

**METALS EMISSIONS FROM THE COMBUSTION  
OF USED OIL FUEL**

**PHASE I, LITERATURE SEARCH AND SURVEY**

Report Prepared for:

Used Oil Recycling Coalition  
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# SECTION 1

## SUMMARY AND RECOMMENDATIONS

### 1.1 STUDY APPROACH

The Used Oil Recycling Coalition (UORC) has contracted with Entropy, Inc. to evaluate metals emissions from used oil combustion. The project is divided into two phases.

- Phase I - [REDACTED] and
- Phase II - [REDACTED] Processes

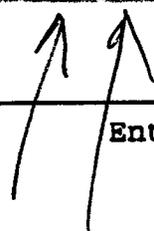
The objective of Phase I is to summarize pertinent data and to determine if there are significant gaps in the available technical information regarding the emissions of lead, barium, cadmium, chromium, arsenic and zinc from the combustion of used oil. The results from Phase I will be used to determine if the Phase II emission testing is needed, and if so, the types of combustion sources and types of used oil that are most important.

An emission inventory for used oil combustion sources was compiled during the Phase I work to evaluate the emission quantities of each source category. This inventory was based on (1) [REDACTED] and (2) [REDACTED] the metals retention and [REDACTED] the various types of combustion processes.

### 1.2 PHASE I CONCLUSIONS

Entropy, Inc. conducted a comprehensive literature search to compile information concerning air emissions resulting from the combustion of used oil in various types of boilers, industrial processes, and space heaters. The literature search included research studies published by government agencies and the private sector, data provided by Coalition members, and information provided by used oil marketers and processors contacted as part of the NORA survey. The general conclusions are listed below.

1. [REDACTED]
2. Due to the lead-in-gasoline phase-down requirements, [REDACTED]



## Summary and Recommendations

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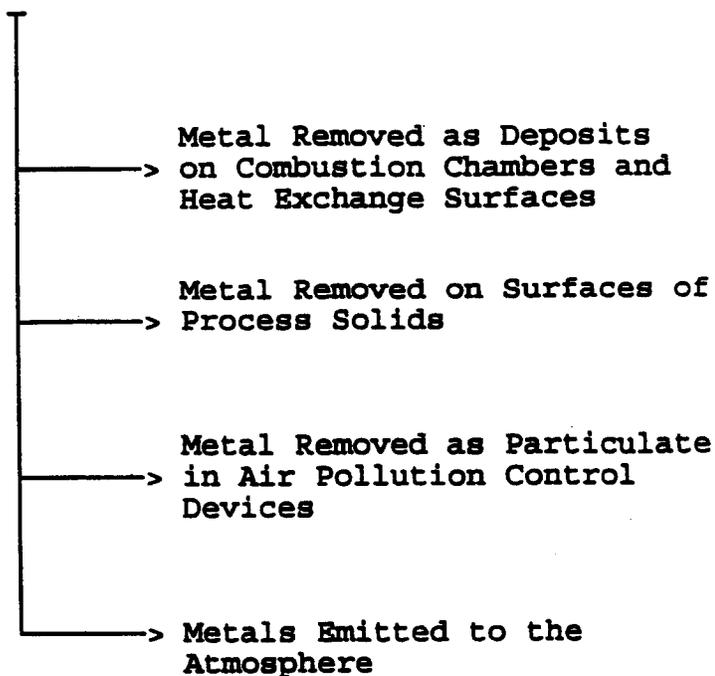
3. Barium levels in used oil have decreased substantially due to changes in the composition of additive packages used for lubricating oils. Average levels in automotive used oils are now in the range of 10 ppm.
4. A large fraction of the metals contained in used oil fuel stays within the combustion chamber and is not emitted into the effluent gas stream. The metals are trapped as deposits on combustion chamber walls and heat exchange surfaces. For industrial boilers, commercial boilers, space heaters, and other small combustion sources, the quantities retained as permanent deposits are probably 50% of the total quantity of metals entering with the fuel.
5. A significant fraction of the metals contained in used oil fuel for industrial furnaces is captured by the product streams processed in the furnaces. These metals are trapped as permanent solid deposits on the surfaces of the product materials and are not emitted to the atmosphere. In the case of cement kilns and asphalt plant rotary driers, metal capture by product materials can range from 20% to 90%. ??  
←
6. Metal-containing particles formed due to vaporization in the combustion chamber and nucleation during gas stream cooling are primarily in the submicron size range. Multi-cyclone collectors are ineffective for this particle size range. The collection efficiencies for the types of wet scrubbers, electrostatic precipitators, and fabric filters used on used oil combustion sources range from approximately 50% to greater than 99%.

The emission studies reviewed as part of this project clearly indicate that the metals contained in used oil fuel are partitioned into several separate streams. In the case of space heaters and boilers, a portion of the metal is retained in the combustion chamber on refractory walls and/or heat exchange surfaces. In the case of industrial furnaces such as cement kilns and asphalt plant driers, a major fraction of the metal is captured by the process solids passing through the furnace. Also, some of the metal is captured as particulate matter in air pollution control systems. The partitioning of metals in combustion processes is illustrated in Figure 1-1.

Typical values of these various partitioning factors have been used to estimate the lead emissions from the various combustion sources included in this scope of this study. The basis for these partitioning factors is provided in Section 5 of this report.

Figure 1-1. Metals Partitioning

Metals Entering  
With Used Oil Fuel



The lead emission estimates are based on source-by-source partitioning factors and total used oil fuel consumption rates. The used oil fuel data is based in part on the NORA sponsored survey of used oil marketers and processors. The respondents to this survey accounted for 570 million gallons of used oil in 1993. Of this total, 204 million gallons were used or sold as fuel. A total of 100 companies responded to this survey.

The NORA survey did not include the used oil fuels burned in space heaters since that fuel is usually burned by the organization generating the used oil and is not processed or marketed by one of the surveyed companies. Entropy has estimated that an additional 69 million gallons of used oil is consumed as fuel in space heaters.

While the NORA survey was one of the largest surveys of used oil marketers and processors that has been conducted, it is

## Summary and Recommendations

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impractical to survey all of the companies processing and burning used oil fuel. Accordingly, Entropy has applied a multiplication factor to the source category data from the NORA survey to estimate total U.S. consumption rates. A multiplication factor of 3.583 has been used for this purpose. This multiplication factor is based on the assumption that 204 million gallons of used oil per year are used as fuel in the United States.

Emission rates of the other metals (weighted average concentrations listed below) not necessarily included in the scope of the study can be estimated by using a ratio of the average metals concentrations used in developing these emission estimates. These metals concentrations are the weighted averages of the 204 million gallons of used oil fuel described in the responses to the NORA survey.

The partitioning of the other metals is not necessarily the same as lead. Accordingly, there are differences in the fractions of each metal which are emitted. These differences may affect the relative importance of each source category as a source of metal emissions.

|          |           |
|----------|-----------|
| Lead     | 40.36 ppm |
| Barium   | 10.0 ppm  |
| Arsenic  | 1.19 ppm  |
| Cadmium  | 1.60 ppm  |
| Chromium | 8.83 ppm  |
| Zinc     | 917.0 ppm |



Based on the emission inventory, several conclusions have been reached regarding the need for further evaluation of metals emissions from the various source categories.

7. Asphalt plants are the largest source of metals emissions. There are indications that the consumption of asphalt is increasing. Metals removal in baghouse controlled asphalt plants is very high. The baghouse is the most efficient control for particulates.

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8. Utility boilers are an important consumer of used oil. More data is needed regarding the types of boilers that burn used oil, the types of fuels used, the types of air pollution control systems on these boilers, and the used oil firing practices. There are no previously conducted air emissions studies on utility boilers regarding the potential metal emissions attributable to the burning of used oil.
9. Industrial boilers used for space heating and steam generation are important due to the quantities of used oil consumed and the limited air pollution control system capability. Emissions data is needed to evaluate the metals retention as deposits in the boilers and to evaluate the fractions removed as part of bottom ash or flyash streams. There is no relevant, up-to-date data concerning metals emissions from industrial boilers.
10. Space heaters are an important category due to the quantities of used oil burned and the lack of air pollution control systems. There is very little available emissions data. However, data collected by the State of Vermont is likely to provide valuable data.
11. The NORA survey data indicates that small commercial boilers, apartment house boilers, and school boilers (collectively termed "residential/commercial" boilers) comprise a small fraction of the overall used oil generated in the United States.
12. Cement kilns and other types of industrial processes do not appear to be significant sources of metal emissions due to high efficiency of metal capture.

### 1.3 PHASE I RECOMMENDATIONS

1. An emission test program should be conducted to characterize emissions from three types of used oil combustion sources: (1) industrial boilers, (2) residential/commercial boilers, and (3) space heaters. The tests should include analyses of the used oil fuel and the accumulation of metals on heat exchange surfaces and refractory within the boilers. The scope of the tests should include lead, barium, cadmium, chromium, arsenic, and zinc.
2. Air pollution emission data obtained as part of the State of Vermont study of space heaters should be requested as soon

Summary and Recommendations

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as it is available to further evaluate the environmental aspects of used oil combustion in space heaters.

## SECTION 2

### PROJECT PURPOSE, TECHNICAL APPROACH, AND SCOPE

#### 2.1 PURPOSE OF THE USED OIL AIR EMISSIONS STUDY

The main objective of the Used Oil Recycling Coalition (UORC) Air Emissions Study is to compile accurate lead air emissions data for a variety of combustion sources utilizing used oil as a fuel.

The Coalition recognizes that other elements and compounds potentially emitted during used oil combustion may also be of concern. Accordingly, a secondary objective of this project is to compile accurate emissions data for other metals including barium, cadmium, chromium (Total and Cr<sup>+6</sup>), arsenic, and zinc.

#### 2.2 PROJECT TECHNICAL APPROACH

The project is divided into two phases. A detailed literature search was conducted as Phase I. As part of this work, tests relevant to used oil combustion were reviewed, and data on trends in used oil composition was obtained. An emission inventory approach was used in compiling the Phase I data. The presently available data concerning used oil composition, quantities burned in combustion processes, and metals partitioning factors were used to estimate pollutant emissions from each category of the used oil combustion process.

The information compiled in Phase I will be reviewed by the Coalition to identify important gaps, if any, in the available technical information. These gaps could be addressed by surveys of Coalition members and/or additional air emission tests of selected combustion sources.

If UORC decides to continue with Phase II, Entropy, Inc. will conduct air emissions tests at facilities firing used oil fuel. These test programs would include the following basic elements.

- Used oil fuel sampling to characterize the as-fired fuel contaminants
- Gas stream sampling at the stack or air pollution control system outlet to measure emissions of contaminants
- Gas stream sampling at the inlet to the air pollution control system to evaluate the metals concentrations in the submicron size range

## Project Purpose, Scope, and Technical Approach

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- Ash and/or solid waste stream sampling to measure concentrations of the contaminants exiting the combustion process as part of non-air waste streams
- Combustion process and air pollution control system operating data generated by plant instruments which document the representativeness of the test

These tests will be conducted in accordance with Site Specific Test Plans and Quality Assurance Plans to be developed by Entropy prior to the field tests. These documents will be submitted to the Coalition and to the U.S. EPA, Emission Inventory Branch for comment to ensure that the test program provides the data needed by UORC.

Following the emission tests, a Phase II report will be prepared. This report will include results from the emission tests and other information compiled by follow-up surveys of UORC member companies. An emission inventory format will be used in evaluating lead emissions and other air contaminant emissions from used oil combustion in various types of processes.

### 2.3 PROJECT SCOPE

The project is strictly limited to the evaluation of metals emissions from the combustion of used oil. These metals include the following.

- Lead
- Barium
- Cadmium
- Chromium
- Arsenic
- Zinc

#### 2.3.1 Scope of Phase I

Phase I was composed of two parallel efforts: (1) a survey of used oil marketers and processors, and (2) a compilation and review of previous air emission data. A survey form designed and distributed by the National Oil Recyclers Association (NORA) was used to obtain data from member companies and organizations actively involved in used oil purchasing, processing, re-refining, and sale. The NORA survey is described in Section 2.3.2.

## Project Purpose, Scope, and Technical Approach

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The used oil combustion air emission data literature search was begun by conducting computerized searches for relevant technical articles, government sponsored research projects, and other publications. The following computerized literature searches were conducted.

- National Technical Information Service (NTIS)
- ARIES (U.S. Government Research Publications)
- Congressional Research Service
  - Lexis-Nexis System
  - JANUS System
- State of California, Publications Department
- Electric Power Research Institute, Documents Department
- U.S. EPA, Pollution Prevention Clearinghouse

This material was supplemented by reference lists and limited searches provided by the American Petroleum Institute, Eastern Research Group, Inc., and the U.S. Army Corp. of Engineers Construction Engineering Research Laboratory. Many of the technical articles, research reports, manuals, and newspaper articles cited in these literature searches were purchased and reviewed. Additional documents were obtained from the Used Oil Recycling Coalition.

The comprehensive literature search revealed that there were a large number of documents concerning used oil in general, but very few were relevant to used oil combustion-related emissions. In an attempt to identify the existence of previous combustion-related studies, Entropy initiated a telephone survey of individuals and organizations active in the used oil business and air pollution control business. A partial list of the organizations contacted is listed below.

- Used oil marketers or processors who responded to the NORA survey (approximately 15 organizations contacted)
- Used Oil Recycling Coalition members listed by the UORC Project Manager
  - Edison Electric Institute
  - American Petroleum Institute
  - Cement Kiln Recycling Coalition
  - Waste Oil Heaters Manufacturing Association
- Electric Power Research Institute (Palo Alto and Washington D.C.)
- Lubricant Manufactures and Consultants
  - Castrol
  - Ozimek, Inc.
  - Pennzoil, Inc.

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- U.S. Environmental Protection Agency
  - Air Energy and Environmental Research Lab
  - Office of Air Quality Planning and Standards
  - Office of Technology Transfer
  - Hazardous Waste Laboratory
  - Documents Room
- Department of Energy, Bartlesville, Oklahoma
- U.S. Military
  - Army Industrial Hygiene Laboratory
  - Air Force Occupational and Environmental Health Lab.
  - Army R&D Engr.Center, Fuels and Lubricants Branch
  - Navy Energy and Environmental Support Activity
- Oil Burner Manufacturers
  - North American Manufacturing Co.
  - Peabody Engineering
  - Coen Co.
  - Todd Combustion, Inc.
- Trade Organizations
  - American Boiler Manufacturers Association
  - Council of Industrial Boiler Operators
  - National Asphalt Paving Association
  - Building Owners and Managers Association
  - Convenient Automotive Services Institute
- Private Companies
  - General Motors
  - Carolina Power & Light
  - Southern California Edison
  - Florida Power & Light
  - Clean Burn, Inc.
- Consulting Firms
  - JACA, Inc.
  - Leo Wietzman
  - Entropy, Inc.

Essentially all of the persons contacted were actively involved in used oil related work, hazardous waste related work, or other subject areas directly relevant to this project. This group of individuals was essentially unanimous in stating that they were not aware of any air emission studies involving the emissions of metals from used oil combustion other than those which were already published and well known.<sup>1-44</sup> All of these individuals were very helpful in that they suggested other people or organizations which may have relevant data. However, few of these leads resulted in the identification of air emissions data that was not already listed on one or more of the computerized searches.

## Project Purpose, Scope, and Technical Approach

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The initial review of the articles and other materials received indicated that there was little information of direct relevance to this study. Accordingly, the scope of the literature search was expanded to include the burning of virgin oils and the burning of hazardous wastes. The primary sources of information in these two categories were published articles and seminar proceedings. The search included the following materials.

- Air & Waste Management Journals (10 years)  
(formally Air Pollution Control Association)
- Air & Waste Management Conference Proceedings (10 years)
- Hydrocarbon Processing (limited number of issues)
- Environmental Progress (6 years)
- Chemical Engineering Progress (6 years)
- Environmental Science and Technology (6 years)
- AIChE Journal (10 years)
- The Chemical Engineer (3 years)
- Oil & Gas Journal (1 year)
- World Oil (2 years)
- Combustion Institute Proceedings (8 years)
- Atmospheric Environment (6 Years)

This search identified several used oil related articles which had escaped listing the in computerized databases due to the types of keywords chosen by the author. Furthermore, several virgin oil and hazardous waste combustion-related articles were obtained.

The remainder of the literature search involved the review of the documents and the references cited in the documents. Several additional documents were obtained in this manner.

Entropy has made a determined effort to identify and obtain technical information concerning air emissions from the combustion of used oil. The authors believe that most of the useful materials have been compiled. However, due to the volume of non-relevant articles, publications, and manuals, it is possible that one or more useful documents has escaped detection. If so, the Phase I report will be amended when the Phase II work is completed.

Overall, the presently available database concerning used oil combustion can be characterized as very limited and out-of-date. Many of the out-of-date studies have been reviewed in detail to demonstrate why they should no longer be used in evaluating metals emissions from used oil combustion.

### 2.3.2 NORA Survey

The National Oil Recyclers Association (NORA) conducted a survey to support the Used Oil Recycling Coalition (UORC) study of used oil air emissions. The survey form was designed by NORA member organizations. A blank version of the form and an accompanying letter from UORC is included in Appendix A of this report. The survey addressed a number of topics of importance to the UORC air emissions study.

- Quantities of used oil handled
- Average metals concentrations in used oil for the year (1993)
- Sources of used oil
- Quantities of used oil sold for combustion purposes

The survey form was mailed by NORA in October, 1993 to used oil marketers, processors, and re-refiners. The 100 respondents accounted for 570 million gallons of used oil. The quantity used or sold as fuel by these respondents was 204 million gallons. This is approximately 25% of the total used oil believed to be sold as fuel based on several previous studies<sup>45,46</sup>.

Some of the 100 responses had significant data gaps, especially concerning the metals concentrations of the used oil. There were also some data entry errors with respect to the total quantities of used oil received, processed, and sold. To the extent possible, Entropy has corrected these errors by contacting the organizations submitting the survey forms. The NORA data is presented in Sections 3 and 4 of this report.

### SECTION 3

## COMPOSITIONS AND QUANTITIES OF USED OILS

Many of the studies concerning air pollution emissions from the combustion of used oil were conducted prior to 1985. Since that time, significant changes in the compositions of used oil could make some or all of the conclusions based on these studies out-of-date and not representative of present-day conditions. Accordingly, Entropy has evaluated shifts in used oil composition.

The 1993 survey conducted by the National Oil Recyclers Association is one of the major sources of information concerning used oil compositions. Additional information was obtained by contacting respondents to the NORA survey, by contacting lubricant additive manufacturers, and by reviewing the technical literature. Several conclusions are based on this survey of used oil compositions and quantities.

- Lead levels in mixed used oil streams have dropped significantly due to the phaseout of lead-in-gasoline which began in 1985. Lead levels during the 1992-1993 time period were between 20 and 60 ppm. Some of the lead which is now present may be attributable to erosion of passenger car and heavy duty vehicle metal parts.
- Barium levels in mixed used oil streams have dropped significantly due to the elimination of barium as a component in lubricant dispersants. Barium concentrations are now less than 10 ppm.
- Zinc levels in mixed used oil have remained approximately the same since the 1985 time period. Zinc is present due to its continued use as a detergent in lubricants manufactured for passenger cars and heavy duty vehicles. Used oil zinc levels range between 400 and 1200 ppm.
- Cadmium and chromium levels continue to be present at pre-1985 levels of 1 to 10 ppm. The presence of these metals are due to wear of metal parts in passenger and heavy duty vehicle engines.
- Arsenic levels continue to be quite low. No information was found concerning the possible sources of arsenic.

### 3.1 TYPES AND QUANTITIES OF USED OILS

Passenger cars, diesel trucks and buses, and heavy duty vehicles are some of the main sources of used oil. These oils include the spent lubricant drained from the crankcase, brake fluid, and transmission fluid. These materials will be collectively termed "crankcase oil," or "vehicular oil" throughout the remainder of this report to be consistent with conventional terminology.

Bider et al.<sup>45</sup> estimated that the total quantity of crankcase oil generated in the U.S. in 1983 was 699,000,000 gallons. This was based on a new lubricant sales of 1,251,000,000 gallons and generation factors which varied from 0.47 to 0.75 (average factor 0.559) for most subcategories of oil included in the overall category. The generation factors take into account losses of lubricant due to combustion within the engine, leakage from the engine, and improper disposal. A summary of the data used by Bider et al. is provided in Table 3-1.

The data presented in Table 3-1 is useful for comparing the relative importance of different used oil generators. This will be used in Section 3.2 in the qualitative evaluation of trends affecting used oil composition.

Industrial oils are the second major category of used oils. The oils used in the industrial sector can be further subdivided into a large number of quite different applications.

#### General Industrial Oils

- Hydraulic
- Gear
- Other (i.e. Refrigeration, Compressor)

#### Industrial Engine Oils

- Marine
- Railroad diesel
- Natural gas turbine

#### Metalworking Oils

- Metal removing
- Metal forming
- Metal treating
- Metal protecting

#### Process Oils

- Transformer

Compositions and Quantities of Used Oil

Table 3-1. Vehicular Oil Generation Rates<sup>45</sup>

| Subcategory of Used Oil Generator | New Oil Sales<br>10 <sup>6</sup> Gals. | Used Oil Generation Factor | Used Oil Quantity,<br>10 <sup>6</sup> Gals. |
|-----------------------------------|--|----------------------------|---|
| <b>On-Road</b>                    |  |                            |   |
| Passenger Cars                    | 441.6                                  | 0.67                       | 295.9                                       |
| Commercial Cars                   | 158.9                                  | 0.66                       | 104.7                                       |
| Trucks and Buses                  | 139.7                                  | 0.59                       | 82.4  |
|                                   |  |                            | 483.0                                       |
| <b>Off-Road</b>                   |  |                            |   |
| Farm                              | 74.1                                   | 0.59                       | 43.7  |
| Construction                      | 68.8                                   | 0.59                       | 40.6  |
| Mining                            | 45.5                                   | 0.59                       | 26.8  |
| Government                        | 11.7                                   | 0.63                       | 7.4   |
| Aviation                          | 12.7                                   | 0.47                       | 6.0   |
|                                   |  |                            | 124.5                                       |
| <b>Hydraulic Fluids</b>           |  |                            |   |
| On-Road                           | 79.0                                   | 0.10                       | 7.9   |
| Off-Road                          | 111.4                                  | 0.75                       | 83.6  |
| Greases & Other                   | 108.0                                  | 0.00                       | 0.0   |
|                                   |  |                            | 91.5  |
| <b>TOTALS</b>                     |  | <b>0.599</b>               | <b>699.0</b>                                |

Bider et al<sup>45</sup> has estimated that the total quantity of these industrial oils reaching the used oil management system is 567,000,000 million gallons. As in the case with the crankcase oils, these estimates are based on new oil sales and an estimated used oil generation factor. The data and assumptions which are the basis of the industrial oil estimate are presented in Table 3-2.

Compositions and Quantities of Used Oil

Table 3-2. Industrial Oil Generation Rates<sup>45</sup>

| Subcategory of Used Oil Generator | New Oil Sales<br>10 <sup>6</sup> Gals. | Used Oil Generation Factor | Used Oil Quantity,<br>10 <sup>6</sup> Gals. |
|-----------------------------------|--|----------------------------|---|
| <b>General Industrial Oils</b>    |  |                            |   |
| Hydraulic Oils                    | 264.0                                  | 0.80                       | 211.2                                       |
| Gear Oils                         | 33.0                                   | 0.606                      | 20.0  |
| Other Oils                        | 124.0                                  | 0.617                      | 76.5  |
|                                   | 421.0                                  |                            | 307.7                                       |
| <b>Industrial Engine Oils</b>     |  |                            |   |
| Railroad Diesel                   | 57.0                                   | 0.20                       | 11.4  |
| Marine                            | 49.0                                   | 0.50                       | 24.5  |
| Gas Turbine                       | 38.0                                   | 0.20                       | 7.6   |
|                                   | 144.0                                  |                            | 43.5  |
| <b>Metalworking Oils</b>          |  |                            |   |
|                                   | 163.0                                  | 0.874                      | 125.8                                       |
| <b>Process Oils</b>               |  |                            |   |
| Transformer                       | 76.0                                   | 0.90                       | 68.4  |
| Other                             | 222.0                                  | 0.1                        | 22.2  |
| <b>TOTAL</b>                      |  |                            | <b>90.6</b>                                 |

It should be noted that the data summarized in Tables 3-1 and 3-2 were based on a comprehensive literature search, a series of telephone interviews, and visits to 25 facilities involved in used oil management. However, the authors concluded that data concerning used oil quantities was "... not well documented.." and it "... was often necessary to make reasonable best estimates.." The authors further state that,

"It is important to emphasize in this section that the accuracy of the used oil generation factors is unknown and believed to be based upon theoretical considerations rather than measured values." Reference 45, page 3-15.

## Compositions and Quantities of Used Oil

The five references used by Bider et al. to estimate the used oil generation rate factors were published in 1974, 1978, 1980, and 1982 (two publications). Accordingly, some of the information is much older than indicated by the 1983 base year of the Bider et al. study.

More recent data was compiled by Temple, Barker, & Sloane (TBS) for the U.S. Environmental Protection Agency. This data indicates that the total crankcase used oil generated for 1988 was 847,000,000 gallons (Bider estimate for 1983 was 699,000,000 gallons). The TBS estimate<sup>46</sup> was based in part on a used oil generation factor of 60.4% which was slightly higher than the value used by Bider et al.<sup>45</sup>. Temple, Barker, & Sloane assumed that the 1988 quantity of used crankcase oil generated by do-it-yourselfers (DIY) was essentially unchanged from the 1983 data of Bider et al. The differences between the estimates of crankcase oil by TBS and Bider et al. were entirely due to non-DIY's.

The industrial used oil generation rates estimated by TBS for 1988 were 504,000,000 gallons which is 8.8% higher than the quantity estimated by Bider et al for 1983. TBS assumed an industrial oil generation factor of 49.9% in calculating the used oil quantity. The industrial used oils represented 40% of the total used oil estimates calculated by Bider et al. and 37% of those calculated by TBS.

Entropy has used the TBS data of 847,000,000 gallons of crankcase oil and 504,000,000 gallons of industrial oil per year since these are the most recent estimates available. A total used oil fuel consumption level of 800,000,000 million gallons has been assumed based on these TBS estimates. This is approximately 59% of the total crankcase oil and industrial oil generation rate.

A number of changes since 1988 are probable. The use of used oil for road oiling has been prohibited in response to regulations promulgated by State and local governments. Accordingly, the quantity of used oil entering the used oil management system and available for either re-refining or energy recovery should have increased. TBS estimated that road oiling accounted for 34,000,000 gallons of used oil use in 1983<sup>46</sup>. Furthermore, there have been improvements in do-it-yourself oil disposal that should have increased the quantities of used oil entering the national used oil management. Accordingly, the total oil quantity figures prepared by TBS and used later in this study may be low.

