0.13	0.12	0.06	0.05	<0.05
Ni	0.3	0.2	0.2	0.1	<0.07
Cu	0.1	0.1	<0.1	<0.1	<0.1
Zn	0.07	0.07	0.03	0.01	<0.01
Cd (μg/ℓ)	2	1	0.4	0.2	0.3

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[a] Experimental conditions: 500 g of calcite-coated, -3/8-in. coal refuse material was packed into a 5-cm-diam by 40-cm-long glass column. Distilled water was passed upward through the column at a rate of 0.5 mℓ/min. Except where noted, element concentrations are μg/mℓ.

or cementitious materials from the refuse itself. This could involve the calcining of powdered refuse and limestone mixtures or perhaps the treatment of the refuse to produce a pozzuolanic material.

Subtask 1.2—Assess Technology to Control or Reduce Trace Element Contamination of Refuse Dump Drainages

The purpose of this subtask is to identify environmental controls to reduce or attenuate undesirable trace elements in the acidic drainages from coal refuse dumps. Our attention in this area has been given to pollution abatement techniques that have proved effective in treating acidic waste waters with compositions similar to coal refuse drainage. These techniques include alkaline neutralization, ion exchange, reverse osmosis, and permanganate oxidation. We have also initiated studies on the effectiveness of using a variety of sorbents, such as clays, soils, and solid coal combustion by-products, on high-sulfur coal refuse leachates. (The latter research was discussed above in Subtask 1.1.) In these studies we are continuing to give greatest emphasis to the control of the dozen or so trace elements that we have identified in our previous studies as being of greatest concern in the drainages from Illinois Basin coal refuse.

During the year, we were able to initiate a small number of cooperative projects with commercial organizations having expertise in water treatment. We supplied the contaminated leachates and performed the before and after chemical analyses, and the commercial organizations treated the supplied solutions. Of the companies contacted, General Mills Chemical, Inc. (Minneapolis, Minnesota) agreed to treat some of our high-sulfur refuse drainage solution using chelating agents; Bio-Rad Laboratories, Inc. (Richmond, California) used ion exchange; Carus Chemical Co. (LaSalle, Illinois) used permanganate oxidation; and UOP Fluid Systems Div. (San Diego, California) used reverse osmosis. Diamond Shamrock Chemical Co. (Redwood City, California), a supplier of ion-exchange resins, expressed an interest in our program and asked to be kept current.

Treatment of Contaminated Refuse Drainage by Alkaline Neutralization

Alkaline neutralization is used extensively to treat acid drainage from coal mines. Although it is well known that alkaline neutralization is very effective in controlling the acid and overall salt compositions of mine waste waters, the degree of control that this method exerts over some of the more highly leachable, toxic trace elements remains to be established. Elaboration of this latter point is the basis for the study that we conducted in this area.

The experiments conducted were basically titrations in which limestone, lime, or lye (NaOH) were added to one liter of contaminated refuse drainage (iron mostly in ferric state) until a predetermined pH value was reached. The solutions (or slurries) were allowed to sit overnight and then filtered, the pH values were measured, and the compositions of the resulting solutions were analyzed. A brief description of the experiments follows.

- Sample 0 was the control.
- Sample 5 was prepared by titrating with slightly more than the chemical equivalent of powdered limestone (31.65 g). The limestone was assumed to be pure $CaCO_3$ and buffering effects were neglected.
- Sample 6 was prepared by adding limestone (175 g) to one liter of waste water until there was no further pH change.
- Sample 2 had an elevated pH by adding about 0.2 g of lime to the 35 g of limestone used initially.

- Samples 4 and 1 both were neutralized using lime. The 14.5 g of lime in No. 4 produced a pH of 7.3, and the 17 g of lime in No. 1 produced a pH of 10.7.
- Sample 7 was neutralized with concentrated NaOH to increase pH without having the attendant calcium salt precipitation problem.

The results from these alkaline neutralization experiments are seen in Table XVII and show the effectiveness of this technique for decreasing trace element concentrations in coal waste leachates. The pH values and iron contents of the treated leachates are within acceptable limits based on the 1977 EPA effluent limitation guidelines for coal preparation plants (Fe $\leq 3.5\ \mu g/m\ell$ averaged over 30 days and pH 6-9). Manganese exceeds the acceptable level of 2.5 to 3 $\mu g/m\ell$ averaged over 30 days in the limestone case, however. This is due to the dissolution of manganese from the limestone during the neutralization of the leachate acid.

As were many techniques discussed in this report, alkaline neutralization was shown to be an effective method for reducing or abating trace element contamination of coal refuse drainage. The projected costs for such a treatment are relatively low ($0.10 to $0.80 per ton of cleaned coal, see Appendix B, Table B-IV). Also the technique is relatively easy to apply, as evidenced by the large number of neutralization plants already in operation to treat acid mine drainage.

However, in spite of the low cost and ease of application, alkaline neutralization has some rather considerable disadvantages. For example, it never really treats the source of contamination (that is, the refuse itself), and hence, its use in treating the drainage from the disposal site may be needed almost indefinitely. Also, although the standard refuse disposal practice involves burying the refuse on top of impermeable liners, such as clay, to channel refuse dump drainage into treatment areas, there is no assurance that drainage will not eventually escape through or around these liners and thus negate the effectiveness of this method. Another consideration that may make alkaline neutralization less attractive involves the costs associated with meeting RCRA requirements. Most certainly, waste materials disposed of in a way that produces contaminated drainage will be classified as hazardous. Thus the apparent low cost of alkaline neutralization may have to be tempered with additional costs needed to meet RCRA permit and performance requirements. Finally, there is little opportunity for by-product recovery during or subsequent to neutralization treatment. Thus the potential for realizing economic gain in this way is quite low.

Although alkaline neutralization as a refuse drainage treatment technique has some rather severe drawbacks, it is nonetheless widely used, highly accepted, and as this program has shown, very effective in controlling trace element contaminants. Undoubtedly, this method will continue to be used widely in the near future to treat contaminated coal refuse effluents.

Treatment of Contaminated Refuse Drainage by Ion Exchange

Bio-Rad Laboratories treated some of our high-sulfur refuse drainage solutions by ion exchange and returned them to us for analysis. The treatment consisted of flowing 250 $m\ell$/min of leachate in two equal fractions over 25 cm^3 resin beds (1.5 x 15 cm column) at a flow rate of about 2 $m\ell$/min. Four resins were used, making eight samples in all. The first fraction of the leachate was sufficient to swamp the two resins that were not strongly acidic. A second fraction of the leachate was too much for all the resins. In the best case, nearly 100 gal. of resin were needed per 1000 gal. of refuse drainage.

Table XVIII provides the catalog description of the resins used and our experiment identification numbers. Table XIX is a summary of the results of the ion-exchange experiments. Trace element analyses for 14 of the more common trace elements of environmental concern in the Illinois Basin are tabulated for the original leachate and for the treated solutions. It is clear from the data in the table that the acidic cation-exchange resins (AG-50W-X8 and AG-MP-50) depressed the pH of the original solution to even lower values. These, however, were most effective in reducing the trace element concentrations.

TABLE XVII

ANALYSES FOR THE CONTROL OF REFUSE DRAINAGE BY ALKALINE NEUTRALIZATION[a]

Sample No.	0	5	6	2	4	1	7
NEUTRALIZING AGENT	**NONE (CONTROL)**	**LIMESTONE**	**LIMESTONE**	**LIMESTONE + LIME**	**LIME**	**LIME**	**NaOH**
pH	1.1	7.1	7.4	6.6	6.6	10.7	5.9
TDS(%)	0.47	3.14	3.20	3.14	3.17	3.07	3.36
Na	2.4	3.8	4.8	3.8	3.8	4.0	9400
Mg	22	66	75	73	60	30	22
Al	18	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
K	2.7	4.3	8.4	3.9	3.5	3.3	4.6
Ca	170	7700	9300	8200	8200	8000	120
Sc							<0.01
Ti	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4
V	0.11	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cr(μg/ℓ)	15	1.0	0.5	2	16	62	1.5
Mn	3.6	6.4	4.4	3.3	1.0	0.1	0.07
Fe	820	0.3	0.3	0.4	0.3	0.3	0.06
Co	2.0	0.82	1.1	0.50	0.58	0.23	0.05
Ni	3.2	1.00	1.8	0.72	0.69	0.32	0.05
Cu	0.53	0.20	0.19	0.22	0.21	0.22	0.01
Zn	3.9	0.14	0.15	0.08	0.10	0.04	0.02
Rb							<0.01
Ag							<0.01
Cd(μg/ℓ)	18	4.6	0.4	<0.2	2.4	<0.2	0.6
Cs							<0.04
La							<0.5
Ce							<0.08
Sm							<0.02
Eu							<0.01
Tb							<0.1
Dy	0.07	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Pb(μg/ℓ)	13.5	<1	<1	<1	<1	<1	<1
Th							<0.02
U							<0.01

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[a] Values in μg/mℓ unless otherwise stated.

TABLE XVIII

EXPERIMENT IDENTIFICATION AND CATALOG DESCRIPTION OF RESINS USED IN BIO-RAD'S ION-EXCHANGE EXPERIMENTS ON HIGH-SULFUR COAL REFUSE LEACHATES

Experiment ID[a]	Dose	Resin	Mesh Size	Resin Description
811	1st	AG-50W-X8	-200 to -400	A strongly acidic cation exchange resin composed of nuclear sulfonic acid exchange groups attached to a styrene divinylbenzene polymer lattice.
812	2nd	AG-50W-X8		
821	1st	AG-MP-50	-200 to -400	A strongly acidic, macroporous cation-exchange resin with nuclear sulfonic acid exchange groups. The resin has an effective surface area approximating 35 m²/dry g or 30-35% porosity.
822	2nd	AG-MP-50		
831	1st	Chelex 100	-100 to -200	A chelating resin that is a styrene divinylbenzene copolymer containing iminodiacetate functional groups, structurally classed with the weak acid cation exchangers by virtue of its carboxylic acid groups.
832	2nd	Chelex 100		
841	1st	AG-501-X8	-20 to -50	A mixed bed resin for deionization with equivalent amounts of AG-50W-X8 H^+ form and AG-1-X8 OH^- form.
842	2nd	AG-501-X8		

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[a] Leachate CTWT-8 was used.

TABLE XIX

SUMMARY OF pH, TDS, AND TRACE ELEMENT COMPOSITIONS RESULTING FROM ION-EXCHANGE TREATMENT OF A HIGH-SULFUR COAL REFUSE LEACHATE[a]

	INITIAL LEACHATE	AG−50W−x8		AG−MP−50		Chelex 100		AG−501−x8	
		811	812	821	822	831	832	841	842
pH	1.8	0.85	1.45	0.88	2.00	2.68	2.54	1.94	2.37
TDS(%)	3.26	2.62	3.12	2.52	3.37	3.15	3.99	1.74	3.33
Al	540	<0.5	26	4	430	110	820	180	440
Ca	170	0.9	7.8	0.6	4.2	290	360	115	330
Cd(μg/ℓ)	400	0.4	140	0.4	90	70	650	110	120
Co	7	<0.05	8	0.4	10	0.3	16	5	9
Cr(μg/ℓ)	160	110	135	270	220	220	410	180	300
Cu	0.15	<0.02	0.15	<0.02	0.15	<0.02	0.06	0.1	0.16
F		2.8	4.4	3.3	5.1	2.5	6.3	3.3	5.2
Fe	6600	0.2	7550	36	8190	4820	7550	3910	7370
K	28	2.3	23	1.2	3.3	30	12	17	0.04
Mn	15	<0.02	13	1	15	0.3	21	8	14
Na	8	2.6	17	120	48	33000	1170	11	9
Ni	17	0.08	13	1	15	0.3	21	8	14
Ti		<0.5	<0.5	1.2	1.2	<0.5	<0.5	<0.5	<0.5
Zn	27	0.06	28	1	31	0.3	65	14	28

[a] All concentrations in μg/mℓ except where noted.

These experiments are preliminary in nature and were designed to demonstrate the applicability of this particular water cleaning technology to the problem of undesirable trace element contamination of high-sulfur coal preparation waste leachates. Therefore, we did not attempt to complete an economic analysis of this control option. Using data from the literature, we estimate that the cost of treating acid refuse drainage with ion exchange would be in the area of $0.29 per ton of cleaned coal (Appendix B, Table B-IV). It is recognized that should we pursue the ion-exchange method for acid mine drainage and refuse leachate clean-up, we should also need to consider (a) the effect of solution pH control on trace element removal, (b) resin type, (c) resin capacities, (d) resin regeneration, and (e) related items, such as capital and operational costs. It is clear, however, that ion exchange can reduce concentrations of trace elements of environmental concern in refuse leachates to acceptable levels (note Expt No. 811 in Table XIX), though the need to further treat these solutions for acid greatly reduces the applicability of the method. This and the known tendency of ion-exchange to overload or plug when the contaminant or suspended solids contents are high lead us to believe that ion exchange might be most applicable as a secondary treatment method to clean up the effluents from some other control process.

Treatment of Contaminated Refuse Drainage by Reverse Osmosis

Reverse osmosis (RO) is a technique that is used widely to desalinate seawater and other types of contaminated drainages produced by agricultural and industrial operations. In this method, a series of semipermeable membranes or filters are used to segregate or isolate dissolved contaminants from the main volume of water. Separation is achieved by forcing water that is relatively free of contaminants through the filter while retaining the contaminants in a concentrated liquor on the upstream side of the filter. This produces clean or product water and contaminated or reject water. There are many variables that can affect the performance of an RO water treatment system, including the composition of the contaminated water, the efficiency and selectivity of the filter material, and the number of times the water is passed through the filter bank.

A series of preliminary, bench-scale experiments to test the effectiveness of RO at cleaning contaminated refuse water were performed for us by UOP Fluid Systems Division. In these experiments, UOP used two types of RO filters designated as Filters 1 and 2 in Table XX. Filter 1 was UOP's RC-100, which is a poly(ether/urea) membrane, and Filter 2 was UOP's PA-300, which is a poly(ether/amide) membrane. The initial feed solution (Sample 01) of contaminated refuse drainage was passed through each of these RO filters. The compositions of the respective product or treated waters from the first pass through each of the filter units are listed under Samples 02 and 03 in Table XX. These data show that both filters were quite effective at reducing the priority trace elements in the refuse leachates to acceptable levels. Filter 1 appeared to be the better of the two for this purpose, but a suspected break in Filter 2 probably negated its anticipated better performance. (Note that as was the case for the ion-exchange studies that we conducted, reverse osmosis did not appreciably affect the pH of the refuse leachates.) The analyses for the combined reject waters from the first passes through both membranes also are listed in Table XX (Sample 04).

In the next stage of the RO experiment, the reject water (now the feed solution) was split and passed through each filter type. This process was continued until the reject water had been successively passed through each filter five more times. The analyses for the now highly concentrated feed solution just before the seventh pass through the filters (Sample 19) and the analyses of the cleaned or product water derived by RO from this concentrated feed (Samples 20 and 21) are in the last three columns of Table XX. These latter data, of course, reveal the perhaps marginal effectiveness of the RO method at treating highly concentrated waste leachates. Here Filter 1 (the RC-100 membrane) still reduced the concentrations of trace contaminants to acceptable levels, whereas the iron and manganese levels of the product water from treatment with Filter 2 exceeded presently established point source levels. The reduction of iron content during the first and seventh passes through each membrane is depicted graphically in Fig. 3.

An important consideration in the use of RO concerns the ratio of the final reject water that will still need further treatment before final disposal and the total amount of water treated. Peak recovery when placing six filters in series is 80-85%. Thus 15-20% of the original volume of drainage will still need to be treated. The magnitude of water is less, but all of the contamination is still present and still needs to be treated.

The work that we have conducted thus far shows that RO, like ion exchange, can be quite effective under some circumstances for treating trace element contamination in coal refuse drainage. RO is marginally effective for highly concentrated leachates (Table XX) and is apparently quite susceptible to membrane fouling by suspended particulates and solids. In addition, it is necessary to further treat the effluents from RO to reduce the acidity to acceptable levels. These considerations suggest that RO, like ion exchange, may function best as a secondary method to polish off effluents from the alkaline neutralization of acid refuse drainages. Data in the literature suggest that the cost of using RO to treat coal refuse drainage would be in the range of $0.20 per ton of cleaned coal (Appendix B, Table B-IV).

Permanganate Oxidation to Treat Coal Refuse Drainage

One of the problems with direct alkaline neutralization of coal refuse drainage to control trace contaminants is that some elements (notably, iron and manganese) are not precipitated from

TABLE XX

TRACE ELEMENT ANALYSES ON REVERSE OSMOSIS EXPERIMENTS

Sample No.[a]	**01**	**02**	**03**	**04**	**19**	**20**	**21**
Pass thru system	1st	1st	1st	1st	7th	7th	7th
Process Position	Feed	Product	Product	Reject	Feed	Product	Product
Filter Type[b]	-	1	2	1+2	-	1	2
Cond (μmhos)	5 090	1 780	1 140	-	21 600	4 710	3 280
pH	2.38	2.39	2.61	2.36	2.02	1.93	2.15
TDS (%)	0.55	<0.01	<0.01	0.73	4.54	0.02	0.05
Al	174	<0.1	0.7	206	1 390	<0.1	8.9
Ca	230	7.4	4.6	280	1 480	3.3	7.9
Cd (μg/ℓ)	102	<1	<1.0	110	680	<1	5
Co	0.8	<0.01	<0.01	0.9	6.4	<0.01	0.05
Cr	0.19	<0.001	<0.001	0.26	1.8	0.005	0.03
Cu	2.0	<0.01	0.01	2.5	20.7	<0.01	0.16
F	4.1	0.1	0.1	4.6	9.9	0.1	0.3
Fe	235	0.08	1.64	298	2 020	0.22	17.8
Mn	73.7	<0.01	0.32	86.6	620	0.02	4.4
Ni	2.1	<0.01	<0.01	2.6	23.3	<0.01	0.16
Zn	7.4	0.01	0.04	8.8	58.3	0.02	0.46

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[a] Concentrations reported as μg/mℓ unless noted otherwise.

[b] See text.

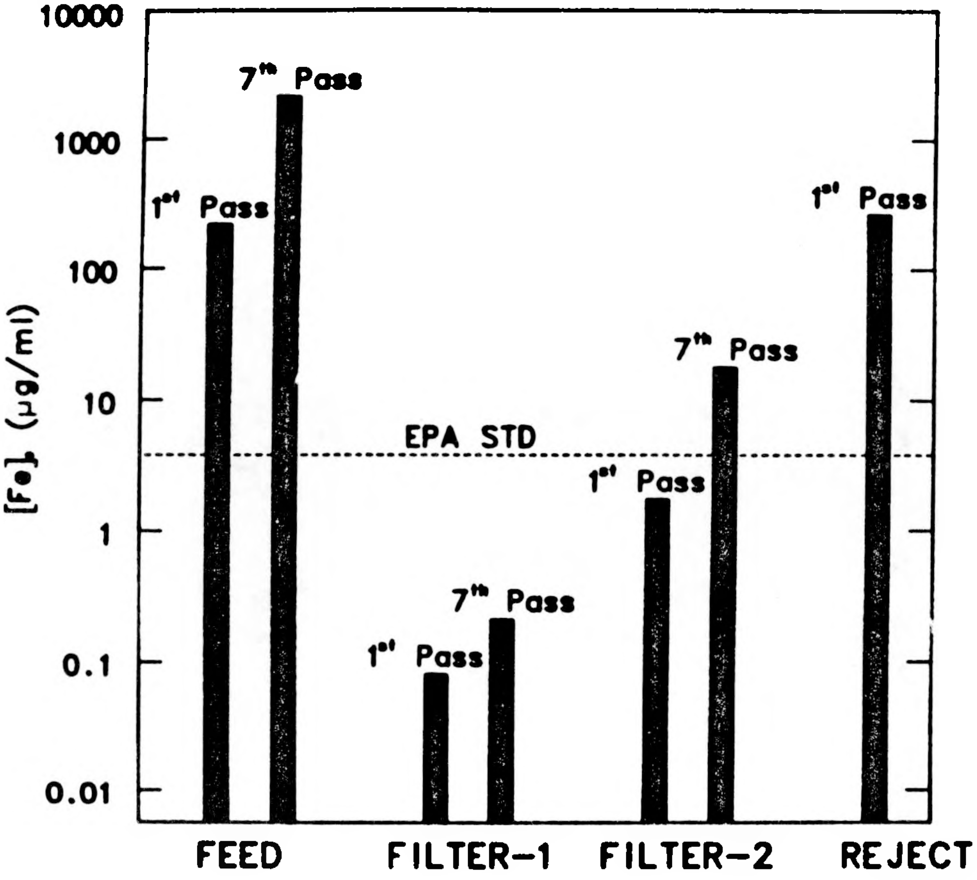

Fig. 3.
Iron levels in aqueous streams of a reverse osmosis system.

solution in their lower oxidation states by merely adjusting the pH to the neutral point. To circumvent this problem, acid drainage treatment facilities usually precede the neutralization step by some type of oxidation reaction to convert such components as Fe^{+2} and Mn^{+2} to higher oxidation states that precipitate from solution in the range of pH 5 to 6.

