

Integrated Science Assessment for Lead

Appendix 1: Lead Source to Concentration

External Review Draft

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DOCUMENT GUIDE

1 This Document Guide is intended to orient readers to the organization of the Lead (Pb) Integrated Science
2 Assessment (ISA) in its entirety and to the sub-section of the ISA at hand (indicated in bold). The ISA consists of
3 the Front Matter (list of authors, contributors, reviewers, and acronyms), Executive Summary, Integrated Synthesis,
4 and 12 appendices, which can all be found at <https://cfpub.epa.gov/ncea/isa/recordisplay.cfm?deid=357282>.
5

6 Front Matter

7 Executive Summary

8 Integrative Synthesis

9 **Appendix 1. Lead Source to Concentration**

10 Appendix 2. Exposure, Toxicokinetics, and Biomarkers

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ACRONYMS AND ABBREVIATIONS

1	AQCD	Air Quality Criteria Document
2	AQS	air quality standards
3	CEC	cation exchange capacity
4	CFR	Code of Federal Regulations
5	CSN	Chemical Speciation Network
6	DI	deionized
7	DO	dissolved oxygen
8	DOC	dissolved organic carbon
9	DOM	dissolved organic matter
10	ED-XRF	energy-dispersive X-ray fluorescence spectrometry
11	EF	enrichment factor
12	EPA	Environmental Protection Agency
13	FAAS	flame atomic absorption spectroscopy
14	FEM	Federal Equivalent Method
15	FRM	federal reference method
16	FTC	freeze thaw cycles
17	HA	humic acid
18	HAP	hazardous air pollutants
19	ICP-MS	inductively coupled plasma mass spectrometry
20	IMPROVE	Interagency Monitoring of Protected Visual Environments
21	Me-L	metal-ligand
22	NAAQS	National Ambient Air Quality Standards
23	NATTS	National Air Toxics Trends Stations
24	NCore	National Core multipollutant monitoring network
25	NEI	National Emissions Inventory
26	OM	organic matter
27	OX	oxide-bound
28	PM	particulate matter
29	PTFE	polytetrafluoroethylene
30	SLAMS	state and local air monitoring stations
31	SPM	suspended particulate matter
32	SS/CAR	specifically sorbed/carbonate-bound
33	STR	soil temperature regimes
34	TSS	total suspended solids
35	TSP	total suspended particulate
36	UFP	ultrafine particle
37		

APPENDIX 1 LEAD SOURCE TO CONCENTRATION

1.1 Introduction

1 This appendix characterizes the current state of atmospheric and environmental science relevant
2 to understanding lead (Pb) exposure and Pb-related health and ecological effects described in subsequent
3 appendices. It builds on previous research reviewed in the 2013 Integrated Science Assessment for Lead
4 (Pb ISA) ([U.S. EPA, 2013](#)) and previous Pb Air Quality Criteria Documents (AQCDs) ([U.S. EPA, 2006a](#),
5 [1986](#), [1977](#)) and emphasizes relevant advances in sources and emissions, fate and transport, sampling and
6 analysis methods, and concentration trends. Because of the large body of literature on the subject, this
7 appendix focuses primarily on new studies from the United States and Canada, with a few exceptions for
8 highly relevant international publications. The scope is not limited to airborne Pb from contemporary
9 emission sources because non-atmospheric processes, as well as legacy sources, are also relevant for
10 understanding the effects of airborne Pb. For example, transport and transformation processes in soil and
11 water after deposition are also relevant. Therefore, current research in other media is also included to
12 promote understanding of airborne Pb in the context of non-atmospheric sources and media.

13 In previous ISAs, an up-to-date review of air emissions, monitoring, and concentration trends has
14 been accomplished through a combination of analysis of U.S. Environmental Protection Agency (EPA)
15 monitoring network data and a synthesis of observations reported in the peer-reviewed literature.
16 Reference data such as total emissions, coverage of network monitors, average concentrations, and
17 concentration trends can become out of date before the document is published because these data are so
18 frequently updated. To facilitate provision of the most current emissions and concentration data from the
19 Pb monitoring network, a set of relevant maps and graphics that have been routinely included in the
20 atmospheric appendix or chapter in previous ISAs are now drawn from a separate document “Overview
21 of Lead (Pb) Air Quality in the United States ([U.S. EPA, 2022](#)).” This Appendix complements the
22 Overview by providing a literature-based synthesis of recent research on Pb sources, fate and transport,
23 measurement, and concentration trends. Section 1.2 provides an overview of sources and emissions of Pb
24 in ambient air and other environmental media. Section 1.3 gives descriptions of the fate and transport of
25 Pb in air, soil, and aqueous media. Section 1.4 describes advances in Pb measurement methods, and
26 Section 1.5 describes ambient air Pb concentrations, including spatial and temporal variability on national
27 and local scales and the size distributions of Pb-bearing particulate matter (PM).

1.2 Sources of Atmospheric Pb

1 Total Pb emissions have steadily decreased for decades, largely due to the elimination of leaded
2 gasoline use in automobiles before 1996 and in later years because of reductions in emissions from metals
3 processing sources ([U.S. EPA, 2013, 2006b](#)). From 1990 to 2020, there has been a steep decline in total
4 U.S. Pb emissions from about 5 kton/year to less than 1 kton/year and a replacement of industrial sources
5 with non-road mobile sources as the dominant category of emissions, which reflects the prominence of
6 leaded aviation fuel as the largest emissions source ([U.S. EPA, 2021b](#)). Total estimated national
7 emissions from the 2017 National Emissions Inventory (NEI) were 668 tons, with 70% from emissions
8 associated with use of leaded aviation gasoline, 20% from industrial sources, and 7.0% from fuel
9 combustion. All other sources combined were estimated to account for less than 4% of total U.S. Pb
10 emissions estimated by the NEI ([U.S. EPA, 2022, 2021a](#)). Up-to-date graphics of total U.S. Pb emissions
11 estimates by source, geographic distribution of Pb emissions estimates, and the 30-year total U.S.
12 emissions estimates trends are available in “Overview of Lead (Pb) Air Quality in the United States”
13 ([U.S. EPA, 2022](#)). Not included in the NEI are Pb emissions estimates from wildland fires or resuspended
14 legacy Pb. Pb emissions from wildland fires were not estimated in the 2017 NEI but there are plans for
15 including them in the 2020 NEI scheduled for release in March 2023 (Section 1.2.4). A preliminary
16 national emissions estimate is approximately 18 tons per year, which would put wildfires as the fourth
17 largest source of Pb, behind piston engine aircraft, industrial processes, and fuel combustion.

18 While emissions inventory data are essential for understanding emissions, there are potential
19 limitations and uncertainties. A comparison across several inventories covering the same area found the
20 inventories sometimes did not include all emission sources, contained data that were not current, and
21 reported emissions that varied considerably within the same year, leading to a recommendation that
22 emissions data would benefit from data sharing, greater uncertainty analysis, and standardization of
23 emissions estimation methods ([Harris et al., 2006](#)). For context, much of the Pb in the U.S. is neither
24 emitted into air nor transported into air from other media. Non-air Pb sources include plumbing ([Santucci
25 and Scully, 2020](#); [Frank et al., 2019](#); [USGS, 2018](#); [Rosen et al., 2017](#); [Stillo and Macdonald Gibson,
26 2017](#); [Hanna-Attisha et al., 2016](#); [Pieper et al., 2015](#); [Brown et al., 2011](#)), mine waste ([Duval et al., 2020](#);
27 [Gutiérrez et al., 2020](#); [Pavlovsy et al., 2017](#)), and food ([FDA, 2022](#); [Martín-Domingo et al., 2017](#);
28 [Ritchie and Gerstenberger, 2013](#); [Guney and Zagury, 2012](#)).

29 Even airborne Pb is only partly produced by contemporary Pb emissions into the atmosphere.
30 Contemporary sources are discussed in this Section, including aviation fuel and airports (Section 1.2.1),
31 industrial emissions (Section 1.2.2), fuel combustion from stationary sources (Section 1.2.3), wildland
32 fires (Section 1.2.4), traffic-related emissions (Section 1.2.5), and volcanoes (Section 1.2.6). Substantial
33 contributions to airborne Pb can also be attributed to historical sources of airborne Pb (Section 1.2.7) and
34 non-atmospheric Pb sources, the Pb from which can in some cases become airborne through the processes
35 of suspension and resuspension (Section 1.3.4). The resulting airborne Pb concentrations observed in

1 ambient air (Section 1.5) can thus potentially be the result of a combination of contemporary atmospheric
2 sources and resuspension of historical atmospheric sources and non-atmospheric sources.

1.2.1. Aviation Gas and Airports

3 Leaded aviation gasoline, or avgas, is the largest national source of Pb emitted into the
4 atmosphere identified by the NEI and is responsible for 70% of atmospheric Pb emissions in the U.S. Pb
5 additives, usually in the form of tetraethyl Pb, prevent engine knocking that could result in sudden engine
6 failure ([U.S. EPA, 2013](#)). Most avgas is considered “100 Low Lead”, which contains 2.12 g Pb/gallon
7 ([ASTM, 2021](#); [U.S. EPA, 2013](#)). In 2017, 208M gallons of avgas were consumed in the U.S. ([FAA,](#)
8 [2020](#)), leading to ~470 tons of total Pb emissions ([U.S. EPA, 2022](#)).

9 At the time of the last review, it was reported that Pb emissions from avgas come in gaseous or
10 particulate forms ([U.S. EPA, 2013](#)). In avgas exhaust PM, Pb is largely composed of lead bromide
11 (PbBr_2) crystals coated with hydrocarbons ([Griffith, 2020](#); [U.S. EPA, 2013](#)). These particles are typically
12 under 100 nm in diameter, although they can form larger agglomerates ([Turgut et al., 2020](#)). Pb particles
13 emitted from piston engine aircraft exhaust have been observed as small as 13 nm diameter, which are
14 significantly smaller than the mode of particles emitted from vehicle exhaust (35 nm) ([Griffith, 2020](#)). Pb
15 had the highest concentration of any element measured by Inductively Coupled Plasma Mass
16 Spectrometry (ICP-MS) in PM_{10} collected directly from aircraft engine exhaust ducts (median Pb value of
17 $4.6 \times 10^6 \text{ ng/m}^3$) and was 40 times more concentrated than the next most abundant element (Na) ([Turgut](#)
18 [et al., 2020](#)). Avgas constituents, including tetraethyl Pb, can evaporate into the headspace of storage and
19 fuel tanks or be exhausted from the engine in the gas phase ([NASEM, 2021](#); [U.S. EPA, 2013](#)). Annual
20 evaporative emissions of Pb from refueling are estimated at 75 kg ([NASEM, 2021](#)).

