

CHAPTER 2. CHARACTERIZING PB AIR QUALITY AND EMISSIONS DATA

This chapter describes the available Pb air quality and emission data used to inform and develop the controls strategies outlined in this RIA. We first describe data sources for air quality measurement. We then provide an overview of data on Pb emission sources contained in available EPA emission inventories. For a more in-depth discussion of Pb air quality and emissions data, see the OAQPS Staff Paper for the Pb NAAQS.¹

2.1 Air Quality Monitoring Data

Ambient air Pb concentrations are measured by four monitoring networks in the United States, all funded in whole or in part by EPA. These networks provide Pb measurements for three different size classes of airborne particulate matter (PM): total suspended PM (TSP), PM less than or equal to 2.5 μm in diameter ($\text{PM}_{2.5}$), and PM less than or equal to 10 μm in diameter (PM_{10}). The networks include the Pb TSP network, the $\text{PM}_{2.5}$ Chemical Speciation Network (CSN), the Interagency Monitoring of Protected Visual Environments (IMPROVE) network, and the National Air Toxics Trends Stations (NATTS) network. The subsections below describe each network and the Pb measurements made at these sites.

In addition to these four networks, various organizations have operated other sampling sites yielding data on ambient air concentrations of Pb, often for limited periods and/or for primary purposes other than quantification of Pb itself. Most of these data are accessible via EPA's Air Quality System (AQS): <http://www.epa.gov/ttn/airs/airsaq/>. In an effort to gather as much air toxics data, including Pb, into one database, the EPA and National Association of Clean Air Agencies (NACAA) created the Air Toxics Data Archive. The Air Toxics Data Archive can be accessed at: <http://vista.cira.colostate.edu/atda/>.

2.1.1 Ambient Pb Measurement Methods

A number of methods are used to collect Pb and measure Pb concentrations in the atmosphere. Most methods use similar sample collection approaches. Ambient air is drawn through an inlet for a predetermined amount of time (typically 24 hours) and the PM is collected on a suitable filter media. After the sample has been collected, the filter may be used to determine the mass of PM collected prior to then being used for determination of Pb. The filter is chemically extracted and analyzed to determine the Pb concentration in the particulate material. The concentration of Pb found in the atmosphere, in $\mu\text{g}/\text{m}^3$, is calculated based on the concentration of Pb in the volume extracted, the size of the collection filter, and the volume of air drawn through the filter.

The primary factors affecting the measurements made are the sampling frequency, duration of sampling, type of inlet used, volume of air sampled, and the method of

¹ U.S. Environmental Protection Agency (2007c), Review of the National Ambient Air Quality Standards for Lead: Policy Assessment of Scientific and Technical Information, OAQPS Staff Paper, Chapter 2, EPA-452/R-07-013, Office of Air Quality Planning and Standards, RTP, NC.

analyzing the filter for Pb content. The following paragraphs describe how these factors affect the Pb measurements.

2.1.2 Inlet Design

In ambient air monitors, a number of inlet designs have been developed that allow certain particle size ranges to be sampled. The inlets use either impaction or cyclone techniques to remove particles larger than a certain size (the size cutpoint) from the sample stream. Three particle size cutpoints are used in ambient Pb measurements including TSP, PM_{2.5}, PM₁₀. The TSP inlet is designed to allow as much suspended particulate into the sampling device as possible while protecting against precipitation and direct deposition on to the filter (nominally 25 to 45 micrometers) (USEPA, 2004c). Sampling systems employing inlets other than the TSP inlet will not collect Pb contained in the PM larger than the size cutpoint. Therefore, they do not provide an estimate of the total Pb in the ambient air. This is particularly important near sources which may emit Pb in the larger PM size fractions (e.g., fugitive dust from materials handling and storage).

2.1.3 Volume of Air Sampled

The amount of Pb collected is directly proportional to the volume of air sampled. Two different sampler types have evolved for PM and Pb sampling – a high-volume and a low-volume sampler. High-volume samplers draw between 70 and 100 m³/hr of air through an 8 inch by 10 inch filter (0.05 m² filter area). Low-volume samplers typically draw 1 m³/hr through a 47 mm diameter filter (0.002 m² filter area). Currently all Federal Reference Method (FRM) and Federal Equivalence Method (FEM) for Pb-TSP are based on high-volume samplers.