During the year, Carus Chemical Company conducted several experiments for us in which permanganate was used as an oxidizing agent to maximize the valences of the components in a contaminated coal refuse leachate. The intent here, of course, was to increase the effectiveness of the alkaline neutralization of the resulting solution. The leachate used had a very high concentration of iron, over 50% of which was in the Fe^{+2} state. The product water produced by the combined oxidation/neutralization process shows clearly that this method can be quite effective at reducing the trace elements in a contaminated refuse drainage sample to acceptable levels. (See Table XXI.)

Because of the success with neutralization alone, we do not have further experiments planned in this area.

Chelating Agents to Control Trace Elements in Coal Refuse Drainage

General Mills Chemical, Inc. participated with us in an investigation of the use of chelating agents to remove heavy metal contaminants from coal refuse drainages. Our collaborators at

TABLE XXI

ALKALINE NEUTRALIZATION/PERMANGANATE OXIDATION OF CONTAMINATED COAL REFUSE DRAINAGE

Parameter[a]	Starting Solution	Carus Sample No. 2[b]
pH	1.8	6.92
TDS (%)	3.25	0.45
Al	540	<0.1
Ca	170	520
Cd	0.4	<0.001
Cr	0.16	0.002
Co	7	<0.01
Cu	0.15	<0.01
Fe	6600	0.055
Mn	15	0.36
Ni	17	<0.01
Zn	27	0.01
Na	8	11.8
K	28	860

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[a] Concentrations reported as mg/ℓ.

[b] pH adjusted to 7 with $Ca(OH)_2$.

General Mills treated a contaminated refuse leachate that we sent them with several commercial and experimental chelating agents. We have not yet received the results of these experiments from them. However, they have reported that because of the highly acidic leachates (pH ~ 2), none of the agents tested were effective in reducing the metals content.

No further experiments are planned in this area.

Subtask 1.3—Define Options for Controlling Trace Element Releases in the Drainages From Coal Refuse

The purpose of this activity is to assess the results and major implications stemming from the research that we have conducted thus far on environmental control technologies for trace element contamination of coal refuse drainages. Also in this subtask we will delineate those areas where more work needs to be done, either to complete our understanding of the various environmental options or to solve specific control problems. The foregoing discussions have emphasized the technical feasibility and some advantages, disadvantages, and tradeoffs that need to be considered when choosing among environmental control methods for preventing or treating contaminated coal refuse drainage. The major issues in this regard for many of the control methods considered in this report appear in Tables XXII and XXIII. For the most part, the comparison grid in the executive summary condenses the information discussed in the previous sections. Arranging the information in a grid illustrates the complexity involved in choosing from among the various control possibilities.

TABLE XXII

MATRIX GRID SUMMARY OF ENVIRONMENTAL CONTROL OPTIONS FOR CONTAMINATED COAL REFUSE DRAINAGE

Parameter	Calcining	Preleaching	Lime Codisposal	Fly Ash Codisposal	Soil Codisposal
Cost[a]	high	very high[b]	moderate	mod. to high	moderate[c]
Effectiveness[d]	excellent	good	good	good	good
Process complexity	high	high	low	low	low
Treatment duration[e]	short	short	short	short	short
By-product potential	high	high	none	none	none
Permanency	excellent	good[f]	?[f]	?[f]	?[f]
Likely RCRA Classification	nonhazard	?[f]	?[f]	?[f]	?[f]

[a] Ranges from a high of >$5 (1978)/ton of cleaned coal to a low of $0.20/ton of cleaned coal.
[b] Can be justified only by development of by-product recovery technology.
[c] Site specific.
[d] Ability to prevent or abate contaminated drainage.
[e] Short means days to months.
[f] Must be confirmed by further experiments.

The relative costs of the control methods under consideration vary from quite high (perhaps as much as $5.00 per ton of cleaned coal for refuse calcining) to fairly low ($0.20 per ton of cleaned coal for alkaline treatment of refuse drainage). In general, the costs tend to reflect the complexity of the control processes. It is noteworthy that the most costly types of controls are also potentially the most permanent and environmentally desirable solutions to the refuse disposal problem. As indicated in the tables, several of the refuse drainage control methods that we are studying have some potential for by-product recovery. This factor could significantly reduce the overall pollution control cost.

Our research suggests that each of the control techniques listed in Tables XXII and XXIII is quite effective over short periods of time. One of the major areas that remains to be defined for many of the methods under consideration is the long-term effectiveness or permanency of the proposed solutions. Answers to this question are being sought from scale-up experiments that more closely simulate actual waste dump conditions than the small scale laboratory experiments that we have been working with.

The last item of importance on the tables concerns the possible constraints imposed by RCRA on the handling and disposal of coal refuse materials. Wastes classified as hazardous by RCRA will involve a maze of paperwork and conformance to regulations that will be quite expensive to negotiate. This consideration alone may represent the singlemost important cost in refuse disposal. The RCRA posture with regard to large volume wastes is still being defined; consequently

TABLE XXIII

MATRIX GRID SUMMARY OF ENVIRONMENTAL CONTROL OPTIONS FOR CONTAMINATED COAL REFUSE DRAINAGE

Parameter	Alkaline Neutralization	Reverse Osmosis	Ion Exchange
Cost[a]	low	moderate	moderate
Effectiveness[b]	good	good	good
Process complexity	moderate	high	high
Treatment duration[c]	very long	very long	very long
By-product potential	none	some[d]	some[d]
Permanency	poor	poor	poor
Likely RCRA Classification	hazardous	hazardous	hazardous

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[a] Ranges from a high of >$5 (1978)/ton of cleaned coal to a low of $0.20/ton of cleaned coal.
[b] Ability to prevent or abate contaminated drainage.
[c] Very long means indefinitely.
[d] By-product is potable water.

we cannot yet identify the probable RCRA classification for many of the waste treatment schemes that we are studying.

The nature of the tradeoffs to be made among the various control options for disposal of acidic coal refuse materials is beginning to emerge. The methods that potentially provide the most effective and permanent means of abating trace element contamination of refuse drainage (calcining and preleaching) are also the most costly and complex methods to use. The control techniques that are designed to retain contaminants within the refuse disposal site, such as codisposal with various agents, are effective for attenuating the trace element compositions of refuse drainages for at least short durations, but some of these may lack long-term effectiveness. Acceptability for nonhazardous RCRA disposal requirements is another questionable aspect. Finally, the methods to treat refuse drainage (alkaline neutralization and reverse osmosis) appear to be quite attractive because of their relatively low costs and effective trace element reduction, but these are methods fraught with other potential problems. These include indefinite treatment duration, possible contaminant escape, and cost to meet RCRA permit and performance requirements for hazardous wastes.

Future work in this program will provide further elucidation of the technical feasibilities and cost/benefit tradeoffs of these and other environmental control options for contaminated coal refuse drainage.

TASK 2—IDENTIFY TRACE ELEMENTS OF ENVIRONMENTAL CONCERN IN HIGH-SULFUR COAL PREPARATION WASTES FROM THE APPALACHIAN REGION

Subtask 2.1—Assess Trace Element Structure and Mineralogy in Representative Refuse Samples

The emphasis of this subtask is to determine sufficient detail about the structure and mineralogy of selected samples of Appalachian Region refuse (and coal) to establish an understanding of the trace elements of greatest environmental concern and to aid in the selection of appropriate environmental control for trace element contamination of refuse dump effluents. Chronologically, there are several parts to this activity: sample selection and collection; trace element and mineralogical characterization of the bulk refuse samples; and detailed delineation of the mineralogy of specific trace elements of interest.

Sample Collection From Homer City Coal Cleaning Plant

We are pursuing our originally stated intent, that of trying to obtain some samples from the new multistream coal preparation plant at Homer City, Pennsylvania. We have received formal approval from the Pennsylvania Electric Company (PENNELEC) for a visit to their facility to collect these samples. As soon as all the necessary details can be attended to, we will proceed. Meanwhile, we have been characterizing the structure and behavior of a low-sulfur coal cleaning plant refuse from the Appalachian Region (Plant G).

Structural Studies of Appalachian Region Coal Refuse

We have completed our assessment of the bulk mineralogy and trace element compositions of several refuse fractions from Appalachian Plant G refuse. Average mineral compositions from x-ray diffraction analyses of three refuse fractions, two coarse and one fine, from this plant are compared with average values from similar analyses of Illinois Basin Plant B refuse in Table XXIV.

TABLE XXIV

MINERAL COMPOSITIONS OF COAL REFUSE SAMPLES

Mineral	Plant G[a] Average Wt %	Plant B[b] Average Wt %
Kaolinite	11	7
Illite	19	11
Quartz	22	17
Pyrite/Marcasite	<1	26
Calcite	1	0
Mixed Clay	6	17
Gypsum	1	1

[a] Low-sulfur refuse.
[b] High-sulfur refuse.

The mineralogy of the Plant G refuse is notably different from that of the Illinois Basin refuse materials that we have been studying. There is very little detectable pyrite or marcasite in the Plant G refuse (<1 wt%), and the clay minerals and quartz represent over 60 wt% of the refuse composition. Small amounts of calcite and gypsum compose the remainder of the detectable mineral matter in the refuse. Therefore, the acid-generating potential of the Plant G material should be very low. An unusually large fraction of the total mineral composition of the Plant G refuse (20 to 25 wt%) was either microcrystalline or amorphous and could not be analyzed by x-ray diffraction methods.

The trace element analyses for the Plant G refuse samples are now complete, and those data are tabulated in Table XXV. Using a portion of the available analytic data, we have compared the trace element make-up of the Plant G refuse with that from a high-sulfur (Plant B) Illinois Basin coal refuse. This is done in Table XXVI. Here it is seen that the most notable difference is in the iron content, with Plant G having 2% and Plant B having 11%. This, of course, is a reflection of the low, iron sulfide mineral content in this sample of Appalachian coal waste. Except for copper, the trace elements are also lower for the Eastern coal. The relatively higher aluminum and silicon values in the Plant G refuse reflect the higher clay and quartz concentrations. From an environmental viewpoint, the Plant G refuse contains potentially troublesome quantities (>50 μg/g of refuse) of aluminum iron, manganese, nickel, and zinc.

The trace element/mineral associations of the Plant G refuse will be reported next year.

Subtask 2.2—Determine Environmental Behavior of the Trace Elements in Refuse Samples

The activities in this subtask are an extension of the environmental weathering and leaching studies, which we conducted previously on Illinois Basin refuse, to selected samples of refuse from the Appalachian Region. The purpose of the research in this subtask is (1) to develop an understanding of the environmental behavior of the trace elements in selected Appalachian Region refuse (and coals) under typical waste dump or storage conditions and (2) to identify the trace elements of greatest environmental concern in these materials. This work, as well as our previous work on the leachability of Illinois Basin refuse, is directed toward defining the technology needs for controlling or preventing trace element contamination of the aqueous drainage from the thousands of refuse dumps, culm banks, and coal storage piles located in the Eastern and Midwestern United States.

Environmental Assessment of a Low-Sulfur Refuse From the Appalachian Region

Static and dynamic leaching tests have been conducted on the Plant G refuse material. These studies were designed to simulate the weathering and leaching behavior of the refuse materials and to yield data on those potentially troublesome trace elements that may be released into the environment. We have identified aluminum, iron, manganese, nickel, and zinc as residing in the Plant G refuse in quantities >50 μg/g of refuse and therefore likely to be released in concentrations high enough to be of environmental concern.

Static leaching tests were performed on 50-g portions (-20 mesh) of Plant G refuse derived from the two coarse fractions. These portions were leached with 200 mℓ of water in a system open to air and at room temperature for periods of up to 42 days. The detailed pH and trace element analyses of these samples appear in Table XXVII. Note that the pH remained fairly constant around 4 for the first 2 wk but at 42 days it had decreased to 3. This decrease probably occurred by a gradual depletion of the small amount of neutralizing capacity naturally present in the refuse material in the form of calcite.

TABLE XXV

TRACE ELEMENT AND MINERAL CONTENT OF COAL WASTE FROM APPALACHIAN PLANT G

SAMPLE	40	41	42
(1)			
IDENTITY	GOB A CORS	GOB B CORS	FN GOB
LOCALE	PLANT G	PLANT G	PLANT G
DATE OBTND	06/23/76	06/23/76	06/23/76
PCT H2O	4.54	4.60	20.14
PCT LTA	84.82	81.52	78.35
PCT ORIGNL	100.00	100.00	100.00
SIZE,KG	59.70	60.80	42.80
CHNS ANAL			
NITROGEN	.28	.42	.46
SULFUR	.60	.64	.88
MINERALOGY			
KAOLINITE	11.18	11.34	11.16
ILLITE	19.03	19.61	19.46
QUARTZ	23.87	19.76	21.31
PYRITE	-1.00	-1.00	-1.00
CALCITE	1.03	.49	1.92
MIXED CLAY	7.47	3.49	6.29
GYPSUM	1.52	1.58	.76

SAMPLE	40	41	42
ELEMENT	RAW BASIS	RAW BASIS	RAW BASIS
(2)			
LI PPM H A	119.00	132.00	114.00
BE PPM H A	3.00	2.60	1.80
B PPM L E	56.00	56.00	52.00
F PPM R O	600.00	560.00	550.00
NA PCT H A	.17	.11	.12
MG PCT H A	.52	.57	.52
AL PCT H A	9.56	9.25	8.88
SI PCT R O	20.20	20.45	20.10
P PPM R O	160.00	150.00	150.00
CL PPM R N			
K PCT H A	2.07	2.05	1.99
CA PCT H A	.12	.15	.14
SC PPM R N	15.80	17.20	14.60
TI PCT R N	.73	.67	.56
V PPM R N	116.00	116.00	109.00
CR PPM H A	87.00	104.00	88.00
MN PPM H A	93.90	96.75	99.50
FE PCT H A	1.89	2.05	2.10
CO PPM R N	9.00	11.00	14.00
NI PPM L E	55.00	46.00	45.00
CU PPM H A	43.00	53.00	47.00
ZN PPM H A	72.00	69.00	65.00
GA PPM R N	22.20	19.30	17.00
GE PPM L E	-8.00	-8.00	-8.00
AS PPM R N	14.20	20.30	18.60
RB PPM R N	121.00	134.00	141.00
Y PPM L E	21.00	19.00	18.00
ZR PPM L E	160.00	130.00	99.00
MO PPM L E	-8.00	-8.00	-8.00
CD PPM H A	.20	.40	.40
SN PPM L E	-8.00	-8.00	-8.00
SB PPM R N		2.95	1.52
CS PPM R N	9.22	9.58	7.52
LA PPM R N	58.30	52.40	38.00
CE PPM R N	74.50	85.80	76.50
SM PPM R N	6.09	5.50	4.34
EU PPM R N	1.13	1.50	1.14
TB PPM R N	.80	1.56	.94
DY PPM R N	5.82	5.68	5.18
YB PPM R N	2.83	2.46	3.00
LU PPM R N	.59	.56	.13
HF PPM R N	5.73	4.82	4.76
TA PPM R N	1.46	1.14	.78
W PPM R N			1.33
PB PPM H A	22.00	20.00	27.00
TH PPM R O	15.60	15.80	13.90
U PPM R O	4.32	4.40	3.84

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[1] PLUS OR MINUS INDICATES VALUE GREATER OR LESS THAN THAT GIVEN. NUMBERS 6 OR LARGER ARE MESH SIZES. OTHERS ARE IN INCHES.

[2] LETTERS INDICATE HOW SAMPLE WAS PREPARED AND ANALYZED
R=RAW SAMPLE
L=LOW TEMPERATURE ASH
H=HIGH TEMPERATURE ASH
N=NEUTRON ACTIVATION ANALYSES
A=ATOMIC ABSORPTION
E=EMISSION SPECTROSCOPY
O=OTHER

TABLE XXVI

TRACE ELEMENT COMPOSITIONS OF COAL REFUSE SAMPLES

Element[a]	Average Plant G[b]	Average Plant B[c]
Al (%)	9.2	5.1
Si (%)	20.2	13.6
Mn	97	144
Fe (%)	2.0	11
Co	12	30
Ni	49	71
Cu	48	35.4
Zn	69	149
As	17.7	94

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[a] Compositions reported as µg/g unless otherwise noted.
[b] Low-sulfur refuse.
[c] High-sulfur refuse.

TABLE XXVII

STATIC LEACHING OF LOW-SULFUR APPALACHIAN PLANT G WASTE

Sample No.[a]	1	2	3	4	5
Time (Days)	0.01	1	4	16	42
pH	3.9	4.3	4.3	4.1	3.0[b]
TDS (%)	0.10	0.13	0.09	0.10	0.23[b]
F	1.4	2.0	2.3	2.6	3.1
Na	18	20	29	25	29
Mg	240	250	270	260	320
Al	29	25	28	40	280
K	90	130	135	170	165
Ca	580	810	850	840	960
Cr(µg/mg)	49	7	9	7	300
Mn	6	7	8	8	12
Fe	15	16	16	11	31
Co	1.5	1.5	2	2	3
Ni	3	4	4	5	6
Cu	3	1	1	2	6
Zn	4	5	6	7	15
Cd(µg/mg)	30	31	27	46	25

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[a] Values in µg/g of waste unless noted otherwise.
[b] See text.

Column leaching studies were also done on Plant G refuse, in which 500 g of material (-3/8 in.) was packed into glass columns 5 cm in diameter by 40 cm long. Distilled water at a flow rate of 0.5 mℓ/min was passed upward through the columns. For two samples (GL-23 and GL-24), the flow of water was stopped after approximately 3 liters had passed through, and the columns were allowed to dry out. Intermittently, these aired columns were moistened during a 2-wk period to simulate the wet and dry periods encountered by a refuse pile. At the end of the 2-wk period, water flow was resumed as before until a total leach volume of 10 ℓ had passed through the column. The behavior of the material as reflected by the pH and the TDS values as a function of water volume passed through the column is shown in Fig. 4. At no time did the pH reach the low levels of the high-sulfur Illinois Basin refuse, but there was much more acid-generating capability in the Plant G refuse than might have been anticipated from the low pyrite/marcasite content of the material. It is possible that pyrite in an amorphous, subcrystalline form not detectable by x-ray diffraction analysis is an active generator of acid. Perhaps a clearer understanding of the acid-generating capacity of this refuse will come about as we continue our studies of trace element/mineral associations.

In an effort to assess which elements present in the Appalachian Plant G refuse are of environmental concern, an analysis of the data from the leaching experiments was made according to the procedure described in Ref. 5. Using the (MATE) criteria established by the EPA and including a dilution factor (100X)indicative of the natural dilution of process effluents by surface or ground waters, we determined an adjusted MATE value. That value for each element was divided into the leachate concentration of that element to ascertain the relative environmental hazard of that refuse constituent. For this purpose elemental concentrations were chosen when 100 mℓ of water had passed through 500 g of waste. When a hazard factor is near or greater than 1, the potential of an element to cause an environmental problem is signaled. Table XXVIII indicates that the elements aluminum, copper, iron, manganese, nickel, and zinc are of environmental concern in the Plant G coal refuse.

Further evaluation of the environmental behavior of Plant G refuse will be included in future reports on this project.

PERSONNEL

A large number of LASL personnel besides the authors participated in the programmatic effort during the year. Their work and contributions are gratefully acknowledged.