## Compositions and Quantities of Used Oil

The diversity of the data included on the NORA survey responses indicates that the various used oil streams generated by the automotive and industrial sectors of the economy are not entirely blended into a common used oil stream. Many of the marketers and processors predominately receive, process, and sell crankcase oil. A lesser number receive, process, and sell exclusively industrial oil. It is probable that these oil streams remain separate even when they are burned in combustion processes. Accordingly, it is necessary to assume that there are entirely different types of used oil being burned in different types of sources and in different plants in any given type of industrial process category.

### 3.2 COMPOSITIONS OF USED OILS

#### 3.2.1 Oil Supply - Used Oil Management Systems

It is important to examine the oil supply and used oil management systems in order to confirm trends which have been observed in the composition of used oil streams. A general model will be used in evaluating the extent to which the factors affecting used oil composition are known. This general model is shown in Figure 3-1. It oversimplifies the oil supply and used oil management systems. However, it will be useful in evaluating the types of contaminants which end up in used oil burned in combustion processes.

The term "application" used in the general model (Figure 3-1) means the point of use of the oil. Example applications include passenger cars, industrial hydraulic oil, and industrial transformer oil.

Some of the materials which become contaminants in the used oil streams are introduced as additives to the lubricant (stream 2 of Figure 3-1). Also, lesser quantities of these materials enter with base stocks (stream 1) used to formulate the lubricant.

Once the lubricant is used in certain applications, it accumulates wear metals (stream 4).

- Chromium
- Cadmium
- Lead
- Antimony
- Iron
- Tin

# Compositions and Quantities of Used Oil

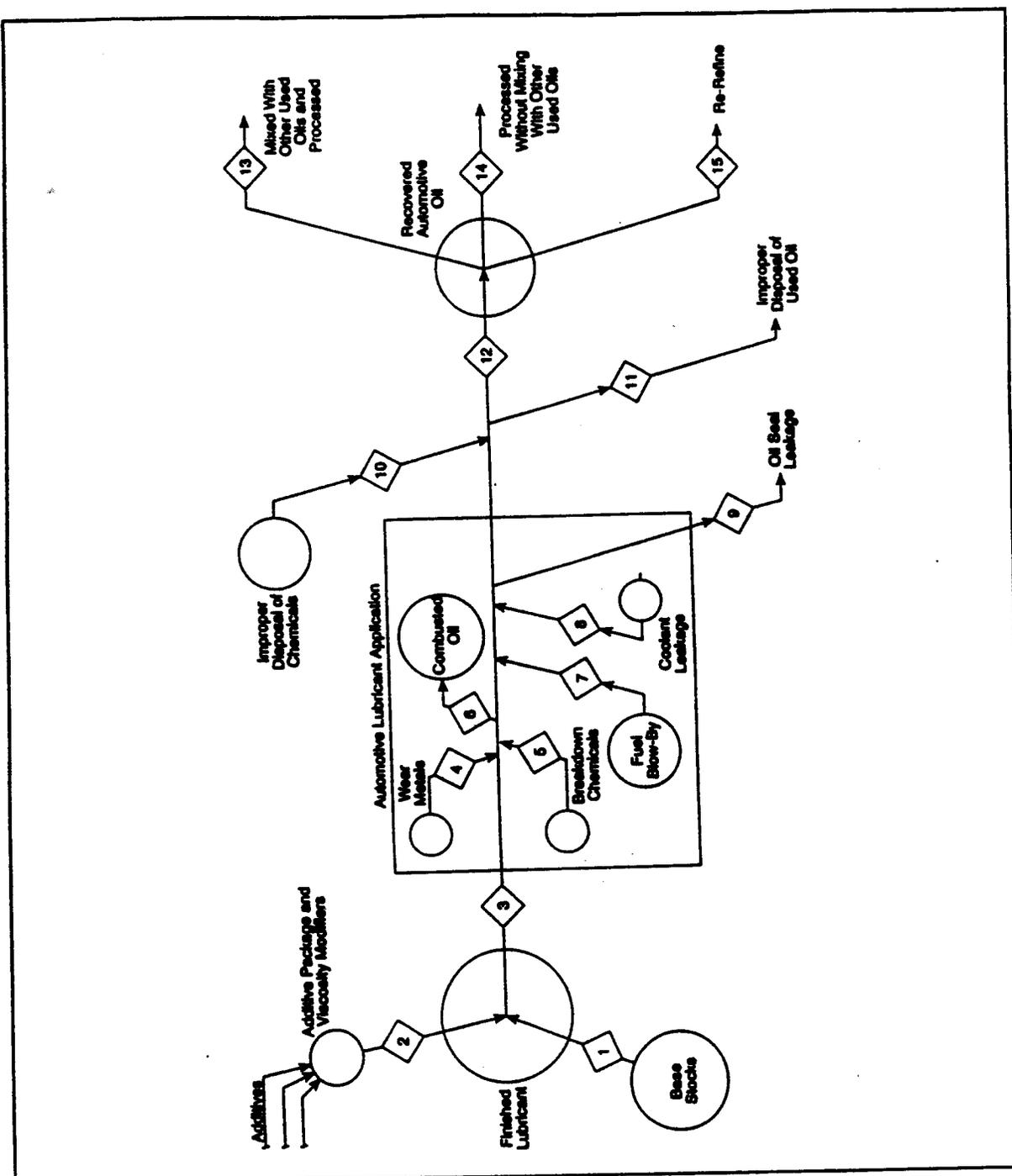


Figure 3-1. Factors affecting used oil composition

## Compositions and Quantities of Used Oil

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It should be noted that most published lists of wear metals have been restricted to chromium, cadmium, and iron. In the Phase I report, lead has been added to this list. This is due to the persistent lead levels in used oils despite the lead-in-gasoline phasedown and since one reference<sup>47</sup> reviewed as part of this study indicated that lead from engine bearings and oil filter components could contribute to used oil lead levels. Antimony and tin have been added to the list for the same reasons as lead.

These metals are important pollutant precursors in the combustion of used oil. The extent of wear metals accumulation depends on the composition of the oil-wetted materials and on the ability of the lubricating oil to maintain necessary properties at the oil drain intervals.

In combustion processes, the lubricant can become contaminated due to the blow-by of pollutants from fuel combustion. This route-of-entry (stream 7 of Figure 3-1) has been important primarily for lead. The lubricant can also be contaminated by leakage of coolant. Materials within the coolant entering the lubricant oil can accelerate rust and corrosion and thereby aggravate the accumulation of wear metals.

One of the roles of the lubricant is to prevent leakage through seals. However, even with the advances in lubricant formulation, it is likely that slight seal leakage is occurring in many applications. This is shown as stream 9 of Figure 3-1.

A portion of the used oil is improperly disposed in landfills and in open dumping. This is shown as stream 11 of the model. This is most common for automobile crankcase oil.

The remaining used oil stream from the application is collected, processed, and re-used. A portion of the oil from the application is collected and either used for fuel or re-refined. The oil used for fuel (stream 14) is often treated to remove water and sludge prior to combustion. The re-refined oil (stream 15) is extensively processed to generate an oil stream which meets the specifications of new oil in one or more lubricant applications. Some of the oil generated by a given application enters the general used oil management system (stream 13) where it is combined with used oil from many different types of applications. In these cases, the unique chemical composition of the used oil stream is blurred due to the mixing of used oil streams from other applications.

### 3.2.2 Crankcase Used Oils

The used oil drained from passenger vehicles and heavy duty vehicles includes the crankcase oil and smaller quantities of other oils such as hydraulic oil (primarily brake fluid), transmission oil, and diesel engine oil. These materials are recovered by service stations, rapid-change services, department stores, and do-it-yourselfers. It is logical to assume that the various types of oil enter the used oil management system together. The general model presented in Figure 3-1 will be used to characterize important used oil characteristics and trends which have a direct impact on the environmental consequences of used oil combustion.

#### Lubricant Formulations

Lubricants are composed of one or more base stock oils and a number of additives. These additives generally comprise 10 to 20% by weight of the finished lubricant<sup>48,49</sup>.

The overall purpose of crankcase lubricants is to improve engine durability, enhance fuel economy, and reduce environmental emissions. The main lubricant material is the base oil itself. However, there are considerable variations in the base oil composition and characteristics depending on the source of the oil. None of the base oils are sufficient without additives for modern passenger cars or heavy duty diesel engines. As engine technology has advanced and environmental requirements have tightened, there has been a steady advancement in lubricant formulation. This is indicated by the chronology of lubricant milestones shown in Figure 3-2.

There are several major categories of additives used in formulating an engine lubricating oil.

- Detergents
- Dispersants
- Inhibitors
  - Anti-wear additives
  - Anti-forming additives
  - Anti-oxidation additives
  - Friction modifiers
  - Pour point depressants
- Viscosity modifiers

## Compositions and Quantities of Used Oil

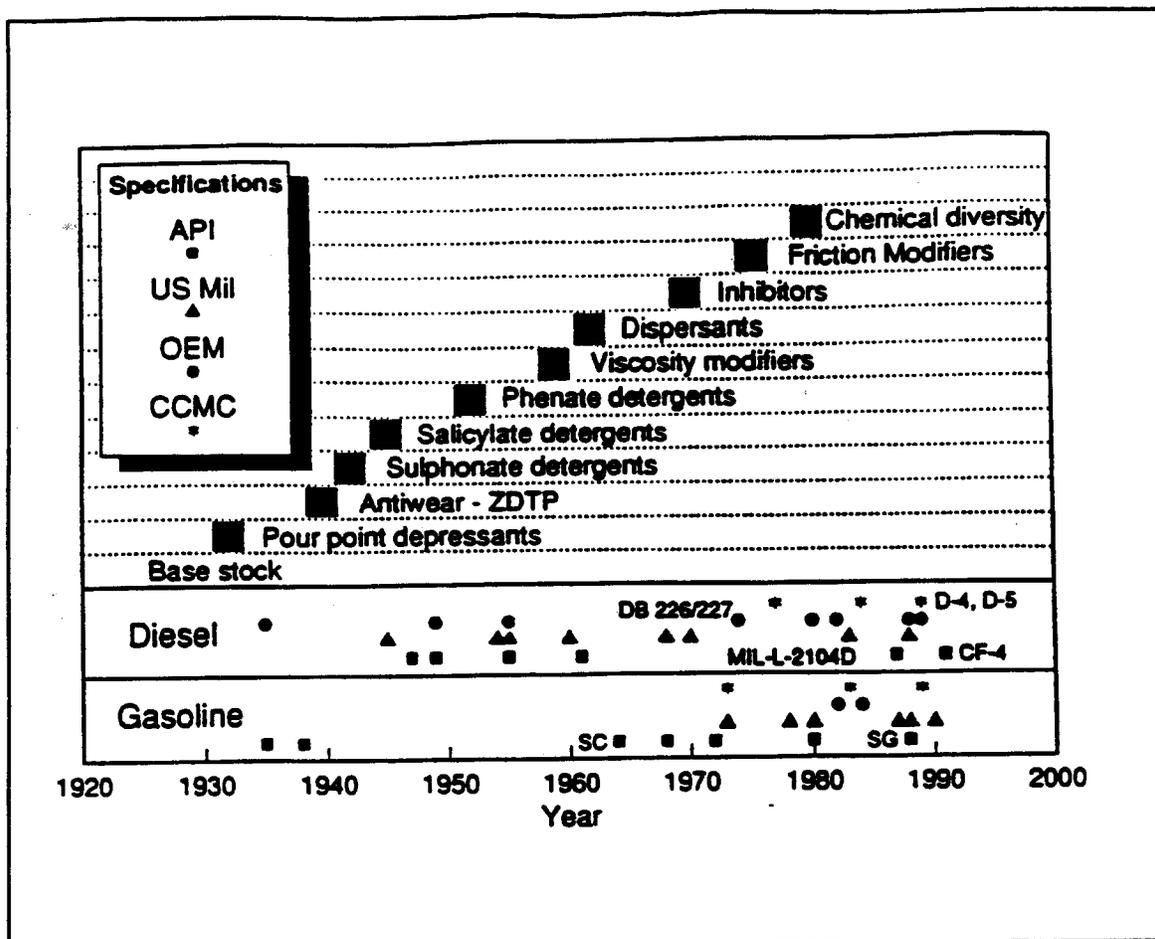


Figure 3-2, Development of Lubricant Additives and the Introduction of Specifications<sup>19</sup>

Detergents are oil soluble compounds which reduce or remove engine deposits. The compounds which could begin to accumulate on metal surfaces have a greater affinity for the detergent molecule than the base oil molecules<sup>19</sup>. Therefore, they are kept in solution and do not begin to accumulate on metal surfaces. Detergent molecules can retard the small (< 20 microns) particle agglomeration by forming a film around the particle<sup>19</sup>. Detergents also provide anticorrosion and antirust protection<sup>19</sup>.

Detergents are composed of a high molecular weight hydrocarbon tail to ensure solubility in the base oil and a polar head with a

## Compositions and Quantities of Used Oil

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metal cation. The most common metal cations are calcium, magnesium, and sodium<sup>9</sup>. Barium and other heavy metals are no longer used<sup>9</sup>.

Dispersants are chemically similar to detergents. However, they do not have metal cations<sup>9</sup>. They adsorb on the surfaces of particles to keep them in suspension and, thereby, reduce engine deposits. They are much more capable of controlling particles than detergent molecules.

Inhibitors are used to reduce engine wear, corrosion, rust, and oxidation. They interrupt free radical oxidation chain reactions and form protective films on metal surfaces. One of the main inhibitors is zinc dithiophosphate (ZDTP)<sup>9</sup>. Other types of chemicals used as inhibitors include sterically hindered phenols for free radical oxidation reaction control, phosphorus compounds for friction reduction, silicon compounds for antiform protection, and sulfurized fatty acids for friction reduction<sup>9</sup>.

Viscosity modifiers are used to produce multigrade oils which perform properly over a wide temperature range. High molecular weight polymers are used for this purpose<sup>9</sup>. The most common types are polyhydrocarbons and polymethacrylates<sup>9</sup>.

As indicated in Figure 3-3, the various additives are usually pre-blended into a performance additive package which is then combined with viscosity modifiers and base oils by the lubricant producer.

There can be between five and fifteen separate compounds comprising the performance additive package. The resulting motor oil composition is shown in Table 3-3<sup>9</sup>.

Table 3-3. Motor Oil Composition<sup>9</sup>

|                        | Passenger Car<br>Motor Oil | Heavy Duty<br>Diesel Oil |
|------------------------|----------------------------|--------------------------|
| Base Stock             | 84.8                       | 85.2%                    |
| Performance<br>Package | 9.2                        | 10.8                     |
| Viscosity<br>Modifier  | 6.0                        | 4.0                      |

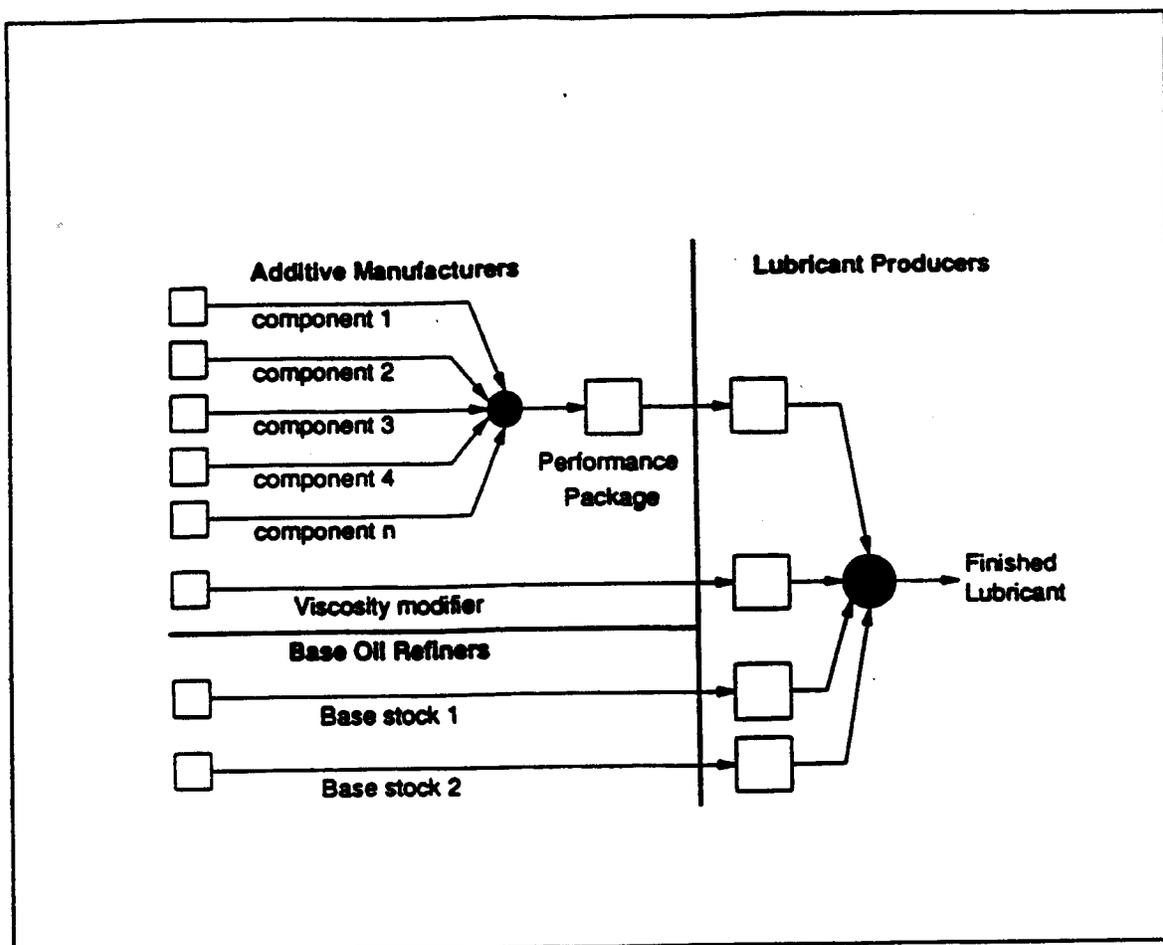


Figure 3-3. Component and Package Formulation<sup>49</sup>

One of the most important conclusions drawn from the review of the lubricant formation data is that barium and other types of heavy metals are no longer used for passenger car lubricating oils<sup>49</sup>. This was also indicated in data gathered by a major lubricant manufacturer and reported to the API as part of this study<sup>50</sup>. It is further confirmed by the very low barium levels reported by numerous respondents to the NORA survey which has been summarized by Entropy. The barium levels in the used oil were less than 10 ppm. These low barium levels are probably due to small quantities of barium in the base stock oil used in making the lubricant oil.

## Compositions and Quantities of Used Oil

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The zinc levels in the final lubricating oil average 0.12 to 0.13% by weight (1200 to 1300 ppm by weight)<sup>50,51</sup>. These levels appear to be relatively similar to those detected in the past.

### Base Oil Stocks

As indicated in Table 3-3, the base oil comprises a large fraction of the total passenger car motor oil. Accordingly, the concentration of metals and other pollutant precursors is important for these materials. No data was found which directly relates to the metals content in these base stocks. Accordingly, they will be evaluated qualitatively by assuming that they are similar in composition to a distillate oil. Data for distillate oil are presented in Table 3-4.

Table 3-4. Average Concentrations of Contaminants in Distillate Oil<sup>52</sup>

| Component | Concentration (ppm) |
|-----------|---------------------|
| Ash       | 25                  |
| Chlorine  | 100                 |
| Sulfur    | 2400                |
| Barium    | 0.5                 |
| Cadmium   | 1.3                 |
| Chromium  | 1.3                 |
| Lead      | 1.8                 |
| Zinc      | 3.6                 |
| Arsenic   | 0.8                 |

It is apparent that there are slight quantities of lead and barium entering the lubricant with the base stocks if these are similar to distillate oil. Furthermore, cadmium and chromium in the base oil stock could contribute to the 1 to 10 ppm levels observed in automotive used oil.

### Fuel Combustion Product Blow-by

It is generally believed that the piston blow-by of lead compounds from gasoline were primarily responsible for the high lead levels in used oils drained from passenger vehicles. The lead-in-gasoline phase down requirements mandated by Federal regulations promulgated on March 7, 1985 have had a direct and

## Compositions and Quantities of Used Oil

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significant impact on the quantities of lead which can penetrate to the used oil stream. The required reductions in average gasoline lead levels are summarized in Table 3-5.

Table 3-5. Lead Phase-Down Requirements

|               | Lead-in-Gasoline<br>grams/gallon |
|---------------|----------------------------------|
| Pre-July 1985 | 1.1                              |
| July 1985     | 0.5                              |
| January 1986  | 0.1                              |

Since 1986, the maximum limit has been less than 10% of the pre-July 1985 value. Furthermore, lead will be prohibited entirely as of 1995.

If this were the only source of lead, the average lead content of passenger vehicle used oil should have decreased approximately 91% since 1985. In fact, the reduction has been slightly less than this value based on the data obtained in the NORA survey, follow-up contacts with re-refiners and marketeers, and data in the published literature. This is summarized in a later section.

### Wear Metals

During use, the lubricating oil accumulates small quantities of metals which are removed due to erosion or corrosion. The types of metals which can be found depend on the composition of the engine parts. The most common wear metals include iron, chromium, and cadmium<sup>45,51</sup>. Other wear metals may include lead, antimony, and tin.

Iron, chromium, and cadmium are typical constituents of the engine piston and crankcase parts. Antimony, tin, and lead are probably due primarily to bearings and filter parts. For example, terne-plated filters can contain all three of these metals. Wilson reports that some main bearings can have lead contents in the range of 21 to 27%, while other bearings can have lead contents in the range of 0.2 to 11%<sup>53</sup>.

Due to the presence of lead and other contaminants in the wear materials, the phase-down of lead in gasoline will not completely eliminate lead in the used oil stream.

### Lubricating Oil Leakage

Lubricating oil that is lost due to leakage from seals is lost to the entire used oil management system. Due in part to the improvements in lubricating oil seal additives, the fraction of oil lost in this manner is approximately 0.5% of the total according to Capon and Haycock<sup>49</sup>.

### Crankcase Used Oil Characteristics

The lead concentration of used oil has decreased substantially during the last 10 years. This is indicated by the relatively low levels of 20 to 90 ppm reported in essentially all of the survey forms returned by respondents to the NORA survey. For example, the average lead concentration of used oil received by Evergreen Oil was 64.14 ppm for the September 1992 to September 1993 time period<sup>54</sup>.

The trend in the lead content of the used oil during the last 10 years is shown in Figure 3-4. This figure displays the tested lead data from a variety of references<sup>45,55-62</sup> over this time period. The high levels prior to 1986 were due primarily to the blow-by lead-containing gasoline. The present levels are probably due primarily to metal wear and slight quantities of lead-containing used oil remaining in tanks and other inventories.

The barium content has also decreased substantially over the past 10 years. This is due to the replacement of barium as a dispersant in lubricating oils. The decrease is indicated by the data from a variety of studies<sup>45,56-59,61</sup> conducted over the 1980 to 1993 time period as shown in Figure 3-5.

The zinc content has remained relatively constant over the last ten years, probably due to the continued use of this material as a detergent additive to the lubricating oil. The zinc data is compiled in Figure 3-6.

The lead and barium concentrations shown in Figures 3-4 and 3-5 are generally supported by data obtained by SAIC<sup>61</sup> for the U.S. Environmental Protection Agency. SAIC collected a limited number of samples and analyzed for metals in the TCLP filtrate. The results for the six categories which are relevant to crankcase oil are summarized in Table 3-6.