2.1.4 Sampling Frequency

The frequency of Pb sampling used in the U.S. varies between one sample every day (1 in 1 sampling) to the more common frequency of one sample every 6 days (1 in 6 sampling). Semi-continuous methods for the measurement of ambient metals (including Pb) are currently being explored which would allow for more frequent sampling (as frequent as 1 sample per hour), but much more work is needed on these methods before they can be deployed in a network setting.

More frequent sampling reduces the uncertainty in estimates of quarterly or annual averages associated with temporal variations in ambient concentrations. However, the costs of sampling and analysis are directly tied to sample frequency. As such, it is necessary to evaluate the reduction in measurement error versus the increase in sampling and analysis costs when selecting the required sampling frequency. A discussion of the observed temporal variation of Pb measurements is given later in this section.

2.1.5 Sample Analysis

After the samples have been collected on filters and the filters have been weighed, the filters are analyzed for Pb content. A number of analytical methods can be used to analyze the filters for Pb content including x-ray fluorescence analysis (XRF), proton-

induced x-ray emission (PIXE), neutron activation analysis (NAA), atomic absorption (AA), or inductively-coupled plasma mass spectrometry (ICP/MS) (CD, pp. 2-80 to 2-81). A detailed discussion of these methods was given in the 1986 CD (USEPA, 1986), and the reader is referred to that document for more information on these analytical methods. A search conducted on the AQS database² shows that the method detection limits for all of these analytical methods (coupled with the sampling methods) are very low, ranging from 0.01 $\mu\text{g}/\text{m}^3$ to as low as 0.00001 $\mu\text{g}/\text{m}^3$, and are more than adequate for determining compliance with the current NAAQS.

2.1.6 Pb-TSP

This network is comprised of state and locally managed Pb monitoring stations which measure Pb in TSP, i.e., particles up to 25 to 45 microns. These stations use samplers and laboratory analysis methods which have either FRM or FEM status. The FRM and FEM method descriptions can be found in the U.S. Code of Federal Regulations, Section 40 part 50, Appendix G. Sampling is conducted for 24-hour periods, with a typical sampling schedule of 1 in 6 days. Some monitoring agencies “composite” samples by analyzing several consecutive samples together to save costs and/or increase detection limits.

2.1.7 Monitor Locations

The locations of Pb-TSP sites in operation between 2003 and 2005 are shown in Figure 2-1. The state and local agencies which operate these sites report the data to EPA’s AQS where they are accessible via several web-based tools. EPA’s series of annual air quality trends reports have used data from this network to quantify trends in ambient air Pb concentrations. The most recent Trends report for Pb-TSP can be found at <http://www.epa.gov/airtrends/lead.html>.

A review of the Pb-TSP network's coverage of the highest Pb emitting sources (as identified in the current version of the 2002 NEI) was conducted as part of preparing this document. This review indicates that many of the highest Pb emitting sources in the 2002 NEI do not have nearby Pb-TSP monitors. This review indicates that only 2 of 26 facilities (both Pb smelters³) identified as emitting greater than 5 tpy have a Pb-TSP monitor within 1 mile. The lack of monitors near large sources indicates we are likely currently underestimating the extent of occurrences of relatively higher Pb concentrations. Additionally, none of the 189 Pb-TSP are located within a mile of airports identified in the NEI as an airport where piston-engine aircraft operate (i.e., aircraft that still use leaded aviation fuel). However, there are historical data for 12 Pb-TSP monitoring sites operating within 1 mile of such airports (going back to 1993). Nine of these sites reported maximum quarterly mean values (for 1993-2002) that ranged from 0.03 to 0.06 $\mu\text{g}/\text{m}^3$ and across all 12 sites, the maximum quarterly mean values ranged from 0.004 to 0.15 $\mu\text{g}/\text{m}^3$.

² EPA’s AQS can be accessed at <http://www.epa.gov/ttn/airs/airsaqs/>

³ Primary and secondary smelters were the source types given particular priority at the time of the last Pb NAAQS review (USEPA, 1990; USEPA, 1991).