21 Around a single airport at which leaded fuel is used, Pb in air is highest around the runways
22 ([Rahim et al., 2019](#); [Turgut et al., 2019](#); [Feinberg et al., 2016](#); [Feinberg and Turner, 2013](#)). EPA uses an
23 estimate of 7.34 g of Pb emissions during a single take-off and landing cycle to estimate airport Pb
24 emissions in the NEI ([U.S. EPA, 2013](#)). Touch-and-go operations are commonly practiced during pilot
25 training and account for up to 23% of flights, depending on the airport ([U.S. EPA, 2020c](#)). Touch-and-go
26 operations generally remain near the airport and can involve the aircraft circling overhead for hours,
27 potentially contributing lead to the local environment near airports used for training ([U.S. EPA, 2020c](#)).
28 Aircraft run-up, the series of checks performed by pilots immediately prior to take-off, contributed up to
29 82% of the 3-month average Pb concentration at one airport modeled by EPA ([U.S. EPA, 2020c](#)). Aircraft
30 engine run-up has been identified as one of the most important emission sources for ground-level Pb
31 concentrations and was estimated at one airport to burn approximately 15.3 g/second of fuel and
32 50 g/second of fuel for a single- and multiple-engine aircraft, respectively ([U.S. EPA, 2013](#); [Carr et al.,](#)
33 [2011](#)). Median run-up times measured at one airport were 40 and 63 seconds for single- and multiple-
34 engine aircraft, respectively ([U.S. EPA, 2020c](#)). These times correspond to a three-month average Pb

1 concentration of $0.092 \mu\text{g}/\text{m}^3$, though this measurement was only conducted for one three-month period
2 at one airport ([U.S. EPA, 2020c](#)). Model-extrapolation analyses for airports using leaded avgas estimated
3 3-month average Pb concentration at the site of maximum impact for some airports with high landing and
4 take-off (LTO) activity, to be $<1\text{-}475 \text{ ng}/\text{m}^3$ at one airport ([U.S. EPA, 2020c](#)) and $10\text{-}20 \text{ ng}/\text{m}^3$ at another
5 airport ([Feinberg and Turner, 2013](#)). Fuel consumption estimated at one airport during taxi ranged from
6 $1.6 \text{ g}/\text{second}$ to $5.1 \text{ g}/\text{second}$ for single- and multiple-engine aircraft, respectively ([U.S. EPA, 2013](#); [Carr](#)
7 [et al., 2011](#)). The time an aircraft spends in taxi can have a significant influence on Pb concentrations, as
8 taxi time can vary greatly ([Feinberg et al., 2016](#)). Taxiing was responsible for 12% of total Pb emissions
9 reported in one study, with about half of those emissions occurring when the aircraft was idle and
10 awaiting clearance for takeoff ([Feinberg et al., 2016](#)). These studies indicate runways to be the primary
11 hot spot for Pb emissions at airports that use avgas.

12 There are 13,117 airports and over 5,000 heliports in operation in the United States ([NASEM,](#)
13 [2021](#)). Remote states, such as Montana and Alaska, rely heavily on air transportation. Alaska contains
14 nearly 10% of the total amount of airports in the United States ([NASEM, 2021](#)). Several studies have
15 observed lower Pb emissions when air traffic is lower ([Rahim et al., 2019](#); [Zahran et al., 2017](#)). There
16 have also been observations of significantly decreased Pb concentrations near airports during precipitation
17 compared to when it is dry ([Rahim et al., 2019](#)).

18 Soil and air Pb concentrations decrease with distance from an airport. Soil samples collected from
19 an Oklahoma airport were analyzed for Pb, for which elevated soil Pb concentrations were generally
20 observed within 500 m from an airport ([McCumber and Strevett, 2017](#)). However, a few sites
21 demonstrated higher soil Pb concentrations more than 500 m from an airport, suggesting influence from
22 other sources such as industrial, historical, or non-air sources. This study also identified hot spots (10-
23 $170 \text{ mg Pb}/\text{kg}$) near fueling centers, suggesting avgas as the primary source ([McCumber and Strevett,](#)
24 [2017](#)). Twenty four-hour average air Pb concentrations ranging from $17\text{-}70.6 \text{ ng}/\text{m}^3$ were reported and
25 remained above background levels ($10 \text{ ng}/\text{m}^3$) up to 900 m from a Santa Monica airport ([Carr et al.,](#)
26 [2011](#)), illustrating the possible dispersion of Pb emissions from avgas. At the same airport, $\text{PM}_{2.5}$ Pb
27 concentrations dropped from $24 \text{ ng}/\text{m}^3$ to $\sim 6 \text{ ng}/\text{m}^3$ after shortening the runway by 450 m ([Hudda et al.,](#)
28 [2022](#)). This 75% reduction in airborne Pb concentration was attributed to a 50% decrease in aviation
29 operations following the shrinking of airport size ([Hudda et al., 2022](#)). Ambient air Pb was measured in a
30 U.S. EPA one-year monitoring study at 17 U.S. airports for a full year ending in December of 2013 ([U.S.](#)
31 [EPA, 2015](#)). Monitoring was required for a set of airports with estimated Pb emissions between 0.50 and
32 1.0 tons Pb per year that also met additional criteria including the dominant use of one runway and the
33 level of piston-engine aircraft activity. Airport Pb concentrations monitored depend on level of piston-
34 engine aircraft activity, the patterns of runway use, meteorology, and the placement of the monitor
35 relative to the run-up area, and other factors. Maximum 3-month average Pb concentrations ranged from
36 0.1 to $0.33 \mu\text{g}/\text{m}^3$ and exceeded $0.15 \mu\text{g}/\text{m}^3$ at 2 of the 17 airports ([U.S. EPA, 2015](#)).

1.2.2. Industrial Sources

1 The 2013 Pb ISA summarized emissions inventory data and source apportionment results that
2 attributed substantial amounts of airborne Pb to the metals industry ([U.S. EPA, 2013](#)). Studies identifying
3 primary smelting mainly of other metals from various ores, secondary smelting—mainly of Pb batteries,
4 and steel manufacturing as important contributors to Pb emissions were reviewed. Observations from
5 several publications of downwind airborne Pb concentrations and PM Pb content from the last remaining
6 primary Pb smelter in the U.S. were summarized, as well as emissions from smelters for other metals that
7 continue to operate. Other industrial emissions contributed less than the metals industry at the time of the
8 2013 ISA, and there were few publications and little discussion of their emissions and contribution.

9 According to the 2017 NEI, industrial sources now account for about 20% of U.S. emissions,
10 making them the second largest category of sources after aviation fuel ([U.S. EPA, 2021a](#)). Roughly half
11 of industrial emissions are from metal industries, both ferrous and non-ferrous in approximately equal
12 amounts, and emissions sources include smelters, steel mills, foundries, and metal fabrication operations.
13 The other half of industrial emissions is not related to metals processing and includes industries such as
14 glass and cement manufacturing. Recent research on industrial Pb sources and emissions is largely limited
15 to a few high-profile areas. Since publication of the 2013 Pb ISA, there have been several studies
16 published on various aspects of Pb emissions from smelters, but there are few recent studies on other
17 industrial Pb emissions, whether metals related or otherwise.

18 The highest ambient air Pb concentrations in the United States are observed near metal industry
19 sources ([U.S. EPA, 2022](#)). Historically, some large Pb smelters have been among the largest single
20 sources of U.S. Pb emissions. Together they dominated local Pb emissions and accounted for a large
21 fraction of national Pb emissions after the removal of Pb from gasoline ([U.S. EPA, 2013](#)). Recent studies
22 have also continued to identify specific smelters as major urban Pb sources ([Wang et al., 2011](#)).
23 Numerous previous field studies have documented Pb emissions from smelters as well as elevated
24 ambient air Pb concentrations in the vicinity of primary smelters and soil Pb concentrations decreasing
25 with distance from smelters ([Bowers et al., 2014](#); [U.S. EPA, 2013](#)). Recent research generally confirms
26 these earlier observations by also showing that soil Pb concentrations decreased with distance from North
27 American smelters and that isotope ratios consistent with smelter emissions could be identified in soil
28 some distance from the smelter ([Widory et al., 2018](#); [Félix et al., 2015](#)).

29 One major focus of recent research has been Pb size distributions of smelter emissions. A
30 bimodal particle size distribution with maxima at 0.18 µm and 9.9 µm was consistently observed over
31 several years of sampling in the vicinity of a large copper (Cu) smelter in Hayden AZ ([U.S. EPA, 2013](#);
32 [Csavina et al., 2011](#)). [Csavina et al. \(2014\)](#) confirmed that airborne Pb followed a bimodal particle size
33 distribution in the vicinity of industrial operations that had both mining and smelting operations in both
34 Arizona and Australia and suggested that the finer particles (<1 µm) were produced from smelters and the
35 coarser particles were from windblown dust sources like mine tailings, crushing and grinding operations,
36 and regional or nearby urban sources. Pb isotope ratios were used to show that fine particles smaller than

1 1 µm aerodynamic diameter in the vicinity of large smelters were mainly due to emissions from the
2 smelter, while the isotope signature of coarse particles near the smelter were more similar to PM from a
3 regional background aerosol or nearby urban environments ([Félix et al., 2015](#); [Csavina et al., 2014](#)). For
4 similar mining operations in the absence of a smelter, only a coarse mode was observed ([Csavina et al.,](#)
5 [2014](#)). Fugitive emissions of airborne dust studied using a Bayesian framework also reinforced that
6 substantial Pb emissions are associated with coarse PM. Pb associated with airborne dust from loading
7 and storage areas were estimated from time-dependent airborne Pb concentration measurements in
8 multiple locations in the vicinity of a Pb-Zn smelter in Trail, British Columbia, and found to make
9 significant contributions to Pb emissions ([Hosseini and Stockie, 2016](#)). As substantiated by results from
10 multiple smelters, large smelting operations can be a large local source of airborne Pb in both fine and
11 coarse PM, and Pb emissions from smelters can also have a broad area of impact because of their
12 concentration in the fine particle size range ([Csavina et al., 2014](#)).

13 There have also been advances in describing the physical and chemical properties of Pb in
14 smelter emissions. Previous speciation data from smelter emissions reviewed by [U.S. EPA \(2006b\)](#) and
15 [Skeaff et al. \(2011\)](#) are qualitative or semi-quantitative in nature ([Skeaff et al., 2011](#)). [Skeaff et al. \(2011\)](#)
16 set as their objective the development of a quantitative chemical speciation of stack particulates from
17 three Cu smelters with a mass balance as close to 100% as possible using x-ray diffraction, scanning
18 electron microscopy, and electron probe microanalysis. Acceptable mass balances were achieved, and Pb
19 accounted for 7.5% to 14% of PM by weight across the three smelters. Although insoluble PbSO₄ was
20 consistently the dominant form of Pb ([Skeaff et al., 2011](#)), another study found that in the vicinity of a
21 smelter in Hayden AZ, the PM size range most enriched in Pb overlapped with the most hygroscopic PM
22 mode ([Youn et al., 2016](#); [Sorooshian et al., 2012](#)).

1.2.3. Fuel Combustion

23 Fuel combustion contributes ~45 tons of Pb/year to the atmosphere (7% of total emissions) and
24 consists of coal (33% of total fuel combustion), biomass (16%), natural gas (14%), or oil sources (31%)
25 ([U.S. EPA, 2021a](#)). Previous reports have provided extensive background on the role of Pb in coal
26 combustion. Pb is found in coal in varying amounts (5–35 mg Pb/kg) ([U.S. EPA, 2013, 2006b](#)). Fly ash, a
27 byproduct of coal combustion, is composed primarily of silicon and oxygen ([Zierold and Odoh, 2020](#);
28 [U.S. EPA, 2006b](#)). Pb in fly ash is enriched 2–10 times compared with that in parent coal ([Zierold and](#)
29 [Odoh, 2020](#); [Wang et al., 2019](#)) for concentrations in fly ash samples ranging from 25.3–308 mg Pb/kg
30 ([Wang et al., 2019](#)) or 1.4–2120 mg Pb/kg depending on the source ([Zierold and Odoh, 2020](#)). In a study
31 performed in Colorado and the Appalachian Basin, 54% of Pb from parent coal was found in fly ash
32 particles at concentrations of 41.8 mg Pb/kg ([Swanson et al., 2013](#)).