# Compositions and Quantities of Used Oil

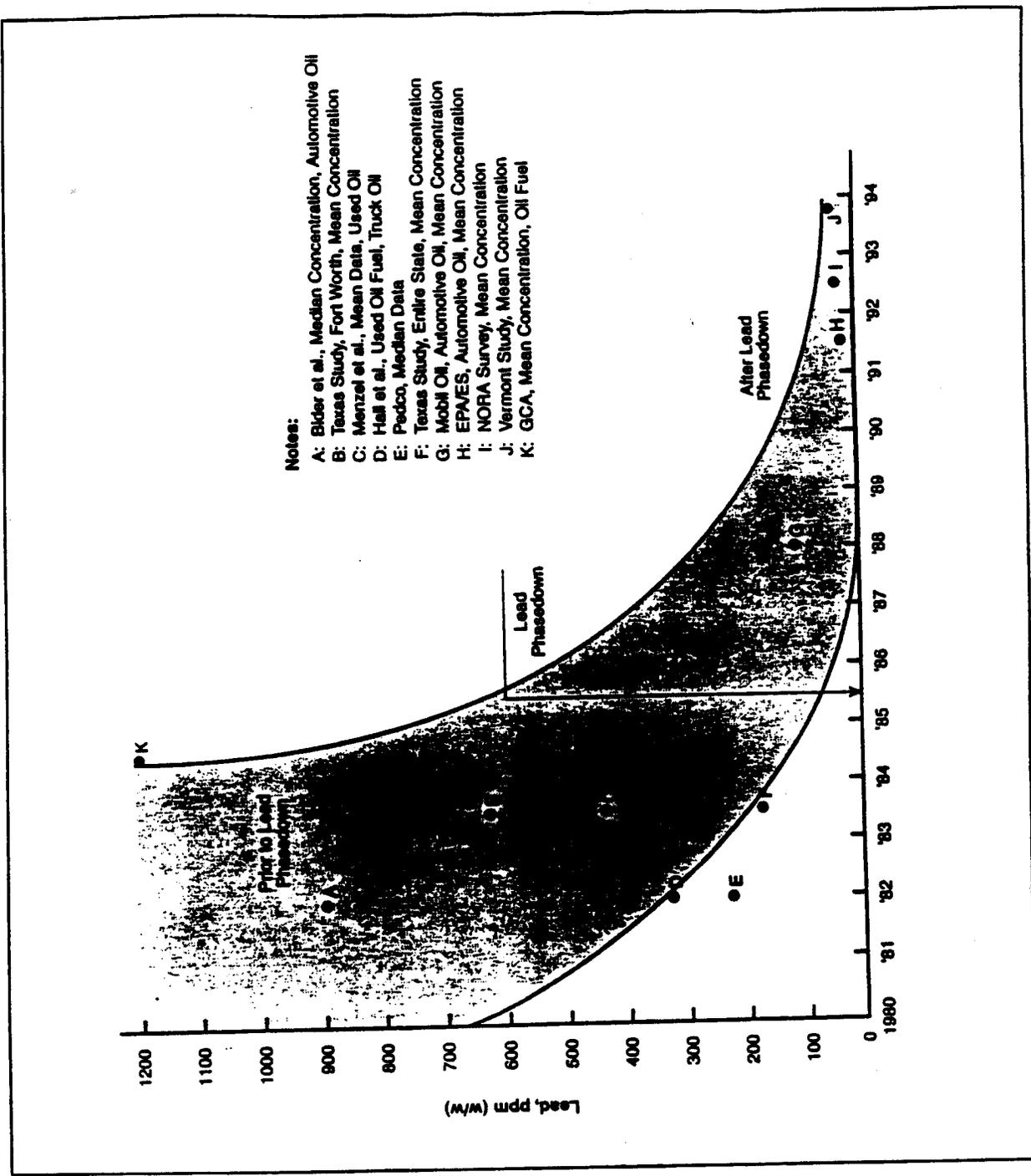


Figure 3-4. Lead Content of Used Oils

Compositions and Quantities of Used Oil

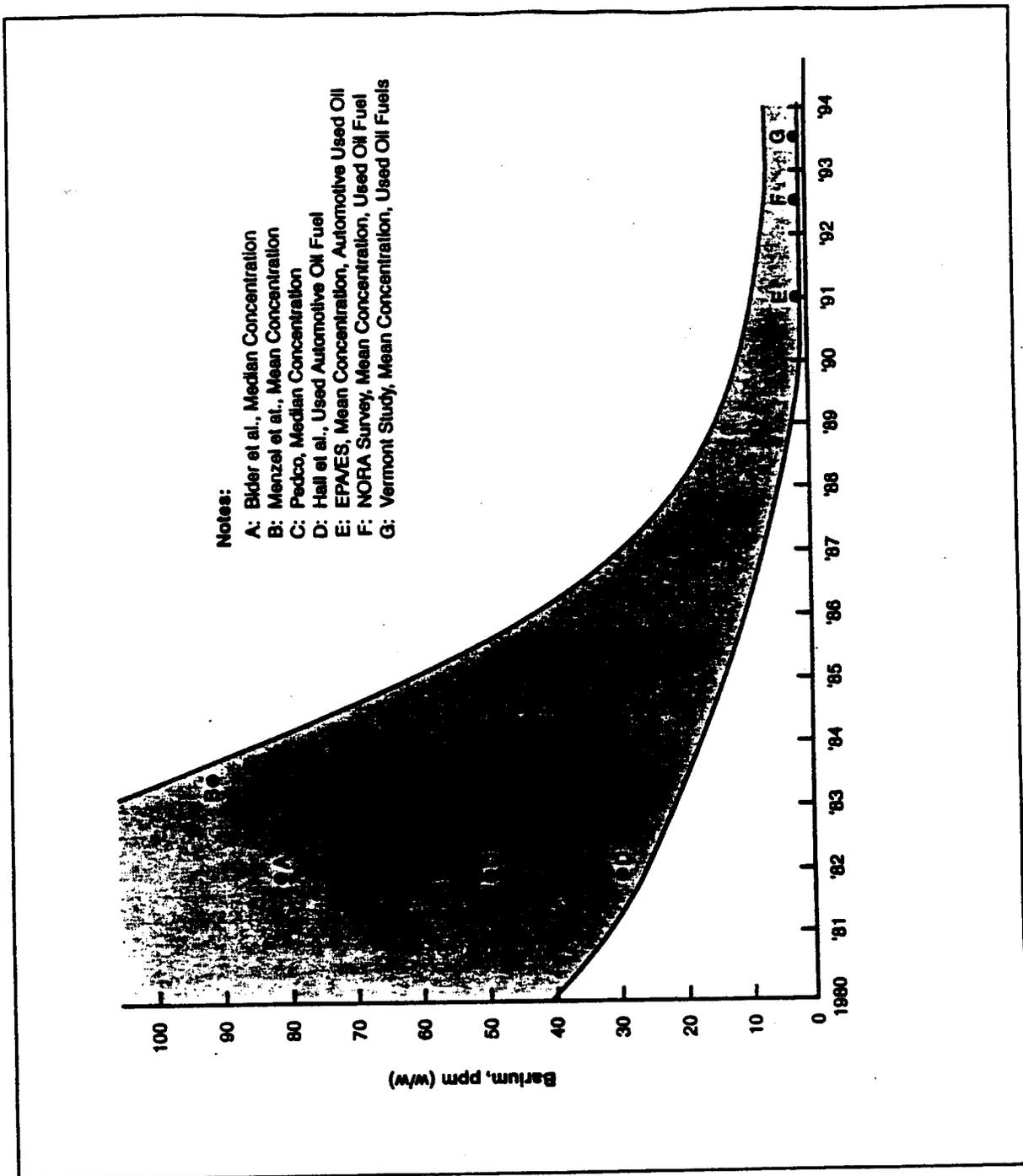


Figure 3-5. Barium Content of Used Oils

Compositions and Quantities of Used Oil

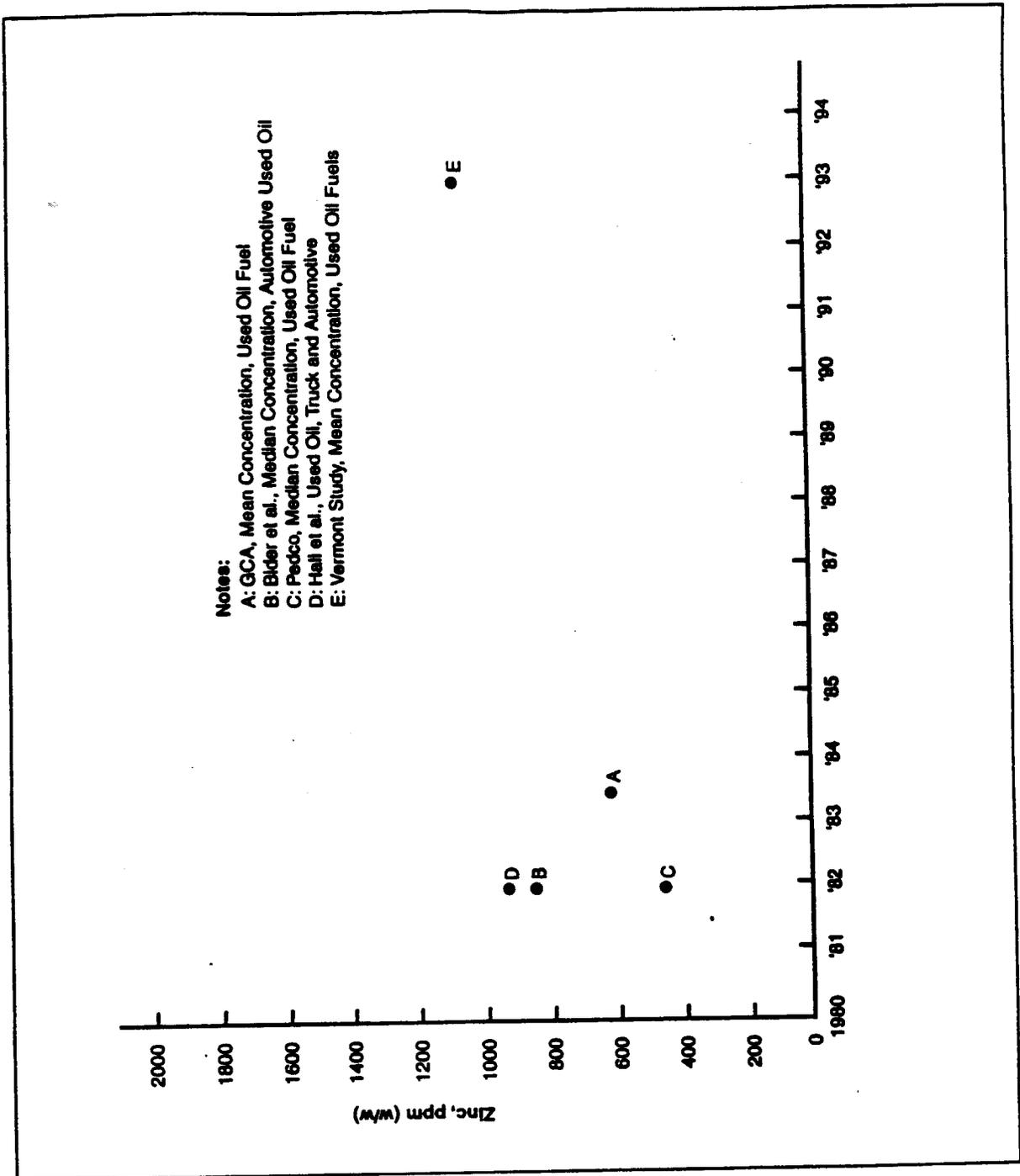


Figure 3-6. Zinc Content of Used Oils

## Compositions and Quantities of Used Oil

SAIC sampled both the vehicles and the used oil storage tanks. The tanks were sampled to determine if the used oil were contaminated during storage.

The data reported in this study only concerned the material that could pass through the filter. The contractor did not attempt to analyze any solids retained on the filters. Eliminating the solids analyses reduced the overall project analytical costs. However, this approach probably reduced the indicated metals concentrations since some of the materials are present as solids in the used oil. Quality assurance checks of twenty-one samples, did, in fact, indicate that some of the solid levels had moderate-to-high levels of barium and lead. Accordingly, some of the data shown in Table 3-6 may indicate lower-than-actual metals concentrations.

Table 3-6. EPA Analysis, 1989<sup>61</sup>

| Source of Samples                     | Metals Concentration in<br>TCLP Filtrate, ppm <sup>1</sup> |      |     |     |      |
|---------------------------------------|--|------|-----|-----|------|
|                                       | As   | Ba   | Cd  | Cr  | Pb   |
| Automotive Crankcase                  | < 1  | < 10 | 1.3 | 5.7 | 33.2 |
| Automotive Crankcase<br>Storage Tanks | < 2.4  | 13.2 | 1.8 | 4.4 | 95.4 |
| Diesel Trucks/Buses                   | 3.0  | 8.1  | 0.5 | 2.6 | 2.3  |
| Diesel Truck/Buses<br>Storage Tanks   | 4.9  | 15.9 | 1.0 | 5.1 | 40.1 |
| Heavy Duty Equipment                  | 2.7  | < 10 | 1.9 | 1.5 | 1.4  |
| Heavy Duty Equipment<br>Storage Tanks | 0.4  | 9.9  | 0.4 | 0.8 | 17.8 |

- Note: Many of the values shown were tabulated from sets of data reported as < minimum detectable. The value of the minimum detectable was used for each of the data points.

The data for the six categories presented in Table 3-6 cannot be averaged to give composite values for used oil generated by the

## Compositions and Quantities of Used Oil

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passenger car, truck and bus, and heavy duty vehicle sectors. The differences in the data for the vehicles and the storage tanks must be reconciled. Also, the data on the quantities of used oil generated in each category would be needed to determine averages. However, the EPA/SAIC data<sup>61</sup>, NORA data, and other supporting data<sup>54,59,63</sup> provide a general indication that the crankcase oils have the following concentrations.

- Lead < 70 ppm
- Barium < 10 ppm
- Arsenic < 5 ppm
- Cadmium < 2 ppm
- Chromium < 10 ppm

### 3.2.3 Industrial Used Oils

The compositions of industrial used oils are substantially different from that of crankcase used oil. The additives used in preparing the lubricating oils are different. Also, there is no fuel-related contamination of the oil streams.

#### Transformer Oils

Transformer oils are used for heat protection. Straight mineral oils with a very high dielectric strength are used. There are no "performance additive packages" as in the case of passenger car lubricants. A transformer oil processing chart is provided in Figure 3-7.

As much as 90% of the total transformer oil is recovered as a used oil<sup>45</sup>. This oil is relatively clean when drained from the transformers since contaminants such as water and high metals levels would have severely reduced the dielectric strength and thereby contributed to short circuits. The transformer oil is replaced when routine tests indicate slight increases in contaminant levels which could lead to transformer failure.

Historically, the main contaminant of environmental concern with transformer oil is PCB. This is due to the use of PCB-containing dielectric fluids during the 1960's and 1970's. However, the presence of PCBs has decreased substantially in used transformer oil due to the prohibition of the replacement of transformer oil with PCB oils in 1979.

Compositions and Quantities of Used Oil

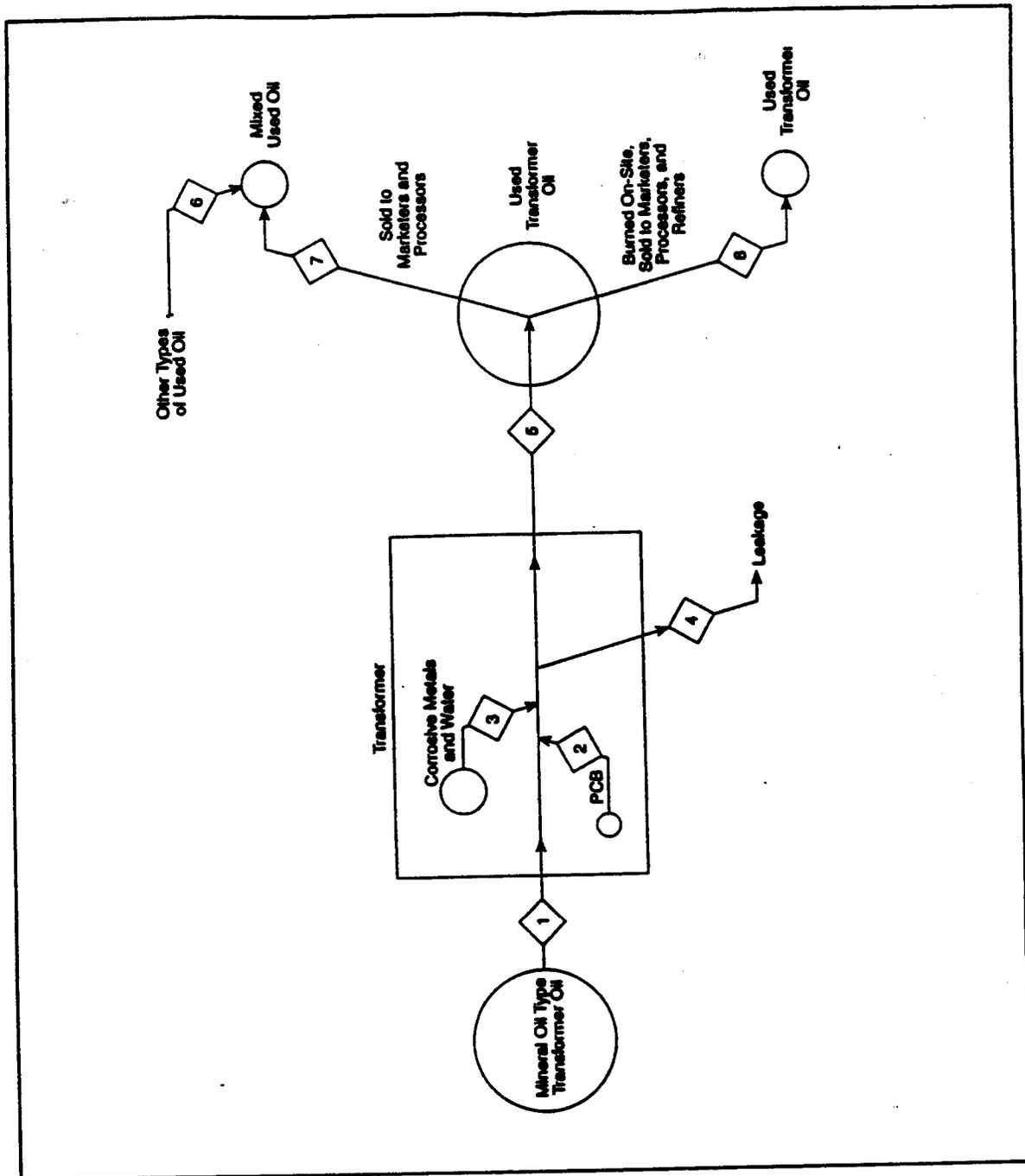


Figure 3-7. Transformer Oil

## Compositions and Quantities of Used Oil

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The slight metal contamination is due to corrosion within the transformer. Water accumulation is due to the condensation of moisture which enters with air infiltrating into the transformer tank.

Overall, transformer oil is very clean. For example, data provided by the EPA/SAIC study and by a utility company<sup>64</sup> is shown in Table 3-7.

Table 3-7. Transformer Oil Composition

|          | EPA/SAIC <sup>61</sup> | Reference 64 |
|----------|------------------------|--------------|
| Arsenic  | < 1 ppm                | < 2 ppm      |
| Cadmium  | < 0.25 ppm             | < 1 ppm      |
| Chromium | < 1 ppm                | < 2 ppm      |
| Lead     | < 1 ppm                | < 3 ppm      |

In most cases, the transformer oil is kept in a separate burn tank and burned by the utility power company operating the transformers. However, in some cases, the transformer oil is mixed with other types of used oil prior to burning. Also, the transformer oil can enter the general used oil management system when it is sold to used oil marketing, processing, and re-refining companies.

Franklin Associates, Inc.<sup>45</sup> estimated that 76 million gallons of transformer oil were used in 1983. There are no more recent estimates available.

### Metalworking and Hydraulic Oils

Metalworking oils are used in a wide variety of cutting, grinding, rolling, and stamping operations in manufacturing facilities. The types of oils include straight mineral oil and emulsified oils. The emulsified oils have high water contents<sup>45</sup>. The total quantity of oil purchased for use as metalworking oil was 163 million gallons in 1983<sup>45</sup>. However, the total quantity of used oil generated could be much larger due to the addition of water to create emulsified oil.

Hydraulic oils are used as the working fluid in hydraulic equipment. The oil must minimize wear of the moving parts and minimize rust. Fire resistant hydraulic oils are being generated

## Compositions and Quantities of Used Oil

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by emulsifying the oil with 60 to 95% water. The total quantity of oil generated in 1983 was 264 million gallons<sup>45</sup>.

There is very little data available concerning the contaminant levels in metalworking and hydraulic oils. The data provided in the EPA/SAIC study is presented in Table 3-8. The metals concentrations are relatively low since metal wear is minimal in these types of oil applications. Also, there are no fuel-related contaminants.

Table 3-8. Metals Concentrations in Hydraulic Oils and Metalworking Oils, ppm<sup>61</sup>

|          | Hydraulic         | Metalworking |
|----------|-------------------|--------------|
| Arsenic  | < 1.0             | 2.1          |
| Barium   | 25.9 <sup>1</sup> | < 10.0       |
| Cadmium  | 1.4               | 0.3          |
| Chromium | 0.9               | 0.7          |
| Lead     | 1.5               | 1.6          |

1. Note: High average value is due to one very high concentration sample.

The majority of these used oils are sold to marketers, processors, and re-refiners. Some of this material is processed separately, rather than entering the mixed used oil system.

### Marine Oils

The primary source of used marine oil is the bilge/ballast water that is pumped ashore when the ship enters port. The used oil is usually processed to reduce water and sludge and then burned on-site for energy recovery. In military bases, the used oil may also include materials generated by aircraft fueling and maintenance. However, low flashpoint jet fuels such as JP-4 are not added to the used oil since this would be unsafe to burn in conventional boilers.

The only available data for marine used oil were included in the EPA/SAIC study. This is summarized in Table 3-9.

Compositions and Quantities of Used Oil

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Table 3-9. Marine Used Oil Composition, ppm<sup>61</sup>

|          | Foreign<br>Cargo<br>Ships | Storage<br>Tanks | Miscellaneous |
|----------|---------------------------|------------------|---------------|
| Arsenic  | < 1.0                     | < 1.0            | < 1.0         |
| Barium   | < 10.0                    | < 10.0           | < 10.0        |
| Cadmium  | < 0.3                     | 0.3              | 0.7           |
| Chromium | 2.4                       | 0.7              | 1.3           |
| Lead     | 7.4                       | 33.1             | 38.6          |

Other Industrial Oils

The other industrial oils include gear lubricants, turbine oils, and compressor oil, railroad diesel engine oil, aviation oils, and natural gas engine oils. There is limited data available for some of these types of used oil. Data from the EPA/SAIC study is presented in Table 3-10. There is no data available concerning any trends in the composition of these oils which would influence the overall contaminant levels in the national used oil management system.

The data presented in Table 3-10 is based on a very few number of samples. Furthermore, only the filtrates of these samples were analyzed. Accordingly, this data may not accurately characterize the composition of these categories of industrial used oils.

Table 3-10. Composition of Other Industrial Used Oils<sup>61</sup>

|          | Railroad<br>Diesel | Natural<br>Gas<br>Compressor | Aviation |
|----------|--------------------|------------------------------|----------|
| Arsenic  | 2.7                | < 1.0                        | < 1.0    |
| Barium   | < 10.0             | < 10.0                       | < 10.0   |
| Cadmium  | 0.3                | 0.3                          | 0.7      |
| Chromium | 2.3                | < 1.0                        | 1.4      |
| Lead     | 2.1                | 2.5                          | 448.1    |

## Compositions and Quantities of Used Oil

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### Summary of Contaminants in Used Industrial Oils

Ten of the 100 respondents to the NORA survey reported receiving between 50% to 100% of their used oil from industrial sources. The mean concentrations of metals were as follows:

|          |      |     |
|----------|------|-----|
| Lead     | 35.8 | ppm |
| Chromium | 16.8 | ppm |
| Cadmium  | 1.83 | ppm |

With the exception of the lead data, these concentrations are relatively similar to the data generated as part of the EPA/SAIC study<sup>61</sup>. The high lead concentrations calculated using the NORA data may be due to the processing of crankcase oils with industrial oils at many of the facilities that submitted NORA forms.

Obviously, more data is necessary to characterize industrial used oil composition at the present time. This data is especially important since industrial oils account for 31 to 40% of the total used oil generated and since it is probable that some of industrial used oils are not mixed with crankcase used oils prior to combustion or re-refining.

### 3.3 SUMMARY - USED OIL COMPOSITION

Based on the presently available information, the data summarized in Table 3-11 will be used to represent the composition of used oils generated by both the automotive and industrial sectors. This data has been calculated using the weighted average concentrations reported on the NORA survey. This data appears to be consistent with the EPA/SAIC data and with the general trends in gasoline and lubricant formulations. The data has been compared with the data of Bider et al. to illustrate general changes in used oil composition since 1983.

## Compositions and Quantities of Used Oil

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Table 3-11. Concentrations of Contaminants in Used Crankcase and Industrial Oils

|          | Mean Concentrations (ppm)                |  |
|----------|--|--|
|          | NORA Survey <sup>1</sup><br>(See Note 1) | Bider et al. <sup>45</sup><br>(See Note 2) |
| Metals   |  |  |
| Arsenic  | 1.19                                     | 19.5                                       |
| Barium   | < 10 <sup>3</sup>                        | 136.5                                      |
| Cadmium  | 1.6                                      | 4.0  |
| Chromium | 8.8                                      | 37.7                                       |
| Lead     | 40.4                                     | 554.7                                      |
| Zinc     | 917.0                                    | 706.9                                      |

- Notes: 1. Weighted average concentrations for data provided concerning 204,058,970 gallons of processed used oil used as fuel, 1993
2. Mean concentrations, used oil burned as fuel Table 46, page 5-5, 1983 base year
3. Based on EPA/SAIC data<sup>61</sup>, samples analyzed in

1989

The low lead values reported in the NORA survey and supporting information are due to a sharp drop in the lead concentration in gasoline and the reduction in the total sales of leaded gasoline. The lower barium concentrations are due to the changes in the additive formations for automotive lubricating oils. Reasons for the apparent decreases in the arsenic, cadmium, and chromium concentrations are unknown. Possible explanations for these the changes shown in Table 3-11 include the following.

- The previous data reported by Bider et al<sup>45</sup>. was inaccurate or nonrepresentative.
- The NORA survey results are not representative of the entire used oil management system.
- Changes in engine design or lubricant formulation have reduced the rates of metal wear.

## SECTION 4

### METAL EMISSIONS FROM USED OIL COMBUSTION

Metals compounds such as lead are not destroyed in combustion processes. They are transformed and partitioned into (1) solids retained in the combustion device, (2) flyash generated during combustion, (3) solid phase product streams (as in asphalt plants and cement kilns), and (4) vapor phase materials which are mixed with the effluent gas stream. Most of the vapor phase material nucleates to form flyash particles while the gas stream cools in heat recovery portions of the boiler. The separation of metals into the different material streams is termed "partitioning." One of the main objectives of this section is to evaluate existing data on metals partitioning during used oil firing.

Once in the particulate form, the metals can be efficiently removed in air pollution control systems. A second objective of this section is to compile data concerning factors that influence the collection efficiency for metal compounds.

Used oil is burned in a variety of combustion sources. For example, the heat input rates for space heaters are less than 0.5 million Btus per hour while the heat input rates for many utility sized oil-fired boilers can be more than 700 million Btus per hour. Many of the combustion variables which potentially affect metals partitioning and particulate removal efficiency vary widely over the different types and sizes of the combustion processes. Important variables include gas stream temperatures, gas stream oxygen concentrations, gas stream residence times in high temperature zones, and entrained solids content in the gas streams. Accordingly, differences in the air emissions from the various combustion processes are expected.

Previous air emissions studies conducted for sources burning used oil and similar fuels have been reviewed as part of the Phase I literature search and evaluation. Conclusions include the following.

- There are very few air emission studies of used oil combustion systems.
- The studies that have been conducted have concerned fuels which are not representative of present-day used oil. Some of the tests were conducted on malfunctioning units or under unusual operating conditions.

## Air Emissions from Used Oil Combustion Processes

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- Metals compounds vaporize initially in the used oil burner flames. The metal vapors may include elemental metals, metal oxides, metal chlorides, and other chemical forms. Each of these materials nucleates once the combustion gas stream cools to the dewpoint temperature for the material.
- The metal compounds condense homogeneously and heterogeneously to form submicron metal-containing particles. The metal compounds may also condense on large size solids being processed in the combustion device or on the heat exchange surfaces in the combustion device.
- Some of the metal-containing particles can build-up on combustion chamber surfaces and/or heat exchange surfaces in the combustion device. Metals retention in used oil combustion systems can range from 20 to 80% of the total metals feed rate.
- Air pollution control devices are capable of removing more than 99% of the metal-containing particles generated in asphalt plants, cement kilns, and other types of industrial furnaces.
- The fraction of low volatility metals such as chromium and barium emitted from combustion systems is lower than the fraction of lead emitted.

### 4.1 GENERAL ASPECTS OF USED OIL COMBUSTION

#### 4.1.1 Regulatory Requirements

The U.S. Environmental Protection Agency promulgated recycled used oil management standards (40 CFR Part 279) on September 10, 1992. As part of this rulemaking, used oil fuel was categorized as "on-specification" or "off-specification" based on the allowable maximum concentrations included in Table 1 of §279.11 (Table 4-1). If the concentration of any constituent exceeds the applicable value in this table, the used oil is categorized as off-specification and, therefore, is subject to all of the requirements of Subpart G (§279.60 through 279.67).