Administrative Advisors: R. D. Baker, R. J. Bard, and R. C. Feber
Analytic Advisors: G. R. Waterbury and M. E. Bunker
Neutron-Activation Analyses: W. K. Hensley and M. E. Bunker
Atomic Absorption Spectrophotometry and Wet Chemisty: E. J. Cokal, L. E. Thorn, and W. H. Ashley
Spectrochemical Analysis: O. R. Simi, J. V. Pena, and D. W. Steinhaus
Electron and Ion Microprobe: W. F. Zelezny, N. E. Elliot, W. B. Hutchinson, W. O. Wallace, R. Raymond, and R. C. Gooley
X-Ray Diffraction Analyses: R. B. Roof
Optical and SEM Microscopy: R. D. Reiswig and L. S. Levinson
Statistical Evaluation: R. J. Beckman

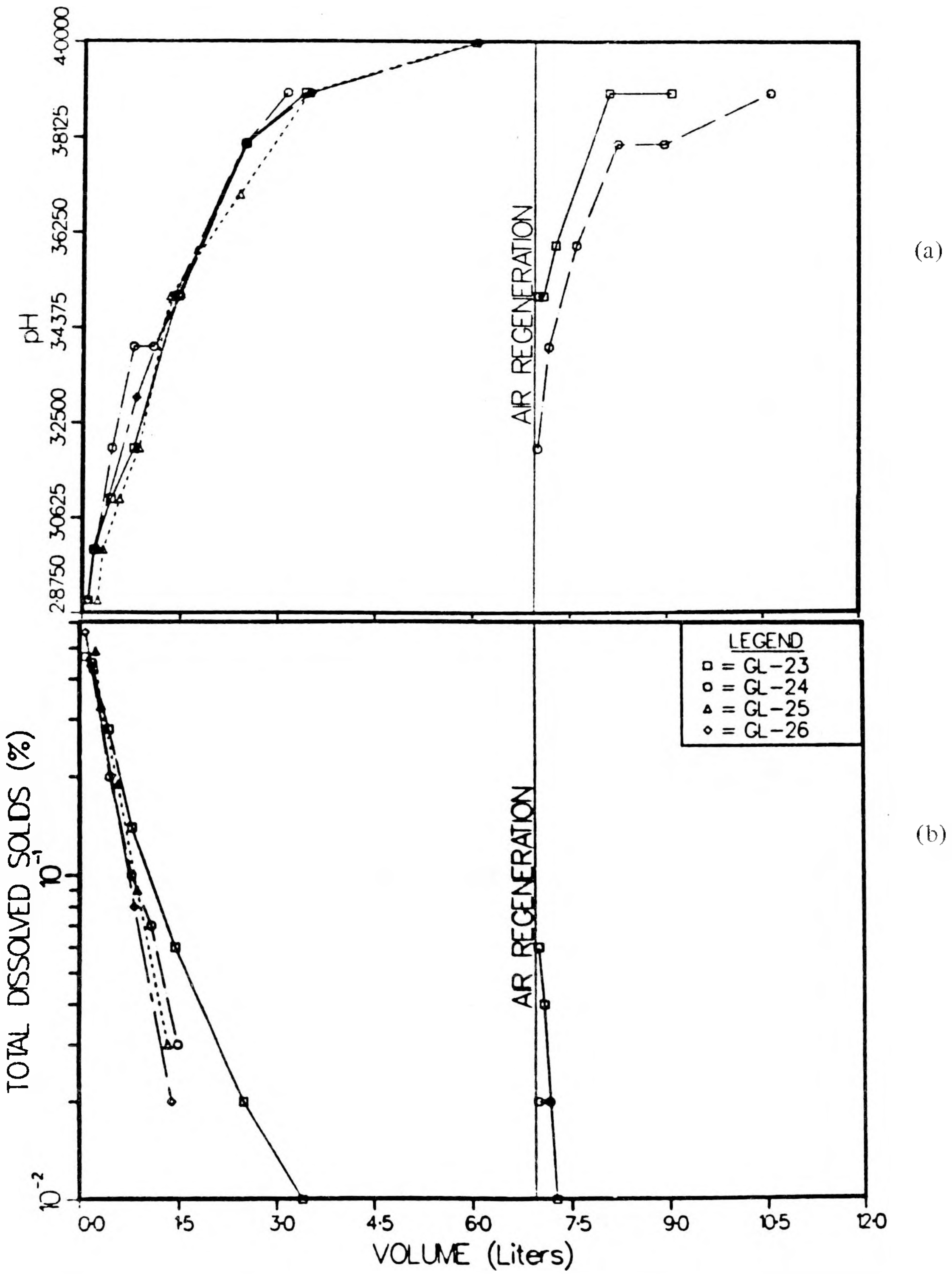

Fig. 4.

Leachate pH and TDS versus volume for column leaching study of Plant G refuse.

TABLE XXVIII

MEG/MATE ANALYSES OF PLANT G COAL REFUSE LEACHATE

	PLANT G		
ELEMENT	LEACHATE[a] LEVEL, PPM	ADJUSTED MATE[b] VALUE, PPM	HAZARD FACTOR[c]
Ni	3.8	1	4
Fe	50	25	2
Mn	10	10	1
Cu	4.8	5	1
Al	90	100	0.9
Zn	7.2	10	0.7
Cd	0.033	0.1	0.3
Ca	320	1 600	0.2
Co	1.9	25	0.07
Mg	250	8 700	0.02
K	26	2 300	0.01
Cr	0.12	25	0.004
F	1.4	380	0.003
Na	16	80 000	0.0002

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[a] Column leach; 100-mℓ aliquot; 500 g of refuse.
[b] 100x MATE value for liquids.
[c] Leachate value/adjusted MATE value.

APPENDIX A

COLUMN LEACHING STUDIES OF CALCINED REFUSE

TABLE A-I

EXPERIMENT IDENTIFICATION FOR DYNAMIC LEACHING STUDIES OF CALCINED REFUSE

Experiment No.	Sample[a]	Sample Size (kg)
GL-12	Control	1.5
GL-18	Calcined refuse	1.5

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[a] Minus 3/8 in. Plant B refuse used.

TABLE A-II

ANALYSES FOR DYNAMIC LEACHING STUDIES OF CALCINED REFUSE

Experiment No. GL-12

Parameter[a]	Sample No.								
	1	3	4	7	8	10	18	20	23
Vol (ℓ)	0.040	0.580	1.290	2.365	3.345	4.175[b]	7.735	8.290	9.855
pH	1.3	1.6	2.1	2.2	2.5	2.6	1.7	1.8	2.2
TDS(%)	8.63	3.13	1.02	0.55	0.08	0.11	3.87	1.87	0.33
Na	25	8	4	3	1	1	3	1	0.9
Mg	500	170	43	29	7	28	56	30	4
Al	1600	520	130	61	7	4	450	230	22
K	51	11	13	6	4	3	4	5	4
Ca	530	440	260	210	52	47	230	140	35
Sc	3.0	0.78	0.14	0.03	<0.01	<0.01	0.77	0.31	0.01
Ti	<0.4	<0.4	<0.4	<0.4	0.16	<0.4	<0.4	<0.4	<0.4
V	1.44	0.71	0.42	0.21	0.04	0.04	0.38	0.16	0.07
Cr(μg/ℓ)	790	420	85	38	<4	<5	260	160	13
Mn	130	19	8	3	0.7	0.5	7	4	0.9
Fe	15000	5300	1700	930	200	170	7600	3300	650
Co	36	13	4	3	0.4	0.2	4	2	0.4
Ni	51	19	8	3	0.7	0.5	7	5	0.9
Cu	10	1.5	<0.9	<0.06	<0.07	<0.09	4	1	<0.09
Zn	76	38	13	6	1	1	11	6	1
Ga	<0.2	<0.2	<0.2	<0.2	<0.2	<.2	<0.2	<0.2	<0.2
As	7.9	1.3	0.58	0.65	0.08	0.07	2.8	0.56	0.1
Br	<0.04		<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Rb	<2	<2	<2	<2	<2	<2	<2	<2	<2
Mo	<5	<4	<9	<1	<1	<1	<1	<1	<1
Ag	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cd(μg/ℓ)	430	130	85	35	4	19	41	30	22
Cs	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
La	1.3	0.60	0.26	<0.5	<0.5	<0.5	0.2	0.2	<0.5
Ce	5.3	1.4	0.53	0.44	0.09	<0.08	1.1	1.0	0.2
Sm	0.83	0.34	0.12	0.06	<0.02	<0.02	0.19	0.14	<0.02
Eu	0.35	0.11	0.03	0.02	<0.01	<0.01	0.08	0.04	<0.01
Tb	0.47	0.02	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Dy	0.14	0.08	0.03	0.43	0.04	0.02	0.12	0.03	0.01
Yb	0.25	0.11	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03
Lu	0.05	0.02	<0.01	<0.01	<0.01	<0.01	0.02	0.01	<0.01
Hf	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Ta	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
W	0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Pb(μg/ℓ)	1100	210	60	45	12	51	110	30	47
Th	1.7	0.34	0.04	<0.02	<0.02	<0.02	0.43	0.15	<0.02
U	0.46	0.31	0.19	0.01	<0.01	<0.01	0.14	0.06	0.01

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[a] Values in μg/mℓ unless otherwise stated.

[b] Water flow was stopped at this point, air was passed through the column for 4 wk, then water flow was resumed.

TABLE A-III

ANALYSES FOR DYNAMIC LEACHING STUDIES OF CALCINED REFUSE

Experiment No. GL-18

Parameter[a]	Level in Material[b] (ppm)	Sample No.					
		1	2	3	6	19	26
Vol(ℓ)		0.060	0.155	0.765	2.795	13.740	18.630
pH		3.9	3.8	4.1	5.7	4.9	5.7
TDS(%)		1.91	1.48	0.57	0.03	0.06	0.01
Na	1140.	350.	260.	67.	2.	7.	1.
Mg	4900.	940.	710.	1780.	4.	8.	0.5
Al	115000.	170.	85.	3.	<0.1	<0.8	<0.1
SiO_2	168000.						
P	560.						
K	24500.	2300.	1600.	710.	13.	33.	2.
Ca	3200.	590.	610.	690.	45.	58.	3.
Sc	29.3	0.08	0.03	<0.01	<0.01	<0.01	<0.01
Ti	5490.	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4
V	118.	0.17	0.09	<0.01	<0.01	<0.01	<0.01
Cr($\mu g/\ell$)	100000.	55.	14.	2.	<1.	<8.	<3.
Mn	191.	75.	61.	16.	0.3	2.	0.1
Fe	190000.	680.	610.	180.	3.	25.	2.
Co	69.9	7.	5.	0.5	<0.02	0.8	<0.05
Ni		7.	5.	0.6	<0.05	1.7	<0.1
Cu	73.	<0.08	<0.08	<0.02	<0.02	<0.17	<0.05
Zn	296.	10.	4.	0.2	<0.01	3.	0.1
Ga	28.6	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
As	108.	0.13	0.08	<0.02	<0.02	<0.02	0.03
Br	<0.1	0.1	<0.04	<0.04	<0.04	<0.04	<0.04
Rb	371.	3.57	2.11	<2.	<2.	<2.	<2.
Mo		<1.	<1.	<0.2	<0.2	<1.	<1.
Ag	<0.1	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cd($\mu g/\ell$)	290.	7.	0.5	0.9	0.04	3.	<0.05
Cs	15.8	0.09	0.04	<0.04	<0.04	<0.04	<0.04
La	98.6	1.92	0.87	0.14	<0.5	<0.5	<0.5
Ce	229.	2.35	1.57	0.39	<0.08	0.85	<0.08
Sm	29.6	0.66	0.30	<0.02	<0.02	<0.02	<0.02
Eu	3.21	0.19	0.14	0.02	<0.01	<0.01	<0.01
Tb		<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Dy		0.40	0.31	0.05	<0.01	<0.01	<0.01
Yb	8.68	0.13	0.06	<0.03	<0.03	<0.03	<0.03
Lu	1.11	0.02	0.02	<0.01	<0.01	<0.01	<0.01
Hf	8.48	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Ta	1.66	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
W	8.15	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Pb($\mu g/\ell$)	12000.	10.	14.	<5.	<5.	<42.	<13.
Th	25.4	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
U	9.55	0.64	0.27	0.01	<0.01	0.01	<0.01

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[a] Values in $\mu g/m\ell$ unless otherwise stated.

[b] Water flow was stopped at 4.0ℓ, air was passed through the column for 4 wk, then water flow was resumed.

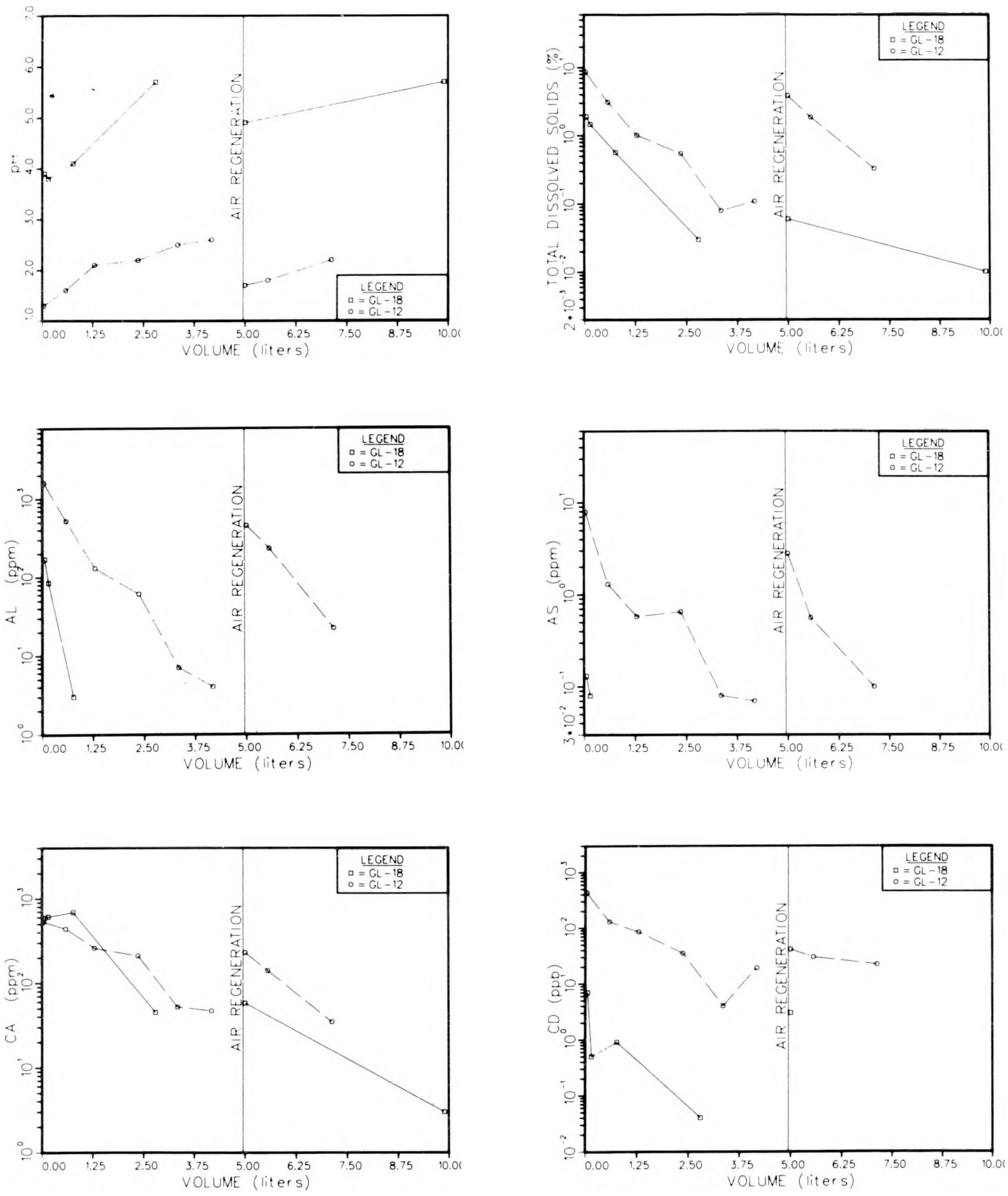


Fig. A-1.

The pH, TDS, and trace element concentrations for dynamic leaching experiments with calcined refuse.

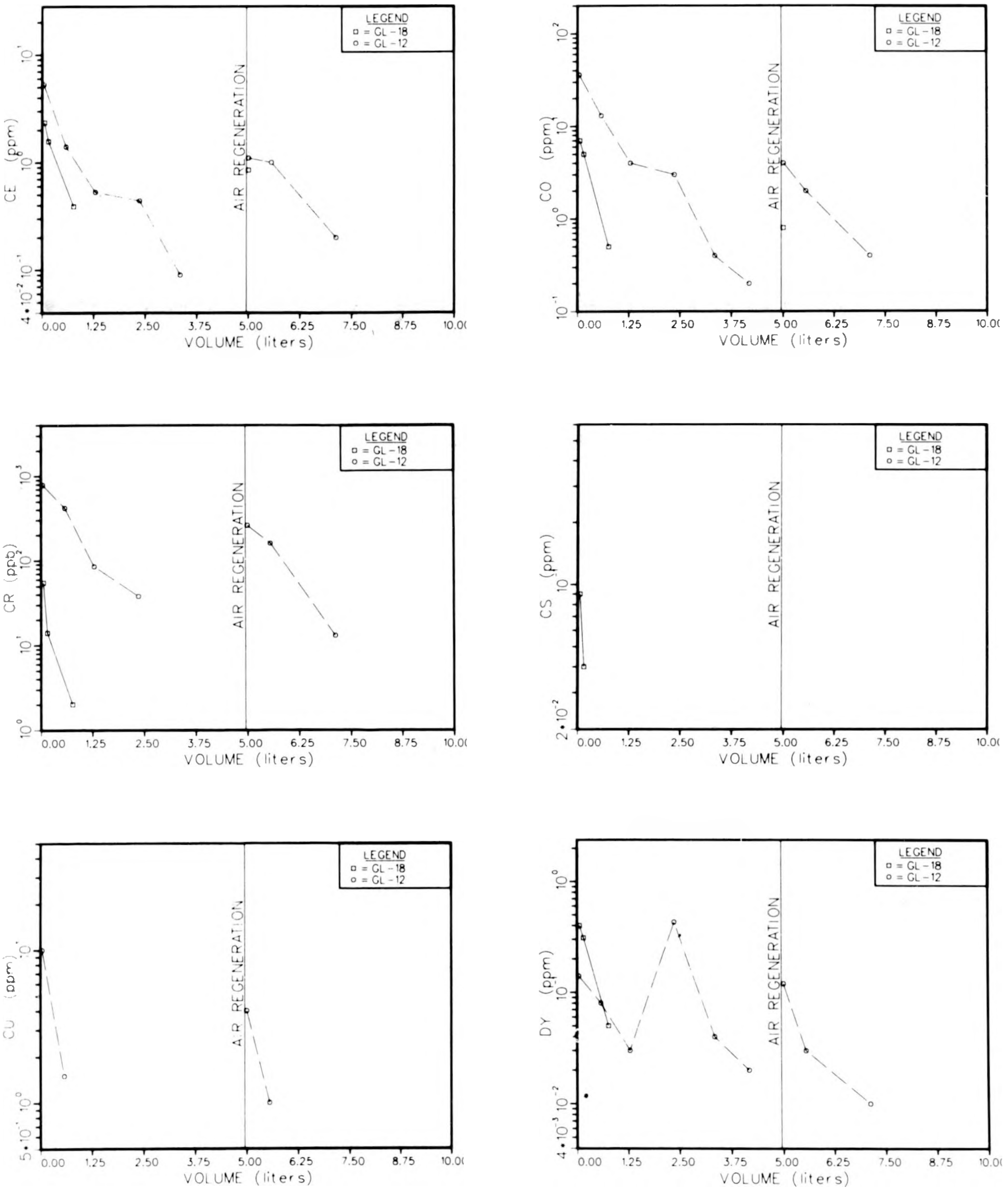
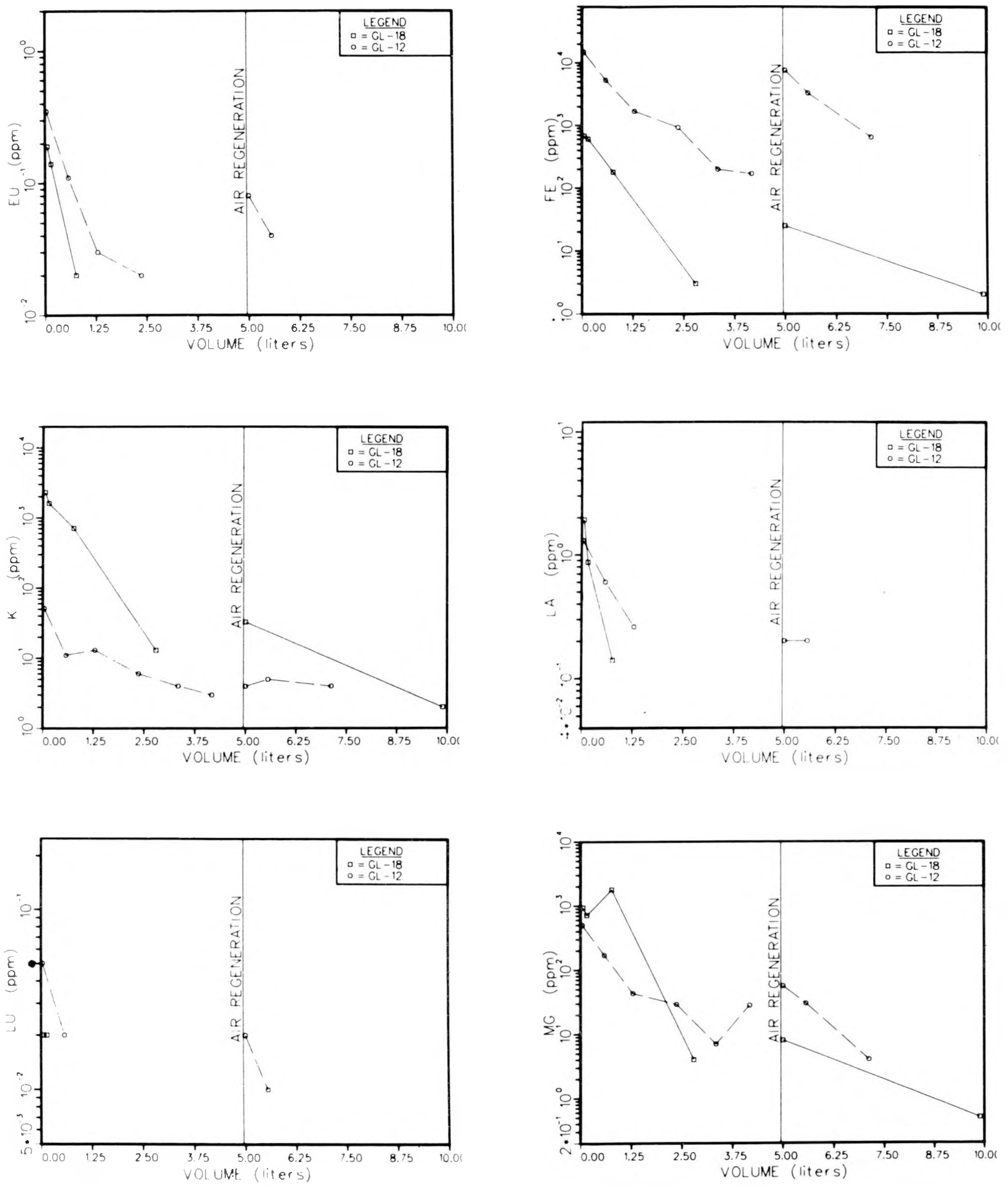