33 Pb emissions from coal-fired power plants have decreased by 36% since 1993 due to pollution
34 control measures and plant closures, though power plants can still dominate local Pb emissions ([U.S.](#)

1 [EPA, 2020b](#); [Zierold and Odoh, 2020](#); [Gingerich et al., 2019](#)). In New Mexico, average Pb concentrations
2 in PM_{2.5} samples were 0.65 ng/m³ (range 0.20–1.04 ng/m³), with approximately 0.44 ng/m³ attributed to
3 the two nearby coal-fired power plants ([Gonzalez-Maddux et al., 2014](#)). Another study identified elevated
4 Pb concentrations on rock samples taken near power plant sites, with Pb the most enriched of the 15
5 elements analyzed by X-ray fluorescence (XRF) spectroscopy ([Nowinski et al., 2012](#)). Pb concentrations
6 were higher for the skyward facing side of the rock compared with the interior, suggestive of atmospheric
7 Pb deposition ([Nowinski et al., 2012](#)). In general, Pb concentrations are not correlated with the amount of
8 electricity generated at an individual plant ([Bray et al., 2017](#)), complicating predictions of Pb emissions
9 from coal-fired power plants.

10 Petroleum-fueled power plants emit ~6.4 g Pb/1000 L of fuel oil burned ([U.S. EPA, 2006b](#)).
11 Though there are uncertainties surrounding the concentration of Pb in crude oil ([U.S. EPA, 2013](#); [Murphy](#)
12 [et al., 2007](#)), New York City had average Pb concentrations of 3.40 ng/m³ (range 1.22–10.98 ng/m³) in
13 2009 which land use regression models associated with residual oil burning ([Ito et al., 2016](#)). Used motor
14 oil, which may be burned in personal space heaters, contains some Pb ([Murphy et al., 2007](#)). Fuel
15 extraction also contributes to elevated ambient air Pb concentrations. Several studies near the Athabasca
16 Oil Sands in Alberta, Canada report ambient air Pb concentrations ~0.35 ng/m³, though some of this Pb
17 may come from long-range or regional transport ([Graney et al., 2019](#); [Landis et al., 2019](#)), though oil
18 fields of this size are not present in the United States. Biomass fuel consumption has average Pb
19 emissions of 0.56 mg Pb/kg fuel ([U.S. EPA, 2006b](#)). Residential wood burning releases airborne Pb at
20 concentrations of 3.3–12.2 mg Pb/kg wood and 2.89–30.3 mg Pb/kg wood for woodstoves and fireplaces,
21 respectively ([U.S. EPA, 2013](#)). Pb-containing particles are ubiquitous in urban areas, indicating
22 widespread emissions from combustion sources ([Murphy et al., 2007](#)). A mode for Pb urban aerosol was
23 identified at 200 nm, though Pb was also observed in 50 nm particles, the smallest particle size detected
24 by single particle mass spectrometry ([Murphy et al., 2007](#)).

1.2.4. Fires

25 Another probable source is Pb deposited historically in forests and remobilized during wildfires,
26 as well as Pb in anthropogenic structures and vehicles mobilized when wildfires burn infrastructure. As
27 described in the beginning of Section 1.2, the 2020 NEI scheduled for release in March 2023 plans to
28 include Pb emissions from wildfires ([U.S. EPA, 2020a](#)), with preliminary emissions of approximately 18
29 tons per year, and this would put wildfires as the fourth largest source of Pb in the United States, behind
30 piston engine aircraft, industrial processes, and fuel combustion. Particulate matter from fires is mostly
31 carbonaceous, but also contains other elements in low concentrations, including Pb. Preliminary
32 emissions testing results indicate that more Pb is emitted from smoldering emissions than flaming
33 emissions, and that current emission factors are substantially lower than previous literature observations,
34 probably because of lower Pb levels in the environment due to the phase-out of leaded gasoline. Because
35 fires are the largest source of primary PM in the United States ([U.S. EPA, 2021a, 2019](#)), even trace level

1 emissions make fires a potentially important source of airborne Pb. Limited studies have observed Pb
2 concentrations attributed directly to smoke from wildfires. Aside from the plan to add Pb emissions from
3 wildfires to the NEI, recent evidence of Pb from fires is not typically emissions based. Without reliable
4 emissions data, studies often focus on air, ash, and soil concentrations to evaluate Pb from fires. This
5 Section is therefore organized as follows: air concentration studies from the previous 2013 Pb ISA,
6 followed by new air concentration studies, and ending with ash and soil measurement studies.

7 The 2013 Pb ISA noted a handful of air concentration studies that have found elevated Pb
8 concentrations in PM₁₀ and PM_{2.5} during biomass burning episodes ([U.S. EPA, 2013](#)). [Qureshi et al.](#)
9 [\(2006\)](#) observed a spike in Pb-PM_{2.5} at 42 ng/m³ in Queens, NY during a fire event in Quebec. This 24-
10 hour spike was considerably larger than the 3-month average (July to September) of 5.1 ng/m³. Another
11 study quantified the increase of Pb-PM₁₀ measured in Finland at 1.7–3.0 times higher during forest fire
12 emissions from a fire in Russia compared to the reference concentration of 3.5 ng Pb /m³ ([Anttila et al.,](#)
13 [2008](#)). Similarly, [Golobokova et al. \(2020\)](#) observed air above Lake Baikal in Siberia before and during
14 large wildfires. They found levels of Pb were double the base level during the wildfires, with base level
15 concentrations averaging 0.16 ng/m³ and fire concentrations averaging 0.33 ng/m³. [Isley and Taylor](#)
16 [\(2020\)](#) evaluated trace element and Pb isotope compositions in aerosols from four wildfires near Sydney,
17 Australia. They found Pb concentrations pre-fire up to ~120 ng/m³ and concentrations during and post-
18 fire up to ~210 ng/m³. They attributed 94% of the Pb mass to anthropogenic pollutants, namely historical
19 Pb from previous emissions. These four studies found elevated Pb measurements, up to 8 times higher,
20 during days with fire emissions present. The concentrations varied depending on location, with more
21 isolated locations, such as shipboard on Lake Baikal and rural Finland, measuring lower concentrations
22 compared with locations with legacy Pb, such as Sydney, Australia and New York City, NY. Despite
23 these differences, the relative increase is similar across studies.

24 Also examining Pb attributed to PM_{2.5}, [Boaggio et al. \(2022\)](#) analyzed Pb air concentrations on
25 smoke-affected days across 13 years. Their results disagree with the previous studies when looking at Pb
26 from fires over a longer period of time (13 years). They found Pb to be insignificantly different on smoke
27 days compared with non-smoke days apart from during wildfires that burned substantial infrastructure.
28 The median percent change for Pb comparing smoke to non-smoke days was found to be 2.73% lower,
29 but the maximum was 4071% higher at the station that received smoke from the 2018 Camp Fire which
30 destroyed ~18,000 structures. Another study detected Pb-bearing particles in the coarse mode (PM_{10-2.5})
31 during the Camp Fire ([Sparks and Wagner, 2021](#)). After the burning of the Notre Dame Cathedral in Paris
32 which contained approximately 460 tons of Pb, an increase in particulate Pb concentrations from 0.050 to
33 0.105 µg/m³ was observed 50 km downwind of the fire ([van Geen et al., 2020](#)). These results emphasize
34 the importance of accounting for Pb mobilized from burning infrastructure and vehicles during more
35 destructive wildfires, typically occurring in the wildland-urban interface.

36 Pb found from burning of anthropogenic structures was also seen in ash studies. [Burton et al.](#)
37 [\(2016\)](#) looked at ash following a fire in California in 2009 and found Pb was higher in ash samples

1 collected from burned residences and buildings compared with soil and sediment Pb concentrations.
2 Similarly, [Alexakis \(2020\)](#) found the median values of Pb in residential ash (78 mg Pb/kg) were 1.5 times
3 higher than those found in wildland ash (53.5 mg Pb/kg) after a fire in western Attica. The residential ash
4 sample concentrations of Pb ranged up to 205 mg Pb/kg. Additionally, [Campos et al. \(2016\)](#) studied Pb
5 levels in burnt soils after a wildfire in Portugal and found unburnt soil concentrations ranging from
6 approximately 40–50 mg Pb/kg with burnt soil concentrations ranging from approximately 55–150 mg
7 Pb/kg. These two studies find ash concentrations of Pb similar to those found by [Alexakis \(2020\)](#), who
8 noted elevated concentrations in residential ash. Soil Pb concentrations ranged from 30–9,000 mg Pb/kg
9 within 1 km of the Notre Dame Cathedral fire in Paris, whose roof and spire were composed of 460 tons
10 of Pb ([van Geen et al., 2020](#)). This study also observed elevated concentrations in the direction of
11 prevailing winds during the fire ([van Geen et al., 2020](#)). The 2013 Pb ISA also cites a study focusing on
12 wildfire ash by [Odigie and Flegal \(2011\)](#) that found measurements of Pb in ash following the Jesusita Fire
13 in 2009 ranging from 4.3 to 51 mg Pb/kg. Another study from the same group measured Pb and other
14 trace metals remobilized by the Williams Fire in 2012 and found Pb concentration in ash ranging from 7
15 to 42 mg Pb/kg ([Odigie and Flegal, 2014](#)). Both studies concluded the Pb was primarily of anthropogenic
16 origin remobilized by the fires.

17

1.2.5. Traffic and Roads

18 In 2006, the major sources of Pb emissions from on-road mobile sources were fuel combustion
19 and vehicle wear ([U.S. EPA, 2006b](#)). After the phase-out of Pb as an anti-knock agent in gasoline for on-
20 road automobiles in the 1990s, Pb emissions from tailpipes declined rapidly. As a result, the relative
21 contribution of non-tailpipe emissions, such as resuspension of Pb in soil and road dust into air, brake,
22 and tire wear, has increased. The 2013 Pb ISA found a significant source of Pb in non-tailpipe emissions
23 from wheel weights. [Aucott and Caldarelli \(2012\)](#) estimated that $13.8 \pm 5.0\%$ of the deposited mass of
24 wheel weights are dispersed each year through abrasion and grinding by traffic ([U.S. EPA, 2013](#)).
25 However, since 2013, wheel weights have been banned in many states. In addition to wheel weights, tire
26 abrasion (mean of two tire samples in Korea=13 mg Pb/kg tire) and brake wear (30.5 mg Pb/kg brake dust
27 from light duty vehicles in Korea) also contribute to Pb emissions ([Jeong et al., 2022](#); [U.S. EPA, 2013](#)).
28 When comparing non-exhaust emission sources, asphalt had the highest Pb concentration (738 mg Pb/kg)
29 followed by road paint (88 mg Pb/kg) ([Jeong et al., 2022](#)). A study that used material metal
30 concentrations, traffic volume, emissions factors, and sales data to estimate the quantity of Pb emitted
31 from brake wear and tires in Stockholm, Sweden in 2005 estimated that 24 kg (0.026 ton) of Pb were
32 emitted from brake wear each year, compared with 2.6 kg (0.0029 ton) of Pb from tire tread wear; an
33 estimated 549 kg (0.61 ton) was emitted from brake wear in 1998 ([U.S. EPA, 2013](#); [Hjortenkrans et al.,
34 2007](#)). Other studies report Pb from asphalt (738 mg Pb/kg) and road paint (88 mg Pb/kg) as other non-
35 tailpipe emissions from vehicles ([Jeong et al., 2022](#)).