Table 4-1. Used Oil Fuel Specifications (40 CFR Part 279)

| Constituent | Maximum Allowable      |
|-------------|------------------------|
| Arsenic     | 5 ppm                  |
| Cadmium     | 2 ppm                  |
| Chromium    | 10 ppm                 |
| Lead        | 100 ppm                |
| Flash Point | 100 °F                 |
| Halogens    | 4,000 ppm <sup>1</sup> |

1. Note: See §279.10 concerning used oils with halogen levels greater than 1,000 ppm.

Off-specification used oil can only be burned in boilers, industrial furnaces, space heaters, and other combustion processes that satisfy the requirements of §279.12. These combustion processes include but are not limited to the following.

1. Industrial furnaces
  - Cement kilns
  - Lime kilns
  - Aggregate kilns
  - Phosphate kilns
  - Coke ovens
  - Blast furnaces
  - Smelting, melting, and refining furnaces
  - Pulping liquor recovery furnaces
2. Boilers
3. Space heaters
4. Hazardous waste incinerators

To qualify as a boiler, the unit must recover thermal energy from the combustion gases and generate steam, heated fluids, or heated gas. The combustion chamber and the primary energy recovery section must be an integral part of the design. This requirement disqualifies modular type two-chamber incinerators and multi-chamber incinerators. However, the definition includes a wide range of boilers used for space heating (residential, commercial and industrial boilers) and for steam generation (industrial and utility).

Space heaters qualify as long as they are used to burn only the

## Air Emissions from Used Oil Combustion Processes

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used oil generated on-site or received from household do-it-yourselfers. Space heaters must have a maximum heat input capacity less than 500,000 Btus/hour, and the exhaust must be vented directly to the atmosphere.

Only organizations with an EPA identification number can receive and burn off-specification used oil for combustion. Records concerning the customers and quantities of off-specification oil must be maintained by the used oil marketer.

The burning of on-specification used oil is not subject to the provisions of Subpart G. Marketers and processors must analyze the fuel and maintain records for a minimum of three years. There are no restrictions on the type of combustion process that can be used for on-specification used oil combustion.

### 4.1.2 Types of Combustion Processes

All of the combustion processes addressed in the Phase I project meet the requirements in §279.12. The common design characteristic of all of these processes is that used oil is fired through a burner mounted within a combustion chamber. There are substantial differences in (1) the design of the combustion chamber, (2) the gas temperature-time profiles in the combustion chamber, (3) the gas residence times in the overall system, and (4) the extent of contact with process solids in the combustion zone. The main types of combustion processes are introduced in this section. They are discussed in more detail in section 4.2

#### Space Heaters (Units meeting §297.23)

A single oil fired burner is used in a steel shell combustion chamber. Room air is heated by circulation through an indirect air-gas heat exchanger<sup>65</sup>. The circulating fans are used in a forced draft mode to maintain slightly positive static pressures on the air side of the heat exchanger. This is necessary to ensure that any leakage through cracks in the heat exchanger dilutes the combustion gas rather than allowing combustion gases to fumigate occupied areas. The combustion chamber is operated at a slight negative pressure (draft) of at least 0.02 inches of water. Highly negative pressures are indicative of air infiltration. This can reduce combustion efficiency. Positive pressures in the combustion chamber are unacceptable since the combustion gases could be leaking into occupied areas. The static pressure (draft) in the combustion chamber is controlled by a barometric damper in the stack.

Heat input is limited by the U.S. EPA regulation (§279.23) to a

## Air Emissions from Used Oil Combustion Processes

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rate of 500,000 Btus/hr which is equivalent to approximately 3.5 gallons of used oil per hour. The combustion gases are vented directly to the atmosphere. There are no air pollution control devices.

Space heaters are used to burn used oil generated on-site or collected from household do-it-yourselfers. Usually, the used oil is burned as is, without processing to remove water and sludge and without blending with virgin oil<sup>58</sup>. Used oils contaminated with chlorinated solvents should not be burned since they will cause corrosion. Low flash point wastes, such as gasoline, should also not be used since they could cause explosions<sup>66</sup>.

### Apartment House, Commercial, Hospital, and School Boilers

For simplicity, these units will be collectively termed "residential/commercial" or "commercial" boilers. They are used strictly for space heating. Heat input capacities can be as high as 15,000,000 Btus/hr<sup>58</sup>. There are two types of units commonly used: (1) cast-iron, and (2) firetube boilers. In the cast-iron furnaces, a single oil fired burner is mounted in either a steel shell or a refractory lined combustion chamber. Hot water can be generated by forcing water to pass through a series of metal sections above the combustion chamber. In firetube boilers, an oil burner is mounted in a small combustion chamber near the bottom of the boiler. Hot combustion gases are forced to pass through several banks of boiler tubes immersed in boiler feedwater. Steam is accumulated at the top of the firetube boiler. Neither type of residential/commercial boiler is usually equipped with air pollution control systems.

### Industrial Boilers

These provide steam for space heating or for process operations. One or more oil burners are mounted in water tube lined refractory combustion chambers. Most small units do not have air pollution control systems. Most moderate-to-large installations have low efficiency multi-cyclone collectors. Heat input capacities range from 10,000,000 to more than 400,000,000 Btus/hr. All new or modified units with a capacity greater than 10,000,000 Btus/hr (approximately 70 gallons of oil per hour) are subject to 40 CFR Part 60, Subpart Dc if they commenced construction or were modified after June 9, 1989. Subpart Dc limits sulfur dioxide emissions (due to sulfur in the fuel) but has no substantial limits on particulate emissions when burning oil. New or modified units greater than 100,000,000 Btus/hr. are subject to 40 CFR Part 60, Subpart Db or Da if they commenced

## Air Emissions from Used Oil Combustion Processes

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construction or were modified after June 19, 1984 (Db) or September 19, 1978 (Da). Both regulations have strict particulate emission regulations that are applicable to oil-fired units. Essentially all of the units are subject to State and local regulations included in State Implementation Plans (SIP).

### Marine Boilers

These are conceptually similar to small-to-moderately sized industrial boilers. They are designed for more rigorous conditions and are more compact. However, oil combustion and pollution generation are relatively similar to other water tube type boilers such as those used on moderate to large industrial boilers. These boilers do not have air pollution control systems, and they are not subject to the air pollution control regulations for stationary sources.

### Utility Boilers

These are large facilities designed to produce steam for electrical generation and/or space heating. A large number of burners are mounted in a water tube lined refractory combustion chamber. A series of heat recovery systems is used to maximize thermal efficiency of the unit. Air emissions from utility boilers are strictly regulated in accordance with the applicable SIP and/or NSPS regulation. All new or modified utility boilers are subject to the NSPS (40 CFR Part 60, Subpart Da) if they commenced construction or were modified after September 19, 1978.

### Asphalt Plants

Used oil is burned in asphalt plants to heat and dry the aggregate prior to the addition of asphaltic binder. A single oil burner is mounted in a short rotary drier. The aggregate passes through the drier and is showered across the path of the hot combustion gas using a large number of flights (scoops) mounted on the interior surface of the drier. The heat input capacities of the burners are approximately the same as small industrial boilers. Most asphalt plants are equipped with high efficiency fabric filters or venturi scrubbers for particulate control. Some have low efficiency particulate scrubbers. Particulate emissions from asphalt plants are subject to the regulations included in the SIP and the NSPS (40 CFR Part 60, Subpart I). The NSPS for asphalt plants applies to all units

constructed or modified after June 11, 1973. Accordingly, it applies to essentially all operating units.

### Cement and Lime Kilns

These are very large refractory lined combustion chambers designed to dry, calcine, and pyroprocess (cement plants) the kiln feed. A single large oil burner is used for heat input. Gas temperatures in the kiln exceed 3000°F. Gas residence times at high temperature are very long. High efficiency particulate control systems are used on cement and lime kilns. Particulate emissions from cement and lime kilns are subject to SIP regulations and NSPS regulations (40 CFR Part 60, Subparts F and HH). The NSPS for cement kilns applies to all units which commenced construction or were modified after August 17, 1971. The effective date for the lime kiln NSPS was May 3, 1977.

### Steel Mill Processes

Used oil can be fired in a variety of oil burners as part of blast furnaces, sintering machines, or basic oxygen furnaces. These are all large processes which operate at high gas temperatures. Emissions from used oil combustion are mixed with metallurgical fume and other process related emissions. These processes are controlled with high efficiency air pollution control systems. All combustion sources and metallurgical processes in steel mills are subject to the applicable SIP regulations. In addition, the basic oxygen furnaces and electric arc furnaces are subject to NSPS regulations (Subparts N, Na, AA, and AAa) promulgated between June 1973 through August 1983.

### Pulp Mill Recovery Boilers

The main purpose of the recovery boiler is to recover the sodium chemicals necessary to digest wood pulp. The lignins and other alkaline-soluble organics separated from the cellulose fibers during digestion are concentrated in the chemical-containing black liquor. These non-cellulose organics provide the main fuel for the recovery boiler. Used oil can be used to supplement the black liquor guns used to operate the boiler. Recovery boilers are equipped with high efficiency electrostatic precipitators. Recovery boilers have heat input rates equivalent to large utility boiler systems. Gas temperatures and gas residence times are relatively long. Pulp Mill Recovery Boilers are subject to 40 CFR Subpart BB which requires high efficiency particulate control, low total reduced sulfur (TRS) emissions, and continuous emission monitoring. Subpart BB applies to recovery boilers that have been constructed or modified since September 24, 1976.

#### 4.1.3 Quantities of Used Oil Being Burned in the Major Categories of Combustion Processes

The NORA survey provides the most up-to-date data concerning the types of combustion processes which fire used oil as fuel. The total quantity of used oil fuel sold by the responding marketers, processors, and blenders was 204,058,970 gallons during the twelve months proceeding the survey. The quantities sold to each of the major categories of combustion processes are based on data included by the 100 respondents to the NORA survey. These data are summarized in Table 4-2.

Asphalt plants were the dominant used oil consumer. They utilize approximately 42.8% of the total used oil sold as fuel. The next largest consumer was industrial boilers at 13.9% of the total. Commercial boilers were not significant based on the quantities of used oil fired.

The used oil fuel sales data in Table 4-2 does not include crankcase and industrial oils that are consumed at the oil point-of-use. These oil streams are not handled by used oil marketers, processors, and blenders. There is no direct data concerning the quantities of point-of-use used oil burned. It is possible that used oil combustion at sources such as car service center space heaters and utility power stations (transformer and turbine oil) are considerably larger than indicated by the NORA survey.

#### 4.1.4 Metal-Containing Particle Formation

Metals enter used oil due to (1) the presence of low levels in the base oil stocks used to produce the oil, (2) additives to lubricating oil, and (3) the erosion and corrosion of metal parts contacted by the oil during use. Some of the metals are present as small particles, and some are present as cations on dissolved organic compounds. During combustion, it is reasonable to assume that all of the metals vaporize in the oil burner flame. Oxidation of the metals to form metal oxides is probable in the burner flames and in the combustion chambers. It is also possible that chloride compounds in the waste influence the volatility of metals such as lead<sup>68,69</sup>.

Table 4-2. Sales of Used Oils for Fuels  
(NORA Survey, 100 Respondents)

| Industrial Category  | Used Oil Combustion |              |
|--|---------------------|--------------|
|  | Gallons/Year        | % of Total   |
| Asphalt Plants   | 87,462,094          | 42.8         |
| Cement/Lime Kilns  | 9,798,500           | 4.8          |
| Utility Boilers  | 25,780,000          | 12.6         |
| Industrial Boilers   | 28,300,085          | 13.9         |
| Commercial Boilers<br>(Apartment House,<br>Hospital, School,<br>and Store) | 351,000             | 0.2          |
| Steel Mills  | 24,115,000          | 11.8         |
| Pulp & Paper Mills   | 7,921,500           | 3.9          |
| Marine Boilers   | 10,605,250          | 5.2          |
| Other  | 9,725,340           | 4.8          |
| <b>TOTAL</b>   | <b>204,058,970</b>  | <b>100.0</b> |

The resulting elemental metals and metal compounds will convert from the vapor phase to the solid phase as the combustion gas stream cools during recovery of thermal energy<sup>70-74</sup>. The temperature at which the various metal species convert from the vapor state depends on their vapor pressure. A vapor pressure curve for a variety of metal species of interest in used oil combustion is provided in Figure 4-1. Each line on this figure presents the vapor pressure data for a single metal compound or elemental metal. As the temperature decreases, the concentration of the species that can remain as a vapor in the gas stream decreases.

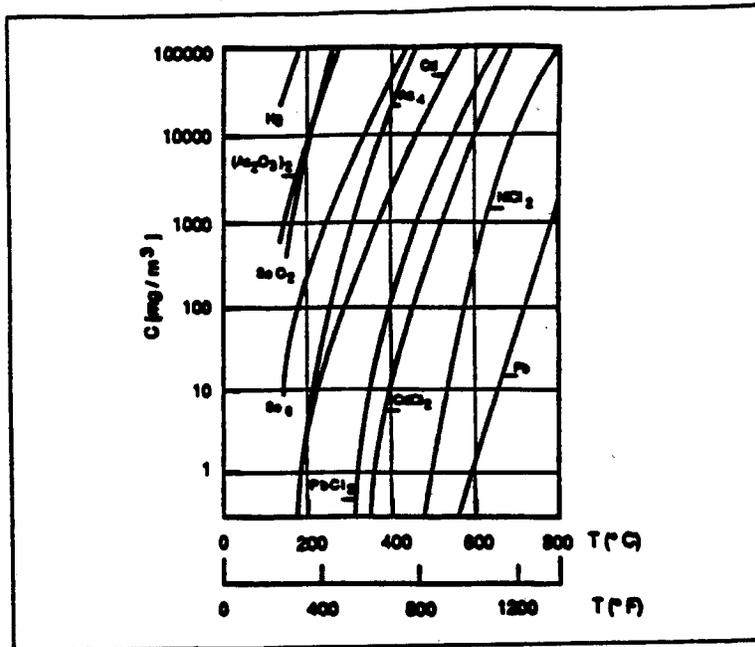


Figure 4-1. Vapor Pressures of Various Metal Species of Importance in Used Oil Combustion<sup>67</sup>

The metal species should nucleate one-by-one as they approach the temperature at which their concentration in the vapor state in the gas stream exceeds the equilibrium vapor pressure indicated by the vapor pressure - temperature curve. The vapor phase metal species can condense in three interrelated ways.

- Condensation on the surfaces of existing particles (termed heterogeneous nucleation)
- Condensation as particles composed almost entirely of the vapor phase material (termed homogeneous nucleation)
- Condensation on the surfaces of particles deposited on the surfaces of the combustion chamber and heat recovery equipment

These mechanisms are related since the particles formed by homogeneous nucleation can become sites for heterogeneous nucleation. Particles formed by both nucleation mechanisms can be deposited on the surfaces of the combustion system due to inertial impaction and Brownian displacement.

## Air Emissions from Used Oil Combustion Processes

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These mechanisms are relevant to this study primarily because they yield particles which are usually very small. Particles generated by homogeneous nucleation are often in the range of 0.05 to 0.2 microns. Heterogeneous nucleation occurs primarily on the surfaces of particles in the 0.1 to 0.5 micron range. Particles generated by other mechanisms are usually in a much larger size range of 0.5 to > 20 microns.

The particles created by heterogeneous and homogeneous nucleation are predominately in the size range of 0.05 to 0.5 microns. This is the particle size range in which some types of air pollution control systems are least efficient. For example, venturi scrubbers (used on asphalt plants) have a minimum efficiency between 0.1 and 0.3 microns. Electrostatic precipitators (used on cement kilns, lime kilns, recovery boilers, and some oil-fired utility boilers) have a minimum efficiency between 0.1 and 0.5 microns.

The overall process of vaporization of metal species in high temperature zones, followed by nucleation onto the surfaces of very small particles as the gas stream cools, shifts the metals to the small, difficult-to-control size range. Small particles have high concentrations of volatile metal species while large particles have relatively low concentrations. This process is termed "enrichment" of the small particle size range.

Conversely, metal species and inorganic materials which are not very volatile tend to accumulate primarily in the large size ranges.

The various metal species of interest in used oil combustion processes have been grouped into three categories with respect to their volatility (Table 4-3). The metal species in the highly volatile group are the most difficult to collect in air pollution control systems.

Table 4-3. Volatility categories<sup>73</sup>

High Volatility

Mercury

Moderately Volatility

- Arsenic trioxide
- Arsenic
- Cadmium
- Cadmium oxide
- Cadmium chloride
- Lead chloride
- Lead oxide
- Lead
- Zinc
- Zinc chloride
- Nickel chloride

Low Volatility

- Barium
- Chromium
- Nickel
- Tin

The categories of volatility provide a general indicator of the behavior of elemental metals and metal compounds in combustion processes. A highly simplified drawing of a combustion process is shown in Figure 4-2 to illustrate how metal species entering with the used oil fuel are partitioned into various gas and solid streams. Highly volatile materials such as mercury will stay predominately in the vapor phase while passing through the air pollution control systems. Accordingly, the collection efficiency will be low (stream 5 slightly smaller than stream 4). Most of the materials in the moderate volatility category will nucleate to form particles in the submicron size range while the gas stream passes through the heat recovery portion of the combustion system. These particles will be captured with medium-to-high efficiency by control systems such as fabric filters, electrostatic precipitators, and venturi scrubbers. Accordingly, stream 8 should be substantially less than stream 7. Furthermore some of the moderately volatile species will be captured by the process solids (cement kilns, asphalt plant driers) and exit the combustion system in stream 13.

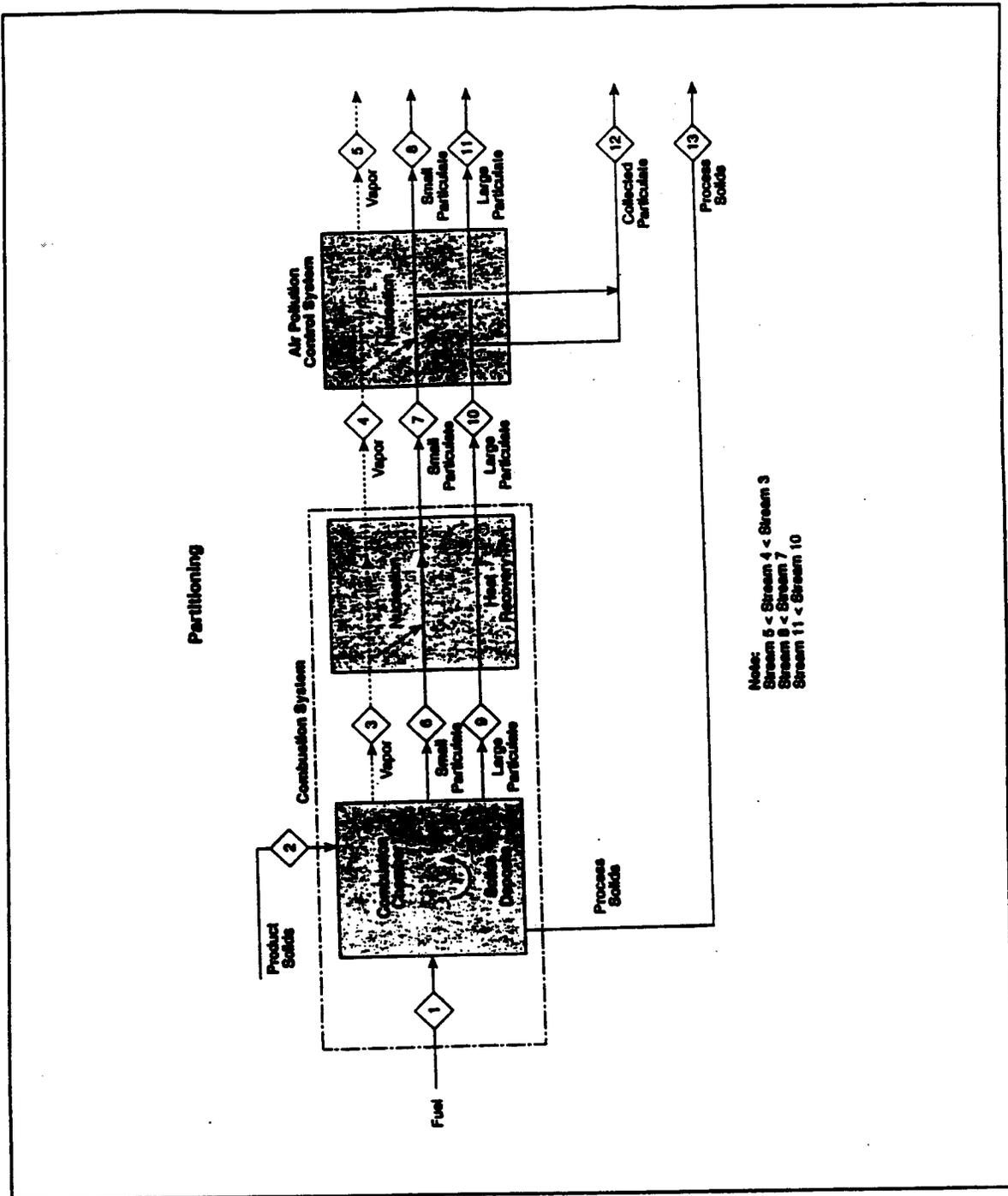


Figure 4-2. Partitioning of Metal Species

The low volatility species will vaporize incompletely. The vapors of these species will quickly nucleate on the surfaces of large diameter flyash and process solids in the combustion chamber. The fraction of the material which does not vaporize forms predominately large sized particles. Accordingly, most of these low volatility materials become part of the large diameter particulate (stream 9) and the process solids (stream 13). However, some small sized particulate can also form due to homogeneous and heterogeneous nucleation in the combustion chamber and due to oil droplet vaporization.

#### 4.2 SPACE HEATERS

Space heaters are the simplest type of combustion system. A single oil burner is used to atomize and burn the used oil in a small combustion chamber. All of the major domestic manufactures now use low pressure air-low pressure oil atomizing systems. This design type accounts for approximately 6500 units sold in the U.S. each year. There is a least one manufacturer who is still making vaporizing pot units, but it is believed that the total production rate is less than 100 units per year<sup>75</sup>.

Prior to 1983, the space heater market was dominated by vaporizing pot units. These were produced primarily by Kroll Manufacturing and Lanair. Both manufacturers converted over to air atomizing designs from 1982 to 1983 because of the following<sup>75</sup>.

- National Fire Protection Association (NFPA) restrictions concerning floor mounted heating units in car service centers
- NFPA concerns about manual lighting of the burners
- High maintenance requirements involved in cleaning sludge from vaporizing pots on a daily basis

Due to the normal service life of these units and NFPA restrictions, most of the vaporizing pot type space heaters are probably no longer in service. They have been replaced with oil atomizing type designs. All of these units have a combustion gas-to-room air indirect heat exchanger and an air circulation fan to recover the heat. Space heaters burning used oil must have a heat input rating of less than 0.5 million Btus per hour due to U.S. EPA regulations (40 CFR Part 260, §279.23).

Entropy has obtained three air emissions studies concerning space heaters. All of these were conducted in the 1982 to 1983 time frame prior to the introduction of present-day space heaters. Therefore, most of this data is no longer relevant. Furthermore, used oil composition has changed significantly since 1983 due to the lead-in-gasoline phasedown and due to advances in lubricant additive formulation.

Due to these changes, these early studies have not been summarized completely. Only the information which is relevant to present day space heaters is discussed in the following sections.

#### 4.2.1 EPA/Battelle Columbus Laboratories Study<sup>57,76</sup>

This test program involved a single atomizing burner design (Dravo Hastings Thermoflo, Model 20-WO) and a vaporizing pot space heater (Kroll Model W400L). The vaporizing pot unit was discontinued by the manufacturer shortly before these tests were conducted. The atomizing burner unit used low pressure atomizing air and was similar to presently available designs.

Air emissions tests were conducted using used automobile crankcase oil and used diesel truck crankcase oil. The concentrations of metal contaminants in these used oil fuels are summarized in Table 4-4.

The oils used in this study had lead levels which are 7 to 80 times higher than present lead concentrations in used oil. The barium concentrations in this old study also were well above present barium concentrations.

Mass balances for various metals were determined by comparing metals emission rates and used oil metal feed rates. Metals emissions rates were calculated from the gas flow rates, and the metals concentrations data were determined by two separate techniques: the SASS train and a dilution tunnel. The metal feed rates were calculated using the used oil firing rate and the metals concentrations.

The mass balance results indicated that metals emissions were less than input rates in most cases. For example, the zinc emission rates during tests of both types of used oil were approximately one-half of the total zinc input rates. These results suggest that a fraction of each of the metals is retained as solids deposits within the combustion chamber and on the heat exchange surfaces.

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Table 4-4. Used Oil Fuel Analyses and Mass Balances for Metals, ug of Metal/Gram of Oil

|    | Automobile Crankcase Oil |                                 |            | Truck Crankcase Oil |                                 |            |
|----|--------------------------|---------------------------------|------------|---------------------|---------------------------------|------------|
|    | Used Oil<br>ppm          | Percentage Emitted <sup>1</sup> |            | Used Oil<br>ppm     | Percentage Emitted <sup>1</sup> |            |
|    |                          | SASS Train                      | Dil. Train |                     | SASS Train                      | Dil. Train |
| Pb | 3320                     | 26.6                            | 15.4       | 290                 | 93.8                            | 38.6       |
| Cd | 2                        | 50                              | 25         | 1                   | 100                             | 30         |
| Cr | 11                       | 272 <sup>2</sup>                | 18.2       | 3                   | 33                              | 13         |
| As | N.D.                     | N.D.                            | N.D.       | N.D.                | N.D.                            | N.D.       |
| Zn | 1130                     | 35.9                            | 24.1       | 940                 | 58.9                            | 22.4       |
| Ba | 29                       | 27.6                            | 24.1       | 48                  | 0.104                           | 16.7       |

- Notes:
1. Calculated from data presented on page B-7 of reference 76
  2. Indicates chromium contamination of samples from sampling equipment or analytical problems

The lead emissions during the automobile used oil test had an especially large retention value. The emission percentage was 26.6% which is equivalent to a retention factor of 73.4%. It is possible that analytical errors involving the automobile used oil lead concentrations or the particulate emissions lead concentrations were in error.

Qualitative estimates of lead retention inside air atomizing space heaters are in the range of 20 to 50% of the lead<sup>77</sup>.

Despite the apparently questionable results, the study provides indications that a fraction of the lead and other metals in used oil is retained in the space heater. According to one of the manufacturers, it is necessary to remove these deposits several times a year in order to maintain proper heat exchange efficiency<sup>77</sup>.

The chromium data shown in Table 4-4 indicates that the stainless steel components in the SASS train can create artificially high results. Similar problems were observed for nickel which is another major alloying element in stainless steel. These

problems were eliminated in the EPA multi-metals sampling train developed by the U.S. EPA and Entropy several years after this study was conducted by Battelle Columbus.