LEGEND
□ = GL-18
○ = GL-12
AIR REGENERATION
CE (ppm)
CO (ppm)
CR (ppb)
CS (ppm)
CU (ppm)
DY (ppm)
VOLUME (liters)
0.00
1.25
2.50
3.75
5.00
6.25
7.50
8.75
10.00
$4 \cdot 10^{-2}$
10^{-1}
10^{0}
10^{1}
10^{2}
10^{3}
$2 \cdot 10^{-2}$
$5 \cdot 10^{-1}$
$4 \cdot 10^{-3}$
10^{-2}

Fig. A-1. (cont)



EU (ppm)
FE (ppm)
K (ppm)
LA (ppm)
LU (ppm)
MG (ppm)
VOLUME (liters)
0.00
1.25
2.50
3.75
5.00
6.25
7.50
8.75
10.00
AIR REGENERATION
LEGEND
□ = GL-18
○ = GL-12

Fig. A-1. (cont)

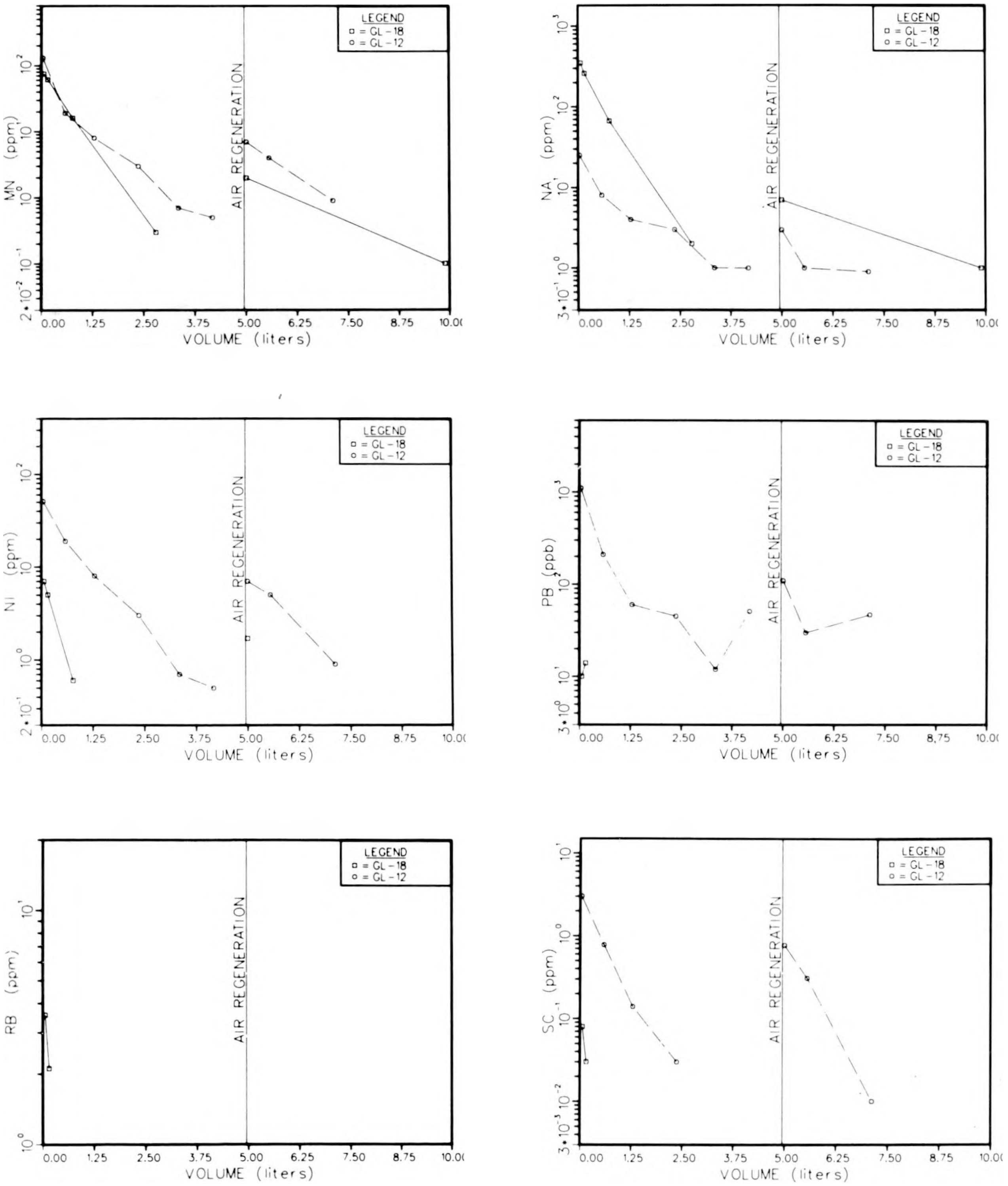

MN (ppm)
NA (ppm)
NI (ppm)
PB (ppb)
RB (ppm)
SC (ppm)
AIR REGENERATION
LEGEND
□ = GL-18
○ = GL-12
0.00
1.25
2.50
3.75
5.00
6.25
7.50
8.75
10.00
VOLUME (liters)
2•10⁻²
10⁻¹
10⁰
10¹
10²
10³
3•10⁻¹
2•10⁻¹
3•10⁰
3•10⁻³
10⁻²

Fig. A-1. (cont)

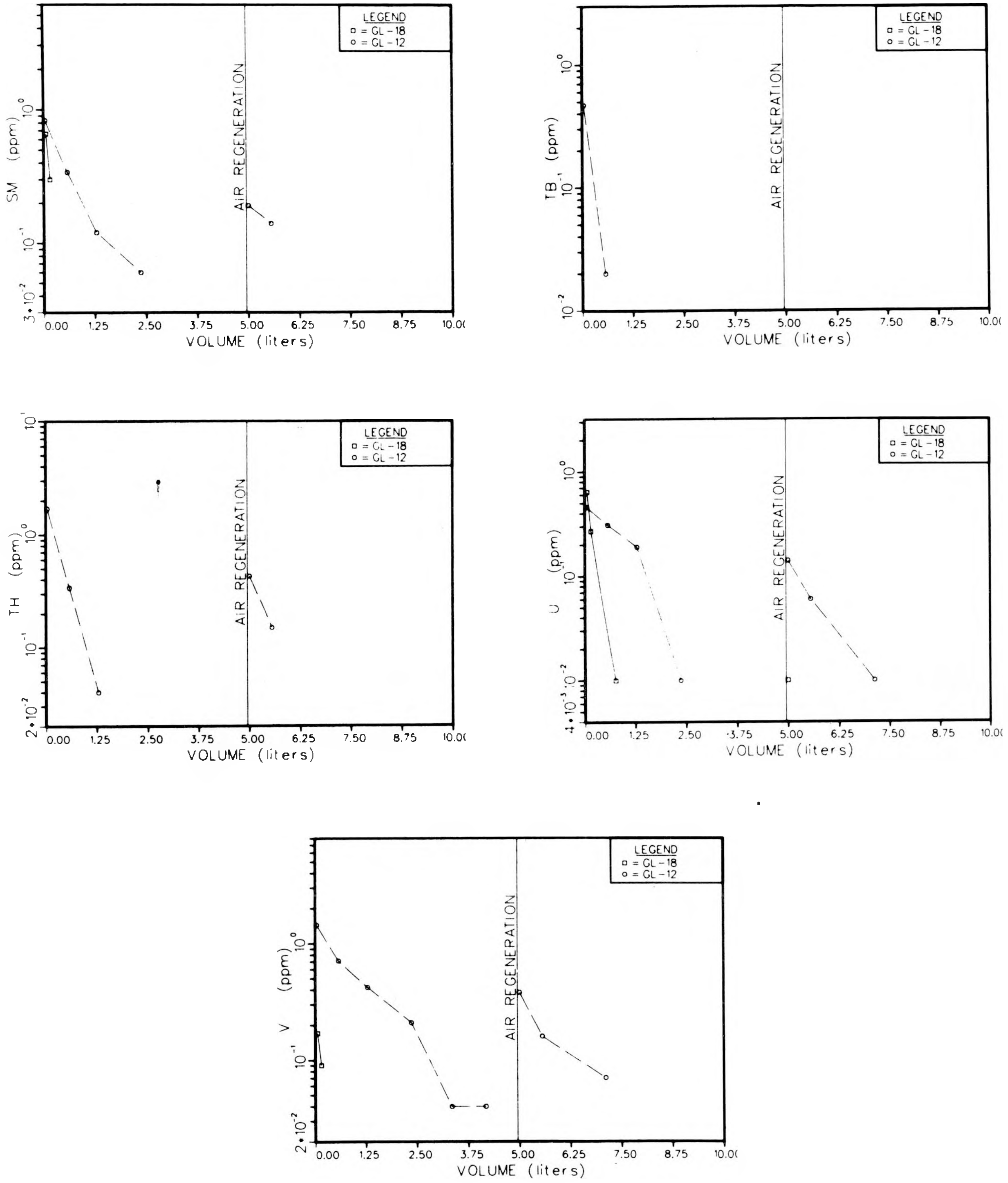

SM (ppm)
10^0
10^-1
3•10^-2
TB (ppm)
10^0
10^-1
10^-2
TH (ppm)
10^1
10^0
10^-1
2•10^-2
U (ppm)
10^0
10^-1
10^-2
4•10^-3
V (ppm)
10^0
10^-1
2•10^-2
AIR REGENERATION
LEGEND
□ = GL-18
○ = GL-12
0.00
1.25
2.50
3.75
5.00
6.25
7.50
8.75
10.00
VOLUME (liters)

Fig. A-1. (cont)

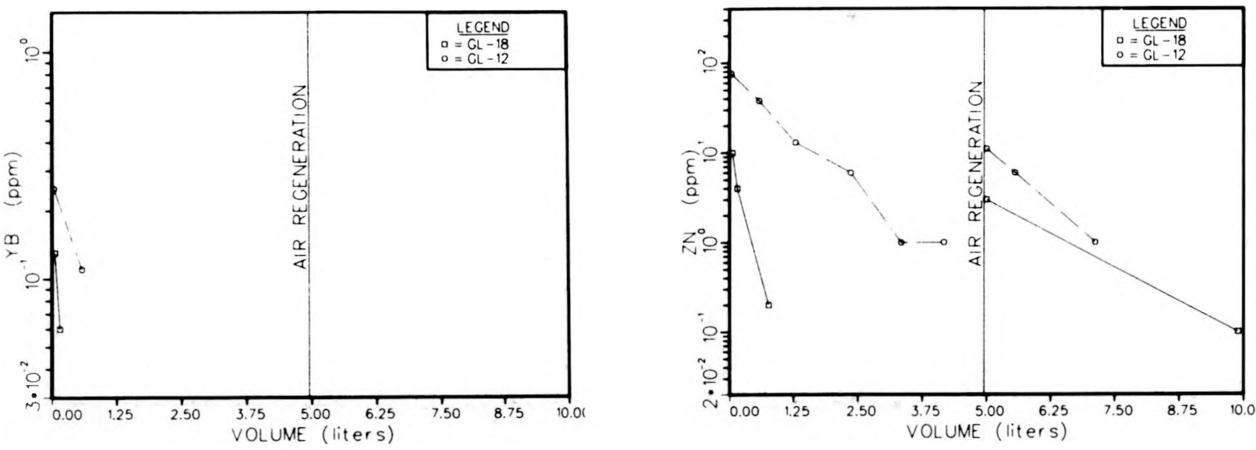

LEGEND
□ = GL-18
○ = GL-12
AIR REGENERATION
YB (ppm)
10^0
10^{-1}
$3 \cdot 10^{-2}$
ZN (ppm)
10^2
10^1
10^0
10^{-1}
$2 \cdot 10^{-2}$
0.00
1.25
2.50
3.75
5.00
6.25
7.50
8.75
10.00
VOLUME (liters)

Fig. A-1. (cont)

APPENDIX B

PRELIMINARY COST COMPARISONS FOR SELECTED ENVIRONMENTAL CONTROL OPTIONS FOR CONTAMINATED COAL REFUSE DRAINAGE

INTRODUCTION

Our assessment of environmental control technology for Illinois Basin coal cleaning wastes has proceeded to the point where a preliminary cost comparison of various pollution-abatement alternatives is in order. Cost data on many of the options are scattered throughout the literature, but they lack a consistent time base (constant-dollar figures) and the vital relationship between solid waste composition and pollutant concentration in the leachate. This latter information is available in our FY 1977 Annual Report[3] and provides the link for an across-the-board comparison.

BASE CASE

To provide a consistent basis for comparison, which could be related to existing data, three hypothetical coal cleaning plants were postulated. For purposes of comparison, all were assumed to have the same production capacity, ratio of waste to cleaned coal, landfill disposal area, and annual rainfall. Also, the active life of the waste disposal site was considered to be the same. To take advantage of published data on costs of landfill disposal, values for these parameters, other than rainfall, were selected from the National Academy of Science/National Academy of Engineers (NAS/NAE) Mine Waste Disposal Report (Ref. 6, pp. 78-79). They were

- production—2 070 00 tons/yr cleaned coal,
- solid waste sent to disposal pile—621 000 tons/yr,
- disposal area—250 acres, and
- active life of disposal area—20 yr.

An average annual rainfall of 35 in./yr was arbitrarily selected. The time base was selected as March 31, 1978, the latest date for which engineering cost indexes were available.[7]

GENERAL GUIDELINES

Costs for complete process operations must be based on a knowledge of the size and design of equipment, labor requirements, electric power needs, cost of consumable materials, and material-handling requirements. The amount of information accessible for each of the seven environmental control processes considered here varied a great deal in relevance and detail. In some instances, a so-called conceptual engineering design was required. In others, proportioning of capacities, updating of the costs, or both was all that was required or feasible. These variations are explained under the heading for each process.

These guidelines were followed consistently.

(1) A capital recovery factor of 0.2588 was used to convert capital costs to an annualized basis. This is consistent with the NAS/NAE report (Ref. 6, p. 139) and takes into account a 10% depletion allowance and normal straight-line, 10-yr depreciation, but no investment tax credit. If anything, it overstates the annualized capital charges.

(2) The above annualized capital charges were lumped together with all other annual charges to calculate $/ton of coal and for discounting in calculating present value data.

(3) All discounted cash flow computations, sinking-fund computations, etc. were performed using standard tables for 10% discrete compound interest and the appropriate time span (usually 20 years).[8]

(4) A delivered cost of lime of $45.00/ton was used uniformly in all calculations. The price of lime is a widely varying quantity, presently ranging from $32 to $42/ton in bulk quantities, FOB.[9] Transportation charges vary also, but the $45/ton price is an average for hauling and unloading over a distance of some 50 to 60 miles.

(5) For all pretreatment and pile treatment options a net weight fraction (nwf) of FeS_2 was calculated from the following relationship.[6]

$$[(\text{Pyrite wf} + \text{Marcasite wf})/119.9 - 5\times(\text{Calcite wf}/100)]\times 119.9 = \text{nwf } FeS_2. \qquad \text{(B-1)}$$

The waste from hypothetical Plants A, B, and C, corresponding to Illinois Basin Plants A, B, and C, had the following net weight fractions of FeS_2: 0.184, 0.260, and 0.294, respectively. Lime requirements were calculated by multiplying the above figures by 0.937; this yielded tons of lime required for complete neutralization of the acid generated by these sulfides in one ton of waste.

DATA SOURCES AND COMPUTATIONAL PROCEDURES

Alkaline Neutralization of Coal Refuse Drainage and Clarification of Effluent

Lime neutralization cost data for acid drainages appear in various prior reports, one of which is the Brown's Creek, Lost Creek Pollution Abatement Study (Ref. 10, p. 83). However, the information is insufficient to permit wide variation in the parameters of effluent flow and iron concentration in determining plant capital and operating costs. Therefore a plant design based on a reactor-clarifer (Ref. 11, pp. 19-51), which returns the slurry precipitate to the active pile, was evaluated. Cost was estimated from the clarifier settling area and 1955 standard cost data (Ref. 12, p. 69) that was updated according to standard proportioning procedures.*

Because it was thought desirable to evaluate a wide variety of input data and system parameters, a computer program was prepared to determine the size and cost of a neutralizer-clarifier for a variety of leaching conditions from piles of coal cleaning wastes. All costs above the base landfill disposal case are calculated, with the exception of costs for retaining, impounding, and channeling the pile effluent from rainwater percolation. These must be considered in any posttreatment process but are very site specific so that no realistic "average figure" seems credible. The following paragraphs serve as documentation for the computer program called LANDFIL (listed in Appendix E), as well as detailing design and cost calculation methods and assumptions for the lime neutralization posttreatment.

Input data to the computer program are (1) parts per million of Fe^{+3} and Fe^{+2} in the effluent; (2) annual rainfall in the area of the pile, in inches; (3) area of the active pile, in acres; and (4) fraction of rainfall absorbed by the pile. This last figure was assumed to be 0.9999 for an uncovered pile and 0.3333 for a soil-covered, grassed-over pile.

Lime requirement was calculated from stoichiometry assuming 1.5 moles of acid generated per mole of iron in the effluent. This is a reasonable average figure according to the literature (Ref. 14, p. 5). However, the calculation was made more conservative (that is, inclined in the direction

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*The standard proportioning procedure was to multiply the standard cost by the ratio (March 1978 value):(year of computation value) of the appropriate category in the Chemical Engineering Plant Cost Index or the Marshall & Swift Equipment Cost Index.[7,13]

of higher lime use) by using $Ca(OH)_2$ for the weight of lime in the neutralization reaction. The resulting figure of 1.2406×10^{-4} pounds of lime per cubic foot of effluent per ppm of iron was used in the calculation of lime demand for each set of initial conditions.