1 Road dust is loose material that can be collected by sweeping and vacuuming the traveled portion
2 of a road. Also called road sediment or street sediment, it is inclusive of particles associated with non-
3 tailpipe Pb emissions from traffic ([Dietrich et al., 2022](#)). Road dust also contains PM deposited from other
4 sources onto or near roads, and is geochemically related to urban soil ([Alshetty and Shiva Nagendra,
5 2022](#); [Dietrich et al., 2022](#); [Jeong et al., 2022](#)). Road dust emissions are a function of dust load and
6 vehicle traffic (frequency of vehicle passing and average weight of vehicles) ([Alshetty and Shiva
7 Nagendra, 2022](#)), with average road sediment concentration of Pb in Busan South Korea in 2014 reported
8 as 210 mg Pb/kg road dust ([Jeong et al., 2020](#)). Road dust in Philadelphia had mean and median Pb
9 concentrations of 516 mg Pb/kg and 202 mg Pb/kg, respectively, with higher values reported for
10 industrial sites and lower for mixed use sites ([O'Shea et al., 2020](#)), while road dust in Toronto had a
11 median Pb concentration of 63 mg Pb/kg (range 21–220 mg Pb/kg) ([Wiseman et al., 2021](#)). [Deocampo et
12 al. \(2012\)](#) observed high spatial variability for Pb concentrations in Atlanta road dust, describing a median
13 road dust Pb concentration of 63 mg Pb/kg in a downtown Atlanta area, but a median of 93 mg Pb/kg and
14 a maximum concentration of 972 mg Pb/kg in a residential area (also in the urban core). They reported
15 significant variation on a scale of tens to hundreds of meters. Most road dust particles are large, with sizes
16 ranging from 10–60 μm ([O'Shea et al., 2021](#)). On average, 13–18% of road dust analyzed in two areas in
17 India was $<10\ \mu\text{m}$, 6–9% was $<2.5\ \mu\text{m}$, and 4–6% was $<1\ \mu\text{m}$ ([Alshetty and Shiva Nagendra, 2022](#)).
18 Road dust and soils can serve as both sources and sinks to one another ([Dietrich et al., 2022](#)).

19 Resuspension of Pb in road dust and soils back to the atmosphere is covered in Section 1.3.4. The
20 relationship between Pb air concentrations and distance to the road is an emerging area of research. [Cahill
21 et al. \(2016\)](#) looked at this question for three size fractions of PM in Detroit in 2010. They found that for
22 coarse PM, Pb concentrations were $\sim 4\ \text{ng}/\text{m}^3$ at 10 meters from the highway, $\sim 1\ \text{ng}/\text{m}^3$ at 100 meters
23 north or south of the highway, and $\sim 1.5\ \text{ng}/\text{m}^3$ 300 meters north of the highway. They deduced that the
24 increase at 300 meters could be attributed to a heavily trafficked road around 380 meters north of the
25 highway. For $\text{PM}_{2.5}$ (0.09 to $2.5\ \mu\text{m}$), they found Pb concentrations were $\sim 4\ \text{ng}/\text{m}^3$ 10 meters north,
26 $\sim 3\ \text{ng}/\text{m}^3$ 100 meters north, and $\sim 2.5\ \text{ng}/\text{m}^3$ 300 meters north of the highway. For very fine PM (0.09 to
27 $0.26\ \mu\text{m}$), Pb concentrations were $\sim 0.75\ \text{ng}/\text{m}^3$ 100 meters south, $\sim 0.25\ \text{ng}/\text{m}^3$ 10 meters north,
28 $\sim 0.95\ \text{ng}/\text{m}^3$ 100 meters north, and $\sim 0.4\ \text{ng}/\text{m}^3$ 300 meters north. Another study found similar near-road
29 concentrations for fine Pb with a mean of $5.23\ \text{ng}/\text{m}^3$ and slightly lower concentrations of coarse Pb with
30 a mean of $1.14\ \text{ng}/\text{m}^3$ ([Silva et al., 2021](#)). Contrary to the previous study, they found Pb to be significantly
31 related to distance to nearest road in coarse concentrations only ([Silva et al., 2021](#)). A third study also did
32 not find a relationship with distance to road in median $\text{PM}_{2.5}$ water soluble Pb ([Oakes et al., 2016](#)), with a
33 slight decline with distance for acid soluble $\text{PM}_{2.5}$ Pb and $\text{PM}_{10-2.5}$ Pb. These studies found Pb associated
34 with PM generally decreases with distance to road. However, the size of this gradient depends on the
35 particle size distribution of Pb and even with consistent size, there could be subtle differences when
36 breaking Pb down into water and acid soluble fractions. For a monitoring site in central Los Angeles
37 located near a major interstate freeway, trends in ambient air Pb concentrations were related to traffic
38 volume. Pb concentrations decreased slightly from 2005 (median 24-hour sample of $0.005\ \mu\text{g}/\text{m}^3$) to 2013-
39 2015 but increased from 2015 (median of $0.001\ \mu\text{g}/\text{m}^3$) to 2018 (median of $0.005\ \mu\text{g}/\text{m}^3$). This was

1 attributed to greater road dust resuspension into air due to increased traffic on the nearby interstate
2 freeway, since traffic near the site was relatively constant before 2013, but increased considerably from
3 2013 to 2018 ([Farahani et al., 2021](#)). For context, this increase is substantially less than the overall decline
4 of airborne Pb concentrations near roads with heavy traffic since leaded gasoline was phased out (Section
5 1.4.3). [Wiseman et al. \(2021\)](#) estimated that 90 ± 23 kg of Pb in road dust was resuspended into air
6 annually in Toronto, Canada, an amount corresponding to 22% of air releases from Toronto industrial
7 facilities. Limitations identified in this study were identified as uncertainties associated with aging of
8 street sweeping equipment, street sweeping frequency, and particle size distribution assumptions.

1.2.6. Volcanoes

9 The 2006 Pb AQCD ([U.S. EPA, 2006b](#)) included an estimate of 540 to 6000 metric tons per year
10 for the range of global Pb emissions from volcanoes ([Nriagu and Pacyna, 1988](#)). More recently, the
11 Masaya volcano in Nicaragua was estimated to emit 1 ton of Pb per year ([Liotta et al., 2021](#)). In two
12 recent studies, Pb concentrations measured at volcanic sources ranged from 0.055 to 0.75 $\mu\text{g}/\text{m}^3$ for
13 samples collected at the main active vent during the 2018 eruption of Kilauea on the island of Hawaii
14 ([Mason et al., 2021](#)), and 0.14 to 0.27 $\mu\text{g}/\text{m}^3$ for samples collected at the rim of a crater of the Masaya
15 Volcano in April 2000 ([Liotta et al., 2021](#)). Concentrations at the upper ends of these ranges are
16 comparable to some of the highest currently observed Pb monitoring network concentrations ([U.S. EPA,](#)
17 [2022](#)). Airborne Pb concentrations associated with the eruption of Kilauea were higher than
18 concentrations at nearby populated areas ([Ilyinskaya et al., 2021](#)). During the week of the 1991 eruption
19 of Mt. Hudson in southern Chile, observed Pb concentrations more than 2000 miles away on King George
20 Island in the Southern Ocean were higher than before or after the eruption ([Evangelista et al., 2022](#)).
21 Highly elevated Pb concentrations associated with the eruption were also observed in lake sediment
22 profiles ([Evangelista et al., 2022](#)).

23 Pb can be emitted in both particulate form ([Liotta et al., 2021](#)), and as a volatile gas at high
24 temperatures ([Edmonds et al., 2022](#); [Liotta et al., 2021](#)), and emissions result in the formation of
25 particulate volcanic plumes downwind of active volcanoes ([Edmonds et al., 2022](#)). Recent research
26 suggests that emissions of Pb from volcanoes might be underestimated. [Ilyinskaya et al. \(2021\)](#) observed
27 that deposition of Pb and other metals were depleted more rapidly from the volcanic plume of Kilauea
28 than more widely studied species such as sulfur, and that Pb concentrations in nearby communities during
29 the 2018 eruption did not change as much as the concentrations of other species. They recognized that
30 volatile metals like Pb, Cd, and Se were emitted as gases in high temperature volcanic vents and formed
31 soluble chlorides, sulfates, and sulfides that were rapidly removed by wet deposition in the vicinity of the
32 source by the rapidly condensing water in the humid environment created by the high abundance of water
33 vapor emitted from the vent or otherwise present in the humid environment near the source ([Ilyinskaya et](#)
34 [al., 2021](#)). They contrasted this with more refractory elements such as Mg and Fe that are not emitted as
35 gases and noted that Pb was depleted from the volcanic plume 100 times faster than these elements

1 ([Ilyinskaya et al., 2021](#)). This is consistent with results from [Liotta et al. \(2021\)](#), who observed
2 enrichment of Pb in rainwater in comparison to volcanic rock at the Masaya Volcano in Nicaragua. There
3 is some evidence of differences in volatility of Pb emitted from different volcanoes ([Liotta et al., 2021](#)).
4 [Ilyinskaya et al. \(2021\)](#) concluded that emissions of Pb and other volatile metals from volcanoes, as well
5 as their concentration and deposition in the immediate vicinity of volcanoes might be underestimated
6 ([Ilyinskaya et al., 2021](#)).

1.2.7. Legacy Sources

7 Contemporary U.S. emissions of airborne Pb described in Sections 1.2.1 through 1.2.6 do not
8 provide a complete picture of all contributions to ambient air Pb, because Pb emitted from past sources
9 can become resuspended. Current air emissions are considerably smaller than historical emissions.
10 Numerous studies of historical records have been reconstructed from sediment and peat cores as well as
11 long-term soil concentration measurements from many North American locations including Virginia, the
12 Northeast United States, the St. Lawrence Valley, and northern Alberta (Section 1.3.3.4). Most of these
13 studies show evidence of decreasing Pb concentrations after the 1970s due to elimination of leaded
14 gasoline and reductions in industrial emissions ([Balascio et al., 2019](#); [Shotyk et al., 2016](#); [Sarkar et al.,](#)
15 [2015](#); [Richardson et al., 2014](#); [Pratte et al., 2013](#)). An exception was a sinkhole near Lake Marion SC,
16 where sediment Pb concentrations increased continuously during the past 60 years ([Edwards et al., 2016](#)).
17 This recent research adds to an even larger body of literature summarized in the 2013 Pb ISA and
18 previous AQCDs that atmospheric Pb concentrations and atmospheric deposition have decreased steadily
19 since the 1970s ([U.S. EPA, 2013](#)). Pb isotope ratios from some of these studies suggest that historical
20 sources are an important if not dominant contributor to Pb in North American soil and sediments.