The emissions data for the air atomizing type space heater indicated that the unit was operating well during the test. Table 4-5 shows that the carbon monoxide concentrations ranged from 16.7 to 25 ppm.

Table 4-5. General Test Conditions, Air Atomizing Burner Type Space Heater<sup>57</sup>

| Test Conditions                          | Automobile Crankcase Oil | Diesel Truck Crankcase Oil |
|--|--------------------------|----------------------------|
| Stack Gas Temperature, °F                | 642                      | 630                        |
| Oxygen Concentration, %                  | 4.8                      | 4.8                        |
| Excess Air, %                            | 27.9                     | 28                         |
| Carbon Monoxide Concentration, ppm       | 25                       | 16.7                       |
| Unburned Hydrocarbon Concentration, ppm  | 3.1                      | 2.1                        |
| Particulate Emissions, mg/M <sup>3</sup> | 224.4                    | 223.4                      |

#### 4.2.2 British Columbia Research Corporation Study<sup>78</sup>

Emission tests on 6 used oil-fired space heaters were conducted by the British Columbia Research Corporation for the Government of the Northwest Territories (Canada). Five Lanair type vaporizing pot heaters and a low pressure air atomizing type space heater were tested. U.S. EPA Reference Method 5 was used to sample the combustion gases. The filters were digested in nitric acid, filtered, and analyzed by inductively coupled plasma - mass spectroscopy (ICP-MS). Only the test conducted on the air atomizing unit is relevant to the UORC study.

The metals emission rates and the used oil metals concentrations for the air atomizing unit are summarized in Table 4-6. The retention factors for the metals could not be calculated since neither the burner heat input rating nor the oil firing rate were included in the report.

## Air Emissions from Used Oil Combustion Processes

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The overall particulate emission rate for the air atomizing unit was higher than the value determined by Hall et al.<sup>57,76</sup>. The British Columbia Research Corporation determined a particulate emission rate of 590 mg/M<sup>3</sup>. The value measured by Hall et al. was 224 mg/M<sup>3</sup>. The high emission rates in the Canadian tests may be due in part to excessive air levels. The measured oxygen concentration during this test was 11.8%. This is well above the normal range for an oil-fired space heater.

Table 4-6. Metals Emission Data<sup>78</sup>

| Metal    | Concentration in Used Oil mg/L | Emission Rate mg/M <sup>3</sup> |
|----------|--------------------------------|---------------------------------|
| Lead     | 200                            | 2.62                            |
| Chromium | 5.0                            | 0.36                            |
| Cadmium  | 1.6                            | 0.09                            |
| Arsenic  | No Data                        | 0.11                            |
| Barium   | No Data                        | No Data                         |
| Zinc     | 990                            | 3.48                            |

High excess air rates reduce the combustion chamber temperatures and thereby reduce the oxidation rates of organics which in turn can contribute to particulate emissions.

### 4.2.3 Pereira et al.<sup>79</sup> Study of Five Space Heaters in England

In 1983, Pereira et al. conducted a detailed study of space heaters at three vehicle service centers, a medical supply company (small boiler), and a research laboratory. The types of space heaters at each location were not clearly stated. However, based on general comments in the report, it appears that these units were air atomizing systems.

Cascade impactor tests were conducted as part of this program to determine the particle size distribution of lead and other pollutants emissions. The less than 3.5 micron fraction of the particulate matter emitted contained 97% of the total lead emissions. The less than 0.6 micron fraction contained 74% of the total lead emissions. This is consistent with the expected homogeneous and heterogeneous nucleation mechanisms discussed earlier in this section. It is apparent that lead volatilizes

## Air Emissions from Used Oil Combustion Processes

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and nucleates even in the relatively limited temperature and residence time environment of a space heater.

The retention of lead within the combustion chambers was high in all five units tested. As indicated in Table 4-7, the percentages of metals emitted were 18% to 38%.

Table 4-7. Percentages of Lead Emitted<sup>79</sup>

| Test Site | Test No. | Percentage of Lead Emitted |
|-----------|----------|----------------------------|
| 1         | A        | 33                         |
|           | B        | 28                         |
|           | C        | 24                         |
| 2         | D        | 30                         |
|           | E        | 24                         |
| 3         | F        | 18                         |
|           | G        | 24                         |
| 4         | H        | 34                         |
|           | I        | 27                         |
| 5         | J        | 38                         |
|           | K        | 35                         |

Very high lead levels were found in the deposits in the combustion chambers. At test site 1 (laboratory), the lead deposits in the combustion chamber were 32% of the total. The lead levels in the heat exchanger and the exhaust box decreased to 12.7% and 9.8% respectively. High lead levels were also detected in the boiler scrapings from test sites 2 and 3.

The data of Periera et al. is generally consistent with the lead retention results from the study of Hall et al. In both cases, a major fraction of the lead remained in the combustion chamber. This means that air emissions are reduced proportionally.

The total particulate emissions from the five units tested during the Periera et al. study ranged from 250 to 438 mg/M<sup>3</sup> in all but one of the units. These particulate emissions may be due in part to non-optimum combustion conditions since the carbon monoxide concentrations in one of the tests (site 1) were in the range of 300 ppm. Carbon monoxide data was not included for the other test sites.

The lead concentrations in the used oils burned during this test program were very high. The lead concentrations ranged from 1200 ppm to more than 7000 ppm. In contrast, used oil analyses summarized in Section 3 indicate that U.S. used oils are typically in the range of 20 to 60 ppm. Accordingly, the tests conducted by Periera et al.<sup>79</sup> are not representative of the emissions from present-day space heaters in the U.S. firing "typical" used oils. Also, the proportion of the lead that is retained in the unit may be a function of the lead content of the used oil fuel.

#### 4.2.4 Vermont Study

The State of Vermont Department of Environmental Conservation Division in conjunction with the U.S. EPA and private organizations is presently conducting an air emissions study of used oil fired (automotive crankcase oil) space heaters in Vermont<sup>80</sup>. The project has been divided into several phases including the compilation of existing data, collection and analysis of used oils, dispersion modeling of emissions, and testing of emissions. It is anticipated that six space heaters will be tested for selected metals and other pollutant emissions. All of the units will be less than five years old. The test data will probably be available in late Spring, 1994. This study should provide data relevant to present-day space heaters and used oils.

#### 4.2.5 Summary - Space Heater Air Emissions

Most of the previously published data concern vaporizing pot type designs which are no longer sold extensively. Furthermore, all of the published studies involved used oils having lead concentrations ranging from approximately 200 to more than 7000 ppm. These concentrations are not representative of present used oil fuels used in the U.S.

The limited data that is available indicates that only a fraction of the lead in the used oil is emitted to the atmosphere. Between 20% to 80% of the lead may be retained in combustion chambers, heat exchangers, and flues. High lead deposits have been found in all of these areas of space heaters. Considerably more data is needed to determine the typical partitioning of lead between the combustion gas stream and internal deposits. It is especially important to determine the extent to which these deposits can serve as a "sink" or "source" of lead emissions during short term emission tests of space heaters.

The particle size distribution of lead-containing particles generated in space heaters is probably quite small. Most of the

lead emitted to the atmosphere is in the submicron range. More data is necessary to confirm the distribution of lead and other metals as a function of particle size.

#### 4.3 RESIDENTIAL/COMMERCIAL BOILERS

These boilers are used for space heating at apartment houses, businesses, hospitals, and schools. The boilers generate hot water or steam. The heat input capacities range from 0.4 million Btus/hr to 25 million Btus/hr<sup>45</sup>. There are three basic design types.

- Cast-iron
- Firetube
- Water tube

Cast-iron boilers derive their name from the water heating sections used for recovering heat from the flue gases. Small, round cast-iron units are larger but nevertheless similar in appearance (externally) to old-style wood-fired stoves used for residential heating. They have a series of hollow sections above and around the combustion chamber for the circulation of water. The hot combustion gases pass upward through the center of the unit and around the hollow sections. Round cast iron boilers have the following components which are arranged vertically.

- Ashpit
- Firepot
- Crown section
- Intermediate sections
- Dome section

Water circulates from section to section through a set of push nipples connecting the sections. After passing through the boiler, the combustion gases enter the outlet breeching and are discharged to the atmosphere through a flue.

Many cast-iron boilers are constructed in a square or rectangular shape. The water circulating sections are stacked vertically above the combustion chamber. The hot gas combustion products are forced to flow upwards and around the sections. The large cast-iron boilers can have an externally mounted header for steam near the top of the unit and two return drums at the bottom of the boiler.

All of the cast-iron boilers are used to generate either hot water or low pressure steam. This type of boiler is used for

## Air Emissions from Used Oil Combustion Processes

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small space heating applications. They are used without air pollution control systems.

Firetube boilers are used for small-to-moderately sized commercial systems. For steam generating applications, the firetube boilers can generate steam up to a pressure of 250 psig. In all firetube boilers, an oil burner is mounted in a combustion chamber. The hot gases are forced through one or more banks of boiler tubes which are surrounded by boiler feedwater. Steam accumulates in a steam drum mounted at the top of the boiler.

One common type of firetube boiler is the Scotch boiler. There are one or more oil burners, each of which discharges the combustion gases into a cylindrical furnace ("firetube") which passes across the bottom of the boiler. With oil-fired units, it is common to refractory line the furnace in order to maximize combustion efficiency. The combustion gases then pass upwards and through a series of baffled sections of firetubes. Steam accumulates at the top of the cylindrical unit.

Firetube boilers are usually not equipped with air pollution control systems. However, new units with heat input capacities greater than 10 million Btus/hr that were built or modified after June 9, 1989 are subject to 40 CFR Part 60, Subpart Dc (New Source Performance Standards). Also, most existing units are subject to air quality regulations included in federally enforceable State Implementation Plans. The main impact of these regulations are limitations of the oil sulfur content, recordkeeping requirements, and continuous emission monitoring requirements. Particulate emission limitations can usually be achieved by means of proper oil combustion.

Watertube boilers are often used for space heating applications requiring heat input rates greater than 15 million Btus/hr. The used oil is atomized and burned using a set of oil burners mounted in a water-tube lined refractory combustion chamber. The hot combustion gases transfer heat as they pass around and through banks of boiler tubes in the combustion chamber and in the upper elevations of the boiler. Steam accumulates in one or more steam drums mounted at the top of the boiler. Induced draft fans are used to move the combustion gases through the boiler and tube banks. Units designed for commercial applications usually do not have air pollution control systems. However, they are subject to the air quality regulatory requirements discussed above.

Small sized commercial boilers are usually operated in an on-off mode controlled by building temperature monitors. Emissions of

organics (possible adsorbents for metals) are greatest during the start-up and shut-down periods due to the cold gas temperatures in the combustion chamber. Moderate-to-large commercial units often operate with more stable boiler loads due to the size of the building being heated and the economical feasibility of more sophisticated boiler controls.

The oil burners used in this boiler size category include rotary cup atomizers, mechanical atomizers, and air atomizers. In all three cases, the oil is dispersed as fine droplets to facilitate rapid oxidation.

#### 4.3.1 GCA Corporation Emission Test Study

In 1984, the U.S. EPA, Industrial Environmental Research Laboratory sponsored a set of emission tests at six commercial sized boilers. The emission tests were conducted by GCA<sup>62</sup>. The study concerned the destruction efficiencies of chemical contaminants such as chlorinated organics which could be present in used oil. As part of the study, the fate of metals in the used oil were also evaluated. The metals portion of the test program work is relevant to the UORC Phase I project.

In order to control the highly variable concentrations of organic contaminants, GCA used a 4000 gallon oil supply tank. A supply of used oil was purchased, analyzed, and then spiked with four volatile chlorinated organic compounds and four semivolatile chlorinated organic compounds. The resulting concentrations were 3,000 to 5,000 ppm for each compound. This is important since the levels of chlorinated compounds were several orders of magnitude higher than the levels present in typical used oil fuels. Reactions between the chlorinated compounds and metal species in the used oil tank and during combustion could lead to the formation of metal chlorides which are inherently more volatile than the oxide form and the elemental form. Accordingly, the partitioning of the metal species measured in this study may not be representative of commercial boilers firing a more conventional used oil fuel.

A summary of the used oil composition is shown in Table 4-8. Each of the values shown is the average of between two and six separate analyses.

At the time that the test program was conducted, the lead levels were relatively typical of most used oil fuels during the early 1980's. However, these concentrations were a factor of 16 to 40 times higher than present lead concentrations.

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Table 4-8. GCA Used Oil Compositions<sup>62</sup>

| Element | Concentrations of Inorganics in Used Oil, (ppm) |        |                     |        |        |        |
|---------|---|--------|---------------------|--------|--------|--------|
|         | Site A <sup>2</sup>                             | Site C | Site D <sup>2</sup> | Site E | Site F | Site G |
| Cl      | 9,420   | 12,266 | 16,233              | 15,883 | 15,067 | 10,833 |
| As      | 2.87  | 9.00   | 2.77                | 10.60  | 9.85   | 9.05   |
| Cd      | 0.95  | 1.00   | 1.23                | 1.55   | 1.88   | 2.05   |
| Cr      | 3.23  | 8.3    | 4.49                | 7.95   | 8.43   | 7.2    |
| Pb      | 637   | 1,620  | 832                 | 1,535  | 1,390  | 1,545  |
| Zn      | 316   | 821    | 420                 | 753    | 651    | 760    |

- Note: 1. This data was calculated based on raw data included in reference 62.  
 2. Data take into account blending of used oil with virgin No. 2 oil to improve combustion for test purposes.

The metals emissions tests were conducted using a modified U.S. EPA Method 5 sampling train which is similar to the multi-metals train that is now accepted as the EPA reference method. The metals were analyzed by Inductively Coupled Plasma (ICP). The emission test data is summarized in Table 4-9.

The emissions data indicate that the operating conditions were nonrepresentative for most of the boilers. The flue gas oxygen concentrations for most of the boilers (sites A, C, D, E) were very high. The oxygen concentrations at site E were extremely high with an average concentration of 16.7% and several of the test runs at levels between 18.8% and 19.9%. Typical concentrations are 4 to 10%. Furthermore, the stack temperatures at these same test sites were extremely low. For example, the average stack temperature at site D was 268°F, and the temperature at site E was only 199°F! Typical stack temperatures range between 250 and 500°F. The oxygen and temperature data suggest either severe air infiltration into the combustion system or extremely poor burner adjustment. Both problems would hinder combustion of organic vapors and thereby contribute to soot formation and excessive particulate emissions. Due to the severity of the problems, most of the data obtained during this study is not representative of normal, well operated commercial boilers.

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Table 4-9. GCA Test Data for Commercial Boilers<sup>62</sup>

|                                     | Emission Test Site |        |       |        |        |        |
|-------------------------------------|--------------------|--------|-------|--------|--------|--------|
|                                     | A                  | C      | D     | E      | F      | G      |
| Fuel, gal./hr.                      | 3.22               | 24.05  | 13.14 | 14.6   | 23.2   | 23.1   |
| Gas flow, DSCM <sup>3</sup> /hr.    | 382                | 1,543  | 1,479 | 2,342  | 1,460  | 1,652  |
| Stack Temp., °F                     | 357                | 437    | 268   | 199    | 575    | 203    |
| Oxygen, %                           | 9.5                | 10.2   | 12.4  | 16.9   | 8.8    | 7.1    |
| CO, ppm                             | 220                | 47     | 350   | 28     | 28     | 48     |
| Particulate lbs/10 <sup>6</sup> Btu | 0.17               | 0.41   | 0.38  | 0.39   | 0.28   | 0.39   |
| As, ug/M <sup>3</sup>               | 11.2               | 655    | 26.1  | 106    | 251    | 286    |
| Cd, ug/M <sup>3</sup>               | 31.2               | 102    | 8.3   | 182    | 350    | 81     |
| Cr, ug/M <sup>3</sup>               | 62.2               | 166    | 112   | 230    | 205    | 263    |
| Zn, ug/M <sup>3</sup>               | 5,150              | 33,700 | 3,134 | 12,100 | 26,800 | 27,000 |
| Pb, ug/M <sup>3</sup>               | 9,680              | 72,400 | 5,390 | 20,300 | 49,800 | 51,000 |

Note: Site A, cast-iron boiler, mechanical atomizer, serving an office building  
 Site C, Horizontal return tube type firetube boiler, rotary cup atomization, serving a greenhouse  
 Site D, Scotch firetube boiler, air atomization, serving an office building  
 Site E, Scotch firetube boiler, rotary cup atomization, serving a greenhouse  
 Site F, Scotch firetube boiler, air atomization, serving a greenhouse  
 Site G, Scotch firetube boiler, air atomization, serving a greenhouse

The reduced temperatures and increased soot deposits on internal surfaces would also affect metals retention in the combustion chambers and heat exchange areas of the units. Metals in the

## Air Emissions from Used Oil Combustion Processes

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used oils being fired could be readily adsorbed in the "cold" carbonaceous deposits and, therefore, make the emission rates during the short term test appear to be lower-than-actual.

The metals retention data for the six tests have been calculated using the used oil firing rates, used oil compositions, combustion gas flow rates, and average metals emission rates reported by the contractor. This data is shown in Table 4-10.

Table 4-10. Metal Mass Balance Calculations, GCA Study<sup>62</sup>

| Metal    | Percentage of Metal in Used Oil<br>Emitted at Six Emission Test Sites |                  |    |                  |                  |    |
|----------|---|------------------|----|------------------|------------------|----|
|          | A   | C                | D  | E                | F                | G  |
| Arsenic  | 14  | 137 <sup>1</sup> | 31 | 47               | 47               | 66 |
| Cadmium  | 114 <sup>1</sup>  | 192 <sup>1</sup> | 22 | 553 <sup>1</sup> | 345 <sup>1</sup> | 83 |
| Chromium | 67  | 38               | 83 | 136 <sup>1</sup> | 45               | 76 |
| Lead     | 53  | 84               | 21 | 62               | 66               | 69 |
| Zinc     | 57  | 77               | 25 | 76               | 76               | 74 |

1. Indicates higher metal emission rates than metal input rates

The metal mass balance data is too weak to support any conclusions regarding metal retention in commercial boilers. There is too much data scatter, and the results do not clearly match the volatilities of these elements.

The high cadmium results indicate one or more of the following problems.

- Cadmium release from internal deposits
- Cadmium contamination from the sampling train
- Inaccurate cadmium analyses in the used oil
- Inaccurate emission test results

The emissions tests were conducted using the appropriate U.S. EPA reference methods and using applicable quality assurance procedures for the emission sampling and sample analysis. However, the operating conditions of the unit being tested were not fully described. Basic combustion parameters like oxygen

concentration and stack temperature were abnormal in some cases. Basic combustion parameters such as combustion chamber draft were not recorded. The emission tests were conducted under the assumption that short term emission tests can characterize emissions of species that are both retained and released from internal surfaces. No attempt was made to confirm that equilibrium conditions were established with respect to the internal mass transfer mechanisms prior to the tests. Overall, this study was extremely well executed from the standpoint of the air emission tests. However, the units being tested were apparently (1) fired under highly suspect operating conditions, (2) fired with fuel having highly unusual levels of chlorinated compounds, and (3) evaluated using short term tests with unknown retention conditions.

It should be noted that some qualitative information concerning lead behavior can be obtained from the results. Particle size analyses performed during some of the tests (sites A, G, and E) have been plotted in Figure 4-3. In all three cases shown, more than 80 percent of the lead emissions were in particles less than 1 micron in diameter. This indicates that lead vapor is nucleating homogeneously and/or heterogeneously to form submicron particles in the commercial boiler.

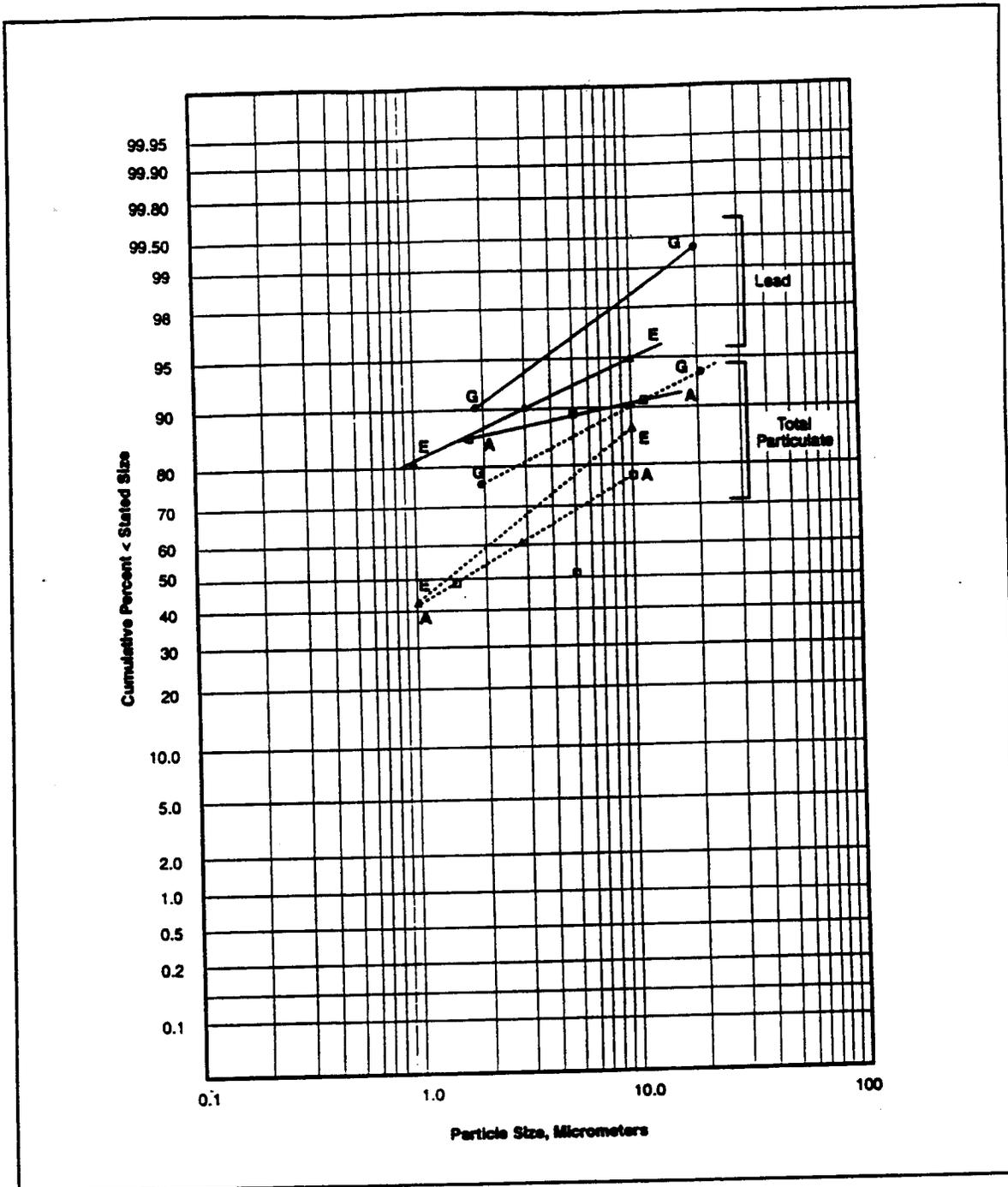


Figure 4-3. Particle Size Distributions of Lead Containing Particles<sup>62</sup>

#### 4.3.2 Summary - Commercial Boilers

Entropy identified and reviewed only one available air emissions test program for commercial boilers. This emission data is not representative of boilers firing present-day used oils. Some of the units appeared to operating improperly during the tests. Due to limitations of the data, it is not possible to accurately evaluate partitioning of the metals between the flyash and boiler deposits. The particle size data suggests that the lead emissions are predominantly in the submicron range.

#### 4.4 INDUSTRIAL BOILERS

Industrial boilers range in capacity from 5 million Btus/hr to more than 250 million Btus/hr. Small units are usually firetube boilers. These can have capacities up to approximately 15 million Btus/hr. Watertube type boilers are generally used for heat input capacities greater than 15,000,000 Btus/hr to more than 400,000,000 Btus/hr. Both types of boilers have been described in Section 4.3.

Industrial boilers used for space heating usually operate throughout the heating season. Boiler loads are much more stable than commercial boilers. Facilities having more than one boiler in the heating plant can adjust for steam demand changes by starting or shutting down one of the boilers. This normally allows the boilers that are operating to be near full load and thereby operate in the most thermally efficient manner. Any oil-fired boiler capable of firing No. 5 or No. 6 oil can be readily adapted to burn used oil. Some burner system modifications would be necessary to adapt a No. 2 burner for used oil.

There is no information available concerning the extent to which industrial boiler operators blend used oils with virgin oil prior to firing. Also, it is not known whether or not used oil firing occurs during start-up.

The types of air pollution control systems used on industrial boilers depend on the heat capacity of the boiler. Existing boilers are subject to fuel burning source particulate regulations (included in the SIPs) which require lower emissions from large units.

Also new or substantially modified boilers greater than 10 million Btus/hr (most industrial boilers) are subject to the New Source Performance Standard 40 CFR Part 60, Subpart Dc

(applicable after June 9, 1989). Units greater than 100 million Btus/hr are subject to Subpart Db (applicable Jun 19, 1984).

Large industrial boilers (greater than 100 million Btus/hour) which are fired entirely with oil do not usually need electrostatic precipitator or fabric filter particulate control systems. The efficiency of electrostatic precipitators is limited by the low resistivity of the carbonaceous particles and by the inherent difficulties in collection plate rapping at units with low inlet mass loadings. The efficiency of fabric filters can be adversely affected by acidic soot emissions which can blind portions of the bags and cause acid-related bag holes and tears.

Industrial boilers which fire coal and oil usually have high efficiency electrostatic precipitators or fabric filters. The ash generated by coal combustion mitigates the problems associated with the oil ash particles. Accordingly, the particulate removal efficiencies are usually above 99% overall and 90 to 95% in the difficult-to-control size range of 0.1 to 0.5 microns where metal concentrations are greatest.

Small industrial boilers are usually controlled by multi-cyclone collectors which achieve overall particulate removal efficiencies of 50 to 90%. However, they are completely ineffective for particles less than approximately 2 microns in diameter. For this reason, multi-cyclone collectors would not substantially reduce emissions of lead-containing particulate.

#### 4.4.1 GCA/DOE Data

Limited data concerning lead emissions from used oil fired industrial boilers were summarized by Surprenant et al.<sup>61</sup>. This data was drawn from a series of test programs conducted in the 1970 to 1973 time period. This data can not be considered representative of present day used oil firing for the following reasons.

- The emission studies were conducted prior to the development of U.S. EPA Reference Test Methods.
- Analytical techniques for metals have improved substantially.
- Used oil lead levels have decreased dramatically since the reduction in leaded gas consumption in the early 1980's and the lead phasedown in 1985.