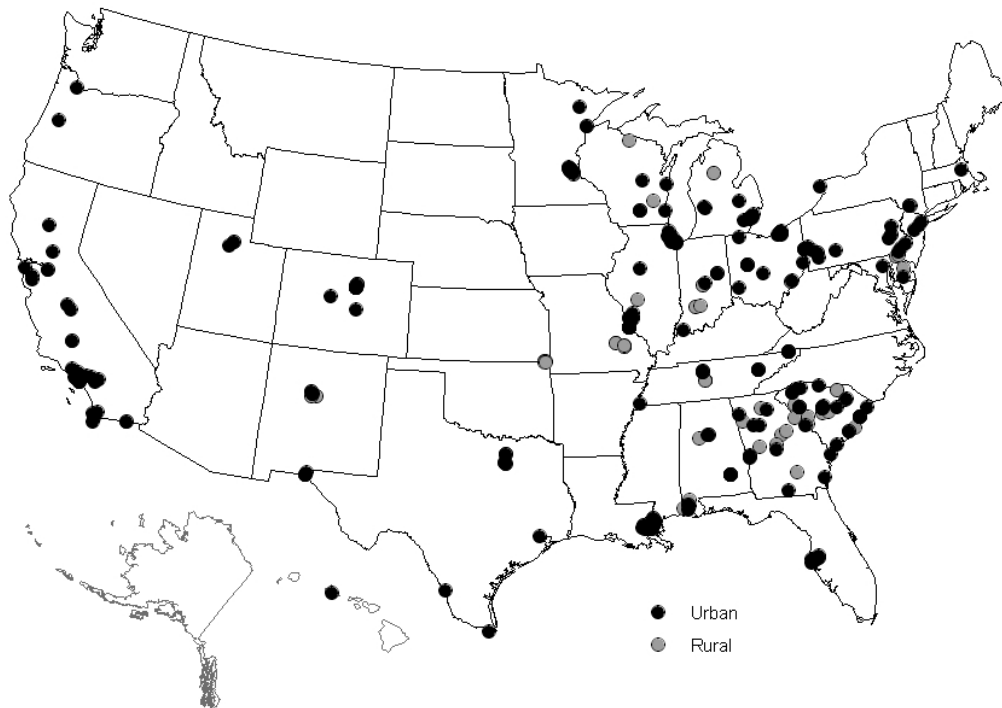


Figure 2-1. Pb-TSP monitoring sites: 2003-2005.

The number of sites in the Pb-TSP network has decreased significantly since the 1980s. The number of sites in the network reached its highest point in 1981 (946 sites). About 250 sampling sites operated during 2005. This decline in the number of Pb-TSP sites is attributable to the dramatic decrease in Pb concentrations observed since the 1980s and the need to fund new monitoring objectives (e.g., PM_{2.5} and ozone monitoring). Lead-TSP sites in lower concentration areas were shut down to free up resources needed for monitoring of other pollutants such as PM_{2.5} and ozone.

2.2 Air Quality Modeling

As part of the Agency's national air toxics assessment (NATA) activities, a national scale assessment of hazardous air pollutants including Pb compounds has been performed twice over the past few years (USEPA 2006c, 2002c, 2001a). These two assessments included the use of the NEI for the years 1996 and 1999, respectively, with atmospheric dispersion modeling to predict associated annual average Pb air concentrations across the country. A national scale assessment is not yet available based on the 2002 NEI. A number of limitations are associated with the 1996 and 1999 ambient concentration estimates and the underlying emissions estimates.

Historical studies show that Gaussian dispersion models, such as ASPEN, typically agree with monitoring data within a factor of 2 most of the time. In the case of Pb in the NATA assessment, model estimates at monitor locations were generally lower than the

monitor averages for Pb, suggesting that the modeling system (i.e., emissions estimates, spatial allocation estimates, dispersion modeling) may be systematically underestimating ambient concentrations. This may be particularly true for Pb as metals tend to deposit rapidly with distance from the source according to their particle size and weight. The model-to-monitor analysis is described in detail at <http://www.epa.gov/ttn/atw/nata1999/99compare.html>. The modeling system underestimation may also be due in part to a lack of accounting for emissions re-entrainment (these "re-entrained" particles may be observed by the monitors, but they are not accounted for in the emissions inventory, and thus would not contribute to the model estimate). For more details on the limitations of the 1999 NATA national scale assessment, see <http://www.epa.gov/ttn/atw/nata1999/limitations.html>.