Clarifier area was calculated from the Coe and Clevenger formula[15] using the largest area calculated from settling-rate data. The solids were assumed to settle to 12% of the original volume before being removed as underflow from the clarifier. The rationale and method of calculation are explained in detail in Ref. 15. Total dissolved solids were assumed to be eight times the iron concentration and to this was added the lime requirement for calculation of solids in clarifier underflow. The cost of the clarifier was determined by linearizing the two major portions of the standard cost curve (Ref. 12, p. 69). This approximation is reasonably good in the ranges 100-1000 sq ft and 1000-8000 sq ft of clarifier surface. For output in excess of 8000 sq ft the calculated cost is clearly too low, especially because more than one clarifier would be needed in such high ranges. However, in the worst case of active pile application studied, the requirement of 8000 sq ft was not reached until after seven years had passed so that discounting the future cost of additional clarifiers tends to offset the low-cost prediction.

Clarifier underflow is returned to the active pile by a pipeline assumed to be 1500 ft in length with an effective hydraulic head of 250 psi. The computer program sizes the pipe and pump depending on flow volume and calculates the cost of pipe, pump, and the pumping energy according to standard formulas using a maximum velocity of 1 ft/s* (see Ref. 11, pp. 6-45 and Ref. 16, pp. 92 and 177).

Operating costs consisted of labor costs plus power costs. Power was calculated for the clarifier and the slurry pump. Labor was estimated at $40.00/day for a part-time operator/maintenance man; this estimate may be too low.

Output of program LANDFIL provides information on the calculated lime requirement in tons/day, clarifier area in sq ft, annualized capital cost in dollars, and annual operating cost (exclusive of lime) in dollars. As previously mentioned, the output also included annual cost of lime. The program was run for each of the 20 yr for each of the three hypothetical plants, using laboratory data on ppm of iron versus the total water-to-waste ratio as input. For year one, a 12.5-acre uncovered pile is assumed. For years 2-20, covered piles increasing by 12.5 acres per year are calculated and first year lime costs are added because there is always 12.5 acres of open pile being worked. This provides a precise lime cost and slightly understates the annual operating cost. Average figures for each of the four 5-yr periods were calculated. Finally, a sinking-fund payment, applied to each year of operation, was calculated from the average cost for the last 5 yr (highest average annual costs). The sinking fund was calculated at a 10% rate of return to provide the necessary trust fund corpus at the end of the 20-yr active live. Annualized total cost and cost per ton of cleaned coal were obtained by summing annualized capital cost, annual operating cost, annual lime cost, and annual sinking-fund payments.

Ion Exchange and Reverse Osmosis Costs to Treat Refuse Drainage

Time did not permit careful cost analysis of more expensive reverse osmosis and ion-exchange posttreatment options. However, Source 62062 in the Brown's Creek and Lost Creek study (see Ref. 11, pp. 83-87) was found to be roughly comparable to Plant C in effluent composition, though differing in flow volume. For a preliminary comparison of costs, it was deemed sufficient to use the ratios of costs derived from the earlier study. The cost estimates for these two processes should be considered only as very crude approximations.

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*Through errors, the velocity and cost were calculated for different wall thicknesses of pipe. Velocity was calculated for thin-wall 5s-pipe, and cost was calculated for the thicker wall 40 st-pipe corresponding to the assumed pressure. In some instances this may result in a pumping energy cost that is 5 to 10% too low. This cost is only a small proportion of the total cost however.

Codisposal of Lime and Coal Refuse

Costs for directly adding 25 and 50% of stoichiometric amounts of lime needed if all the pyrite was converted to acid were calculated on delivered lime cost only. Because of its fine particle size, it was assumed that the lime would not add appreciably to the bulk of the pile but would fill the voids between the waste material. Only a negligible amount of energy and labor above that already devoted to pile construction would seem to be required. There is good theoretical reason, based on the comparatively small exposed surface of pyrites in coal waste piles, to believe that even less lime may serve to deactivate or neutralize the acid-leaching processes.

Codisposal of Fly Ash and Coal Refuse

Capp and Adams have reported successful attempts to reclaim the surface of spoil banks and coal waste piles for vegetative propagation by large additions of alkaline, power-plant fly ash.[17] The amount varied with conditions of the waste and overburden, but the authors stated that the fly ash they used had about one-twelfth the neutralizing capacity of limestone. This unmodified fly ash therefore had one-eighteenth the neutralizing capacity of lime. We also considered limestone-modified fly ash, which was assumed to have one-twelfth the neutralizing power of lime.

Two scenarios for fly ash use were considered. In the first, the power plant was located 15 road miles from the coal cleaning plant; in the other it was within 1500 ft of refuse landfill. In both scenarios, it was assumed that no market existed for the fly ash. It was assumed that (based on the base-case costs) the cost to the power plant for disposal to landfill was $1.50/ton of fly ash. Truck loading charges were set at $2.00/ton for the first scenario. To this was added a hauling charge of $1.80/ton, an unloading charge of $0.50/ton, and an additional operating cost of $0.50/ton; a total of $4.80/ton of fly ash added to the pile for the first scenario. For the second scenario, it was assumed that the power plant would deliver the fly ash with its own conveyer system to the landfill area, without charge. The only charge would be $0.50/ton of fly ash for additional operating expense at the landfill. This results from the very large additional volume of material that must be distributed and compacted. More machines, fuel, and labor are necessarily required for any of the fly-ash scenarios than for straight wastefill. Calculations of the cost were made only for one level of addition, namely, fly ash equivalent in neutralizing power to direct addition to the pile of 25% of the theoretical amount of lime needed.

Codisposal of Local Soils With Coal Refuse Materials

The model system used was a soil with alkalinity corresponding to 5% by weight of $CaCO_3$. It was assumed that the worst case of oxidation before disposal would be 10% of the FeS_2 content. Calculations were made for sufficient soil to negate 11% oxidation. For the three plants (A, B, and C) in the study the mass-of-soil per mass-of-waste ratios were 0.678, 0.957, and 1.082, respectively. In other words, burying coal cleaning waste from Plant A would require 0.678 tons of a 5% alkaline soil to neutralize any acid formed before disposal and to immobilize or attenuate any further reaction or release of metal ions from one ton of waste. The costs would be somewhat lower if the soil was more highly alkaline; however, a certain minimum amount would be required to attain the densification and compaction that is deemed essential to this process. Therefore, the Plant A costs given are probably about as low as one might expect for any plant, regardless of the soil alkalinity.

Mixing soil with waste during the disposal process would have the advantage of filling the voids between the larger refuse particles with small particles of soil. On a volume basis, the theoretical maximum ratio required for this purpose would be approximately 0.68 soil/waste. In practice, this theoretical maximum would never be reached, so very good densification should be possible with a 50 to 75 wt% addition. One could expect the permeability of the resulting mixture to be much lower than that of normally compacted waste alone. The combination of the neutralizing and immobilizing power of the soil, together with the lowered permeability, could be sufficient to prevent significant ground-water pollution.

Calcining to Immobilize Refuse Contaminants

Pretreatment by calcining to approximately 1000°C is an attractive method for immobilizing the labile contaminants in coal refuse materials. The landfill requirements and cost would not change much from those of uncalcined refuse materials because the calcined mass and scrubber slurry (sulfur dioxide removal from the calciner effluent) would occupy about the same disposal volume as the original coal cleaning waste. Another important point is that an amount of lime proportional to the FeS_2 present in the refuse would be required for sulfur dioxide removal from the stack gases. Optionally, half the lime in the scrubber may be replaced with limestone.

For calculational purposes, we assumed that the heat of combustion of the residual coal and the pyrite and marcasite constituents in the refuse would be sufficient to maintain operating temperature in the kiln once the temperature was reached using auxiliary fuel to heat the kiln. The assumption was based upon the high thermal efficiency of modern kilns but may not hold if the heat of fusion of the glassy materials formed is substantial.

Capital costs were approximated only roughly. Based on 20 days/yr of operation, the throughput of waste would require three rotary kilns of commercial maximum size for Plant A and four for Plants B and C. Baghouse and sulfur dioxide scrubbers account for the remainder of the major equipment items. Capital cost was estimated at $7 000 000 for Plant A, $8 500 000 for Plant B and $9 000 000 for Plant C. However, these costs could be 50% or more low without significantly affecting the ultimate cost figures because the lime for sulfur dioxide neutralization is two-thirds or more of the total cost. However, if a combined limestone/lime neutralization system is used, annual and unit costs may be significantly reduced.

Calcining costs depend on the proportion of sulfur immobilized in the residue and also upon calcining temperature. The predicted costs given are the minimum that might be expected. They could be 50 to 100% higher, depending upon the ultimate process chosen.

Water Flow Through Waste Landfill Pile

The magnitude of the annual volume of leach water from the landfill may be observed from the calculated data in Table B-I. To compare this with our laboratory column leachings, consider that 4 129 125 tons of water are predicted to flow through the pile by the 20th yr and 12 420 000 tons of water will have accumulated in the pile, which means that the cumulative flow in the 20th yr is equivalent to only 220 mℓ of leach water having passed through a laboratory column packed with 1500 g of coal cleaning waste. After the pile is complete, the cumulative flow increases at a rate of 17.7 mℓ/yr so that the two-liter mark, a point at which we find the leachate still loaded, corresponds to about 100 yr after the pile has ended its active life.

Lime/Waste Ratio Required for Various Control Processes

The lime/waste ratio for various processes is shown in Table B-II. Note that the posttreatment will require considerable additional lime beyond the 20-yr active life, but this amount has not been calculated. The amount of fly ash needed for codisposal neutralization of waste acidity is given in Table B-III.

CONTROL OPTION COSTS

Tables B-IV, B-V, and B-VI present total cost data for 8 control processes and 13 total variations, in different forms. Table B-IV compares each on a basis of unit cost in dollars per ton of cleaned coal shipped. Table B-V presents the same data in terms of annual costs over the 20-yr working life of the disposal area. Costs in Tables B-IV and B-V include charges for treatment required after the active life of the pile has expired. Table B-VI compares the options in terms of net present value of cost, calculated at 10% cost of capital. To the person unfamiliar with the terminology of finance, these figures may be considered as the total number of dollars that would need to be paid in a lump sum in 1978 to assure pollution abatement for the life of the project (and beyond, if necessary). Figures in all three of these tables do not include the basic cost of landfill, which is indicated on each table, or the cost of sealing the disposal site.

TABLE B-I

LANDFILL GROWTH AND MASS FLOW OF WATER THROUGH WASTE PILE DURING 20 YR ACTIVE LIFE OF PILE[a]

Year	Flow Through Pile (Tons of Water)		Annual Total Flow (tons)	Cumulative Data			
	Uncovered	Covered		Water Flow (tons)	Waste (tons)	Water/Waste	
						Mass Ratio	Liters/1.5kg
1	49 550	0 [b]	49 550	49 550	621 000	0.07979	0.0532
2	49 550	16 517	66 066	115 616	1 242 000	0.09309	0.0621
3	49 550	33 033	82 583	198 198	1 863 000	0.10639	0.0709
4	49 550	49 550	99 099	297 297	2 484 000	0.11968	0.0798
5	49 550	66 066	115 616	412 913	3 105 000	0.13298	0.0887
6	49 550	82 583	132 132	545 045	3 726 000	0.14628	0.0975
7	49 550	99 099	148 649	693 693	4 347 000	0.15958	0.1064
8	49 550	115 616	165 165	858 858	4 968 000	0.17288	0.1153
9	49 550	132 132	181 682	1 040 540	5 589 000	0.18618	0.1241
10	49 550	148 649	198 198	1 238 738	6 210 000	0.19947	0.1330
11	49 550	165 165	214 715	1 453 452	6 831 000	0.21277	0.1418
12	49 550	181 682	231 231	1 684 683	7 452 000	0.22607	0.1507
13	49 500	198 198	247 748	1 932 431	8 073 000	0.23937	0.1596
14	49 550	214 715	264 264	2 196 695	8 694 000	0.25267	0.1684
15	49 550	231 231	280 781	2 477 475	9 315 000	0.26597	0.1773
16	49 550	247 748	297 297	2 774 772	9 936 000	0.27926	0.1862
17	49 550	264 264	313 814	3 088 586	10 557 000	0.29256	0.1950
18	49 550	280 781	330 330	3 418 916	11 178 000	0.30586	0.2039
19	49 550	297 297	346 847	3 765 762	11 799 000	0.31916	0.2128
20	49 550	313 814	363 363	4 129 125	12 420 000	0.33246	0.2216

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[a] Assumes 35 in./yr annual rainfall and 250 acres ultimate landfill area

[b] No covered portion in first year.

TABLE B-II

LIME REQUIREMENTS FOR VARIOUS TREATMENTS OF COAL WASTE

	TON OF LIME/TON OF WASTE		
Process	**Plant A**	**Plant B**	**Plant C**
Calcining-Lime Neutralization	0.1722	0.2433	0.2571
Calcining-Lime and Limestone Option	0.0862	0.1218	0.1377
25% Lime to Pile	0.0431	0.0609	0.0689
Lime Neutralization of Drainage—1st yr.	0.0003	0.0019	0.0001
Lime Neutralization of Drainage—2nd yr.	0.0004	0.0025	0.0001
Lime Neutralization of Drainage—10th yr.	0.0010	0.0090	0.0004
Lime Neutralization of Drainage—20th yr.	0.0015	0.0229	0.0008

TABLE B-III

FLY ASH DEMAND REQUIREMENTS FOR CODISPOSAL TREATMENT OF COAL WASTE[a]

	TON OF FLY ASH/TON OF WASTE		
Type of Fly Ash	**Plant A**	**Plant B**	**Plant C**
Unmodified	0.7758	1.0962	1.2402
Limestone Modified	0.5172	0.7308	0.8268

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[a] Equivalent to case where 25% of the theoretical amount of lime is used.

TABLE B-IV

COSTS OF VARIOUS DRAINAGE TREATMENT/PREVENTION PROCESSES

Process	Dollars/Ton of Cleaned Coal[a]		
	Plant A	Plant B	Plant C
Pretreatment			
Calcining-(60% Fixation of SO_2)	3.30[b]	4.44[b]	4.94[b]
Calcining-(Lime-Limestone SO_2system)	2.14[b]	2.80[b]	3.08[b]
Codisposal			
25% of Theoretical Lime Requirement	0.64	0.90	1.02
50% of Theoretical Lime Requirement	1.28	1.81	2.05
Unmodified Fly Ash (Equivalent to 25% Lime) (Mine 15 mi. from power plant)	3.72[c]	5.26[c]	5.95[c]
Unmodified Fly Ash (Equivalent to 25% Lime) (Mine adjacent to power plant)	0.39	0.55	0.62
Limestone-Mod. Fly Ash (Equivalent to 25% Lime) (Mine 15 mi. from power plant)	2.48[c]	3.51[c]	3.97[c]
Limestone-Mod. Fly Ash (Equivalent to 25% Lime) (Mine adjacent to power plant)	0.25	0.36	0.41
Local Soils and Subsoils (Equivalent to 4% Lime)	0.81	1.15	1.30
Effluent Treatment			
Lime Precipitation/Clarification (First five years of active pile)	0.08	0.83[d]	0.05
Lime Precipitation/Clarification (Last five years of 20-yr active pile)	0.10	1.11[d]	0.06
Reverse Osmosis			0.20
Ion Exchange			0.29

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[a] All costs would be added to a basic landfill disposal cost of \$0.46/ton of cleaned coal shipped. All costs are adjusted to March 1978 value. See text for assumptions and qualifications regarding costs.

[b] Major cost is for lime or limestone in scrubbing system.

[c] Major cost is for transporting the fly ash, which is assumed to be free.

[d] Highly active waste.

TABLE B-V

ANNUAL COSTS OF VARIOUS DRAINAGE TREATMENT/PREVENTION PROCESSES

Process	Annual Cost ($k)/2.07 MM Annual Tons of Cleaned Coal[a]		
	Plant A	Plant B	Plant C
Pretreatment			
Calcining-(60% Fixation of SO_2)	6 826	9 201	10 219
Calcining-(Lime-Limestone SO_2 system)	4 420	5 802	6 375
Codisposal			
25% of Theoretical Lime Requirement	1 326	1 872	2 118
50% of Theoretical Lime Requirement	2 651	3 744	4 234
Unmodified Fly Ash (Equivalent to 25% Lime) (Mine 15 mi. from power plant)	2 312	3 268	3 697
Unmodified Fly Ash (Equivalent to 25% Lime) (Mine adjacent to power plant)	240	340	385
Limestone-Mod. Fly Ash (Equivalent to 25% Lime) (Mine 15 mi. from power plant)	1 542	2 178	2 465
Limestone-Mod. Fly Ash (Equivalent to 25% Lime) (Mine adjacent to power plant)	161	227	257
Local Soils and Subsoils (Equivalent to 4% Lime)	1 677	2 380	2 681
Effluent Treatment			
Lime Precipitation/Clarification (First five years of active pile)	172	1 725	106
Lime Precipitation/Clarification (Last five years of 20-yr active pile)	202	2 292	121
Reverse Osmosis			407
Ion Exchange			602

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[a] The basic landfill disposal cost, adjusted from 1974 to 1978 dollars using the Marshall and Swift Equipment Index for Mining, is $962 000 per year. All costs are adjusted to March 1978 values. See text for assumptions and qualifications regarding costs.

TABLE B-VI

NET PRESENT VALUE OF COSTS FOR VARIOUS DRAINAGE TREATMENT/PREVENTION PROCESSES

	Net Present Value of Cumulative Cost[a] ($k)		
Process	**Plant A**	**Plant B**	**Plant C**
Pretreatment			
Calcining-(60% Fixation of SO_2)	58 086	79 299	86 965
Calcining-(Lime-Limestone SO_2 system)	37 614	49 371	54 251
Codisposal			
25% of Theoretical Lime Requirement	11 275	15 932	18 016
50% of Theoretical Lime Requirement	23 550	31 865	36 032
Unmodified Fly Ash (Equivalent to 25% Lime) (Mine 15 mi. from power plant)	19 679	27 807	31 460
Unmodified Fly Ash (Equivalent to 25% Lime) (Mine adjacent to power plant)	2 050	2 897	3 277
Limestone-Mod. Fly Ash (Equivalent to 25% Lime) (Mine 15 mi. from power plant)	13 120	18 539	20 973
Limestone-Mod. Fly Ash (Equivalent to 25% Lime) (Mine adjacent to power plant)	1 367	1 931	2 185
Local Soils and Subsoils (Equivalent to 4% Lime)	14 268	20 257	22 811
Effluent Treatment			
Lime Precipitation/Clarification	1 568	15 981	946
Reverse Osmosis			3 170
Ion Exchange			5 077

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[a] Present value of basic landfill operation, adjusted to March 1978 value, is $8 187 000. All costs are adjusted to March 1978 value. See text for assumptions and qualifications regarding costs.

APPENDIX C

COLUMN LEACHING STUDIES OF LIMESTONE/REFUSE MIXTURES

TABLE C-I

EXPERIMENT IDENTIFICATION FOR DYNAMIC LEACHING STUDIES OF LIMESTONE/REFUSE MIXTURES

Experiment No.	Limestone Location	Sample[a]
GL-12	(None - Control)	1500 g refuse
GL-14	Intermixed	1300 g refuse 220 g limestone
GL-15	Layered at outlet	1300 g refuse 229 g limestone
GL-16	Layered at inlet	1300 g refuse 221 g limestone
GL-17	Layered at outlet	1300 g refuse 220 g limestone (-20 mesh)

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[a] Minus 3/8 inch Illinois Basin Plant B refuse used throughout; minus 3/8 inch limestone unless noted.