21 Leaded gasoline has been a major contributor to Pb in the environment, particularly in roadside
22 and urban soils. An estimated 5.4 million metric tons of Pb additives were used in leaded gasoline in the
23 United States between 1927 and 1994 ([Mielke et al., 2010](#)), peaking between 1968 and 1972 at more than
24 200,000 metric tons per year ([U.S. EPA, 2013](#)). Pb additive use subsequently declined by 92% from 1970
25 to 1990 due to health concerns and leaded gasoline was finally banned in the United States in 1996.
26 Numerous studies have identified leaded gasoline as a prominent source in sedimentary and other
27 historical records of atmospheric Pb pollution ([U.S. EPA, 2013](#)). Recent studies investigating Pb isotope
28 ratios continue to show that leaded gasoline was the principal source of atmospheric Pb ([Pratte et al.,](#)
29 [2013](#)) and the dominant source of Pb in samples of North American sediments ([Pratte et al., 2013](#)) and
30 forest soils ([Richardson et al., 2014](#)). In the United States, emissions were concentrated in urban areas,
31 with emissions in 90 urban areas estimated to account for about 30% of total U.S. automotive Pb
32 emissions in 1982 ([Mielke et al., 2011](#)). In a detailed recent study in a mid-size southern U.S. city, current
33 roadside soil concentrations decreased since the peak of leaded gasoline usage but remained higher than
34 geologic background ([Wade et al., 2021](#)).

1 The United States was a leading producer of Pb in the previous century with major mining and
2 smelting operations. High Pb concentrations in soils near smelters and other industrial operations have
3 been observed in numerous studies ([U.S. EPA, 2013](#)). The last primary Pb smelter in the United States
4 closed in Herculaneum, MO in 2013 ([Sullivan and Green, 2020](#)). Recent studies also found high Pb
5 concentrations in soil near closed smelters in Tar Creek OK, Pueblo CO, and Eureka NV ([Diawara et al.,
6 2018](#); [Chaffee and King, 2014](#); [Zota et al., 2011](#)). Pb concentrations in residential soil samples in Tar
7 Creek OK ranged from 12 to 2436 mg Pb/kg and averaged 201 mg Pb/kg ([Zota et al., 2011](#)), and Pb
8 concentrations in samples from Pueblo CO ranged from 12 to 10,011 mg Pb/kg and averaged 366 mg
9 Pb/kg ([Diawara et al., 2018](#)). Pb from closed smelters was also the dominant source of Pb in lake
10 sediments near Tacoma WA ([Gawel et al., 2014](#); [Gray et al., 2013](#)) and attic dust in El Paso TX ([Van Pelt
11 et al., 2020](#)). [Wang and Kanter \(2014\)](#) identified 229 former Pb industrial sites in urban areas of the
12 United States.

13 Pb-based paint was banned in the United States in 1978. However, 15.3 million homes, 14% of
14 the homes in the United States, have significantly deteriorated Pb-based paint according to the
15 Department of Housing and Urban Development’s American Healthy Homes Survey. The proportion of
16 houses with deteriorating Pb-based paint increases with the age of the housing, accounting for 86% of
17 U.S. houses built before 1940. Regionally, a greater fraction of houses in the Northeast and Midwest
18 contains Pb-based paint than in the South and West. Housing with Pb-based paint is also unevenly
19 distributed on a local scale, with a greater fraction of poor and non-white families living in houses with
20 Pb-based paint ([HUD, 2011](#)). A recent county-level geospatial analysis of Pb paint hazard in homes and
21 childcare facilities found potential Pb hazard hotspots coincided with areas of higher populations of non-
22 white children ([Baek et al., 2021](#)). Peeling and deteriorating paint is a source of high Pb concentrations in
23 yard soil and house dust ([HUD, 2011](#)). In streetside, residential, and other soil samples from Durham NC,
24 soil Pb concentrations ranged from 6 to 8825 mg Pb/kg, with the highest Pb concentrations observed
25 within 1 m of pre-1978 residential foundations and both foundation and yard soil Pb concentrations
26 considerably higher around older houses ([Wade et al., 2021](#)). In urban and industrial areas and near
27 heavily trafficked roads, historical air emissions together with Pb from deteriorating paint comprise a pool
28 of legacy Pb in urban soil ([Wang et al., 2022](#); [Obeng-Gyasi et al., 2021](#)). The potential for the
29 contribution of legacy Pb to ambient air through suspension and resuspension is discussed in Section
30 1.3.4.

1.2.8. Other Sources

31 Exposure to Pb from community gardens and consumer products is mainly through other media,
32 but Pb from these sources can briefly become airborne. Pb in food from residential and community
33 gardens has been the subject of numerous recent studies. Although additional recent research also
34 indicates that soil Pb can be a concern for urban gardens ([Engel-Di Mauro, 2021](#); [Clarke et al., 2015](#);
35 [Kaiser et al., 2015](#); [Wiseman et al., 2015](#)), there are ongoing research efforts to improve urban gardening

1 by reducing Pb contamination in garden produce ([Egendorf et al., 2021](#); [Taylor et al., 2021](#); [Gallagher et](#)
2 [al., 2020](#); [Harada et al., 2019](#); [Fitzstevens et al., 2017](#); [Brown et al., 2016](#); [Schwarz et al., 2016](#); [Splithoff](#)
3 [et al., 2016](#); [Kaiser et al., 2015](#)).

4 Pb is also a concern in a variety of consumer products. Batteries were responsible for 92% of Pb
5 consumed in the United States in 2021([USGS, 2022](#)). Many of the source oriented Pb monitoring sites in
6 the national monitoring network for Pb ([U.S. EPA, 2022](#)) are near secondary smelters for battery
7 recycling. Recent research has focused on trends for recycling used batteries from the United States and
8 Europe in other countries. It has been estimated that there are more than 10,000 informal industrial sites
9 for processing used Pb-acid batteries in low- and middle-income countries, especially in East Asia, South
10 Asia, and Africa ([Ericson et al., 2017](#)). Informal industry is defined as industry characterized by a lack of
11 adherence to regulation, including zoning and pollution controls ([Ericson et al., 2017](#)). In a study of soils
12 from 15 recycling plants and one battery manufacturing site in 7 countries in Africa, mean soil Pb
13 concentrations ranged from 480 to 140,000 mg Pb/kg and averaged 23,200 mg Pb/kg inside plant sites,
14 and ranged up to 48,000 2600 mg Pb/kg and averaged 2600 mg Pb/kg in soil samples from communities
15 surrounding the plants([Gottesfeld et al., 2018](#)). By amount of Pb consumed, ammunition ranks second
16 after batteries as an end use for Pb in the United States ([USGS, 2022](#)). The mean estimate of Pb
17 concentrations in soils from shooting ranges in 10 studies was twice as high as Pb concentrations from
18 non-residential Superfund sites ([Frank et al., 2019](#)). Recent research has advanced our understanding of
19 the ranges of particle size, solubility, bioaccessibility, and chemical forms of Pb in gunshot residue
20 particles from shooting range soils ([Schindler et al., 2021](#); [Sanderson et al., 2012](#)). There is a large body
21 of research on the environmental and health consequences of the use of Pb in ammunition ([Arnemo et al.,](#)
22 [2016](#)). Other consumer products that are sources of Pb are contaminated ceramic cookware, food, toys,
23 cosmetics, antiques, and herbal medicines ([Frank et al., 2019](#)).

1.3 Fate and Transport of Pb Emitted into Air

24 Knowledge of Pb transport within and between diverse media, including air, surface water, soil,
25 and sediment, provides a foundation for understanding the various pathways leading to atmospheric Pb
26 exposure, as well as the atmospheric contribution to total Pb exposure discussed in Appendix 2
27 (<https://cfpub.epa.gov/ncea/isa/recordisplay.cfm?deid=357282>). Pb emitted into the
28 atmosphere can be distributed into soil, water, and other media, leading to human and ecosystem contact.
29 Understanding Pb transport in soil, water, and other media is also necessary for assessing impacts of
30 atmospheric Pb relative to non-atmospheric sources such as wastewater discharges or mobilization from
31 waste materials. Sections 1.3.1, 1.3.2, 1.3.3, and 1.3.4 summarize our understanding of fate and transport
32 of Pb in air, soil, water, and urban media, respectively.

1.3.1. Fate and Transport in Air

1 Pb is mainly emitted in particulate form, and the fate of airborne Pb is strongly influenced by the
2 whether it is primarily emitted in the form of ultrafine combustion particles as observed for aviation gas
3 exhaust, or coarse particles, as observed for resuspended Pb from soil (Section 1.3.4). As described in
4 Section 1.2.1, Pb is introduced into aviation fuel as the anti-knocking agents tetramethyl and tetraethyl Pb
5 ([Kumar et al., 2020](#)). During engine operation, the organic functional groups of these compounds are
6 oxidized and emitted as water and carbon dioxide. A second additive to the fuel mixture, an alkyl bromide
7 compound, reacts with the Pb present in the combustion mix to form an array of compounds composed of
8 Pb (II), bromide and chloride ions, molecular ammonia, and other, nonvolatile compounds that form
9 particles. These particles are either entrained into the engine exhaust or remain in the engine's crankcase
10 lubricant ([NCBI, 2022](#)). Unreacted tetramethyl and tetraethyl Pb have sufficiently high vapor pressures at
11 ambient and engine operation temperatures to allow for fugitive emissions of these gases ([U.S. EPA,](#)
12 [1986](#)), which go on to photolyze in the presence of atmospheric ultraviolet radiation to form Pb
13 compounds that also contribute to atmospheric PM. These Pb-containing particles are then subject to the
14 same atmospheric processes that transport and remove other forms of PM. As discussed in Sections
15 1.3.1.1 and 1.3.1.2, the transport and deposition of dry particles is defined by size. Depending on the
16 chemical counter-ion, Pb compounds vary in water solubility, determining the degree to which Pb is
17 removed by wet deposition. Figure 1-1 provides a general illustration of the geochemical lifecycle of Pb
18 derived from fuel additives. Resuspension of soil bound Pb has the potential to contribute to airborne
19 concentrations near major Pb sources and is considered in Section 1.3.4.

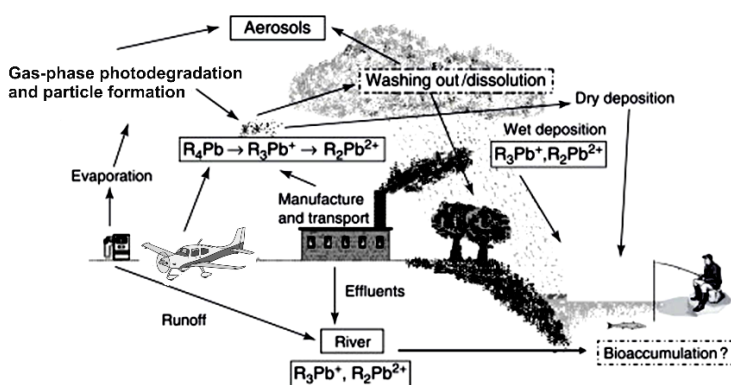


Figure 1-1 The biogeochemical cycle of tetramethyl/tetraethyl Pb

Source: Adapted with permission from [Encinar and Moldovan \(2005\)](#).