For more information on Pb modeling, see section 2.4 of the OAQPS Staff Paper for the Pb NAAQS.⁴ For reasons discussed in section 3.1.1, we did not use an air quality model for this analysis.

2.3 Sources of Pb Emissions to Ambient Air

The primary data source for this discussion is the National Emissions Inventory (NEI) for 2002 (USEPA, 2007a). As a result of Clean Air Act requirements, emissions standards promulgated for many source categories that have taken effect since 2002 are projected to result in much lower emissions at the current time or in the near future. For a more comprehensive discussion of Pb sources, see section 2.2 of the OAQPS Staff Paper for the Pb NAAQS.⁵

2.3.1 Types of Pb Sources

Lead is emitted from a wide variety of source types, some of which are small individually but the cumulative emissions of which are large, and some for which the opposite is true. The categories of Pb sources estimated via the 2002 NEI to emit –as a category- more than five tons per year (tpy) of Pb are listed in Table 2-2. Note that there is often greater inherent uncertainty in reported emissions from small sources than from larger sources, and that a large portion of the lead inventory consists of sources emitting less than than five tons per year of lead.

2.3.1.1 Stationary Sources

The main sources of emissions in the 2002 NEI are comprised primarily of combustion-related emissions and industrial process-related emissions. Point source emissions account for about 66% of the national Pb emissions in the 2002 NEI. The

⁴ U.S. Environmental Protection Agency (2007c), Review of the National Ambient Air Quality Standards for Lead: Policy Assessment of Scientific and Technical Information, OAQPS Staff Paper, section 2.4, EPA-452/R-07-013, Office of Air Quality Planning and Standards, RTP, NC.

⁵ *Ibid.*, section 2.2.

point source emissions are roughly split between combustion and industrial processes, while mobile, non-road sources (e.g. piston-engine aircraft using leaded fuel) account for 29%.

Table 2-1 presents emissions estimates for stationary sources grouped into descriptive categories. Presence and relative position of a source category on this list does not necessarily provide an indication of the significance of the emissions from individual sources within the source category. A source category, for example, may be composed of many small (i.e., low-emitting) sources, or of just a few very large (high-emitting) sources.

Table 2-1. Source categories emitting greater than 5 tpy of Pb

ALL CATEGORIES - Total tons	1371	% of Total
Mobile sources	623	45.44
Iron and Steel Foundries	83	6.05
Primary Lead Smelting	59	4.30
Industrial/Commercial/ Institutional Boilers & Process Heaters	53	3.87
Hazardous Waste Incineration	47	3.43
Secondary Lead Smelting	44	3.21
Municipal Waste Combustors	33	2.41
Military Installations	27	1.97
Pressed and Blown Glass and Glassware Manufacturing	26	1.90
Utility Boilers	23	1.68
Secondary Nonferrous Metals	22	1.60
Portland Cement Manufacturing	18	1.31
Integrated Iron & Steel Manufacturing	17	1.24
Lead Acid Battery Manufacturing	17	1.24
Stainless and Nonstainless Steel Manufacturing (EAF)	17	1.24
Mining	15	1.09
Primary Metal Products Manufacturing	13	0.95
Waste Disposal - Solid Waste Disposal	10	0.73
Primary Copper Smelting	10	0.73
Secondary Aluminum Production	9	0.66
Fabricated Metal Products Manufacturing	9	0.66
Pulp & Paper Production	9	0.66
Transportation Equipment Manufacturing	8	0.58
Electrical and Electronics Equipment Manufacturing	8	0.58
Sewage Sludge Incineration	7	0.51
Nonferrous Foundries	7	0.51
Ferroalloys Production	7	0.51
Industrial Inorganic Chemical Manufacturing	7	0.51
Industrial and Commercial Machinery Manufacturing	7	0.51
Residential Heating	6	0.44
Secondary Copper Smelting	6	0.44
Miscellaneous Metal Parts & Products (Surface Coating)	6	0.44
Commercial and Industrial Solid Waste Incineration	6	0.44
Autobody Refinishing Paint Shops	5	0.36
Coke Ovens	5	0.36
Stationary Reciprocating Internal Combustion Engines	5	0.36
Other	97	7.08

There are some 13,067 point sources (industrial, commercial or institutional) in the 2002 NEI, each with one or more processes that emit Pb to the atmosphere. Most of these sources emit less than 0.1 tpy Pb. There are approximately 1300 point sources of Pb in the NEI with estimates of emissions greater than or equal to 0.1 tpy and these point sources, combined, emit 1058 tpy, or 94% of the Pb point source emissions. In other words, 94% of Pb point source emissions are emitted by the largest 10% of these sources.