This data is of interest only because it demonstrates that the concept of metals retention within the combustion chambers of oil-fired boilers has been recognized for more than twenty years. The fraction of lead emitted was as low as 20% in one of the tests, and it was usually in the range of 40 to 50%.

#### 4.4.2 Summary - Industrial Boilers

Air emissions data for used oil firing in industrial boilers is extremely limited. There are qualitative indications that lead retention in the boiler does occur. However, accurate partitioning factors for lead and other metals cannot be based on the data, all of which is more than twenty years out-of-date.

#### 4.5 MARINE BOILERS

These are similar in size and performance to small industrial boilers. Most marine boilers are firetube designs. Air pollution control systems are not used on these boilers.

While at sea, the marine boilers operate at relatively stable loads, especially if there are several boilers in use. However, very low boiler loads are possible when ships are docked. Marine boilers must be capable of high turndown rates in order to operate at the very low steam demands which exist at such times. High opacity emissions are possible if the unit is not adjusted properly. During these periods, some metal emissions are possible. Occupied areas close to harbors could be impacted by these emissions. However, it should be noted that the relatively "cold" conditions in the boiler at low load should favor metal retention due to the adsorption of metal species vapors on the soot-coated heat exchanger surfaces. Accordingly, only a fraction of the total metals in the used oil would be emitted. No air emission tests were identified concerning marine boilers.

#### 4.6 UTILITY BOILERS

Used oil can be burned in both oil-fired and coal-fired utility boilers. The types of used oil burned include but are not limited to vehicle crankcase oil, transformer oil, and turbine oil.

Utility scale coal-fired and oil-fired boilers are watertube boilers with a large number of burners arranged in rows. Combustion gases pass upwards through a boiler tube-lined combustion chamber and through superheater and reheater tube

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banks used to generate high pressure steam for the turbine. The gases then pass through an economizer section for preheating the boiler feedwater and an air preheater for the combustion air. The utility boilers have heat input rates ranging from 175 million Btus per hour to more than 6,500 million Btus per hour. This is equivalent to a megawatt range from less than 30 to more than 600 megawatts. There are approximately 2,420 operating utility scale boilers<sup>82</sup>.

Coal-fired boilers have one or more oil burners so that the combustion chamber can be preheated prior to the introduction of pulverized coal. These same burners can be used during full load operating periods for the combustion of small quantities of used oil fuel. During these periods, the heat input due to oil is a small fraction of the heat input due to coal. Most of the metals in used oil which are vaporized during combustion nucleate on flyash or bottom ash particles generated due to coal combustion. The partitioning factors for the used oil metals should be similar to those for metals entering with the coal. Partitioning factors for metals in coal are presented in Table 4-11.

Table 4-11. Partitioning of Metals from Coal-Firing<sup>83</sup>

| Metal    | Amount in Flyash % | Amount in Bottom Ash % | Amount in Vapor State % |
|----------|--------------------|------------------------|-------------------------|
| Arsenic  | 82.9               | 15.5                   | 1.6                     |
| Cadmium  | 84.1               | 13.2                   | 2.7                     |
| Chromium | 62.8               | 36.3                   | 0.9                     |
| Lead     | 88.6               | 7.8                    | 3.6                     |

Coal-fired boilers are equipped with high efficiency particulate control systems. These include reverse air fabric filters and electrostatic precipitators. Total removal efficiencies for flyash exceed 99%. Removal efficiencies for the submicron size range where most of the metals are present are in the range of 90 to 99% for electrostatic precipitators. Submicron particle removal efficiencies for fabric filters substantially exceed 99%. Accordingly, the metals emissions resulting from the combustion of used oil in coal-fired utility boilers should be quite low.

In the case of oil-fired units, the used oil can be fired in separate burners or blended with virgin oil prior to firing.

Many oil-fired units are equipped with multi-cyclone collectors for removal of large sized particulate matter. Some units are equipped with high efficiency particulate control systems.

No air emission test data were found concerning utility scale boilers firing used oils. Testing is warranted based on the used oil consumption data indicated by the NORA survey (Table 4-1). Utility boilers are the third largest consumer of used oils supplied by marketers, processors, and blenders.

#### 4.7 ASPHALT PLANTS

Asphalt plants consume approximately 40% of the used oil fuels generated in the U.S. according to the results of the 1993 NORA survey. The National Asphalt Paving Association (NAPA) has estimated that the industry consumes as much as 200 million gallons per year<sup>64</sup>. That consumption rate has increased during the last few years.

Used oil is used in asphalt plants to dry and heat the aggregate prior to its mixing with asphaltic binder. There are two main types of plants: (1) hot mix, and (2) drum mix.

A simplified schematic of a hot mix plant is shown in Figure 4-4. Aggregate of various size gradations is metered out of bins in proportions necessary to produce the grade of asphaltic concrete specified by the customer (usually State DOT). The aggregate must be dried and heated to 250 to 325°F in order to ensure proper coating with the asphaltic binder. Heating and drying are performed in a small rotary drier with a single oil-fired burner. The portion of the rotary drier on the opposite side from the burner has side-mounted scoops to shower the aggregate across the combustion gas stream as the drum rotates. The hot aggregate is then transported by a hot bucket elevator to the hot bins and hot screens. The size proportions are readjusted, and the aggregate is dumped into a pug mill for mixing with the asphaltic binder.

The hot combustion gases entrain fine aggregate dust in the rotary dryer. These particulate emissions are controlled by a pulse jet baghouse or a medium pressure drop venturi scrubber. In many plants, fugitive dust captured from the hot screens and the elevator are also ducted to the particulate control system.

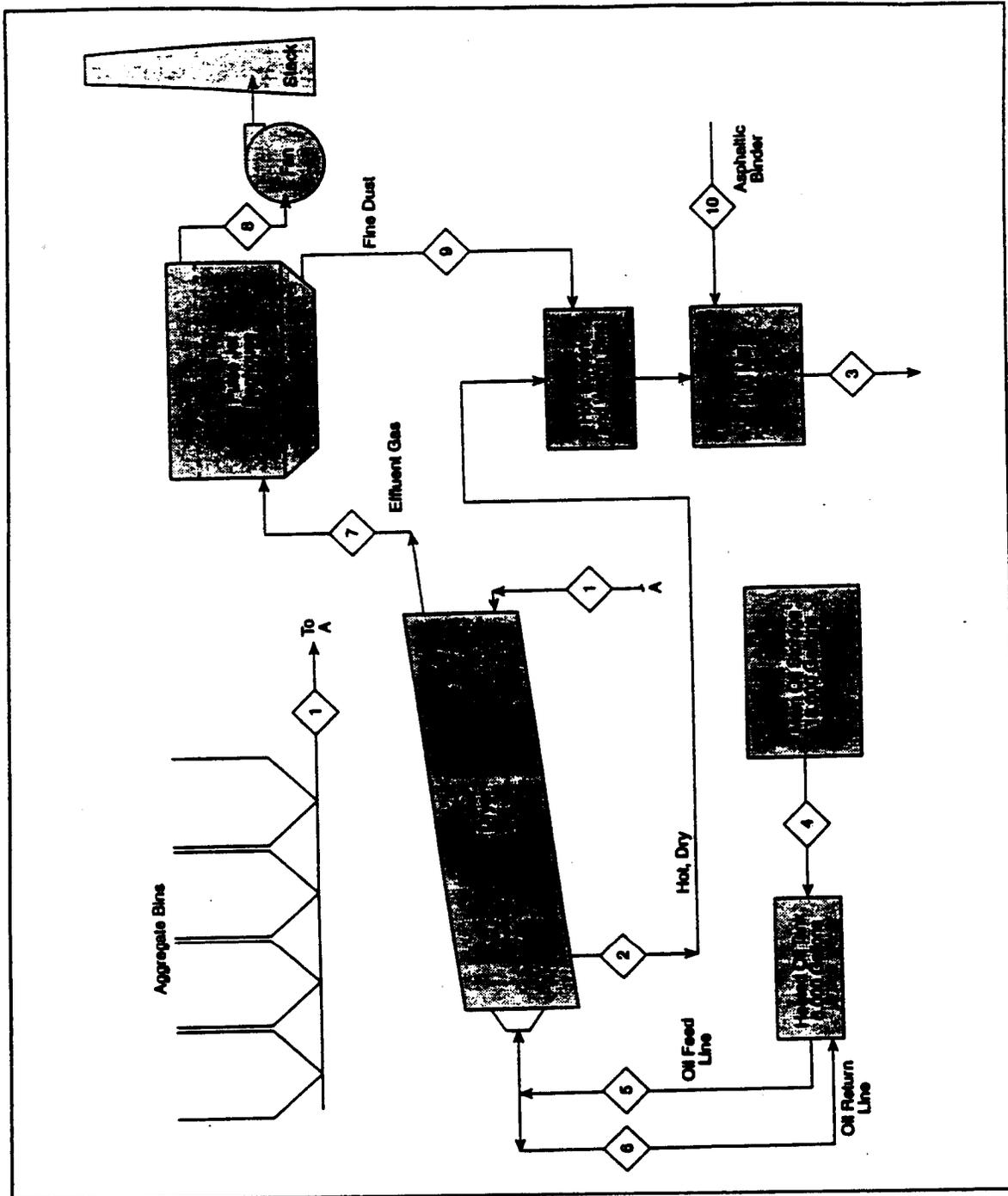


Figure 4-4. Hot Mix Type Asphalt Plant

A drum plant performs aggregate drying/heating and binder application in the rotary drum. There are no hot bins, hot screens, or pug mill. After passing part of the way through the rotary drier, the dried and heated aggregate is coated with binder using an injection pipe which extends in from the cold side of the drier. The placement of the injection pipe is adjusted to minimize volatilization of the asphaltic binder. The hot gases are ducted to a pulse jet baghouse or a medium pressure drop venturi scrubber.

Most asphalt plants have oil burners which can be readily adapted for used oil firing. The used oil increases the metals emissions to the particulate control system. However, it is important to note that the used oil is not the only source of metals. The aggregate itself has low concentrations of many of the metals addressed in this project. These low concentrations are important simply because of the large aggregate dust loadings to the air pollution control system.

There are two air emissions studies of asphalt plants firing used oils. These are summarized in the following sections.

#### 4.7.1 Harris and Schlickenrieder Study<sup>55</sup>

Harris and Schlickenrieder of A.D. Little, Inc. conducted a study of a hot mix plant under the sponsorship of the Massachusetts Department of Environmental Quality Engineering (DEQE). The report states that it was conducted in part because the Commonwealth of Massachusetts had determined that asphalt plants consumed approximately one-half of the used oil generated in the State. Furthermore, the State had determined that much of the data published prior to 1983 was inadequate to support regulatory decisions.

The tests were conducted in accordance with a trial burn plan developed by A.D. Little and reviewed by both the DEQE and the Massachusetts Asphalt Paving Association. U.S. EPA Reference Method 5 was used for particulate sampling, and the metals were analyzed using ICP and atomic absorption (AA). The used oil fuel was a blended mix of two used oil supplies. Prior to the tests, trichloroethylene was added to yield a concentration of 9,400 ppm. The resulting used oil composition is summarized in Table 4-12.

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Plant operations during the test appeared to be normal for the specific facility and representative of the industry in general. There were no apparent sampling or analytical problems with this study.

Table 4-12. Harris and Schlickerieder Used Oil Composition<sup>85</sup>

| Contaminant       | Concentration, ppm |
|-------------------|--------------------|
| Arsenic           | < 10               |
| Cadmium           | 1.0                |
| Chromium          | 7                  |
| Lead              | 997                |
| Trichloroethylene | 8900               |
| PCBs              | 5-7                |

A set of three full emission tests was conducted. The metals data is summarized in Table 4-13. This data indicates that the metal emissions are very low. The mass balance calculations indicate overall removal efficiencies of 85 to 99.88% for the various metals. These high efficiencies are the result of high efficiency particulate removal in the baghouse and the capture of metals on the surfaces of the aggregate in the rotary drier.

Table 4-13. Harris and Schlickerieder Test Data<sup>85</sup>

|                    | Arsenic | Cadmium | Chromium | Lead   |
|--------------------|---------|---------|----------|--------|
| Fuel, ppm          | < 10    | 1.0     | 7        | 997    |
| Lbs/hr.            | 0.0027  | 0.0028  | 0.0178   | 2.67   |
| Stack, Conc. ug/g  | 0.175   | 0.605   | 23.1     | 26.4   |
| Lbs./hr.           | 0.00002 | 0.00007 | 0.0027   | 0.0031 |
| Removal Efficiency | NA      | 97.52   | 85.0     | 99.88  |

The fate of the captured metals is the same regardless of the specific partitioning factors. As shown in Figure 4-4, the baghouse solids are reunited with the aggregate from the drier at the hot screens. The small particle size dust captured by the baghouse is needed to achieve the asphaltic concrete aggregate

size distribution specifications. The metals captured by the aggregate do not pose an environmental hazard since they are incorporated into the hardened asphaltic concrete.

#### 4.7.2 Baker et al. Study<sup>86</sup>

This study was reported in the Background Information Document (BID) for the BIF Regulations (Volume II Industrial Furnaces). However, as of this time, Entropy has not been able to obtain a copy of the draft report (dated 1983) which is cited as the source of emissions data. Accordingly, only the data and information included in the BID can be evaluated.

The air emissions tests were conducted at two plants. One was a 400 ton per hour drum mix plant equipped with a baghouse. This was fired entirely with a processed industrial used oil. The second plant was a 76 ton per hour unit equipped with a scrubber. It was fired with 100% processed crankcase oil. The production data included in reference<sup>86</sup> indicates that both plants were operating normally during the test program.

At both plants, metals concentrations were measured in the used oil feeds, the aggregate feeds, the asphaltic binder, the baghouse inlet dust, and the stack emissions. U.S. EPA Method 5 was used for sampling at the baghouse inlet and the stack.

Baker et al. attempted to measure the metal-specific control efficiencies of the baghouse and scrubber. In order to isolate the fraction of the metals due to the used oil, they analyzed the aggregate dust prior to the drier. Mass balance calculations were then performed using the dust loadings generated in the drier. The results of these calculations are shown in Table 4-14 for Plant #1 and in Table 4-15 for Plant #2.

Mass balance calculations of the type shown in Table 4-14 and 4-15 are vulnerable to a number of problems in obtaining representative samples and accurate analyses of several different types of materials. Furthermore, the aggregate stream is at least one hundred times larger than the used oil stream. Despite these inherent limitations, the data in these two tables do indicate the following.

- Used oils can be major contributors to the metals listed in these two tables.
- The removal efficiencies for all of the metals are relatively similar.
- The removal efficiencies for the metals are different for the scrubber controlled plant.

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Table 4-14. Mass Balance Calculations, Plant 1, Baghouse Control<sup>84</sup>

|          | Metals Input, lbs./hr x 10 <sup>-2</sup> |       |             |                | Baghouse Removal Efficiency |
|----------|--|-------|-------------|----------------|-----------------------------|
|          | Stone                                    | Oil   | Total Calc. | Total Measured |                             |
| Barium   | 0.68                                     | 9.08  | 9.76        | 7.79           | 0-100                       |
| Cadmium  | 0.72                                     | <0.11 | 0.72        | 0.2            | 99-100                      |
| Chromium | 0.41                                     | 110   | 110         | 101            | 100                         |
| Lead     | 1.7                                      | 27.4  | 29.1        | 39.8           | 89-100                      |
| Zinc     | 2.3                                      | 25.6  | 27.9        | 24.2           | 35-97                       |

Table 4-15. Mass Balance Calculations, Plant 2, Scrubber Control<sup>84</sup>

|          | Metals Input, lbs./hr x 10 <sup>-2</sup> |      |             |                | Scrubber Removal Efficiency |
|----------|--|------|-------------|----------------|-----------------------------|
|          | Stone                                    | Oil  | Total Calc. | Total Measured |                             |
| Barium   | 0.27                                     | 3.22 | 3.49        | 0.92           | 99-100                      |
| Cadmium  | <0.03                                    | 0.14 | 0.17        | 0.04           | 56-81                       |
| Chromium | 0.30                                     | 0.32 | 0.62        | 0.97           | 85-93                       |
| Lead     | 0.20                                     | 51.5 | 51.7        | 13.48          | 25-42                       |
| Zinc     | 0.27                                     | 73.4 | 73.7        | 21.0           | 25-28                       |

The metal-by-metal removal efficiencies for the wet scrubbing system are consistent with the presumption that the moderately volatile metals such as lead vaporize in the drier and nucleate as submicron particles. Scrubbers are least able to capture particles in the 0.1 to 0.5 micron range. Baghouses have relatively little particle size - collection efficiency dependence.

#### 4.7.3 Summary - Asphalt Plants

The limited data available indicates that metals in used oil fuels burned in asphalt plant driers are captured with high efficiency at units equipped with well operated baghouses. Emissions of all of the metals may be less than 1% of the total input quantities present in the used oil.

A plant with a wet scrubbing systems may have lead emissions which were 50 to 75% of the total input quantities. This may be due to the presence of lead and other volatile metal species in the difficult-to-control submicron size range. The single study identified concerning wet scrubber equipped asphalt plants is not sufficient to characterize metal collection efficiencies.

Metal removal efficiencies may be high in the asphalt industry due to the characteristics of the combustion process. The metals vaporized in the burner flame have ample opportunity to nucleate on the surfaces of aggregate dust circulating through the combustion gas stream. The aggregate surface temperatures are much lower than the dewpoint temperatures for the metal vapors. In fact, the aggregate temperatures are far below the normal surface temperatures of the boiler tubes and combustion chamber surfaces of boilers. Accordingly, condensation of these vapors is likely on the aggregate dust. Metal emissions are further reduced by fabric filters used in many plants. Since fabric filter efficiency is high even in the submicron range, metal vapor which nucleates homogeneously will also be captured.

There are practical limits to the firing of used oils in asphalt plants. Used oil fuels with high concentrations of chlorinated compounds could cause severe HCl-related corrosion of the ductwork and air pollution control equipment.

#### 4.8 CEMENT KILNS

Portland Cement kilns pyroprocess feed materials (limestone, sand, and iron oxide) to yield tricalcium silicate, tricalcium aluminate, tetracalciumaluminate-ferrite, and a variety of other component compounds. The feed materials are blended in the proper stoichiometric proportions and fed into a large, refractory lined rotary kiln. The kiln is fired by a single burner which can be easily adapted for used oil firing. Fuel costs represent an important fraction of the overall cost of producing portland cement. There are a number of Portland Cement kiln designs.

Wet Process Plants

Dry Process Plants

- Long dry
- Dry with preheater
- Dry with preheater and precalciner

In wet process plants, the feed materials are blended as a slurry. Due to the moisture content of the feed materials, wet process kilns are long and have very high heat demands of 5.5 to 6 million Btus per ton of clinker produced. Due to improvements in grinding and blending techniques during the last 30 years, a variety of dry process were developed. Due to the low moisture levels of the feed material, the kilns were shortened and fuel demands were reduced for dry process mills. Fuel demand for a dry process is 4.5 to 5.0 million Btus per ton of clinker produced.

Preheaters were developed more than 30 years ago to reduce the cost of the rotary kiln. These are direct gas-to-solids heat exchangers mounted on the feed end of the kiln. The feed materials are dried, and the calcination process is started in this stationary equipment. This reduces the size of the rotary kiln. Precalciners were developed during the 1970's for the same reason. They are located between the feed end of the kiln and the preheaters. Feed materials which have been heated in the preheater enter the non-moving calciner where limestone calcination is partially completed. A separate burner is used in the calciner to provide the heat necessary for calcination. Rotary kilns equipped with preheater-precincer systems do not need to be as large since part of the processing of the feed material has already been completed prior to entry to the kiln.

A flowchart of a dry process Portland Cement plant with a preheater type kiln is shown in Figure 4-5. The dry feed materials are ground and blended in a grinding mill (raw mill) and air classifier. Hot combustion gas from the preheater tower usually pass through the raw mill to recover some of the sensible heat from the gas stream. The feed material then enters the system near the top of the preheater tower. It cascades downward through a series of cyclones shown as a block in Figure 4-5. A simplified sketch of the cyclones is shown in Figure 4-6. The feed materials become progressively hotter as they move downward through the preheater tower cyclones. In the kiln, the solids reach temperatures exceeding 2700 to 2800°F as they approach the burner end of the kiln. These temperatures are needed in order to complete the inorganic reactions which yield the mix of chemicals which are collectively termed clinker.

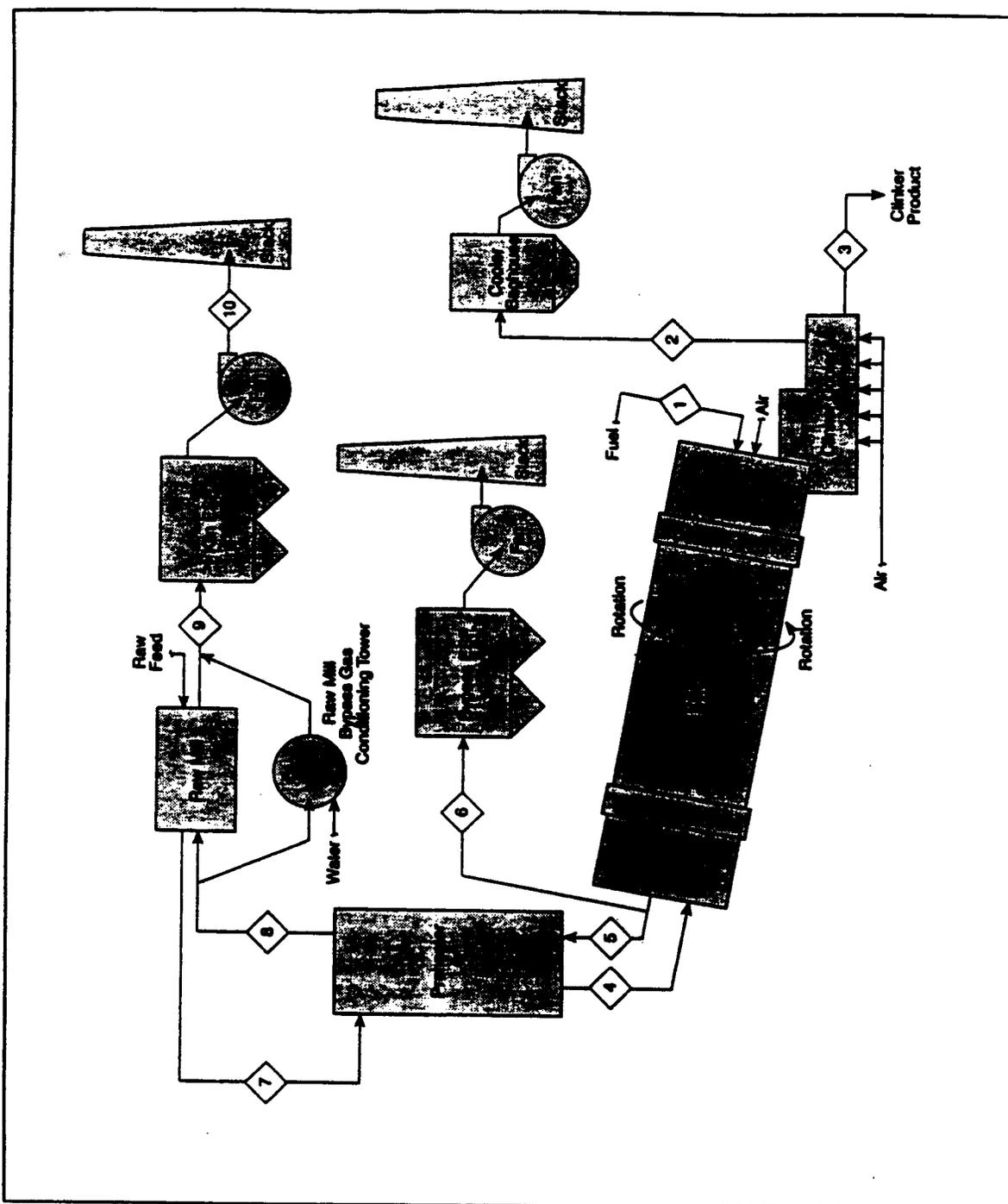


Figure 4-5. Preheater Type Portland Cement Plant

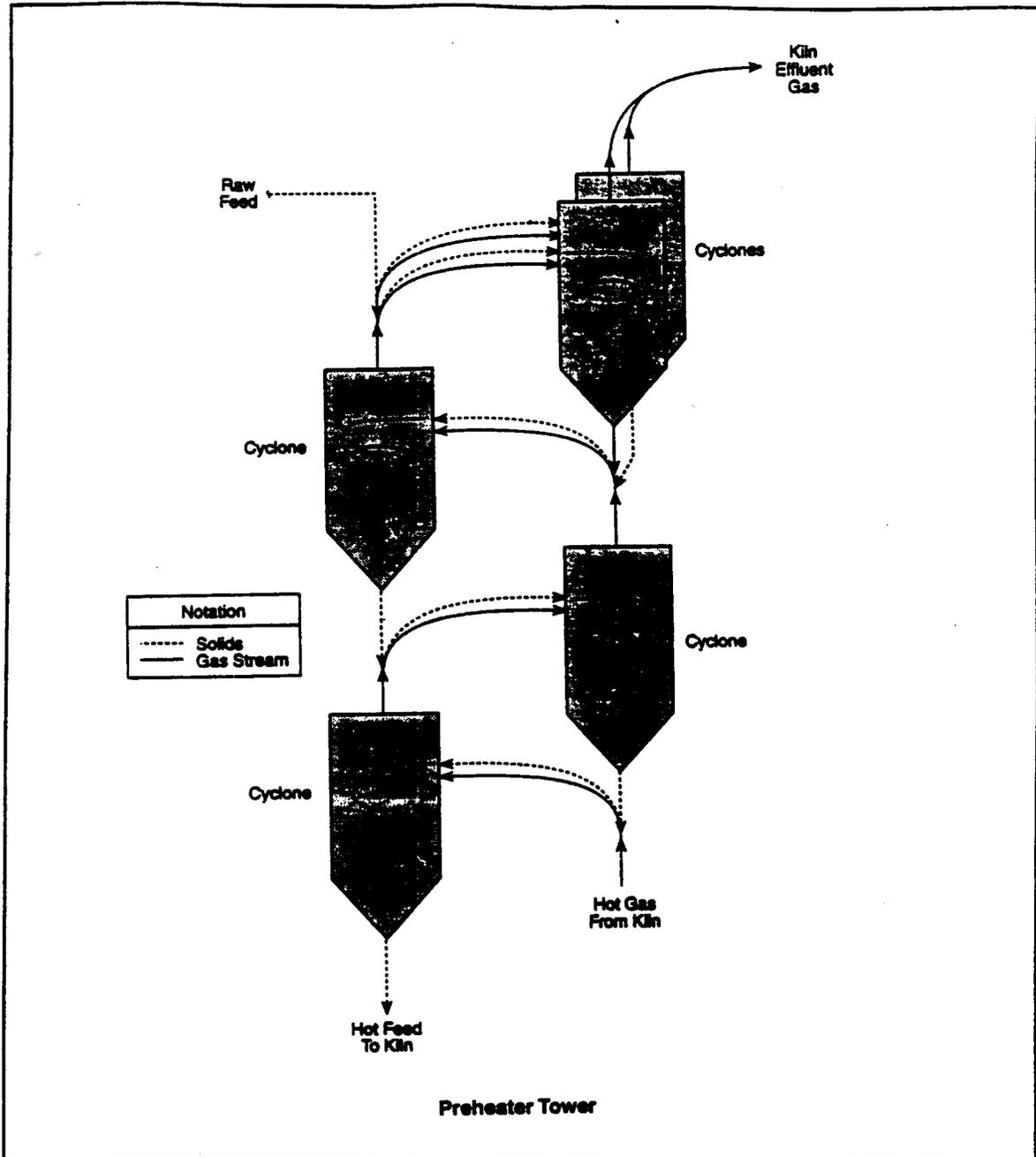


Figure 4-6. Simplified Sketch of Flow Patterns in Preheater Tower Cyclones

The gas stream passes in a counterflow direction from the solids. The gas temperatures reach peak levels of 3000 to 3400°F for several seconds. They remain at high temperatures throughout the kiln and preheater tower. Due to the high temperatures and long gas residence times, essentially all of the metals of interest in the Phase I study are volatilized. As the combustion gas stream cools passing up through the kiln, the nonvolatile elements can be captured by the feed materials and partially processed solids moving in a counterflow direction. Some of the metal species can react with the partially processed feed materials and be immobilized<sup>73</sup>. These materials will exit the kiln with the clinker. The moderately volatile metal species can remain in the vapor state until they reach the preheater tower. As the gas stream cools moving up through the series of cyclones, the metal vapors nucleate heterogeneously on the surfaces of the feed material particles.