1.3.1.1. Atmospheric Transport

1 The 2013 Pb ISA discussed in detail the available studies concerning the variables governing
2 long-range and urban-scale transport of particle-bound Pb in the atmosphere, concluding that Pb was
3 primarily present in submicrometer aerosols, but bimodal size distributions within this size range were
4 frequently observed ([U.S. EPA, 2013](#)). As described in detail in Section 1.2.1, Pb (as PbBr and other
5 halogenated forms of Pb) is primarily emitted by piston-driven aircraft in the ultrafine particle (UFP) size
6 range (<100 nm) and larger particles formed from agglomeration of individual particles in the UFP size
7 range ([Turgut et al., 2020](#)). Particles emitted by aircraft have been observed to be as small as 13 nm in
8 diameter, when emitted ([Griffith, 2020](#)).

9 Consistent with particles from other sources, according to several studies, particle-bound Pb in
10 the fine PM is transported long distances and found in remote areas. The patterns of dispersion can be
11 modeled using Gaussian plume models or Lagrangian and Eulerian continental transport models,
12 indicating that Pb remains a nonvolatile, unreactive particle component. The 2013 Pb ISA also described
13 studies that indicate that small Pb-containing particles can be scavenged by larger, soil-derived geogenic
14 particles which can lead to chemical reactions that alter the composition and hygroscopicity of the
15 composite particle ([U.S. EPA, 2013](#)). This finding was supported by evidence of Pb enrichment of
16 particles originating from Pb-free sources that were deposited in remote locations.

17 There has been little recent research on transport of airborne Pb, and recent studies have
18 continued to focus on transport of Pb associated with particles smaller than 10 μm . Recent research
19 supports previous results that Pb derived from high temperature processes such as smelting is largely
20 emitted in the submicrometer fraction and is capable of being transported over long distances and being
21 deposited in remote environments ([Cullen and McAlister, 2017](#)). On a smaller scale, using a generalized
22 regression model with 4 km^2 sampling grids, [Fortuna et al. \(2020\)](#) demonstrated that Pb content of lichen
23 samples was significantly spatially associated with dispersion modeling outcomes for Pb, and other
24 metals primarily associated with PM_{10} emitted from a coal-fired power plant over an area of 176 km^2 , for
25 a dispersion model developed for a time frame that corresponds to the average age of biomonitor sample
26 material. There is also recent research on chemical transformation of Pb. In Beijing, [Peng et al. \(2020\)](#)
27 reported complete atmospheric transformation of PbO and PbCl_2 from coal combustion to $\text{Pb}(\text{NO}_3)_2$, from
28 a process highly dependent on NO_2 concentration and relative humidity, and especially efficient at
29 relative humidity greater than 60% in the presence of sufficient NO_2 . This observation is important
30 because insoluble Pb is converted into a more soluble and potentially more bioavailable form ([Peng et al.,
31 2020](#)).

32 Although atmospheric lifetime depends on atmospheric conditions, UFPs quickly grow into fine
33 particles, or particles smaller than 2.5 μm ($\text{PM}_{2.5}$), by way of gas-to-particle partitioning or coagulation
34 with other particles into the before removal. Larger particles in the size range from 2.5 to 10 μm
35 ($\text{PM}_{10-2.5}$) and larger are removed more quickly from the atmosphere than $\text{PM}_{2.5}$ by way of gravitational
36 settling and deposition. This results in UFP and $\text{PM}_{10-2.5}$ concentrations having substantially greater

1 spatial variability than PM_{2.5}, with higher atmospheric concentrations typically near their sources and
2 greater spatial variability on urban and neighborhood scales ([U.S. EPA, 2009](#)). More rapid removal
3 processes also result in shorter atmospheric lifetimes and transport distances for UFP and PM_{10-2.5} than
4 the PM_{2.5} size range.

1.3.1.2. Atmospheric Deposition

5 The 2013 Pb ISA summarized atmospheric deposition patterns for Pb ([U.S. EPA, 2013](#)). There
6 has been a sharply decreasing trend in Pb deposition in the United States and globally since the 1970s,
7 corresponding to decreasing ambient air concentrations (Section 1.5.1) and decreasing traffic emissions
8 associated with the removal of Pb from gasoline. In general, fine particulate Pb is mostly soluble and
9 removed from the atmosphere by wet deposition, and coarse particulate Pb is mostly insoluble and
10 removed from the atmosphere by dry deposition. Other factors also influence Pb deposition, however.
11 The pH of precipitation can also play a role because Pb solubility increases with decreasing pH, and
12 precipitation can also scavenge insoluble particulate Pb as an aqueous suspension. Diurnal variations in
13 Pb deposition have been observed and attributed to differences in atmospheric structure, specifically
14 boundary layer height. Several U.S. studies reported substantially greater deposition rates in areas near
15 industrial sources than in non-industrial areas, and ([U.S. EPA, 2013](#)). As a recent example, regional
16 differences in Pb deposition patterns have also been documented in a number of studies ([U.S. EPA, 2013](#))

17 Several recent studies have addressed Pb deposition. [Wu et al. \(2016\)](#) used Pb isotope ratios in
18 lichens and fungi to show that deposited Pb in bioindicators still reflected historical deposition from
19 leaded gasoline exhaust. [Mazari and Filippelli \(2020\)](#) focused on urban atmospheric deposition patterns of
20 Pb and other metals over a wider range of time scales by analyzing soil, bark, and leaves. They oriented
21 sampling locations along an increasingly urban transect and found the highest Pb levels at the most urban
22 locations. Previous observations of decreasing Pb deposition with distance from sources also supported
23 by a recent study showing that previously remediated soil became re-contaminated following aerial
24 deposition from a Pb smelter, with soil Pb concentrations ranging from 25–100 mg Pb/kg within years of
25 remediation ([Bowers et al., 2014](#)). [Stankwitz et al. \(2012\)](#) investigated the effect of elevation on Pb
26 deposition in a forested area of the Northeast United States and found deposition increased with elevation
27 due to increasing precipitation with elevation. They also found the increase was not linear however,
28 instead including two abrupt threshold increases associated with the two most common cloud base
29 altitudes, which in turn corresponded to changes in vegetation. In recent research on cloud processes,
30 [Ebert et al. \(2011\)](#) observed enrichment of Pb in ice nuclei, with a 25 times higher likelihood of Pb in ice
31 nuclei than in interstitial aerosols by number in clouds.

32 Consistent with the 2013 Pb ISA, recent studies continue to show decreasing Pb deposition in
33 various locations. For example, [Pérez-Rodríguez et al. \(2018\)](#) observed a peak in Pb concentrations in
34 peat samples in southern Greenland that contained Pb transported from North America and Eurasia in

1 southern Greenland, and [Wu et al. \(2020\)](#) used bioindicators to monitor decreasing Pb deposition in
2 Guangzhou after 2000. Other research continues to evaluate biomonitors for research on atmospheric
3 deposition of Pb and other trace metals ([Kempter et al., 2017](#)). After deposition, resuspension of Pb in
4 contaminated soil and road dust into air by traffic, construction, and wind is a potentially important
5 contributor to airborne Pb (Section 1.3.4).

1.3.2. Fate and Transport in Soil

6 Knowledge of Pb transport in soil following wet and dry deposition is required to understand risk
7 and exposure to human and ecological receptors following deposition of atmospheric Pb into soil. The
8 2013 Pb ISA summarized the long retention time and low mobility of Pb in soil and confirmed the role of
9 soil as an overall sink for Pb even though atmospheric Pb concentrations peaked several decades ago
10 ([U.S. EPA, 2013](#)). Pb can be deposited onto surface soils in both close proximity to and considerable
11 distances from point sources. Once deposited in soil, subsequent fate and transport through the soil
12 column is influenced by several physicochemical factors, including storage in leaf litter, amount, and
13 decomposition rates of organic matter (OM), composition of organic and inorganic soil constituents,
14 mobile colloid abundance and composition, microbial activity, and soil pH. These physicochemical
15 properties are based on soil forming factors: climate, organisms, parent material, relief, time, and
16 anthropogenic input. Soils that differ in these factors will subsequently have different physicochemical
17 properties and different trends in Pb transport. The 2013 Pb ISA summarized studies that describe the role
18 that each of these physicochemical factors play in Pb fate and transport through soil, and more recent
19 studies confirm conclusions from the 2013 Pb ISA ([U.S. EPA, 2013](#)).

1.3.2.1. Transport into Soil

20 The 2013 Pb ISA ([U.S. EPA, 2013](#)) confirmed findings from the 2006 AQCD ([U.S. EPA, 2006a](#))
21 that Pb is deposited from air onto soils in the vicinity of stationary sources and deposition decreases with
22 increasing distance from the source. As previously discussed, Pb particles of varying size can be emitted
23 into the atmosphere from several types of stationary sources (Section 1.2) and subsequently deposited
24 onto soil (Section 1.3.1.2). Pb-derived spatial distribution of contaminants in soils located in the vicinity
25 of stationary sources, such as non-ferrous smelters, depend on wind direction, size of particles emitted
26 (smaller particles will travel further than larger particles), and mineralogical and chemical composition of
27 particles emitted ([Ettler, 2016](#)). If soluble forms occur in the dust, greater downward leaching can occur
28 in the soil profile following deposition ([Ettler et al., 2012](#)). Pb derived from high temperature processes
29 such as smelting is largely emitted in the submicrometer fraction and is capable of being transported over
30 long distances and being deposited in remote environments (Section 1.3.1.1). [Bing et al. \(2014\)](#)
31 demonstrated the long-range transport capability of Pb emissions from industrial sources by measuring Pb
32 concentrations and isotope ratios in soil profiles from the remote Hailuogou Glacier foreland in the

1 Eastern Tibetan Plateau. Results revealed Pb enrichment in the O and A horizons relative to the C
2 horizon, indicating Pb from recent atmospheric deposition rather than parent material. The binary mixing
3 model using Pb isotope ratios reported that anthropogenic sources contributed to 45.2–61.3 % of Pb in the
4 O horizon and 8.6–34.8% in the A horizon. Furthermore, the isotopic compositions of Pb and air mass
5 trajectory models revealed that the major contributions of anthropogenic Pb in surface soils were from
6 distant sources including Pb ore processing in southwest China and coal combustion in southwest China
7 and South Asia. The study also discussed potential effects of climate change on soil properties that would
8 result in Pb release from soils potentially affecting downstream water quality. [Bińczycki et al. \(2020\)](#)
9 reported similar results, measuring total Pb concentrations and Pb isotope ratios in nine Podzol profiles
10 located in high elevation remote areas of Karkonosze National Park in Poland. Results revealed high
11 concentrations of Pb in surface horizons originating from combustion of coal in Poland and the Czech
12 Republic followed by long-range transport. As described in the 2013 Pb ISA, Pb deposition to soils has
13 decreased since the phase-out of on-road leaded gasoline ([U.S. EPA, 2013](#)). Reduction of Pb surface soil
14 concentrations since the phase-out are variable, however, particularly in high altitude areas where there
15 has been little change in O horizon Pb decreases since the phase-out. [Kaste et al. \(2011\)](#) used ^{210}Pb
16 measurements to estimate the timescale over which Pb in canopy-derived litter is converted into mobile
17 colloid phases that are transported to mineral horizons. Results showed that the Pb is retained in the O
18 horizon for longer periods of time in areas of higher elevations and latitudes. Similar results have been
19 reported by [Zhou et al. \(2019\)](#) and [Stankwitz et al. \(2012\)](#). Longer Pb retention times in surface soils at
20 higher elevations may be due to higher annual precipitation and cloud water depositions as well as slower
21 OM decomposition due to lower temperatures.