TABLE C-II

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIMESTONE/REFUSE MIXTURES

Experiment No. GL-12

	Sample No.								
Parameter[a]	1	3	4	7	8	10	18	20	23
Vol (ℓ)	0.040	0.580	1.290	2.365	3.345	4.175[b]	7.735	8.290	9.855
pH	1.3	1.6	2.1	2.2	2.5	2.6	1.7	1.8	2.2
TDS(%)	8.63	3.13	1.02	0.55	0.08	0.11	3.87	1.87	0.33
Na	25	8	4	3	1	1	3	1	0.9
Mg	500	170	43	29	7	28	56	30	4
Al	1600	520	130	61	7	4	450	230	22
K	51	11	13	6	4	3	4	5	4
Ca	530	440	260	210	52	47	230	140	35
Sc	3.0	0.78	0.14	0.03	<0.01	<0.01	0.77	0.31	0.01
Ti	<0.4	<0.4	<0.4	<0.4	0.16	<0.4	<0.4	<0.4	<0.4
V	1.44	0.71	0.42	0.21	0.04	0.04	0.38	0.16	0.07
Cr(μg/ℓ)	790	420	85	38	<4	<5	260	160	13
Mn	130	19	8	3	0.7	0.5	7	4	0.9
Fe	15000	5300	1700	930	200	170	7600	3300	650
Co	36	13	4	3	0.4	0.2	4	2	0.4
Ni	51	19	8	3	0.7	0.5	7	5	0.9
Cu	10	1.5	<0.9	<0.06	<0.07	<0.09	4	1	<0.09
Zn	76	38	13	6	1	1	11	6	1
Ga	<0.2	<0.2	<0.2	<0.2	<0.2	<.2	<0.2	<0.2	<0.2
As	7.9	1.3	0.58	0.65	0.08	0.07	2.8	0.56	0.1
Br	<0.04		<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Rb	<2	<2	<2	<2	<2	<2	<2	<2	<2
Mo	<5	<4	<9	<1	<1	<1	<1	<1	<1
Ag	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cd(μg/ℓ)	430	130	85	35	4	19	41	30	22
Cs	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
La	1.3	0.60	0.26	<0.5	<0.5	<0.5	0.2	0.2	<0.5
Ce	5.3	1.4	0.53	0.44	0.09	<0.08	1.1	1.0	0.2
Sm	0.83	0.34	0.12	0.06	<0.02	<0.02	0.19	0.14	<0.02
Eu	0.35	0.11	0.03	0.02	<0.01	<0.01	0.08	0.04	<0.01
Tb	0.47	0.02	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Dy	0.14	0.08	0.03	0.43	0.04	0.02	0.12	0.03	0.01
Yb	0.25	0.11	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03
Lu	0.05	0.02	<0.01	<0.01	<0.01	<0.01	0.02	0.01	<0.01
Hf	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Ta	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
W	0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Pb(μg/ℓ)	1100	210	60	45	12	51	110	30	47
Th	1.7	0.34	0.04	<0.02	<0.02	<0.02	0.43	0.15	<0.02
U	0.46	0.31	0.19	0.01	<0.01	<0.01	0.14	0.06	0.01

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[a] Values in μg/mℓ unless otherwise stated.

[b] Water flow was stopped at this point, air was passed through the column for 4 wk, then water flow was resumed.

TABLE C-III

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIMESTONE/REFUSE MIXTURES

Experiment No. GL-14

Sample No.[a]	1	3	6	8	11	18	19	21	25	26	29	30
Vol (ℓ)	0.085	0.195	2.260	3.000	6.090	11.190[b]	11.280	11.570	12.770	13.040	15.380	17.465
pH	2.5	2.6	2.9	4.4	7.4	7.2	3.2	3.6	5.4	6.1	6.9	7.7
TDS (%)	4.72	4.62	0.52	0.34	0.25	0.14	0.65	0.51	0.21	0.21	0.08	0.07
Na	18	12	1.3	1	0.9	0.5	8	2.6	1.1	0.8	0.4	0.9
Mg	630	430	31	19	3.2	1	8	4	3	3	1	1
Al	810	730	9	<0.5	<0.5	1.6	29	13	<0.5	<0.4	<0.4	<0.4
K	0.37	1.2	0.5	0.5	0.7	1	16	4	4	3	2	2
Ca	590	730	530	570	550	308	520	530	460	480	202	194
Sc	1.4	1.1	<0.01	<0.01	<0.01	<0.01	0.09	0.03	<0.01	<0.01	<0.01	<0.01
Ti	<0.4	<0.4	<0.4	<0.4	<0.4	0.8	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4
V	0.48	0.37	<0.01	<0.01	<0.01	0.02	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cr(µg/ℓ)	320	180	4.5	<5	<5	<5	8	<4	<5	<4	<4	<4
Mn	48	49	4	2.4	0.28	0.1	2.3	2.2	1.1	0.8	0.3	0.2
Fe	8100	8500	660	305	3	4	940	700	86	24	3	2
Co	26	24	1.8	0.5	<0.09	<0.10	1.6	0.9	<0.11	<0.08	<0.08	<0.09
Ni	44	43	3.1	1.4	<0.23	<0.26	1.8	1.3	0.4	<0.2	<0.2	<0.2
Cu	4.1	2.4	<0.1	<0.1	<0.1	<0.1	0.5	<0.1	<0.1	<0.1	<0.1	<0.1
Zn	55	49	3.5	1.9	0.23	0.03	1.3	0.9	0.3	0.3	0.03	0.35
Ga	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
As	0.40	0.06	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	0.04
Br	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Rb	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
Mo	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Ag	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cd(µg/ℓ)	320	210	18	4.8	0.28	0.5	21	8.8	0.3	0.1	0.08	<0.02
Cs	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
La	0.47	0.93	0.10	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Ce	1.20	<0.08	<0.08	<0.08	<0.08	<0.08	0.16	0.20	<0.08	<0.08	<0.08	<0.08
Sm	<0.02	0.02	<0.02	<0.02	<0.02	<0.02	0.10	<0.02	<0.02	<0.02	<0.02	<0.02
Eu	0.10	0.07	0.08	<0.01	<0.01	<0.01	0.01	0.01	<0.01	<0.01	<0.01	<0.01
Tb	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Dy	0.21	0.18	0.17	0.01	<0.01	<0.01	<0.01	0.02	<0.01	<0.01	<0.01	<0.01
Yb	0.14	0.10	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03
Lu	0.02	0.03	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Hf	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Ta	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
W	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Pb(µg/ℓ)	30	12	9	<5	<5	16	21	<4	<5	4	<4	26
Th	0.47	0.36	<0.02	<0.02	<0.02	<0.02	0.02	<0.02	<0.02	<0.02	<0.02	<0.02
U	0.27	0.23	<0.01	<0.01	<0.01	<0.01	0.03	0.01	<0.01	<0.01	<0.01	<0.01

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[a] Values in µg/mℓ unless otherwise stated.

[b] Water flow was stopped at this point, air was passed through the column for 4 wk, then water flow was resumed.

TABLE C-IV

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIMESTONE/REFUSE MIXTURES

Experiment No. GL-15

Sample No.[a]	1	2	3	6	11	18	19	22	27	28
Vol (ℓ)	0.090	0.200	1.650	3.825	7.020	14.280[b]	14.395	14.915	20.050	25.135
pH	2.4	2.6	2.6	4.3	5.8	6.2	3.6	4.7	5.4	5.8
TDS(%)	6.08	4.97	0.43	0.34	0.12	0.05	0.94	0.77	0.02	0.03
Na	30	16	0.8	1	0.4	0.4	4	2	0.4	0.5
Mg	470	1940	24	11	4	2	24	17	1.5	1
Al	1000	590	40	<2	<0.4	<0.4	39	0.8	0.7	0.5
K	0.4	48	3.2	5	2	2	8	7.5	2	3
Ca	920	970	370	830	260	96	680	780	100	100
Sc	1.8	1.0	0.08	<0.01	<0.01	<0.01	0.03	<0.01	<0.01	<0.01
Ti	<0.4	<0.4	1.11	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4
V	0.70	0.53	0.09	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cr($\mu g/\ell$)	330	230	24	<16	<4	<1	<8	<8	<7	<5
Mn	60	54	2.4	1.5	0.4	0.2	5	3	0.2	0.2
Fe	10400	9200	610	180	66	22	1400	1100	7	5
Co	30	27	1.6	1.3	<0.08	<0.09	2	2	<0.2	<0.1
Ni	50	48	2.4	1	0.3	<0.2	4	2.5	<0.4	<0.3
Cu	7	3.2	0.32	<0.3	<0.1	<0.1	0.5	<0.2	<0.2	<0.1
Zn	70	54	3.2	0.8	0.3	0.2	8	2	<0.04	<0.03
Ga	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
As	0.31	<0.02	<0.02	<0.02	<0.02	<0.02	0.02	<0.02	<0.02	<0.02
Br	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Rb	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
Mo	<2	<1	<2	<2	<2	<2	<2	<2	<2	<1
Ag	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cd($\mu g/\ell$)	330	200	16	16	0.8	<0.09	8	8	0.2	0.1
Cs	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
La	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Ce	3.2	1.1	0.25	<0.08	<0.08	<0.08	0.43	<0.08	<0.08	<0.08
Sm	0.06	0.31	<0.02	<0.02	<0.02	<0.02	0.05	<0.02	<0.02	<0.02
Eu	0.17	0.11	0.02	<0.01	<0.01	<0.01	0.02	<0.01	<0.01	<0.01
Tb	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Dy	0.20	0.20	0.02	<0.01	<0.01	<0.01	0.01	0.05	<0.01	<0.01
Yb	<0.03	0.13	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03
Lu	0.03	0.24	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Hf	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Ta	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
W	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Pb($\mu g/\ell$)	40	170	16	16	<8	<9	16	<16	<15	<10
Th	0.74	0.43	0.03	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
U	0.34	0.21	0.02	<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01

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[a] Values in $\mu g/m\ell$ unless otherwise stated.

[b] Water flow was stopped at this point, air was passed through the column for 4 wk, then water flow was resumed.

TABLE C-V

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIMESTONE/REFUSE MIXTURES

Experiment No. GL-16

Sample No.[a]	1	3	5	7
Vol (ℓ)	0.100	0.300	1.510	2.815
pH	1.5	1.5	2.2	2.5
TDS(%)	6.27	5.88	0.74	0.34
Na	28	17	2.3	2
Mg	370	350	46	18
Al	1100	1100	110	36
K	38	33	11	4.5
Ca	640	650	220	122
Sc	2.35	2.1	0.12	0.02
Ti	<0.4	<0.4	<0.4	<0.4
V	0.99	0.80	0.28	0.11
Cr(μg/ℓ)	570	660	76	42
Mn	38	37	5	2
Fe	10700	7200	1200	540
Co	24	25	3	1
Ni	40	41	5	2
Cu	7	5	0.23	<0.06
Zn	56	58	7.6	3
Ga	<0.2	<0.2	<0.2	<0.2
As	5.8		0.39	0.28
Br	<0.04	<0.04	<0.04	<0.04
Rb	<2	<2	<2	<2
Mo	0.2	0.6	<1	<1
Ag	<0.01	<0.01	<0.01	<0.01
Cd(μg/ℓ)	300	410	34	15
Cs	<0.04	<0.04	<0.04	<0.04
La	1.5	1.3	<0.5	<0.5
Ce	2.2	2.6	0.37	<0.08
Sm	0.08	0.68	0.03	<0.02
Eu	0.20	0.19	0.03	0.01
Tb	0.05	0.10	<0.1	<0.1
Dy	0.61	0.49	0.04	0.04
Yb	0.30	0.15	<0.03	<0.03
Lu	0.06	0.05	<0.01	<0.01
Hf	<0.02	<0.02	<0.02	<0.02
Ta	<0.05	<0.05	<0.05	<0.05
W	<0.04	<0.04	<0.04	<0.04
Pb(μg/ℓ)	900	640	53	36
Th	1.1	1.1	0.03	<0.02
U	0.38	0.31	0.06	0.01

---

[a] Values in μg/ml unless otherwise stated.

TABLE C-VI

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIMESTONE/REFUSE MIXTURES

Experiment No. GL-17

Sample No.[a]	1	4	7	9	13	18
Vol (ℓ)	0.100	2.040	3.530	5.660	9.435	11.590
pH	3.5	3.8	4.5	5.4	6.5	7.2
TDS(%)	4.01	0.51	0.31	0.26	0.19	0.13
Na	43	2	1	0.8	1	1
Mg	710	39	10	3	4	3
Al	130	<0.3	0.5	<0.3	<0.1	<0.1
K	71	3	10	2	1.5	1.5
Ca	600	600	630	580	510	320
Sc	0.40	<0.01	<0.01	<0.01	<0.01	<0.01
Ti	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4
V	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cr(μg/ℓ)	43	21	<5	<3	<1	<1
Mn	54	5	2	0.6	0.1	0.04
Fe	7800	480	150	61	<0.5	<0.5
Co	27	2.4	0.5	0.3	<0.02	<0.02
Ni	43	3	1	0.3	<0.05	<0.05
Cu	<0.1	<0.1	<0.1	<0.06	<0.02	<0.02
Zn	49	3	1	0.2	<0.01	<0.01
Ga	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
As	0.04	<0.02	<0.02	<0.02	<0.02	<0.02
Br	0.12	<0.04	<0.04	<0.04	<0.04	<0.04
Rb	<2	<2	<2	<2	<2	<2
Mo	<1	<1	<1	<1	<0.2	<0.2
Ag	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cd(μg/ℓ)	140	12	4	0.3	<0.02	0.03
Cs	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
La	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Ce	<0.08	<0.08	<0.08	<0.08	<0.08	<0.08
Sm	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Eu	0.03	<0.01	<0.01	<0.01	<0.01	<0.01
Tb	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Dy	0.24	<0.01	<0.01	<0.01	<0.01	<0.01
Yb	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03
Lu	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Hf	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Ta	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
W	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Pb(μg/ℓ)	27	9	-	-	<5	<5
Th	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
U	0.07	0.01	<0.01	0.08	<0.01	<0.01

---

[a] Values in μg/ml unless otherwise stated.

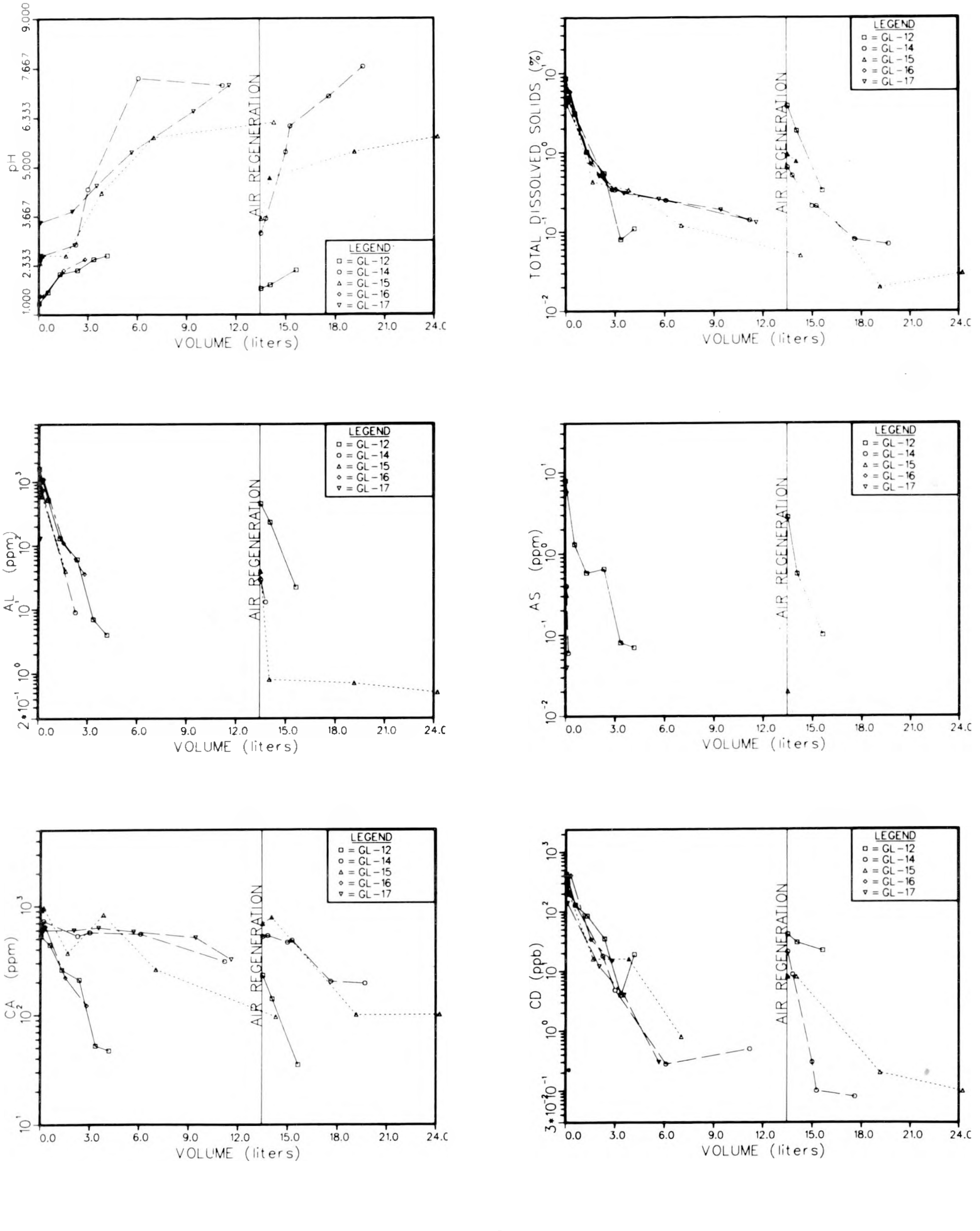


Fig. C-1.

The pH, TDS, and trace element concentrations for dynamic leaching experiments with limestone/refuse mixtures.

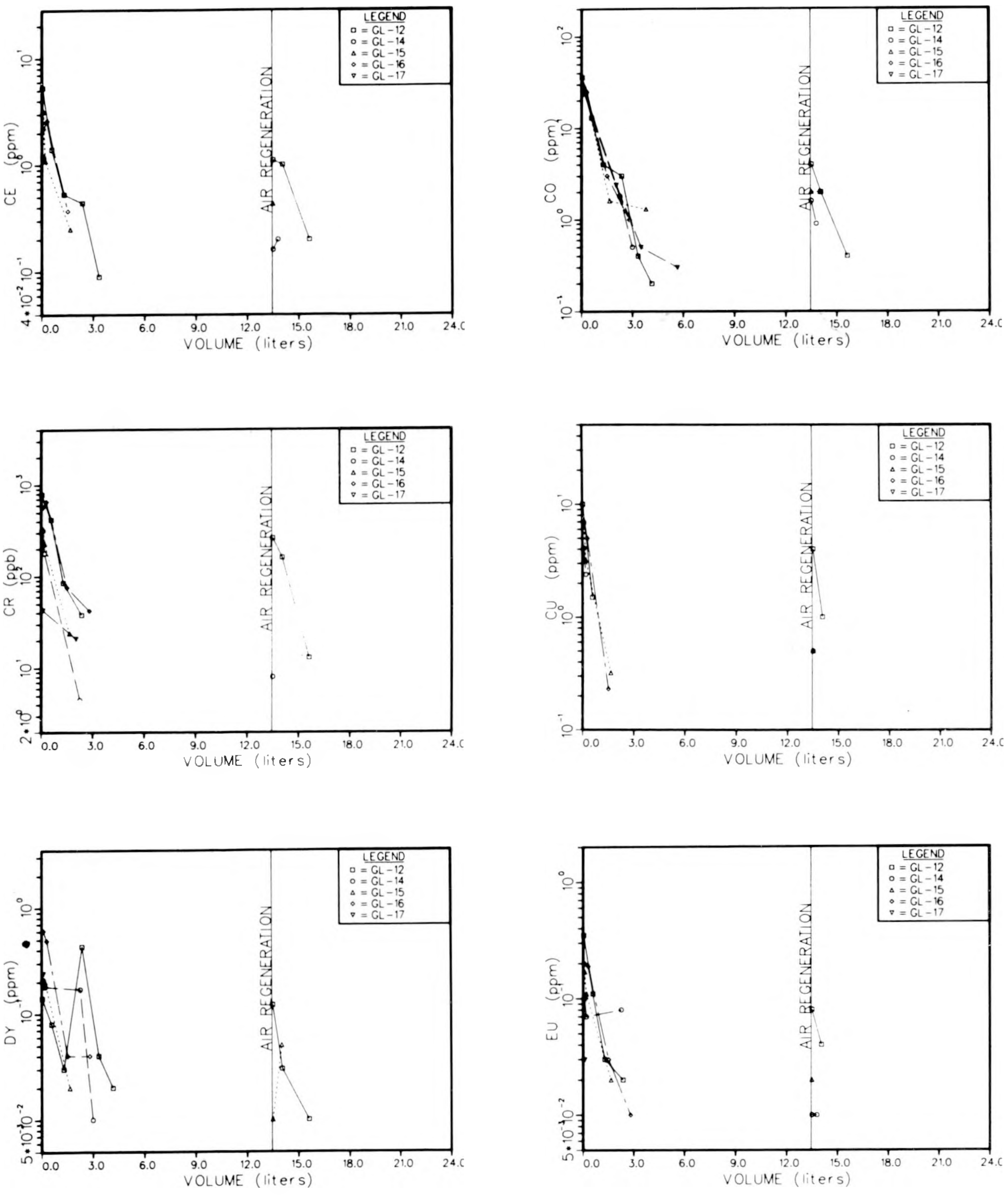
LEGEND
□ = GL-12
○ = GL-14
△ = GL-15
◇ = GL-16
▽ = GL-17
AIR REGENERATION
VOLUME (liters)
0.0
3.0
6.0
9.0
12.0
15.0
18.0
21.0
24.0
CE (ppm)
$4 \cdot 10^{-2}$
10^{-1}
10^{0}
10^{1}
CO (ppm)
10^{-1}
10^{0}
10^{1}
10^{2}
CR (ppb)
$2 \cdot 10^{0}$
10^{1}
10^{2}
10^{3}
CU (ppm)
10^{-1}
10^{0}
10^{1}
DY (ppm)
$5 \cdot 10^{-2}$
10^{-1}
10^{0}
EU (ppm)
$5 \cdot 10^{-2}$
10^{-1}
10^{0}