The metals emitted from the combustion of used oil are partitioned into the clinker stream exiting the kiln, the feed materials passing downward toward the kiln, and the particulate exiting the preheater tower and going to the particulate control system. A high efficiency particulate control system is needed due to the high solids loadings resulting from entrainment of particulate in the preheater tower and raw mill. Both reverse air fabric filters and electrostatic precipitators are used.

The metal vapors which condensed heterogeneously in the preheater tower can be re-volatilized as the feed material particles cascade downward and are exposed to increasingly higher gas temperatures. The process of re-volatilization in the high temperature area near the bottom of the preheater tower or the kiln and nucleation in the cold temperature area at the top of the preheater can set up a loop for concentrating the metal species and compounds such as sodium and potassium. There is no means of escape for this materials unless a portion of the kiln gases are drawn off and treated in a separate air pollution control system. This is termed the alkali bypass system since the main objective is to purge the kiln system of sodium and potassium that can not be present at high concentrations in the clinker product. A major fraction of the metals released during used oil firing can exit the kiln system through the alkali bypass gas stream. An electrostatic precipitator is used for high efficiency collection of the particulate matter in the alkali bypass stream.

Portland Cement plants are well suited for used oil firing. All of the metals in the oil should be vaporized in the firing end of the kiln and captured as nucleated material on the clinker solids

or entrained dust. The entrained dust is treated in one of the two high efficiency particulate control systems. Emissions from the control systems should be low even in the small particle size ranges where most of the metal species reside. The relatively nonvolatile metal species captured in the clinker material remain trapped. Accordingly, the fraction of used oil metals emitted to the air should be very low. The only metal that is not captured efficiently is mercury. Due to its extreme volatility, it can penetrate the preheater tower, the raw mill, and the particulate control system as a vapor.

The metal capture capabilities described with respect to dry process preheater-type kilns also apply in general to the other types of kilns. Overall, cement kilns have a number of inherent characteristics which are valuable with respect to used oil firing.

- Stable combustion conditions
- Infrequent start-up, shut-down cycles
- Long gas residence times at high temperature
- High efficiency particulate control systems

Due to these (and other) characteristics, cement kilns are frequently used for hazardous waste incineration. Most of the emissions data that is available concerns hazardous wastes rather than used oil. However, the partitioning factors and air pollution control efficiencies for the metals should be the same for used oil fired and hazardous waste fired kilns.

#### 4.8.1 EPA BIF Background Information Document<sup>87</sup>

Most of the air emissions tests summarized in this document concern destruction efficiencies for organic compounds. However, one set of tests concerning a wet process cement kiln was included, and this data is summarized in Table 4-16. The source being tested had an electrostatic precipitator for control of the kiln combustion gases. It should be noted that wet process mills do not have an alkali bypass stream. Therefore, any metals exiting the kiln as vapor or as particles will be treated by this particulate control system.

The indicated removal efficiency for lead is 97.7%. The much lower efficiencies for mercury and selenium are probably due to incomplete nucleation of these very volatile elements. It is also possible that these elements are nucleating primarily in the

difficult-to-control size range of 0.1 to 0.5 microns. Electrostatic precipitators are least efficient in this size range.

Table 4-16. Metal Mass Emission Rates<sup>67</sup>

| Metal    | Mass Flow Rates, Lbs./hr. |                 |
|----------|---------------------------|-----------------|
|          | Fuel Total                | Stack Emissions |
| Cadmium  | 0.13                      | 0.005           |
| Mercury  | 0.009                     | 0.001           |
| Lead     | 2.396                     | 0.055           |
| Selenium | 0.08                      | 0.027           |

Note: This is a secondary reference. No data is available concerning plant or control device operating conditions.

4.8.2 Dellinger et al.<sup>73</sup>

The Scientific Advisory Board (SAB) on Cement Kiln Recycling has prepared a comprehensive report on the performance of cement kilns burning hazardous wastes. The portions of the report concerning the partitioning and collection of metals is relevant to the Phase I UORC project.

A general model of metals behavior in cement kilns has been prepared by Dellinger et al. This model was based on the volatilities of the various metal species, the temperature profiles in the equipment, and the cement plant design characteristics. The model has been used to predict metals partitioning into the clinker, the cement kiln dust (CKD) captured by the particulate control systems, and the emissions to the atmosphere. The model results have been compared to air emission data obtained in a variety of earlier test programs.

Comparisons of the model results and the emission data for lead and chromium at two separate plants are shown in Figures 4-7 through 4-10 from the SAB report. This data indicates that most of the lead (moderately volatile) is partitioned to the cement kiln dust which is captured by the particulate control systems.

Air Emissions from Used Oil Combustion Processes

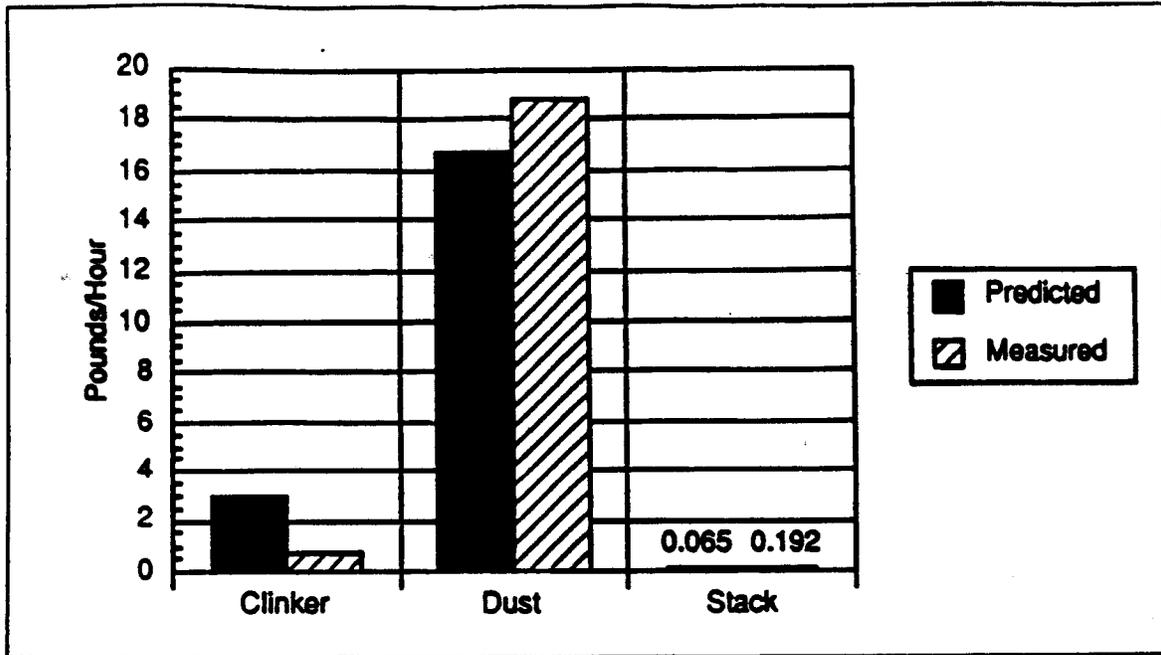


Figure 4-7. Predicted and Actual Lead Partitioning, Plant "P"

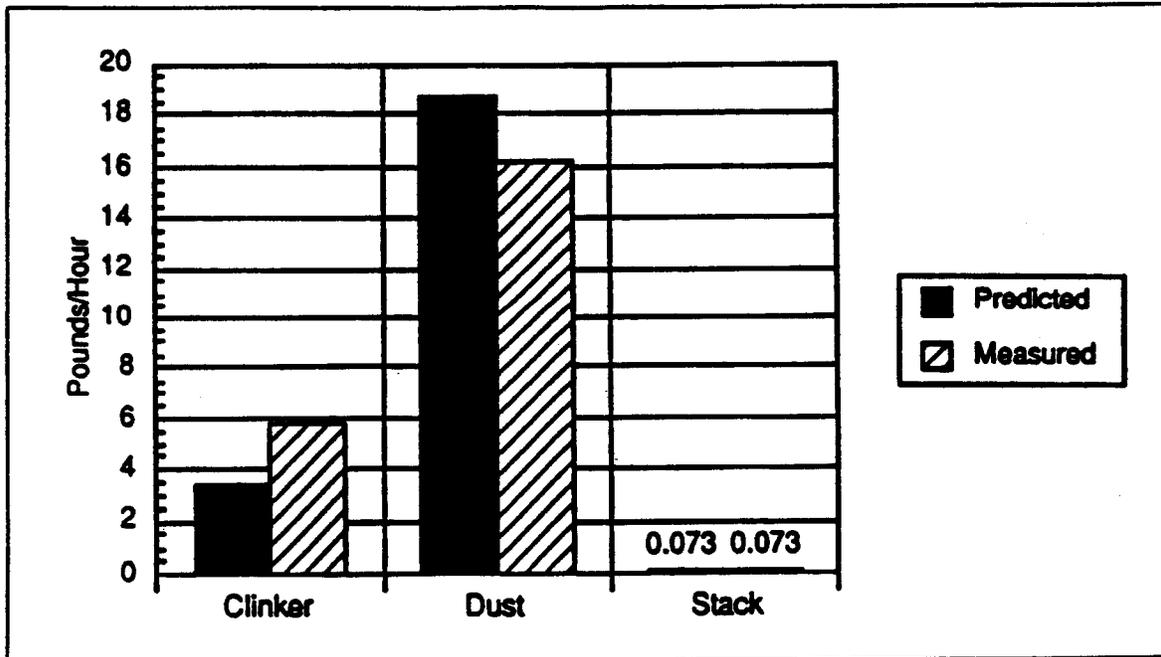


Figure 4-8. Predicted and Actual Lead Partitioning from Plant "O"

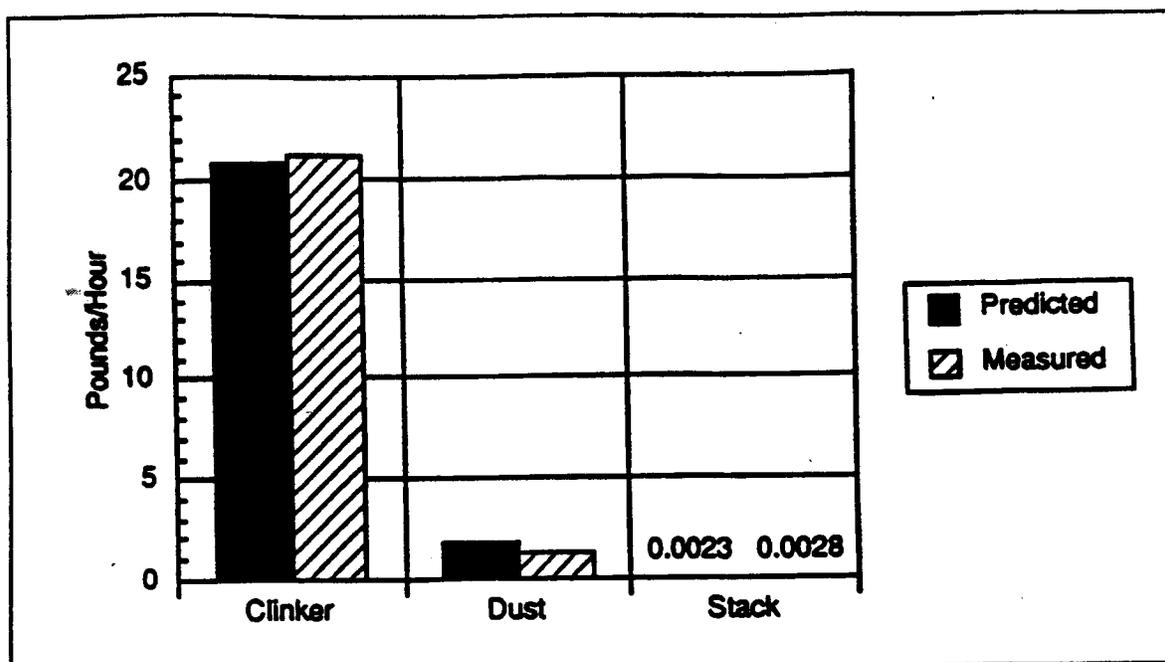


Figure 4-9. Predicted and Actual Chromium Partitioning, Plant "P"

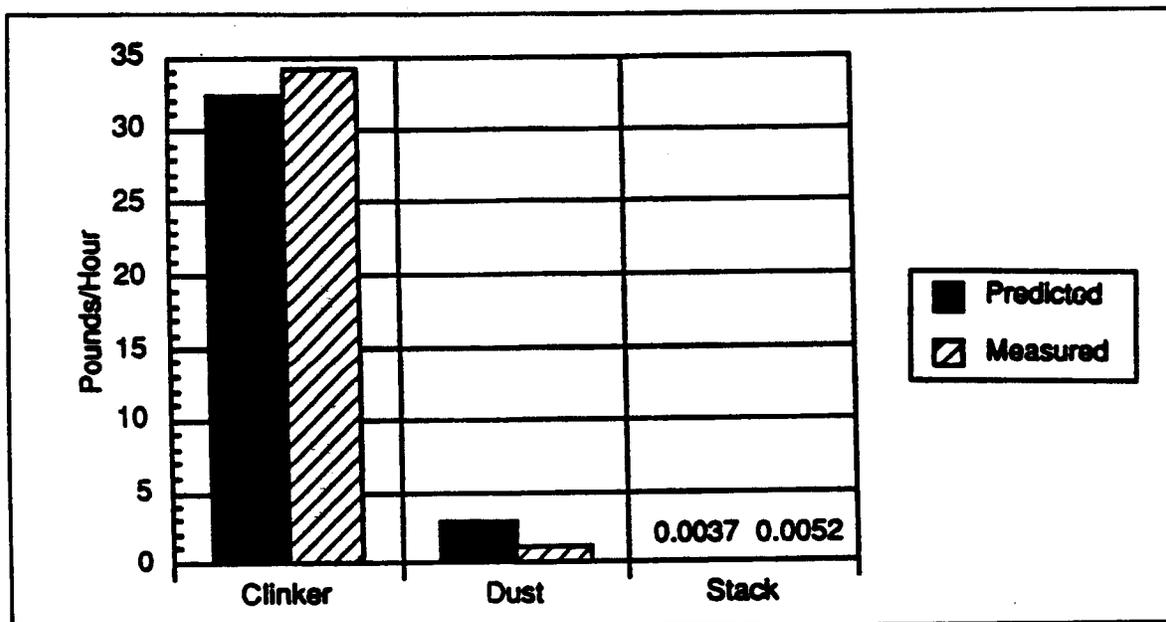


Figure 4-10. Predicted and Actual Chromium Partitioning, Plant "O"

## Air Emissions from Used Oil Combustion Processes

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A relatively small fraction of lead remains within the clinker product. The lead emissions from Plant "P" were approximately 1% of the total lead emissions. The lead emissions from Plant "O" were approximately 0.3% of the total.

Based on the model results, SAB has proposed general emission factors for metals from several types of cement kilns. A partial list of those factors is presented in Table 4-17.

Table 4-17. SAB Emission Factors<sup>73</sup>

| Metal    | Percentage Emitted |                |
|----------|--------------------|----------------|
|          | Preheater Kilns    | Long Wet Kilns |
| Arsenic  | 0.016              | 0.001          |
| Barium   | 0.011              | N.D.           |
| Cadmium  | 0.080              | N.D.           |
| Chromium | 0.007              | N.D.           |
| Lead     | 0.110              | 0.562          |
| Zinc     | 0.033              | 0.057          |
| Mercury  | 5.23               | N.D.           |

There could be plant-to-plant variations in the actual emission rates of metals due to a variety of factors including but not limited to the following.

- Metal feed quantities
- Chlorine feed quantities
- Particulate control system performance

Nevertheless, the cement kiln emissions data evaluated by SAB indicate that very small fractions of lead and the other metals studied in this Phase I project would be emitted from cement kilns.

### 4.8.3 Roth<sup>89</sup>

Roth summarized air emissions data obtained in April 1991 at the Southwestern Portland cement plant in Fairborn, Ohio. The unit tested was a preheater type kiln fired with various combinations of coal, tires, and liquid hazardous wastes. The liquid

## Air Emissions from Used Oil Combustion Processes

hazardous wastes were spiked with metals for approximately 48 hours prior to emission testing and sampling in order to "condition" the process. This was useful in ensuring that changes in the preheater-kiln concentration loops would not affect the metals partitioning and collection efficiency test results.

In preparing the mass balances, Roth did not include the metals contents in the incoming raw feed and in the coal. The overall efficiency of metals removal was determined simply by comparing the total emission values for the main stack, bypass stack, and clinker cooler (Figure 4-5) with the metals input levels from the spiked hazardous waste and the tires. The results of these mass balance calculations are shown in Table 4-18.

Table 4-18. Metals Removal Efficiencies<sup>88</sup>

|          | Condition 1, high metals spiking |                    |        | Condition 2, low metals spiking |                    |         |
|----------|----------------------------------|--------------------|--------|---------------------------------|--------------------|---------|
|          | Stacks <sup>1</sup>              | Input <sup>2</sup> | Eff, % | Stacks <sup>1</sup>             | Input <sup>2</sup> | Eff., % |
| Arsenic  | 0.00034                          | 0.49               | 99.93  | 0.00017                         | 0.14               | 99.88   |
| Barium   | 0.0058                           | 13.08              | 99.96  | 0.0036                          | 13.16              | 99.97   |
| Cadmium  | 0.0011                           | 1.32               | 99.91  | 0.00077                         | 0.48               | 99.84   |
| Chromium | 0.0024                           | 8.28               | 99.97  | 0.0029                          | 8.70               | 99.97   |
| Lead     | 0.0183                           | 14.94              | 99.88  | 0.0078                          | 11.68              | 99.93   |
| Mercury  | 0.0069                           | 0.09               | 92.38  | 0.0079                          | 0.05               | 84.40   |
| Zinc     | 0.0134                           | 37.08              | 99.96  | 0.0084                          | 28.88              | 99.97   |

1. Total stack emissions from three stacks
2. Metals input from hazardous waste, tires, and spiked material

The data presented by Roth<sup>88</sup> are consistent with the metal emission factor estimates presented by SAB<sup>73</sup>. These studies indicate that the emissions of lead and other metals included in the scope of the Phase I UORC study are quite low.

4.8.4 Bolstad and Chehaske<sup>89</sup>

This paper summarized two trial burn tests, one at a wet process mill and another at a dry process mill. Due to a malfunction with the precipitator at the dry process mill, metals emissions data are presented only for the wet process kiln. Bolstad and Chehaske reported that emissions of lead, cadmium, mercury, and selenium increased significantly after co-firing of hazardous waste began. Nevertheless, the overall emissions were relatively small. For example, the emissions of lead increased from 0.0074 pounds per hour during 100% coal firing to 0.055 pounds per hour during co-firing. The emissions of cadmium increased from negligible levels to 0.005 pounds per hour during co-firing.

4.8.5 Hansen<sup>90</sup>

This study concerned the effect of alternative spiking methods on the measurement of hexavalent chromium removal efficiencies. These tests were conducted at the Ash Grove Cement Company plant in Chanute, Kansas. The unit tested was a long, wet process kiln. The removal efficiencies measured during the study are presented in Table 4-19.

Table 4-19. Removal Efficiencies for Chromium<sup>90</sup>

|        | Species          | Input | Emission | Removal Eff, % |
|--------|------------------|-------|----------|----------------|
| Test 1 | Cr <sup>+6</sup> | 9.75  | 0.00039  | 99.996         |
|        | Cr Total         | 19.09 | 0.0014   | 99.992         |
| Test 2 | Cr <sup>+6</sup> | 8.90  | 0.0019   | 99.978         |
|        | Cr Total         | 11.69 | 0.0028   | 99.976         |

The results of this study are entirely consistent with those done earlier. Due to the low volatility of chromium, it is partitioned primarily into the clinker stream. Small quantities are also present in the cement kiln dust. Removal efficiencies for hexavalent chromium and total chromium are very high.

4.8.6 Gossman, Black, and Ward<sup>74</sup>

This study concerned metal sources and partitioning in three kilns at two separate cement plants. Samples were taken during baseline periods and during the co-firing of hazardous wastes.

They determined that the quantities of arsenic entering with the kiln feed and coal were several orders of magnitude greater than that entering with the hazardous waste fuel (<1 to 8.8 ppm). Most of the arsenic became part of the cement kiln dust, and a small amount became trapped in the clinker.

The large majority of the barium entering with the kiln feed, coal, and hazardous waste became part of the clinker. Unlike many other elements, the source of cadmium was almost exclusively the hazardous waste fuel. All of the cadmium became part of the cement kiln dust.

The raw feed dust entrained in the combustion gas streams was considered to be the major source of chromium reaching the particulate control devices. Chromium entering the kiln with the hazardous waste fuel is effectively captured in the kiln and is incorporated into the clinker stream.

The authors conclude that lead has a strong tendency to concentrate in the cement kiln dust. As a volatile material, it can participate in volatilization-nucleation loops. Accordingly, lead emissions to the particulate control systems are subject to variations in input rates when there are operational changes that effect this concentration cycle.

#### 4.8.7 Summary - Cement Kilns

There have been numerous detailed studies of hazardous waste firing in cement kilns. The data provided by these studies regarding metals behavior support the following conclusions.

- The emissions of lead, cadmium, chromium, barium, arsenic, and zinc are very low. Less than 1% of the metals entering with the waste fuel are emitted in the combined effluents from the main kiln stack, the alkali bypass stack, and the clinker cooler stack.
- The high metal removal efficiencies are due to the capture of the low volatility metal species in the clinker and the high efficiency control of cement kiln dust.

It should also be noted that used oils have considerably lower lead, barium, and chloride concentrations than the hazardous wastes tested in the studies discussed. In some cases, the quantities of metals entering with the used oil could be

relatively small compared with the quantities entering with the raw feed and the coal.

#### 4.9 STEEL MILL METALLURGICAL PROCESSES

Used oil fuels can be used in a variety of metallurgical processes, but there is very limited data concerning present used oil firing practices.

Used oil could conceivably be used as an auxiliary fuel in blast furnaces. The use of this fuel could reduce the coke demand and thereby reduce pig iron production costs. However, there are limitations to the used oil firing rates since high lead, zinc, and chloride concentrations could adversely affect the pig iron quality or could harm the blast furnace refractory<sup>87</sup>.

Used oil could also be used in a variety of blast furnace stoves, Basic Oxygen Furnaces, sintering machines, and small heating furnaces. As in the case with the blast furnaces, potential problems with lead, zinc, and chloride would limit the types of used oil which would be acceptable. Industrial oils with low lead and chlorine levels appear to be most applicable for metallurgical processes.

High efficiency particulate controls are used on essentially all metallurgical processes in steel mills. The fabric filters, electrostatic precipitators, and high pressure drop venturi scrubbers must be sophisticated since most of the particulate emissions generated in metallurgical processes are in the difficult-to-control size range of 0.1 to 0.5 microns.

The fraction of the metals emitted due to the burning of used oil would depend on the collection efficiency of the particulate control system and on the hood capture effectiveness of the various tapping, deslagging, and metal transfer operations involved in metallurgical processes.

Entropy has not identified any air emission studies concerning the firing of used oils in metallurgical processes. The available literature is limited to an emission inventory study of two steel mills in Indiana. The study authors concluded that the percentage of lead caused by waste oil combustion increased total emissions from the sources by 15 to 65%. However, the 65% value applied to a source burning a high lead (3400 ppm) content used oil.

## Air Emissions from Used Oil Combustion Processes

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Data is needed concerning the specific types of combustion devices and metallurgical processes firing used oil, the factors which limit the types of used oil burned, and the air emissions resulting from used oil firing.

### 4.10 OTHER COMBUSTION PROCESSES

There are a number of other industrial combustion processes and furnaces that could utilize used oil fuels.

- Pulp mill recovery boilers
- Lime kilns
- Lightweight aggregate kilns
- Sulfur recovery furnaces

Air emissions data during used oil firing is needed for all of these source categories. The most important of these is the pulp mill recovery boilers since the NORA survey indicated that they account for several percent of the total used oil fuel consumption.



## SECTION 5 EMISSION INVENTORY ESTIMATES

The emissions of lead from the various types of used oil combustion processes are evaluated in this section. The emissions have been estimated based on the following types of data.

- Used oil composition (Section 3)
- Used oil consumption for each source category (Section 4)
- Lead retention in combustion processes (Section 4)
- Lead partitioning and collection (Section 4)

The used oil composition is based on the weighted average metals concentrations derived from the NORA survey. The used oil consumption data for each source category has also been taken from the NORA survey (see Table 4-2 of Section 4). The total quantities of used oil burned by each of these source categories has been determined by multiplying the fraction consumed by the total used oil fuel quantity of 800 million gallons per year.