1.3.2.2. Transport within Soil

22 The 2013 Pb ISA described a variety of complex factors influencing Pb retention and distribution
23 in soil, including storage in leaf litter, amount and decomposition rates of OM, composition of organic
24 and inorganic soil constituents, mobile colloid abundance and composition, microbial activity, and pH
25 ([U.S. EPA, 2013](#)). [Zhou et al. \(2020b\)](#) evaluated the Pb adsorption capacity of acidic A and B horizon
26 mineral soils collected from New York and Vermont. Results revealed that Pb was adsorbed more
27 strongly in the A horizon than the B horizon soils across all samples indicating the importance of OM in
28 Pb retention. In addition, soils collected from Vermont were able to selectively adsorb Pb more strongly
29 than the New York samples. This increase in adsorption was attributed to higher pH, cation exchange
30 capacity (CEC), Mn oxide, non-crystalline Fe oxide, and OM contents in Vermont soils.

31 The role of leaf litter as both a contributor to Pb in surface soil and as a sink for Pb from soil in
32 direct contact with leaves was reported in the 2013 ISA ([U.S. EPA, 2013](#)). [Scheid et al. \(2009\)](#)
33 demonstrated that total metals concentrations in leaf litter exposed to manually contaminated soils from
34 the Swiss Federal Institute for Forest, Snow, and Landscape Research increased over the three-year
35 duration of the study, suggesting that leaf litter that may come into contact with Pb-contaminated soil

1 during splashing from rain events can serve as an efficient metal storage pool. [Landre et al. \(2010\)](#)
2 compared the differences between Pb atmospheric inputs measured in bulk deposition with inputs from
3 litterfall and throughfall (water depositing onto soil following collection onto leaves) in a remote forested
4 catchment with limited development in Ontario Canada. Results showed that bulk deposition collectors
5 may underestimate the amount of Pb reaching the forest floor by about 50%. More recent studies reported
6 mixed results regarding the role of leaf litter. [Luo et al. \(2015\)](#) measured Pb in soil, litterfall, and plants in
7 the Gongga Mountain region of Sichuan Province, China and found that both litterfall and atmospheric
8 deposition were main contributing factors to Pb concentrations in the O horizon. In addition, this study
9 also revealed a significant correlation between Pb concentrations in fine roots and the A horizon
10 confirming that fine roots can adsorb and sequester Pb from soil.

11 After Pb is deposited onto surfaces from litterfall and atmospheric deposition, it is transported
12 downward as decomposition slowly transforms buried leaf litter into humus. The fate of Pb in litter and
13 subsequent release to mineral soil horizons occurs over variable timescales that may be strongly
14 influenced by the rate of organic decomposition. [Kaste et al. \(2011\)](#) used measurements of ^{210}Pb
15 throughout soil profiles in coniferous forests in New England and Norway to create a steady-state
16 transport model to quantify the fate of metals in leaf litter during OM decomposition over longer time
17 scales that could be obtained empirically. Results showed the time scale over which canopy-derived litter
18 was converted into mobile organo-metallic colloids ranged from 60–630 years, varying almost an order of
19 magnitude, and was slowest in areas where decomposition was slowest. The results of this study also
20 showed that Pb is retained by the upper litter layer and concentrations increase as litter is buried and
21 decomposes, resulting in Pb that is enriched in the O horizon. [Zhou et al. \(2019\)](#) reported similar results
22 in a study that measured Pb concentrations and pools in forest vegetation, litterfall, organic soil, and
23 mineral soil. In the study, 97.3% of the pools were in litter and organic soil, with Pb concentrations in
24 organic soil being significantly correlated with total OM in both organic and mineral soil, and
25 transportation of Pb to mineral soil was dependent on OM decomposition.

26 Large surface areas with high CEC and negatively charged functional groups make organic and
27 inorganic soil colloids capable of adsorbing Pb and thus play an important role in Pb transport. Physical
28 factors influencing colloid mediated transport of heavy metals include flow rate, medium grain size, and
29 influent concentration ([Xie et al., 2018](#)). Transport of colloids in soil are influenced by flow rate and the
30 physical and chemical properties of the soil porewater and matrix. Soil porewater with a low ionic
31 strength and increased colloid and stationary matrix surface charges are associated with colloid
32 stabilization and maximum Pb-colloid co-migration ([Shang and Li, 2011](#)). Conversely, high ionic strength
33 and lower colloid and surface stationary matrix surface charges are associated with destabilizing colloid
34 conditions where colloids will tend to coagulate and adsorb onto the stationary matrix ([Shang and Li,
35 2011](#)). When colloids are remobilized from the stationary matrix, Pb that is bound to the colloid
36 irreversibly is expected to remobilize along with the colloid. However, Pb that is bound via cation
37 exchange is expected to sorb to the stationary matrix phase following colloid remobilization ([Shang and
38 Li, 2011](#)). [Xie et al. \(2018\)](#) investigated the effects of different colloids on Pb transport under different

1 physical conditions. Results revealed that compared with deionized (DI) water, montmorillonite, loessial,
2 and humic acid (HA) colloids all promoted transport of Pb, with HA having the greatest influence on
3 remobilizing Pb from quartz and sand surfaces with a 33% increase in Pb mobilization through the porous
4 medium compared with colloid-free DI water. The enhanced mobility was attributed to the large number
5 of organic functional groups on the surfaces of HA colloids providing large sites for Pb adsorption.
6 Results also showed that larger matrix grain sizes led to an increase in colloid mobility due to increased
7 outflow of the colloid in more porous media. Higher flow rate decreased Pb adsorption and colloids on
8 quartz surfaces, thus increasing mobility of heavy metals and colloids. [Shang and Li \(2011\)](#) studied the
9 role of rainfall on the migration of Pb-colloid complexes in farmland, floodplain, and Loess platform
10 soils. Pb migration concentrations with colloids in farmland, Loess platform and floodplain columns were
11 respectively, 1.12~2.25, 0.91~1.85 and 0.5~2.01 times more than migration concentration with no
12 colloids. These results confirm the results from [Xie et al. \(2018\)](#), demonstrating that Pb migration is
13 enhanced by colloid-Pb co-migration.

14 The 2013 Pb ISA described the effects of microbial activity on Pb sequestration and transport
15 ([U.S. EPA, 2013](#)). [Perdrial et al. \(2008\)](#) observed bacterial Pb sequestration and proposed a mechanism of
16 Pb complexation by polyphosphate. They also postulated that bacterial transport of Pb could be important
17 in subsurface soil environments. [Wu et al. \(2006\)](#) also concluded that Pb adsorption to the bacterial cell
18 walls may be important with respect to Pb transport in soils. More recent studies suggest that microbial
19 activity may enhance the release of Pb from both organic and mineral soils. [Drozdova et al. \(2015\)](#)
20 studied the effects of both live and dead bacteria on the release of trace elements from both organic and
21 mineral podzols (aqueous solutions in a laboratory system). Results revealed that live bacteria enhanced
22 the release of Pb to solution, particularly in organic soils, while decreasing the release of potassium (K),
23 calcium (Ca), strontium (Sr), Cu, titanium (Ti), manganese (Mn), zinc (Zn), and arsenic (As). The
24 authors' noted that decreases in pH, degradation of dissolved organic carbon (DOC) and metal-organic
25 complexes by microbial activity, element adsorption at cell surfaces, and biological uptake may occur
26 simultaneously in the soil-bacteria suspension to both enhance and decrease the release of trace elements
27 from the soil profiles. In the case of Pb, it is suggested that Pb is released into aqueous solution following
28 bacterial degradation of Pb-organic complexes.

1.3.2.3. Soil Forming Factors and Land Use

29 The physicochemical factors influencing Pb retention and distribution throughout the soil column
30 can vary considerably amongst soils with differences in soil forming factors (i.e., climate, organisms,
31 parent material, relief, time, and anthropogenic input). The 2013 Pb ISA summarized Pb retention and
32 distribution through forest soils as strongly influenced by rate of OM decomposition, depth of soil O
33 horizon, and pH, generally concluding that atmospherically derived Pb will have a longer residence time
34 in organic surface layers that have lower rates of OM decomposition ([U.S. EPA, 2013](#)). Therefore, Pb
35 will be enriched in the O horizon with increased enrichment occurring in forests where climate, elevation,

1 and vegetation (i.e., boreal forests versus deciduous forest) favor slower rates of OM decomposition.
2 Recent literature confirms many of these findings. [Richardson et al. \(2014\)](#) resampled organic and upper
3 two mineral horizons at 16 sites across deciduous and mixed deciduous/coniferous forests in the northeast
4 that were previously sampled in 1980, 1990, and 2002. Results revealed that gasoline derived Pb has
5 leached from the forest floor to mineral soil horizons across the study areas. However, the rate at which
6 Pb is being transported from the forest floor to mineral soils varied across the 16 sites and was slowest at
7 sites with frigid soil temperature regimes (STRs) located in the northern portions of the study area. The
8 decreased Pb response rate and increased retention time in these soils was attributed to slower
9 decomposition rates in frigid STR and more coniferous vegetation compared with other sites, potentially
10 decreasing decomposition rates due to higher lignin content. [Chrastný et al. \(2012b\)](#) compared the
11 leachability of air pollution control residues in deciduous and coniferous organic soil horizons. Results
12 revealed higher Pb sorption onto humified OM from coniferous litter compared with deciduous litter. The
13 increased sorption of Pb in the coniferous organic horizon was attributed to a lower pH and higher portion
14 of fulvic acids compared with the deciduous organic horizon, which was a result of differences in
15 chemical composition and degradability of needles and litter. These results suggest that soil in deciduous
16 forest may be more vulnerable to Pb mobilization compared with soils in coniferous forests. [Chrastný et
17 al. \(2012a\)](#) compared Pb concentrations and mobility in agricultural and forested soil profiles located at
18 varying distances from smelting and/or mining release sources. Total Pb concentrations were generally
19 higher in forested soil profiles compared with agricultural soil profiles. However, Pb in the agricultural
20 soil profile was found to be more mobile, confirming the important role of forest leaf litter in Pb retention.
21 [Du et al. \(2020\)](#) investigated the effects of soil freeze thaw cycles (FTCs) on Pb sorption and desorption
22 behavior in soils vulnerable to alternating periods of freezing and thawing. Results of the study suggested
23 that FTCs tend to increase Pb immobilization by increasing pH with increasing FTCs, which facilitated
24 formation of inner and outer sphere complexes. Adsorption capacity was correlated with carbonate and
25 effects of FTC on Pb adsorption may be more dependent on carbonate and clay content than OM, CEC, or
26 amorphous Fe.