Fig. C-1. (cont)

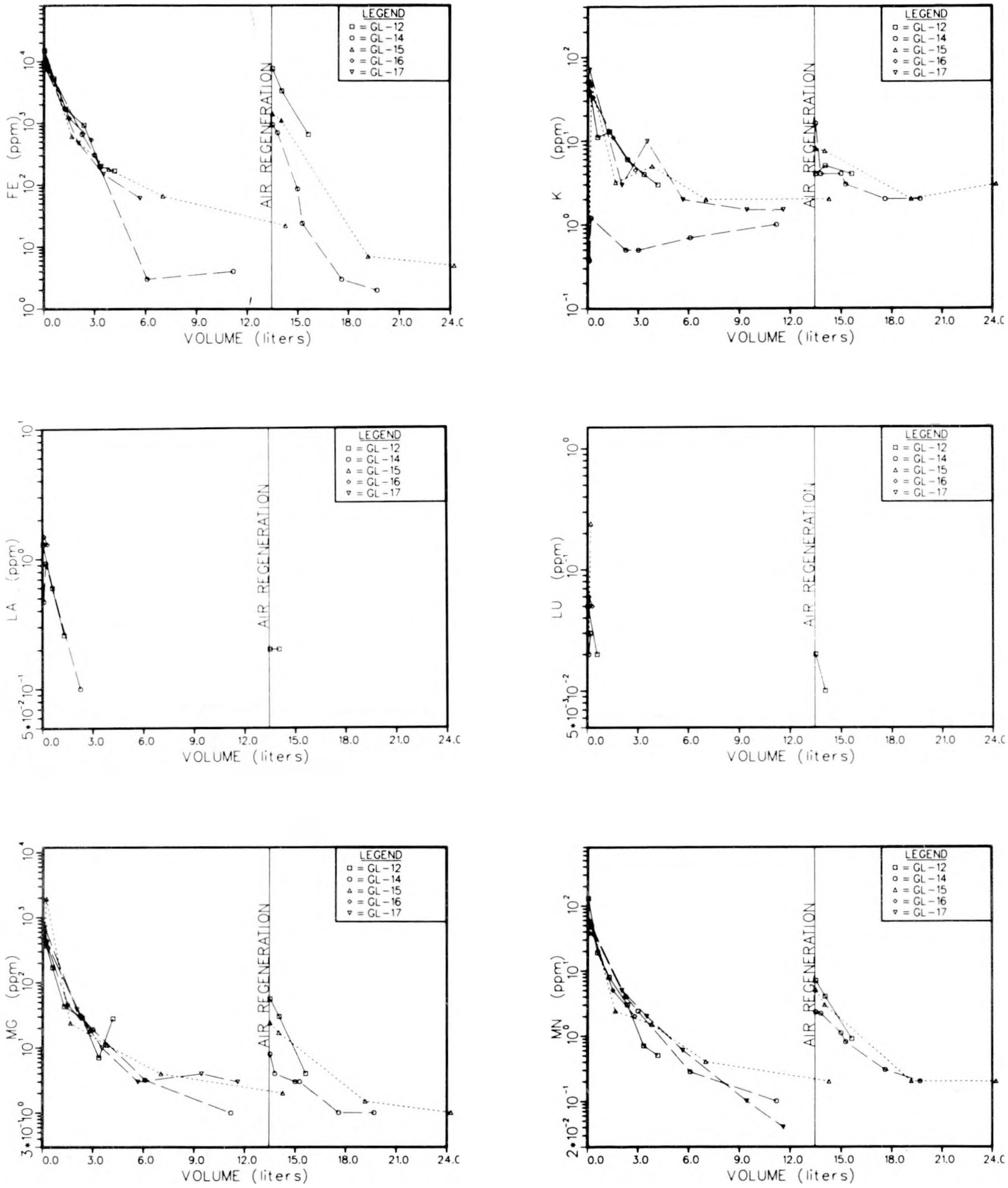

LEGEND
□ = GL-12
○ = GL-14
△ = GL-15
◇ = GL-16
▽ = GL-17
AIR REGENERATION
FE (ppm)
K (ppm)
LA (ppm)
LU (ppm)
MG (ppm)
MN (ppm)
VOLUME (liters)
0.0
3.0
6.0
9.0
12.0
15.0
18.0
21.0
24.0
10^0
10^1
10^2
10^3
10^4
10^{-1}
10^{-2}
$5*10^{-2}$
$5*10^{-3}$
$3*10^{-1}$
$2*10^{-2}$

Fig. C-1. (cont)

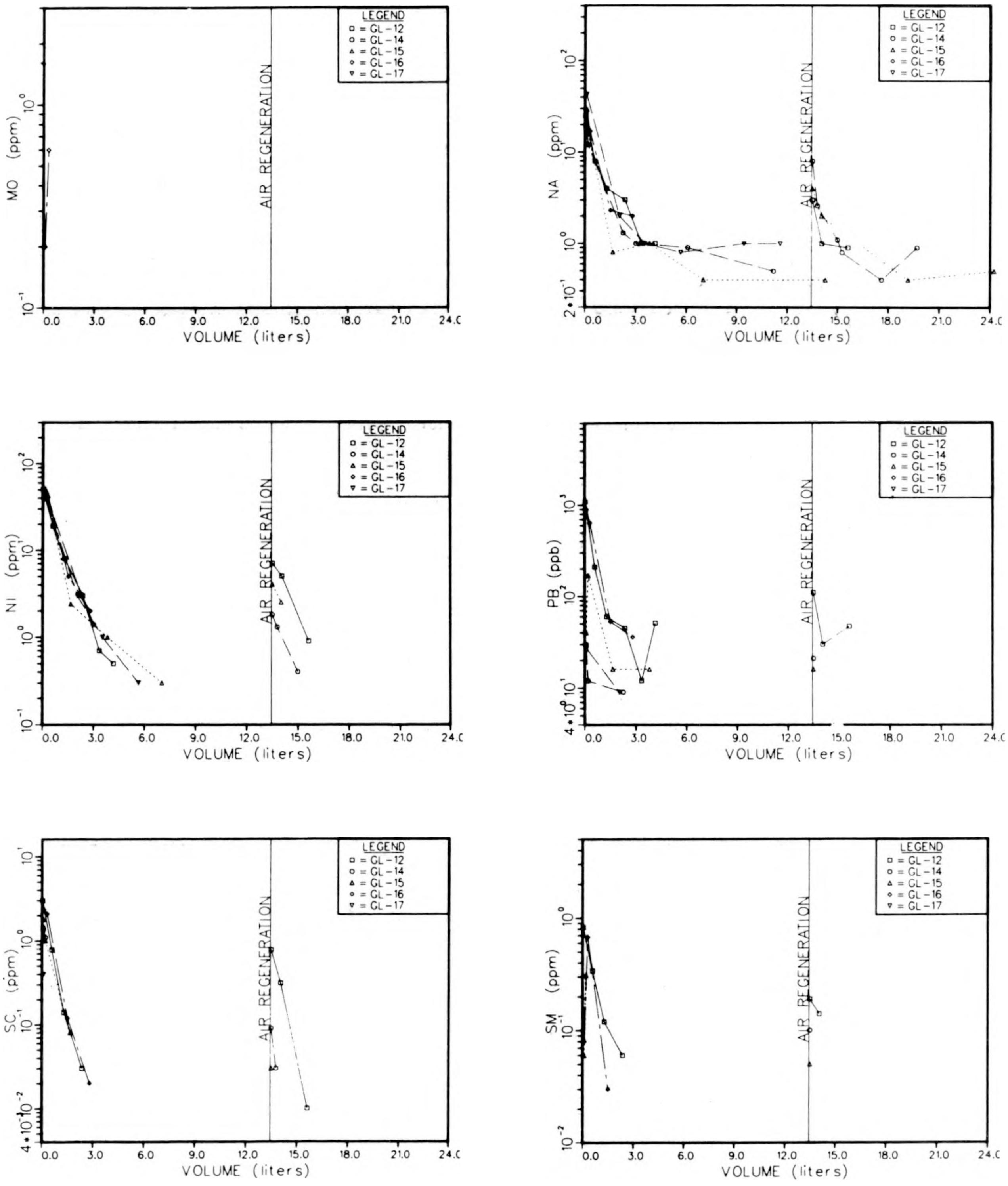
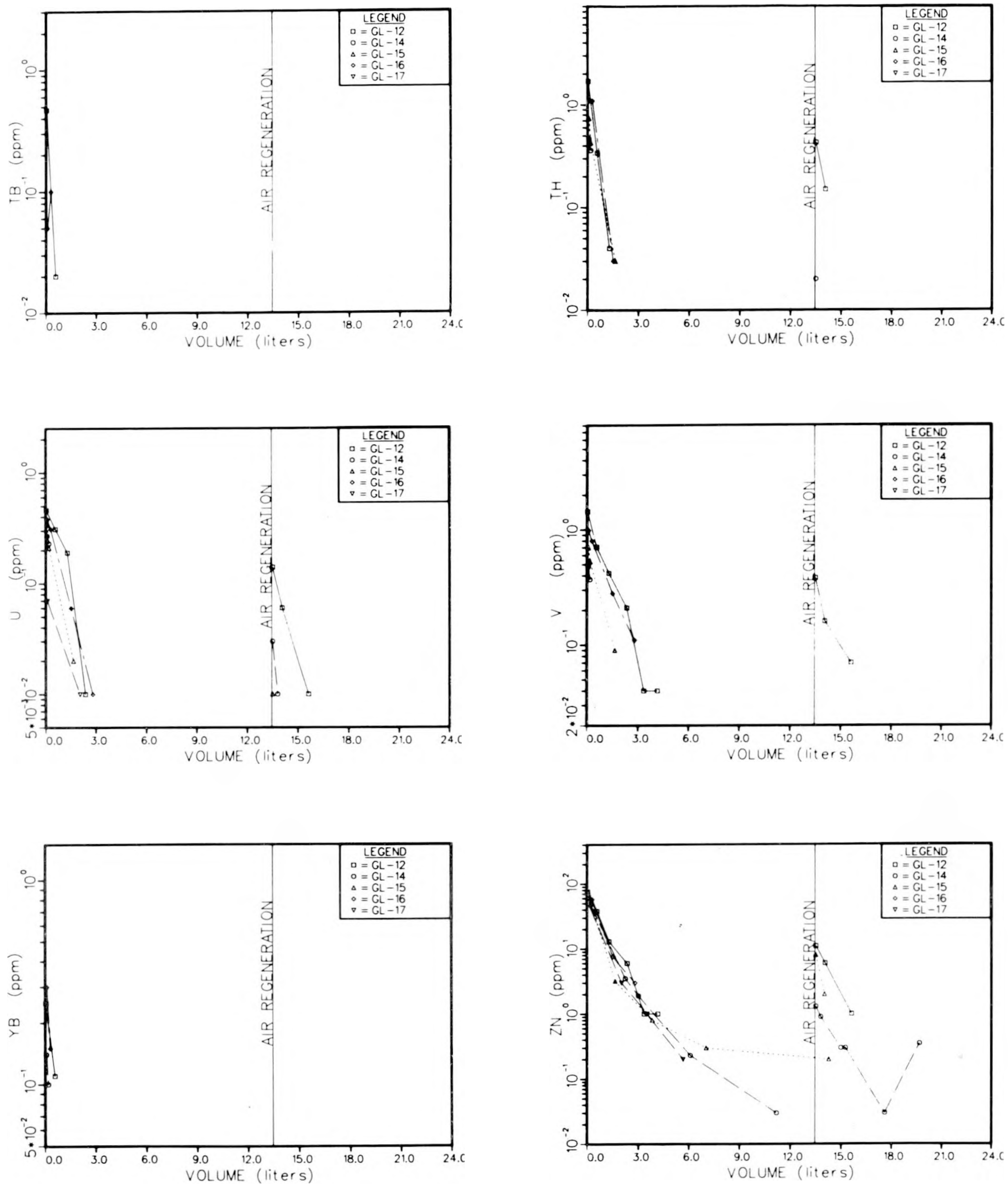

LEGEND
□ = GL-12
○ = GL-14
△ = GL-15
◇ = GL-16
▽ = GL-17
AIR REGENERATION
MO (ppm)
NA (ppm)
NI (ppm)
PB (ppb)
SC (ppm)
SM (ppm)
VOLUME (liters)
0.0
3.0
6.0
9.0
12.0
15.0
18.0
21.0
24.0
10^-1
10^0
10^1
10^2
10^3
10^-2
2•10^-1
4•10^0
4•10^-2

Fig. C-1. (cont)



TB (ppm)
TH (ppm)
U (ppm)
V (ppm)
YB (ppm)
ZN (ppm)
LEGEND
□ = GL-12
○ = GL-14
△ = GL-15
◇ = GL-16
▽ = GL-17
AIR REGENERATION
VOLUME (liters)
0.0
3.0
6.0
9.0
12.0
15.0
18.0
21.0
24.0
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
$5*10^{-2}$
$2*10^{-2}$

Fig. C-1. (cont)

APPENDIX D

COLUMN LEACHING STUDIES OF LIME/REFUSE MIXTURES

TABLE D-I

EXPERIMENT IDENTIFICATION FOR CODISPOSAL OF LIME AND COAL WASTES

Experiment No.	Weight of Waste (g)	Amount of Lime Added (%)[a]
CTWT-11-1	500	0 (control)
CTWT-11-2	500	0.5
CTWT-11-3	500	1.5
CTWT-11-4	500	3
CTWT-11-5	500	10

---

[a] Percentage based on waste. Lime added as slurry, then mixed and dried. Mixtures leached with upward distilled water.

TABLE D-II

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIME/REFUSE MIXTURES

Experiment No. CTWT-11-1

(Control)

Sample No.[a]	1	3	5	7	12	16	18	20
Vol(ℓ)	0.043	0.172	0.344	1.026	1.927	3.581[b]	3.963	4.206
pH	1.8	1.9	1.9	2.4	2.6	3.3	2.2	2.4
TDS(%)	6.54	6.19	4.06	0.56	0.13	0.05	0.34	0.18
F•	12	11	6.5	1.8	1.2	1.3	0.7	0.7
Na	12	8.6	5.6	1.1	0.5	0.6	3.6	2.1
Al	1200	1100	720	74	13	3	42	21
K	4.6	2.4	1.3	1.1	1.2	2.2	5.2	3.5
Ca	380	370	330	100	30	12	60	31
Cr (μg/ℓ)	610	560	450	90	11	3	30	7
Mn	34	33	22	3	0.7	0.2	1	0.5
Fe	13200	12000	7790	1100	230	100	700	380
Co	20	18	12	2	0.4	0.1	0.7	0.3
Ni	30	28	18	3	0.6	0.2	0.8	0.4
Cu	4	4	3	0.34	<0.02	<0.02	0.5	0.20
Zn	48	48	29	4	0.9	0.3	1	0.7
Cd (μg/ℓ)	230	250	170	25	4.6	1.4	7	2.5

---

[a] Concentrations in μg/mℓ unless noted otherwise.

[b] Water flow was stopped at this point, air was passed through the column for 2 wk, then water flow was resumed.

TABLE D-III

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIME/REFUSE MIXTURES

Experiment No. CTWT-11-2

(0.5 wt% Lime)

Sample No.[a]	1	3	5	7	12	16	18	20
Vol(ℓ)	0.055	0.198	0.396	1.166	2.065	4.237[b]	4.658	4.946
pH	2.1	2.2	2.3	2.6	2.9	3.5	2.2	2.6
TDS(%)	5.40	4.33	2.89	0.58	0.19	0.04	0.55	0.14
F	9	9	5.6	1.1	0.5	<0.2	1.2	0.5
Na	4.0	3.1	1.9	0.7	0.5	0.4	2.3	0.8
Al	790	610	420	54	10	0.8	47	8
K	1.4	3.3	0.4	0.4	1.1	1.1	2.6	1.0
Ca	450	510	490	380	240	60	250	70
Cr (μg/ℓ)	480	370	280	60	5	2	50	4
Mn	33	26	18	3	0.6	0.2	2	0.3
Fe	10750	8300	5670	950	260	75	1020	205
Co	18	14	10	1.5	0.3	<0.07	0.8	0.1
Ni	28	21	15	3	0.5	0.2	1	0.2
Cu	4	3	2	0.2	<0.02	<0.02	0.9	0.1
Zn	50	35	23	4	0.9	0.3	2	0.4
Cd (μg/ℓ)	240	200	140	30	5	1	9	3

---

[a] Concentrations in μg/mℓ unless noted otherwise.

[b] Water flow was stopped at this point, air was passed through the column for 2 wk, then water flow was resumed.

TABLE D-IV

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIME/REFUSE MIXTURES

Experiment No. CTWT-11-3

(1.5 wt% Lime)

Sample No.[a]	1	3	5	7	12	16	18	20
Vol(ℓ)	0.048	0.190	0.332	1.064	1.944	3.759[b]	4.104	4.387
pH	2.6	2.6	2.7	3.0	3.2	3.9	2.3	2.6
TDS(%)	1.72	1.54	1.19	0.49	0.29	0.13	1.02	0.37
F	5.9	5.0	3.7	1.6	0.6	0.3	1.8	0.6
Na	7.6	4.9	4.7	1.3	1.1	0.8	1.3	0.5
Al	380	320	240	55	24	6	110	34
K	1.3	3.7	0.9	0.6	1.0	0.8	0.2	0.2
Ca	510	530	550	500	400	220	300	170
Cr (μg/ℓ)	140	120	80	8	<1	<1	100	30
Mn	21	17	13	4	2	0.4	3	1
Fe	2820	2330	3720	540	260	105	1980	710
Co	13	11	8	2	0.8	0.1	1	0.4
Ni	18	15	11	3	2	0.4	3	0.9
Cu	2	1	1	0.2	<0.02	<0.02	2	0.35
Zn	24	20	16	4	2	0.6	6	2
Cd (μg/ℓ)	130	110	90	30	20	3	45	4

---

[a] Concentrations in μg/mℓ unless noted otherwise.

[b] Water flow was stopped at this point, air was passed through the column for 2 wk, then water flow was resumed.

TABLE D-V

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIME/REFUSE MIXTURES

Experiment CTWT-11-4

(3 wt% Lime)

Sample No.[a]	1	3	5	7	12	16	18	20
Vol(ℓ)	0.042	0.168	0.336	1.125	2.076	4.233[b]	4.562	4.830
pH	6.6	6.3	6.5	6.8	7.4	7.9	2.8	3.0
TDS(%)	0.54	0.48	0.37	0.27	0.24	0.15	0.73	0.45
F	0.15	0.15	<0.15	<0.15	<0.15	<0.15	4.8	1.6
Na	7.9	6.4	3.4	1.5	1.1	1.0	4.9	2.0
Al	0.7	<0.6	<0.6	<0.6	<0.6	<0.6	180	54
K	8.9	12.4	6.1	5.3	3.5	3.0	7.9	8.2
Ca	890	870	780	700	620	380	550	540
Cr (µg/ℓ)	<1	<1	23	3	2	<1	19	6
Mn	3	2	2	1	0.6	0.3	12	5
Fe	120	120	69	14	7	3	700	300
Co	0.7	0.6	0.4	0.2	0.2	<0.07	8	3
Ni	1	0.9	0.7	0.3	0.2	0.1	12	4
Cu	0.04	0.04	0.04	0.03	0.02	0.02	0.9	0.2
Zn	0.5	0.4	0.2	0.1	0.08	0.07	14	5
Cd (µg/ℓ)	5.4	4.0	2.0	0.6	0.4	0.6	100	32

---

[a] Concentrations in µg/mℓ unless noted otherwise.

[b] Water flow was stopped at this point, air was passed through the column for 2 wk, then water flow was resumed.