The 800 million gallon consumption estimate is based on the NORA survey and reference 46. This consumption estimate may be in error by  $\pm 20\%$  due to the difficulty in accounting for used oil that does not enter the used oil management system and due to the inherent limitations in used oil consumption surveys. However, the lead emission estimate is being prepared primarily to evaluate the need for additional emission tests for metals emissions in the various source categories. The total used oil consumption estimate does not affect the comparison of the various source categories.

The emission estimates for each source category have been calculated based on estimated partitioning factors and lead control efficiency levels. The supporting data for these estimates is included in Section 4.

### 5.1 LEAD SOURCES

#### 5.1.1 Space Heaters

The lead retention value for space heaters has been assumed to be 50% of the total lead entering with the used oil fuel. 49% of the lead has been assumed to leave the combustion chamber as particulate matter. The remaining 1% has been assumed to be present in the vapor state. Since these units do not have air

pollution control devices, a zero percent removal efficiency for the emitted particles has been used. These partitioning factors and the control efficiency have been entered on Table 5-1.

The NORA survey did not provide information concerning the quantities of used oil burned in space heaters. The organizations involved in used oil marketing and processing would usually not have information on used oil consumption for space heaters since these units operate on used oil generated on-site or locally without processing. For the purposes of this study, Entropy has assumed that 6,500 units are sold each year and that the average life of space heaters is 10 years. Accordingly, the total population of operating space heaters is estimated at 65,000. The used oil consumption has been estimated based on an average heater capacity of 0.3 million Btus/hr. and an average operating time of 500 hours per year. This yields a total used oil consumption rate of 69 million gallons per year for space heaters in the U.S.

#### 5.1.2 Commercial Boilers

The used oil consumption rate for commercial boilers reported in the NORA survey was 351,000 gallons. This has been multiplied by a factor of 3.583 to account for used oil consumers not included in the NORA survey. This results in a total U.S. consumption of approximately 1,300,000 gallons in apartment house, commercial building, and school boilers.

The lead emission rate for this source category is based on a lead retention in the combustion chamber of 50%. As in the case of space heaters, it is assumed that 49% of the lead leaves the combustion system as particulate, and 1% leaves as vapor. A zero percent control efficiency has been assumed for the particulate since the lead-containing particles are too small to be collected by low efficiency control systems. All of the vapor phase lead is assumed to be emitted.

#### 5.1.3 Industrial Boilers

The partitioning factors and control efficiency factors for industrial boilers are identical to those chosen for space heaters and commercial boilers.

The total used oil consumed in the population of sources represented by the NORA survey was 28,300,085 gallons. This has been multiplied by 3.583 to yield a total U.S. consumption of approximately 101,400,000 gallons per year in industrial boilers.

## 5 Marine Boilers

The assumptions used with respect to industrial boilers are also appropriate for marine boilers since these have similar design characteristics and heat input rates. As in the case with industrial boilers, marine boilers do not have effective air pollution control systems for metal-containing particulate.

The total used oil consumption rates for marine boilers indicated on the NORA survey was 10,605,250 gallons. Based on this value, the total U.S. consumption of used oil in marine boilers has been estimated at 28,300,000 gallons.

### 5.1.5 Utility Boilers

The partitioning factors used in this report for utility boilers are considerably different than those for smaller combustion sources. A lead retention factor of zero has been used for the combustion chamber despite the high probability that moderate quantities of lead accumulate on heat exchange surfaces. A zero value is used since utility boilers are routinely cleaned to remove deposits that reduce heat transfer efficiency. The materials used to clean these surfaces are often collected and burned in the boiler when it is back in service. Accordingly, a portion of the metal deposits could be reemitted.

Many of the utility boilers firing used oil, use coal as the main fuel. A portion of the lead released from the used oil nucleates on the bottom ash leaving the boiler. A value of 20% has been used to account for the lead exiting with the bottom ash. Most of the remaining (79%) of the lead nucleates on the surfaces of coal flyash. A very small fraction (1%) may remain as a vapor.

An efficiency of 90% is used as an average value for coal- and oil-fired utility boilers that burn used oil. This value takes into the account the 99% removal efficiencies which should exist for coal-fired units with high efficiency particulate systems and the relatively low efficiencies which should exist for oil-fired units which generally do not require sophisticated particulate control systems due to the relatively low level of particulates emitted from such units.

The used oil consumption rate for utility boilers indicated by the NORA survey was 25,780,000 gallons. This has been multiplied by 3.583 to yield a U.S. consumption of 92.3 million gallons.

#### 5.1.6 Asphalt Plants

The consumption of used oil at asphalt plants indicated by the NORA survey was 87,462,094 gallons. The total U.S. consumption of used oil by asphalt plants has been estimated at 313,400,000 gallons based on the multiplication factor discussed earlier.

The lead emission rate estimate for asphalt plants is based on a lead capture efficiency of 20% on the aggregate passing through the drier. Most of the remaining lead (79%) has been assumed to nucleate on particles entrained in the gas stream. As in the case with the other source categories, it is also assumed that 1% of the lead remains in the vapor state.

A lead-containing particle removal efficiency of 75% has been assumed for the asphaltic concrete industry. This relatively low value takes into account the 99% efficiencies expected for fabric filter controlled units and the 60% removal efficiency estimated for lead-containing particles in wet scrubber controlled plants. The actual performance of asphalt plant wet scrubbers is probably greater than the 60% value chosen. Accordingly, the actual emissions may be lower than indicated in Table 5-1.

#### 5.1.7 Cement Plants

The dominant factor in the case of cement plants is the overall particulate control efficiency for the main kiln gas stream, the alkali bypass gas stream, and the clinker cooler gas stream. The efficiency for metal-containing particulate has been assumed to be 95%. This takes into account the reduced efficiency of electrostatic precipitators in the 0.1 to 0.5% range. In calculating the emissions, it has been assumed that 20% of the metals are captured in the clinker and are not entrained in any of the gas streams.

The total consumption of used oil at cement plants reported in the NORA survey was 9,798,500 gallons. The total U.S. consumption by cement plants has been estimated at 35,100,000 gallons based on the results of the NORA survey.

#### 5.1.8 Steel Mills

An overall collection efficiency of 95% for lead-containing particles has been used for steel mill operations. This factor takes into account the efficiency of the air pollution control

systems and the fugitive emissions around the hoods on the process equipment. In selecting the 95% value, very high (99+%) removal efficiencies were assumed for the particulate control systems. This is reasonable since essentially all of the particles generated in metallurgical processes are small. The control systems are designed to collect the particles in the same size range as the lead-containing particles. The 95% value is needed due to the intermittent operations such as tapping, deslagging, and material transfer where particulate can escape capture in process hoods or building ventilation systems.

The total consumption of used oil by steel mills reported in the NORA survey was 24,115,000 gallons. Based on the 3.583 factor used throughout this section, the total U.S. consumption of used oil at steel mills has been estimated at 84,600,000 gallons.

#### 5.1.9 Pulp Mills

Due to the temperature of the molten smelt formed in the lower section of the pulp mill recovery boilers, it is assumed that all of the lead leaves the unit in the form of particles (98%) and vapor (2%). None of it is probably leaving with the molten smelt. A removal efficiency of 95% has been assumed for the lead containing particulate matter. This relatively low efficiency takes into account the reduced efficiencies achieved by electrostatic precipitators on particles in the submicron size range.

The total consumption of used oil by pulp mills reported in the NORA survey was 7,921,500 gallons. Accordingly, the total U.S. consumption was estimated at 28,300,000 gallons.

### 5.2 LEAD EMISSION ESTIMATES

The lead emission rates were estimated in Table 5-1 based on an average concentration of 40.36 ppm concentrations in used oil. The partitioning of the other metals (weighted average concentrations listed below) is not necessarily the same as lead. Accordingly, there are differences in the fractions of each metal which are emitted. These differences may affect the relative importance of each source category as a source of metal emissions.

|          |       |     |
|----------|-------|-----|
| Lead     | 40.36 | ppm |
| Arsenic  | 1.19  | ppm |
| Cadmium  | 1.60  | ppm |
| Chromium | 8.83  | ppm |
| Barium   | 10.0  | ppm |
| Zinc     | 917.0 | ppm |

## Emission Estimates

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Due to the number of assumptions necessitated by the lack of firm data, these calculations provide only an order-of-magnitude (or "ballpark") estimate of emissions. However, several conclusions can be based on a comparison of the estimates for the various sources. These are discussed in the following sections.

### \* 5.2.1 Space Heaters

Space heaters are an important source of lead emissions due to the limited retention in the combustion chamber and the lack of an air pollution control. Also, emissions from these units could create locally high concentrations of metal-containing particulate due to the limited dispersion characteristics of the stacks.

Information is needed to confirm the metals retention levels of typical air atomizing space heaters presently on the market. Also, data is needed concerning the numbers of operating units, the capacities of these units, and the average operating hours per year. The Vermont Study will provide the needed data.

### 5.2.2 Commercial Boilers

This category appears to be relatively unimportant due to the low consumption rates for used oil. However, these units have very limited emission dispersion capability due to the low stacks, low gas temperatures, and close proximity to densely populated areas.

### 5.2.3 Industrial Boilers

This category of sources is important due to the relative large quantities of used oil consumed and the uncertainty regarding metals retention in the boiler. It is also possible that emission testing would indicate that certain metals such as chromium and barium are generated primarily in large particle size ranges which are collected with modest efficiency by the multi-cyclone collectors used on large industrial boilers. Air emission testing should be conducted at one or more typical industrial boilers.

### 5.2.4 Marine Boilers

Marine boilers do not have as great an environmental impact as industrial boilers since they consume less used oil fuel and

Emission Estimates

Table 5-1. Lead Emission Estimates

| Type of Combustion Process | Used Oil Gals./Yr. x 10 <sup>4</sup> | Lead Partitioning       |                              |                              |                              | On Particles in Gas Stream % | Vapor % | Particulate Coll. Eff. | Lead Emissions Tons/year (Vapor & Uncollected Particulate) |
|----------------------------|--------------------------------------|-------------------------|------------------------------|------------------------------|------------------------------|------------------------------|---------|------------------------|--|
|                            |                                      | Retained as Deposits, % | Retained on Process Solids % | Retained on Process Solids % | On Particles in Gas Stream % |                              |         |                        |  |
| Space Heaters              | 69.0                                 | 50                      | 0                            | 0                            | 49                           | 1                            | 0       | 5.05                   |  |
| Commercial Boilers         | 1.3                                  | 50                      | 0                            | 0                            | 49                           | 1                            | 0       | 0.10                   |  |
| Industrial Boilers         | 101.4                                | 50                      | 0                            | 0                            | 49                           | 1                            | 0       | 7.42                   |  |
| Marine Boilers             | 38.0                                 | 50                      | 0                            | 0                            | 49                           | 1                            | 0       | 2.78                   |  |
| Utility Boilers            | 92.3                                 | 0                       | 20                           | 20                           | 79                           | 1                            | 90      | 1.20                   |  |
| Asphalt Plants             | 313.4                                | 0                       | 20                           | 20                           | 79                           | 1                            | 75      | 9.52                   |  |
| Cement Plants              | 35.1                                 | 0                       | 20                           | 20                           | 79                           | 1                            | 95      | 0.24                   |  |
| Steel Mills                | 86.4                                 | 0                       | 0                            | 0                            | 99                           | 1                            | 95      | 0.75                   |  |
| Pulp Mills                 | 28.3                                 | 0                       | 0                            | 0                            | 99                           | 1                            | 90      | 0.45                   |  |
| Other                      | 34.8                                 | 0                       | 0                            | 0                            | 99                           | 1                            | 90      | 0.55                   |  |
| Total                      | 800.0                                | -                       | -                            | -                            | -                            | -                            | -       | 28.06                  |  |

PRELIMINARY ESTIMATES

since some of the emissions occur away from populated areas. Air emissions data generated for industrial boilers would also be helpful in further evaluating marine boiler emissions.

#### 5.2.5 Utility Boilers

This is one of the most important source categories. The utility boiler sector is a major used oil consumer as indicated by the estimated firing rate of 92.3 million gallons per year. Furthermore, there is uncertainty regarding the extent to which metals are retained in the combustion chambers. There is also uncertainty regarding the metal-containing particulate control efficiency at oil-fired units with only low efficiency particulate control systems.

Air emission testing of one or more utility boilers is recommended. This test should be conducted on oil-fired unit with a low efficiency particulate control system.

#### 5.2.6 Asphalt Plants

Asphalt plants are clearly one of the most important source categories burning used oil. They now utilize approximately 43% of the total used oil fuel supply, and the rate of consumption has been increasing for the last several years based on information supplied by NAPA<sup>42</sup>. There is considerable uncertainty regarding the emissions rates of lead and other metals from asphalt plants controlled with wet scrubbers. The low efficiency estimate used in Table 5-1 was necessary due to the probability that a major fraction of the metal-containing particulate is in the difficult-to-control submicron size range. Air emission testing at one or more asphalt plants is recommended. Hot mix and/or drum mix plants with a venturi wet scrubber should be tested. Particle size distributions of the metal-containing particles should be measured at the inlet and outlet of these units to further evaluate the removal capability of the medium pressure drop venturi scrubbers typically used on asphalt plants.

#### 5.2.7 Cement Plants

The previously generated data concerning hazardous waste firing clearly indicates that metals are retained and collected at high efficiency at cement kilns. No air emissions testing of cement kilns is recommended. If additional data is needed regarding

this source category, it can be obtained from BIF-related air emission tests.

#### 5.2.8 Steel Mills

There are essentially no air emissions data concerning the firing of used oils in this industrial category. Also, there is very little information concerning the types of used oil burned and the specific sources where the fuel is burned. However, all of the metallurgical processes are subject to very strict particulate emissions limitations and are equipped with control devices which are designed to remove submicron, metal-containing particles. The collection efficiencies for metals emitted due to used oil firing are probably very high. For this reason, the maximum mass emissions of used oil-related metals is expected to be low compared to other source categories evaluated in the Phase I study.

#### 5.2.9 Pulp Mills

It has been assumed that the used oil consumed by pulp and paper mills is fired in the recovery boiler. If so, the emissions of metals attributable to used oil would be effectively controlled by the electrostatic precipitator serving the recovery boiler.

### 5.3 RECOMMENDATIONS FOR AIR EMISSION TESTING

There is very little relevant data available concerning used oil burning in boilers, industrial furnaces, and space heaters. The available relevant and up-to-date data indicates that the following three source categories are especially important.

- Asphalt plants
- Utility boilers
- Industrial boilers

Air emission tests should be conducted at one or more facilities in each category. The specific types of tests are discussed in Section 6 of this report.

There are a variety of other issues which could not be addressed based on the presently available information. A partial list of these includes the following.

## Emission Estimates

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- The speciation of  $\text{Cr}^{+3}$  and  $\text{Cr}^{+6}$  in "typical" used oil
- The conversions of the chromium valance states during combustion and in industrial furnaces/processes
- The particle size distributions of low volatility elements such as barium and chromium
- The extent to which low concentrations of chlorine affect the volatility of lead and other metals in used oil being burned.

## SECTION 6

### PHASE II EMISSION TESTING RECOMMENDATIONS

The proposed emission testing programs are introduced briefly in this section. Due to practical and economic limitations, a total of four tests are recommended. These tests include two at asphalt plants, one at a coal-fired utility boiler, and one at an oil-fired industrial boiler. Detailed test program recommendations and protocols will be developed as part of Phase II.

#### 6.1 ASPHALT PLANTS

Air emissions tests should be conducted at a minimum of two asphalt plants. Due to possible differences in metal retention on the aggregate solids, one of the plants tested should be a hot mix plant, and the other should be a drum mix plant. Both plants should be controlled by venturi scrubbers. All five of the following criteria are equally important and should be applied in selecting a facility to be tested.

- The owners should be willing to participate in the tests.
- The venturi scrubbers should be representative of asphalt industry scrubbers and should be in good working order.
- The rotary drier should be typical of other units in the industry and have a representative capacity.
- The unit should be scheduled to operate at the time of the year that UORC/Entropy would prefer to conduct the tests.
- Travel costs associated with the test program should be minimized.

A typical asphalt plant has a capacity of approximately 175 tons of asphaltic concrete per hour. To ensure the representativeness of the test program, the capacities of the units should be between 125 and 225 tons per hour. The units should be controlled by venturi type wet scrubbers operating at static pressure drops of 15 to 25 inches of water. The scrubbers should have liquid-to-gas ratios between 4 and 20 gallons per thousand ACFM. The scrubbers should be in relatively good condition. For several days prior to the test program, the uniformity of the used oil metals concentrations and chlorine concentrations should be checked. Oil samples should be taken in a manner that maximizes their representativeness. This could be accomplished by sampling the oil supply line to the burner or by taking a number of samples from a well mixed burn tank. The samples

## Phase II Emission Test Recommendations

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should be analyzed for lead, barium, cadmium, chromium (total and Cr<sup>6+</sup>), arsenic, zinc, and total chlorides.

The units should operate for several days using the used oil supply to "condition" the ductwork and air pollution control systems. U.S. EPA Reference Method SW 846-0012 (multi-metals) should be used for sampling the stack emissions from the wet scrubber systems. This train uses from four to seven impingers containing solutions of acid hydrogen peroxide and acidic potassium permanganate. These reagents collect gaseous metal emissions effectively. Analyses are performed using inductively coupled argon plasma emission spectroscopy (ICAP), atomic absorption spectroscopy (AA) or graphite furnace atomic absorption spectroscopy (GFAA). The emissions of total chromium and hexavalent chromium should be measured using U.S. EPA Reference Method SW 846-0013.

Metals sampling should also be conducted at the inlets to the wet scrubber systems. This data can be used in conjunction with the stack test data to evaluate the overall metals removal efficiency of the scrubbers. The inlet data can also be used to evaluate the fraction of each metal partitioned to the entrained particulate stream and to the asphaltic concrete. Sampling of the asphaltic concrete should not be attempted since it would be difficult to obtain accurate analyses in the case of the drum mix plant due to the addition of the asphaltic binder in the drum.

As part of the test program, the particle size distributions at the inlet and outlet of the scrubbers should be determined using cascade impactors. Each of the impactor stages should be analyzed for lead, arsenic, and barium. Due to the metal surfaces of the cascade impactors, these tests are not useful for the analysis of low concentrations of chromium and zinc. The weight distributions of lead and arsenic should be used as an indicator of the behavior of moderately volatile metals in the asphalt plant. The weight distribution of barium should be used as an indicator of the behavior of low volatility metals.

Plant operating data should be recorded during the tests to demonstrate that it was operating in a normal manner. This data includes the aggregate temperatures leaving the drum, the production rate, the type of asphaltic concrete produced, and the type of binder used.

### 6.2 UTILITY BOILERS

There are two types of utility boilers of interest: (1) units in which used oil is fired in conjunction with virgin oil, and (2) units in which used oil is fired with coal. Due to practical

## Phase II Emission Test Recommendations

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economic and technical limitations, the testing should be limited to an oil-fired boiler with a low efficiency particulate control system. The potential emissions from a coal-fired boiler with supplemental used oil fuel is relatively low. Furthermore, it would be difficult to identify the very slight impact of the metals entering with the used oil fuel from the metals entering with the coal.

In selecting candidate sites for testing, UORC should consider the following criteria.

- The operating company should be willing to participate in the test program.
- One unit should be 100% oil fired and be equipped with multi-cyclone collectors. The used oil firing rate should be sufficient to allow measurement of possible metals emissions.
- The utility boiler should be representative of other oil fired utility boilers and have not been the subject of testing of high vanadium oils or other materials which may have "conditioned" the heat exchange surfaces.
- The units should fire used oil on a continuous basis, or they should be able to fire used oil for one to two weeks prior to the test program to ensure that all of the internal surfaces are at an equilibrium condition with respect to conditioning.
- Travel costs should be minimized.

Typical oil-fired utility boilers have generating capacities between 100 and 500 megawatts. Prior to the test program, the characteristics of the used oil fired in the unit should be evaluated to confirm that conditions during the emission test are representative of normal, "conditioned" operation.

The emissions tests should be conducted using the same procedures recommended for asphalt plants. Chromium emissions should be measured at the stack using Method SW 846-0013. The other metals should be measured using SW 846-0012. The weight distributions of lead, arsenic, and barium should be measured using cascade impactor sampling on the inlet and outlet of the collector.

Samples of the mechanical collector ash should be obtained and analyzed for the six metals. If possible, samples of internal deposits at various locations in the boiler should be obtained soon after the used oil air emissions tests. This data would be

helpful in confirming the retention of metals in the combustion chamber and on the surfaces of the heat recovery equipment.

The operating conditions of the boiler should be carefully documented during the test. The necessary data includes but is not limited to the following.

- Air pollution control system operating parameters
- Stack opacity as indicated by the continuous emissions monitor and by visible emission observations
- Boiler oxygen (economizer outlet) and carbon monoxide concentrations
- Boiler load
- Boiler gas temperatures
- Types and quantities of fuel fired for approximately two weeks prior to and during the tests
- Soot blowing practices prior to and during the tests

### 6.3 INDUSTRIAL BOILER

The air emission test on an industrial boiler should be conducted. The unit should be designed for burning oil fuel and have a rated heat input capacity between 10 and 100 million BTUs per hour. Some of the other criteria include the following.

- The operating company should be willing to participate in the test program.
- One unit should be 100% oil fired and equipped with multi-cyclone collectors. The used oil firing rate should be sufficient to allow measurement of possible metals emissions.
- The units should fire used oil on a continuous basis, or they should be able to fire used oil for one to two weeks prior to the test program to ensure that all of the internal surfaces are at an equilibrium condition with respect to conditioning.
- Travel costs should be minimized.

The tests would be very similar to those discussed with respect to utility boilers. However, there is no need for testing prior to a control device since it is assumed that metals-containing particles are not collected efficiently in these units.

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**APPENDIX**

**NORA Survey Form and Cover Letter**



# National Oil Recyclers Association

Kathryn McWilliams, *Executive Director*

12429 Cedar Road • Suite 26 • Cleveland, Ohio 44106-3172 • (216) 791-7316 • FAX (216) 791-6047

TO: Used Oil Marketers  
FROM: Kathryn McWilliams  
SUBJECT: Used Oil Management Survey  
DATE: September 23, 1993

As you are aware, the issue of burning used oil for energy recovery has been highly controversial. Critics have charged that substantial quantities of lead — 588,000 pounds per year — are emitted into the air each year. (Among many other questionable assumptions, this so-called "statistic" assumes that all lead in used oil is released into the air.)

The National Oil Recyclers Association ("NORA") believes that the facts on air emissions from used oil should speak for themselves. Consequently, NORA, together with the Used Oil Recycling Coalition, has launched an objective, scientifically valid study of used oil burning in the United States. The Study will be conducted by Entropy, Inc., a highly respected consultant in air emissions research.

A crucial component of this study is up-to-date information on used oil — how much is sold as fuel, who burns it, etc. This information can only be obtained from used

Will you take a few minutes to fill out the enclosed survey? The information you provide will help set the record straight on the facts relating to burning used oil for energy recovery.

Please be as accurate as possible. Please return the survey in the enclosed envelope as soon as possible. The answers you provide will not be available to any governmental agency or to your competitors.

Many thanks for your help. If you have any questions, please call me at (216) 791-7316.

Enclosures

Christopher Harris  
*General Counsel*  
(202) 639-6320, FAX (202) 628-4912

Dan Cowart  
*President*  
(205) 666-8143, FAX (205) 433-3168

**USED OIL MARKETER/PROCESSOR SURVEY**

Send Completed Questionnaire To Entropy (Envelope Enclosed)  
October 15, 1993

Name of Person Completing Form \_\_\_\_\_  
Company \_\_\_\_\_  
Position \_\_\_\_\_  
Telephone # \_\_\_\_\_

**Note: (For each question, use 1992 data or last 12 months, whichever is more accurate or complete.)**

1. How much used oil do you collect or receive each year? \_\_\_\_\_ gallons

2. Do you collect or receive used oil from any of the following sources?

For each source of used oil provide approximate percentage.

Automotive Services Industry \_\_\_\_\_%       Government Facilities \_\_\_\_\_%  
(including public schools)

Other Industry \_\_\_\_\_%       Other \_\_\_\_\_%

3. Do you participate in a Do-It-Yourself ("DIY") collection program?       yes     no

If so, is the program administered by:  
\_\_\_\_\_ industry  
\_\_\_\_\_ local government  
\_\_\_\_\_ state government  
\_\_\_\_\_ other

4. If you participate in a DIY program, what is your estimate of how much DIY used oil you collect each year?

\_\_\_\_\_ gallons

5. How much used oil do you re-refine each year?

\_\_\_\_\_ gallons

6. How much used oil do you sell as fuel each year?

\_\_\_\_\_ gallons

7. How much used oil do you sell to a blender or other processor or marketer each year?

\_\_\_\_\_ gallons

Other than re-refining or direct sale as fuel or to another marketer, processor or blender, what other use is made of your used oil?

Specify \_\_\_\_\_

8. Of the used oil sold as fuel by your company during 1993, what percent is "specification"<sup>1</sup> used oil fuel?

\_\_\_\_\_ %

9. Do you blend used oil with virgin petroleum to bring fuel into "specification"?

yes       no

10. During 1993, what is the approximate average of the levels of the following metals for both specification and non-specification used oil fuels marketed by your company? Also, what is the water content?

Specification

Non-Specification

Lead            \_\_\_\_\_ ppm  
 Chromium     \_\_\_\_\_ ppm  
 Cadmium      \_\_\_\_\_ ppm  
 Arsenic       \_\_\_\_\_ ppm  
 Water         \_\_\_\_\_ %

Lead            \_\_\_\_\_ ppm  
 Chromium     \_\_\_\_\_ ppm  
 Cadmium      \_\_\_\_\_ ppm  
 Arsenic       \_\_\_\_\_ ppm  
 Water         \_\_\_\_\_ %

11. To what types of facilities do you market used oil fuel? (provide approximate percentage of volume of fuel sold)

|   |                          |         | Facility Equipped<br>With Pollution Controls? |                          |
|---|--------------------------|---------|---|--------------------------|
|   |                          |         | YES   | NO                       |
| Asphalt Plants                                | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Cement/Lime Kilns                             | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Steel Mills                                   | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Paper Mills                                   | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Electric Utilities                            | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Other Power Plants                            | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Other Industrial Burners                      | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Green Houses                                  | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Marine Vessels                                | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Residences (including apartment buildings)    | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Other Institutions (hospitals, schools, etc.) | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Other Burners                                 | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |
| Other Processors, Marketers or Blenders       | <input type="checkbox"/> | _____ % | <input type="checkbox"/>                      | <input type="checkbox"/> |

<sup>1</sup> "Specification" used oil fuel is defined by the U.S. Environmental Protection Agency in 40 C.F.R. § 266.40 [recodified at 40 C.F.R. § 279.11].