Chapter 3 of this RIA discusses our methodology for characterizing the relative contributions of stationary point sources (defined in this analysis as sources emitting > 1 ton per year of Pb), area nonpoint sources (defined in this analysis as sources emitting less than 1 ton per year), and mobile sources.

2.3.1.2 Mobile Sources

Thirty-five years ago, combustion of leaded gasoline was the main contributor of Pb to the air. In the early 1970s, EPA set national regulations to gradually reduce the Pb content in gasoline. In 1975, unleaded gasoline was introduced for motor vehicles equipped with catalytic converters. EPA banned the use of leaded gasoline in highway vehicles after December 1995. Currently, tetraethyl lead (TEL) is still added to aviation gasoline (avgas) which is used in most piston-engine powered aircraft. TEL is added to avgas to increase octane, prevent knock⁶, and prevent valve seat recession and subsequent loss of compression for engines without hardened valves. The 2002 National Emissions Inventory (NEI) estimates that lead emissions from the use of leaded aviation gasoline (commonly referred to as avgas) are 491 tons; this accounts for 29% of the air emission inventory for lead. These estimates are based on the volume of avgas supplied nationally, the concentration of lead in avgas and the retention of some lead in the engine and engine oil of these aircraft. The Department of Energy estimates that about 281 million gallons of avgas were supplied in the U.S. in 2002.⁷ In 2006 the volume was about 280 million gallons. The majority of avgas contains up to 0.56 grams of lead per liter (2.12 grams of lead/ gallon); this is referred to as 100 Low Lead (100LL). There is another grade of 100 octane avgas that contains 1.12 grams of lead per liter, but this product is not widely available. Based on newly available information, the retention of lead in the engine and oil of piston-engine aircraft was recently revised from a value of 25% which was more related to lead retention in light-duty vehicles operating on leaded fuel to 5% retention for piston-engine aircraft.⁸ Using these recently available data on lead retention, EPA now estimates that lead emissions from the use of avgas in 2002 were approximately 623 tons or 35% of the national inventory. This estimate is based on all leaded avgas used in the U.S. This estimate does not account for the fact that some lead is emitted in the local area of an airport facility and some lead is emitted at altitude. EPA's method for estimating airport-specific lead inventories is discussed in detail elsewhere.⁹

⁶ Knocking is the sound produced when some of the unburned fuel in the cylinder ignites spontaneously resulting in rapid burning and a precipitous rise in cylinder pressure that creates the characteristic knocking or pinging sound (Chevron 2005 available at: http://www.chevronglobalaviation.com/docs/aviation_tech_review.pdf).

⁷ data available at <http://tonto.eia.doe.gov/dnav/pet/hist/mgaupus1A.htm>

⁸ For more information see the memo to the docket titled "Revised Methodology for Estimating Lead Emissions from Piston-Engine Aircraft Operating on Leaded Aviation Gasoline."

⁹ See memo to the docket titled For more information see the memo to the docket titled "Revised Methodology for Estimating Lead Emissions from Piston-Engine Aircraft Operating on Leaded Aviation Gasoline."

Lead is also present as a trace contaminant in gasoline and diesel fuel and is a component of lubricating oil (CD, pp. 2-45 to 2-48). Inventory estimates from these sources are not currently available. Additional mobile sources of Pb include brake wear, tire wear, and loss of Pb wheel weights (CD, pp. 2-48 to 2-50). Emission rates for Pb from brake wear have been published but inventory estimates have not yet been developed from these data (Schauer et al., 2006). Robust estimates of Pb from tire wear and wheel weights are not available. Currently, Pb from combustion of leaded avgas is the only mobile source of Pb included in the 2002 NEI.