TABLE D-VI

ANALYSES FOR DYNAMIC LEACHING STUDIES OF LIME/REFUSE MIXTURES

Experiment CTWT-11-5

(10 wt% Lime)

Sample No.[a]	1	3	5	7	12	16	18	20
Vol(ℓ)	0.044	0.174	0.304	1.045	1.910	4.002[b]	4.297	4.563
pH	13.1	13.2	13.1	12.9	12.9	12.6	10.7	11.4
TDS(%)	0.50	0.51	0.46	0.47	0.49	0.28	0.36	0.15
F	0.18	0.28	0.20	0.25	0.25	0.12	<0.1	<0.1
Na	6.3	2.9	0.9	0.6	0.6	1.6	16	1.3
Al	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	0.5
K	3.8	1.8	1.7	1.2	1.7	1.0	3.3	0.9
Ca	1100	1400	1100	1100	1200	380	920	360
Cr (µg/ℓ)	<1	<1	<1	<1	<1	<1	<1	<1
Mn	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Fe	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Co	0.05	0.05	0.1	0.1	0.05	<0.05	<0.05	<0.05
Ni	0.1	0.1	0.1	0.06	0.08	<0.03	0.05	<0.03
Cu	0.03	0.04	0.02	0.02	0.03	<0.02	0.03	<0.02
Zn	0.02	0.03	0.02	0.02	0.03	0.01	0.02	<0.01
Cd (µg/ℓ)	<1	<1	<1	<1	<1	<1	2	<1

[a] Concentrations in µg/mℓ unless noted otherwise.

[b] Water flow was stopped at this point, air was passed through the column for 2 wk, then water flow was resumed.

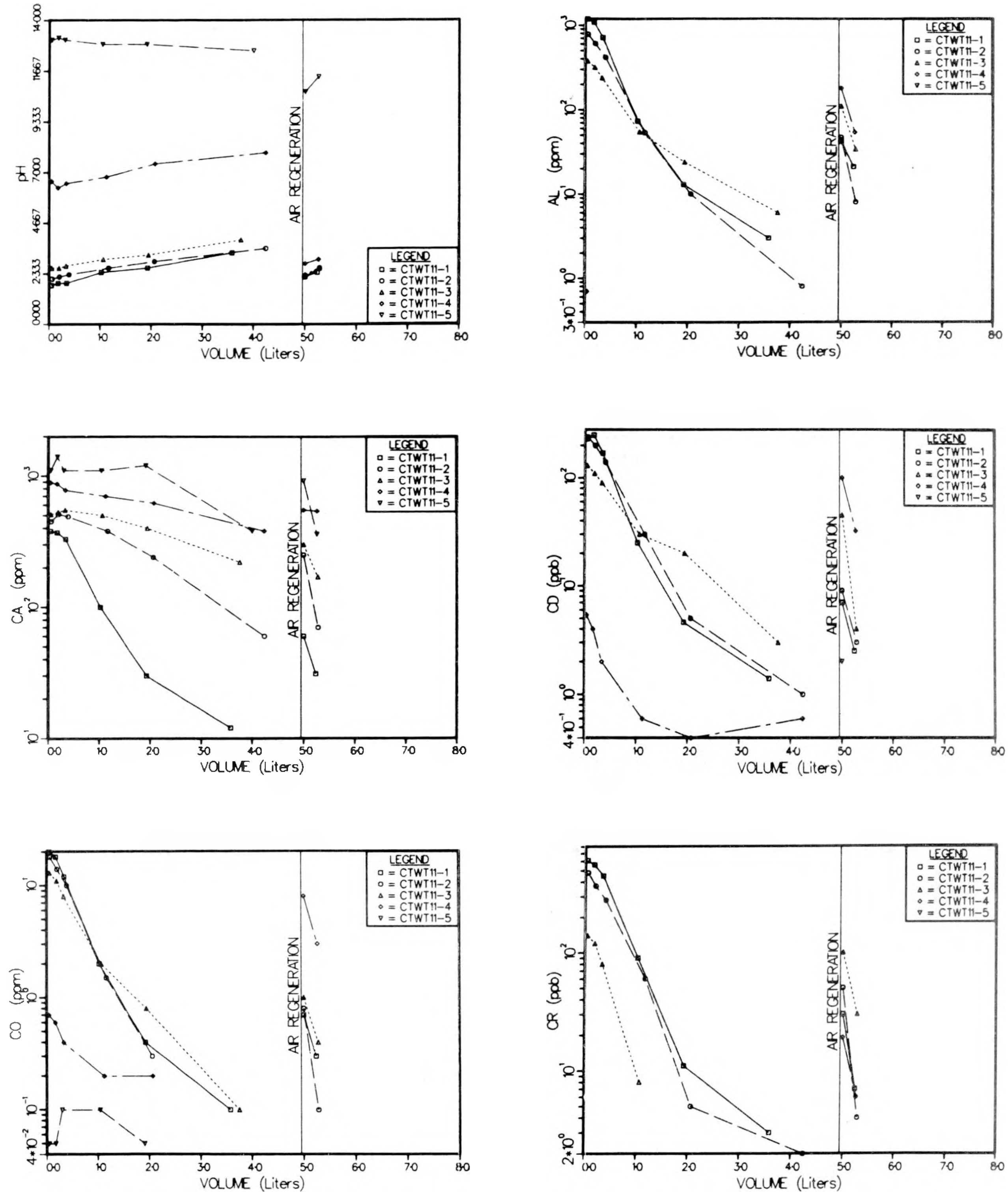


Fig. D-1.
The pH and trace element concentrations for dynamic leaching experiments with lime/refuse mixtures.

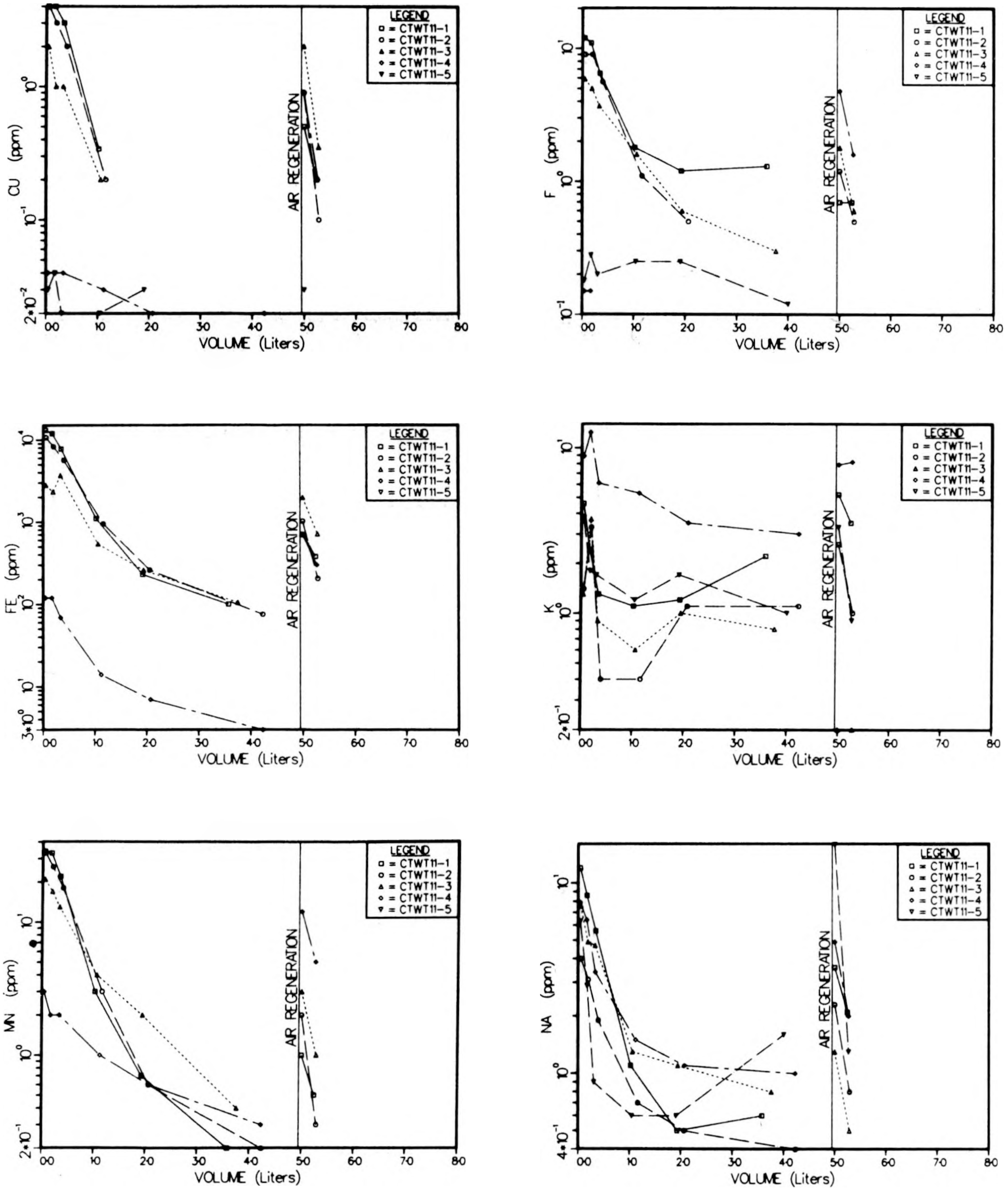

CU (ppm)
2*10⁻²
10⁻¹
10⁰
F (ppm)
10⁻¹
10⁰
10¹
FE (ppm)
3*10⁰
10¹
10²
10³
10⁴
K (ppm)
2*10⁻¹
10⁰
10¹
MN (ppm)
2*10⁻¹
10⁰
10¹
NA (ppm)
4*10⁻¹
10⁰
10¹
AIR REGENERATION
VOLUME (Liters)
0.0
10
20
30
40
50
60
70
80
LEGEND
□ = CTWT11-1
○ = CTWT11-2
▲ = CTWT11-3
◆ = CTWT11-4
▼ = CTWT11-5

Fig. D-1. (cont)

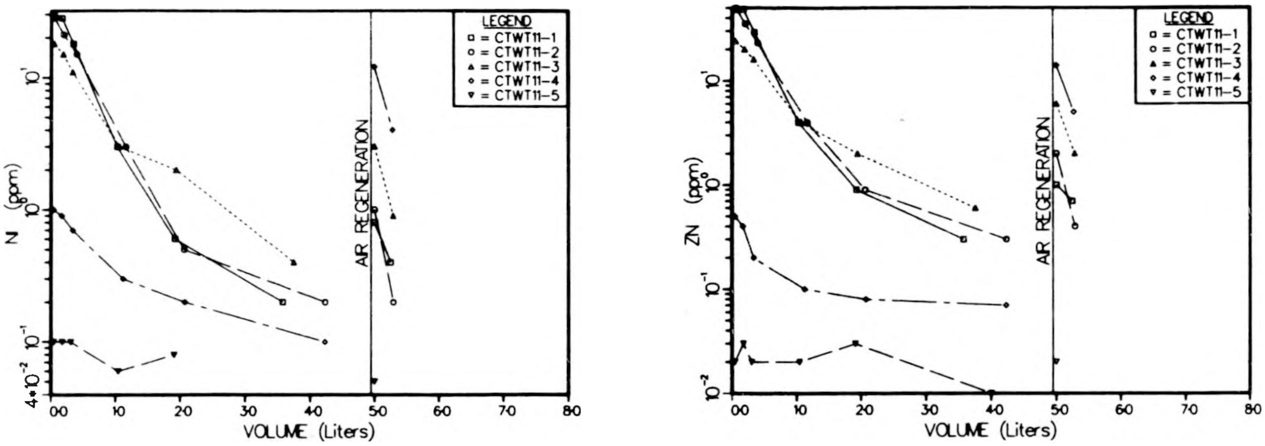

N (ppm)
10^1
10^0
10^{-1}
$4*10^{-2}$
0.0
10
20
30
40
50
60
70
80
VOLUME (Liters)
AIR REGENERATION
LEGEND
□ = CTWT11-1
○ = CTWT11-2
▲ = CTWT11-3
◆ = CTWT11-4
▼ = CTWT11-5
ZN (ppm)
10^1
10^0
10^{-1}
10^{-2}
0.0
10
20
30
40
50
60
70
80
VOLUME (Liters)
AIR REGENERATION
LEGEND
□ = CTWT11-1
○ = CTWT11-2
▲ = CTWT11-3
◆ = CTWT11-4
▼ = CTWT11-5

Fig. D-1. (cont)

APPENDIX E

PROGRAM CODE FOR DETERMINING THE COST OF ALKALINE NEUTRALIZATION OF COAL WASTE DRAINAGES
LASL Identification No. 1065.

```
      PROGRAM LANDFIL(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)
      DIMENSION PPMFE(20),PDLIME(20),   TPYLIME(20),TPDLIME(20),
     1TPDTDS(20),PRILIME(6),COSTLIM(20,6),TTPD(20),CLACOST(20),
     2OPCOST(20),CAPCOST(20),CLARA(20)
C     SET FLAG TO 1 FOR SINGLE IRON CONC. READIN, ELSE 0
    5 READ(5,990)NFLAG
      IF(NFLAG.GT.0) GO TO 10
      DATA PPMFE/10.,20.,50.,100.,200.,400.,500.,600.,700.,
     1900.,1000.,1100.,1300.,1500.,1700.,1900.,2000.,2500.,3000.,3500./
      GO TO 101
   10 READ(5,991)PPMFE(1)
  101 READ(5,992)RAIN,ACRES,FRAAB

  999 FORMAT(1H0,3(E14.6))
      I=1$ACREFT=ACRES*RAIN/12.$CUFT=ACREFT*43560*FRAAB
C     CUFT IS CUBIC FT. EFFLUENT PER YEAR
      IF(NFLAG.GT.0) GO TO 12
   11 DO 15 I=1,20
   12 PDLIME(I)=1.2406E-04*PPMFE(I)
      TPYLIME(I)=PDLIME(I)*CUFT/2000.$TPDLIME(I)=TPYLIME(I)/365.
   13 DO 14 J=1,6
      PRILIME(J)=36.+3*J
   14 COSTLIM(I,J)=PRILIME(J)*TPYLIME(I)
C**** PRILIME IS PRICE OF LIME DELIVERED IN $/TON;COSTLIM IS ANNUAL
C     COST OF LIME
      TPDTDS(I)=8.*PPMFE(I)*CUFT*62.4/730.E09
C**** TONS PER DAY OF TOTAL DISSOLVED SOLIDS
      TTPD(I)=TPDTDS(I)+TPDLIME(I)$CLARA(I)=130.*TTPD(I)
      IF(CLARA(I).LT.100.) CLARA(I)=100.
      IF(CLARA(I).GT.1000.) GO TO 21
      CLARBS=10000.-6.67*(1000.-CLARA(I))
      GO TO 22
   21 CLARBS=10000.+7.*(CLARA(I)-1000.)
   22 ADJCBS=CLARBS*(235./89.)
C**** 235 IS MAR 1978 CHE COST INDEX - CAN BE CHANGED FOR LATER USE
C**** VAR SLURRY IS GPM BOTTOM FLOW FROM SETTLER
      SLURRY=1.70776E-06*CUFT
      CH=250.*SLURRY
C**** MAX TOTAL HEAD OF 250 PSI HAS BEEN ASSUMED FOR COST CALC
      IF(CH.GE.500.) GO TO 301
      PUMPCST=(250.6/115)*3.38*500.
C**** 250.6 IS MAR 78 COST INDEX FOR PUMPS
      GO TO 302
  301 PUMPCST=(250.6/115)*3.38*(500.+4.5*(CH-400.)**.68)
  302 IF(SLURRY.GT.7.67) GO TO 401
C**** 302 IS FOR 1.5 IN PIPE;401-2";402-2.5";403-3";404-4";405-5"
C**** ASSUME 1500 FT TOTAL LENGTH OF PIPE
      DUM=1500*(264/115)
      PIPECST=2.2*DUM
      GO TO 409
  401 IF(SLURRY.GT.12.34) GO TO 402
      PIPECST=2.75*DUM
      GO TO 409
  402 IF(SLURRY.GT.17.97) GO TO 403
      PIPECST=3.3*DUM
```

```
      GO TO 409
  403 IF(SLURRY.GT.27.21) GO TO 404
      PIPECST=4.2*DUM
      GO TO 409
  404 IF(SLURRY.GT.46.) GO TO 405
      PIPECST=5.2*DUM
      GO TO 409
  405 PIPECST=6.8*DUM
      WRITE(6,993)
  409 PPCOST=PUMPCST+PIPECST
      DT1=2.36*ADJCBS$DT2=1.43*DT1$DT3=1.35*DT2
      CLACOST(I)=PPCOST+DT3
      HPPUMP=500.*(SLURRY/3960.)*1.33$HPCLA=CLARA(I)*.015
      EKWH=0.7457*(HPPUMP+HPCLA)*24.*365.$ELECOST=0.035*EKWH
C**** OPCOST IS ANNUAL OPERATING COST EXCLUSIVE OF LIME
C**** CAPCOST IS THE ANNUALIZED CAPITAL COST(PER NAS/NAE BASE CASE)
      OPCOST(I)=365.*40.+ELECOST
      CAPCOST(I)=.2588*CLACOST(I)
      IF(NFLAG.GT.0) GO TO 600
   15 CONTINUE
      WRITE(6,994)RAIN,ACRES,FRAAB
      WRITE(6,995)
      DO 500 I=1,20
      WRITE(6,996)PPMFE(I),TPDLIME(I),CLARA(I),CAPCOST(I),OPCOST(I)
  500 CONTINUE
      WRITE(6,897)
      WRITE(6,997) (PRILIME(J),J=1,6)
      WRITE(6,998) ((COSTLIM(I,J),J=1,6),I=1,20)
      GO TO 899
  600 CONTINUE
      WRITE(6,994)RAIN,ACRES,FRAAB
      WRITE(6,995)
      WRITE(6,996)PPMFE(1),TPDLIME(1),CLARA(1),CAPCOST(1),OPCOST(1)
  850 CONTINUE
      WRITE(6,897)
      WRITE(6,997) (PRILIME(J),J=1,6)
      WRITE(6,998) ((COSTLIM(I,J),J=1,6),I=1,1)
  899  READ(5,990)NEXIT
        IF(NEXIT.GT.0) GO TO 5
  900 CONTINUE
  990 FORMAT(I1)
  991 FORMAT(F10.0)
  992 FORMAT(3(F5.0))
  993 FORMAT(1H0,*WARNING: OVERSIZE SLURRY PIPE*)
  994 FORMAT(1H1,*INPUT DATA*,/,* RAINFALL*,2X,F5.2,* INCHES PER YEAR*,
     1,* AREA OF PILE*,2X,F6.2,*ACRES*,/,* FRACTION ABSORBED:*,2X,F5.4)
  995 FORMAT(1H0,*PPM IRON*,T11,*TONS LIME/DAY*,T26,*CLARIFIER AREA*,
     1T42,*ANN. CAP. COST*,T58,*ANN. OPER. COST*)
  996 FORMAT(3X,F6.0,T11,F8.4,T26,F10.2,T42,F10.2,T58,F10.2)
  897 FORMAT(1H0,*LIME COST AT VARIOUS PRICES, SAME PPMS FE AS ABOVE*)
  998 FORMAT(* *,6F10.2)
  997 FORMAT(1H0,6(4X,F6.2))
      STOP
      END
```

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16. ABSTRACT The report gives third year results of a program to characterize the trace element content of coal waste. In 1978 laboratory experiments were performed to investigate the efficacy of several control options to treat coal wastes at the preparation plant or during disposal. The research revealed that calcining is one of the more effective and permanent means of treating high sulfur coal wastes before disposal to decrease, quite dramatically, the release of environmentally undesirable pollutants into the drainages from disposal sites. Co-disposal of the coal wastes with lime or limestone to neutralize the acid drainage and contain soluble aqueous contaminants within the waste site is also a promising control. Other experiments examined the feasibility of using natural sealants (e.g., clays, soils, calcite, and cements) to isolate the disposal site from its immediate environment. The report discusses the various trade offs for these control options in terms of contaminant reduction, complexity, permanency, and cost. An assessment of coal preparation wastes from the Appalachian region has begun: work on refuse from a single plant indicates significant acid drainage, even with coal wastes with a low percentage (< 1%) of pyrite. Experiments show that Al, Mn, Fe, Ni, and Cu ions are potentially of concern, as their concentrations exceed certain Multimedia Environmental Goal (MEG) values.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group	
Pollution	Drainage	Pollution Control	13B	
Coal	Calcium Oxides	Stationary Sources	08G	
Waste Treatment	Calcium Carbonates	Coal Waste		
Roasting	Sealers	Trace Elements	13H	11A
Chemical Analysis		Acid Drainage	07D	
Sulfur			07B	

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