27 [Burt et al. \(2014\)](#) investigated and compared the effects of different anthropogenic activities on
28 trace metal, including Pb, fate and transport. Surface and subsurface soil samples were collected at
29 locations throughout New York City (NYC) parks (Central Park, Pelam Park, and Van Cortlandt Park)
30 and from areas in the Bronx Watershed for chemical extraction analysis to investigate and compare trace
31 element extent, variability, and relationship between soil properties in the two study areas. Central Park
32 surface samples exhibited higher trace metal concentrations compared with Pelam or Van Cortlandt Park,
33 which may be related to proximity of Central Park sample sites to public roads and a long history of
34 intense human activities (shanties, gardening, piggery, and villages) compared with the relatively
35 undisturbed and mostly wooded Pelam and Van Cortlandt Parks. In the Bronx River Watershed, sum
36 trace metal concentration was significantly higher in sample locations collected from suburban
37 Westchester County compared with the more urbanized Bronx. The authors suggested that that the lower
38 trace element concentrations in the more urbanized area may be attributed to once industrialized land
39 being recently converted to parkland. Together these results suggest that trace element levels may not

1 necessarily be dependent on urbanization, current land use, or vegetation, but may be more reflective of
2 long-term history (type, degree, and age of human disturbances) influencing soil and hydrologic
3 processes.

4 Pb concentration trends in NYC parks decreased with depth confirming Pb airborne deposition
5 from several historical point and non-point sources. Conversely, concentration trends increased with
6 depth in the Bronx Watershed sample locations. These results were likely a result of soil formation in a
7 mantle of construction debris covered by anthropogenically transported soil. In addition, the formation of
8 carbonates from debris materials may have resulted in an increase in pH which increased Pb retention.
9 The sequential extraction analysis revealed that the predominant forms of Pb were the specifically
10 sorbed/carbonate-bound (SS/CAR) and the oxide-bound (OX) fractions, indicating that Pb is
11 predominantly in a form containing the carbonate precipitate, metallic-organic complexes, or metal-
12 oxides with low bonding forces (i.e., easily mobilized fractions). These results are in good agreement with
13 a study of NYC garden soils by [Cheng et al. \(2011\)](#) that also suggested anthropogenic Pb was generally in
14 the highly bioavailable and mobile SS/CAR and OX fractions (i.e., anthropogenic Pb in dust originating
15 from urban soils is more toxic and mobile than naturally occurring Pb). The authors found that the
16 exchangeable and more mobile fractions of Pb were larger in the NYC soil compared with soils found
17 near a Montana smelter, suggesting that the warmer and humid climate in NYC favored chemical
18 weathering and trace element mobility. The distribution of Pb in urban soils and the exchange of Pb
19 between urban soil and other media is further discussed in Section 1.3.4.

1.3.2.4. Summary

20 In summary, recent literature supports the conclusions from the 2013 Pb ISA ([U.S. EPA, 2013](#))
21 regarding Pb fate and transport through soils. Studies continue to report higher concentrations of Pb in
22 soils closer to stationary sources while also demonstrating the potential of Pb being deposited at
23 considerable distances from sources via long-range transport. Once deposited onto soil, Pb is strongly
24 retained in organic surface horizons with subsequent Pb retention and distribution in soil strongly
25 dependent on several physicochemical properties, including storage in leaf litter, amount and
26 decomposition rates of OM, composition of organic and inorganic soil constituents, mobile colloid
27 abundance and composition, microbial activity, and pH. In general, leaf litter, low rates of OM
28 decomposition, neutral pH, and soil constituents rich in charged surfaces such as OM, Fe and Mn oxides,
29 and clay minerals will lead to increased Pb retention and sorption. Conversely, thin organic layers,
30 increased OM decomposition, acidic pH, increases in anthropogenic Pb, and less reactive soil constituents
31 such as quartz will tend to increase Pb leaching from soils.

1.3.3. Fate and Transport in Water and Sediments

1 As discussed in the 2006 AQCD and the 2013 Pb ISA ([U.S. EPA, 2013, 2006b](#)) atmospheric
2 deposition, urban runoff, and industrial discharge are large contributors of Pb to surface waters with
3 greater runoff being linked to larger storm events following a dry period. Evidence from the 2013 Pb ISA
4 also found some evidence of a seasonal effect on runoff with greater runoff following snowmelt or rain-
5 on-snow events. Pb transport and sedimentation within aquatic systems depends upon chemical properties
6 including water pH and salinity, presence of OM and iron (Fe) and Mn oxides, total suspended solids
7 (TSS), as well as mechanical processes including turbidity and flow. Previous research has also shown Pb
8 is relatively stable in lacustrine and riverine sediments but resuspension of sediment into water or
9 dissolution from sediment often occurs during and following storm events and can be a larger source of
10 Pb to the water column and downstream reaches than concurrent atmospheric deposition. New research
11 primarily provides additional support for the 2006 AQCD and 2013 Pb ISA ([U.S. EPA, 2013, 2006b](#))
12 conclusions with additional information on runoff following fire events and seasonality influence on
13 transport and sedimentation. Furthermore, new literature provided information on temporal trends of Pb
14 concentrations in sediments and several studies are summarized in Section 1.3.3.4 to highlight the
15 importance of legacy Pb pools as potential “new” sources of Pb to waterways.

1.3.3.1. Biogeochemistry

1.3.3.1.1. Freshwater Biogeochemical Influences

16 The transport of Pb through freshwater systems is influenced by a variety of biogeochemical
17 factors such as OM content, redox, alkalinity, and seasonality. Since the 2013 ISA ([U.S. EPA, 2013](#)), new
18 information was found for how Pb transport and availability is increased in the presence of higher nutrient
19 levels and under anoxic conditions, while photolysis of OM reduces Pb concentration because it can be
20 bound more to organic molecules. There is also an improved understanding of the mechanisms for how
21 different types of OM (e.g., humic acids, or amount of aromaticity) interact with Pb and how dissolved
22 OM and PM can increase the mobility and solubility of metals in aquatic systems. An increase in DOC
23 leads to a decrease in the amount of Pb bound to PM because Pb instead binds more to dissolved organic
24 matter (DOM). Thus, activities that increase DOM (like surface mining or heavy rain events) can increase
25 the mobility and solubility of metals in aquatic systems ([Guéguen et al., 2011](#)). Similarly, [Chen et al.](#)
26 [\(2019\)](#) found that the solubility and mobilization of Pb increases through the formation of Pb-DOM
27 complexes. As the DOMs become more allochthonous, more humic-like, more aromatic, and optically
28 darker, the active Pb-binding fraction increases ([Chen et al., 2018](#)). Coordination chemistry has shown
29 that Pb predominantly binds to the phenolic and carboxylic group on a salicylic-type structure or two
30 adjacent carboxylic groups on catechol-type structures. [Cabaniss \(2011\)](#) found Pb(II) preferentially binds
31 to strong amine-containing sites which are often located on small molecular weight (MW<1000), and

1 lower aromaticity molecules. There are no highly aromatic molecules acting as strong ligands for Pb(II).
2 Pb(II) binds to phenolic groups but is more strongly bound by amine groups. At low metal loading, Pb(II)
3 is selectively bound to very high molecular weight compounds (>2000 amu). And Pb(II) are bound by
4 molecules with less negative overall charge (average charge of occupied ligand molecule $Z_{\text{bind}} > -1.6$) at
5 relatively low metal loading. At pH 7.0, Pb(II) binds in the order amines > phenols > carboxylates.
6 [Jeremiason et al. \(2018\)](#) found that DOM mobilizes historical deposits of Pb from bog peatlands and Pb
7 and DOC concentrations were correlated in the bog. The key factor is DOM leaching or production
8 leading to Pb redistribution among binding sites in solid peat and the dissolved phase. The amount of Pb
9 mobilized per unit DOC ($\mu\text{g}/\text{mg}$ DOC), was greater in bogwater (0.047; range 0.037–0.067 at individual
10 sites) than lagg water (0.033; range 0.007–0.050). Interestingly, Pb was found to preferentially adsorb
11 onto bacterial cells (organic material) than on clay minerals ([Du et al., 2016](#)).

12 Suspended particulate matter (SPM) and sorption material also influence Pb transport and
13 availability. SPM significantly influences Pb adsorption. Total Pb concentrations in water were higher
14 when the content of the PM in the river water was high ([Milačić et al., 2017](#)). Also, the highest
15 partitioning coefficients observed for Pb, were a consequence of its high affinity to SPM and low Pb
16 solubility in water. Pb binding to SPM in the lower Waikato River in New Zealand is predominantly via
17 Fe-oxide surfaces and can be reliably predicted using surface complexation adsorption modeling
18 ([Webster-Brown et al., 2012](#)).

19 Other metals, nutrients, and inorganic compounds in the sediment and open waters can affect Pb
20 mobilization. High nutrient levels can increase the potentially mobile fractions of Pb ([Kang et al., 2019](#)).
21 A significant negative relationship exists between total phosphorus (TP) and Pb concentrations per unit
22 mass of phytoplankton in lakes ([Gormley-Gallagher et al., 2016](#)). Sulfide also showed a negative
23 relationship with Pb, likely reflecting precipitation of Pb-sulfide complexes in sulfide-rich porewater
24 ([Carling et al., 2013](#)). [Lombardi et al. \(2014\)](#) found that the percent labile Pb (86 %) compared with
25 percent dissolved Pb suggests that most of the Pb was complexed with inorganic compounds. Pb was
26 complexed preferentially with CO_3^{2-} (25 %), NO_3^- (22 %), and OH^- (19 %). Chlorophyll *a* and TSS were
27 also correlated with most Pb fractions. Groundwater may be contaminated with Pb, and this may be due
28 to strong correlations between Pb and Fe or Mn oxides and with total dissolved solids ([Wang et al., 2016](#)),
29 and this study suggests that groundwater contaminant of Pb is due to natural processes and not from
30 surface water contamination.

31 Pb transport and availability is influenced by redox conditions. Pb is typically released from
32 sediments in anoxic environments and adsorbed from the overlying water in an aerobic environment
33 ([Kang et al., 2019](#)). For the exchangeable fraction (characterized by soluble species, species with cation
34 exchanges sites, and carbonate-bound species), Pb increased in aerobic conditions and for both high and
35 low nutrient levels but decreased under anoxic conditions. Sediment absorbs more Pb^{2+} under aerobic
36 conditions. Another study found that Pb can precipitate under either oxic or anoxic conditions, but due to
37 different mechanisms. In a eutrophic lake, [Chen et al. \(2019\)](#) observed that algae degradation may

1 decrease redox state in sediments and sulfide may be released from the degraded algae, which may
2 promote the formation of Pb-sulfide precipitations under anoxic conditions. Whereas in oxic conditions
3 (high redox state), Fe^{2+} and Mn^{2+} oxidation may occur in sediments and result in the adsorption or
4 coprecipitation of Pb. Also, Pb can be confined to an immobile form (organic sulfides) at higher alkalinity
5 in stream sediments, and gradually be released due to chemical weathering (Pearson et al., 2019). Li et al.
6 (2016) found that in acid mine drainage, Pb was found to be associated with the carbonate fraction and
7 under waterlogged conditions, dissolved Pb increased when Fe increased in concentration. In waters with
8 acid mine drainage, Pb was predominantly present in the residual (77.7–85%), followed by oxidizable
9 (9.4–12%) and reducible (5–10%) fractions. Also, the decomposition of OM like cyanobacteria can cause
10 a reduction in the oxidation reduction potential (ORP), which can result in an increase in Pb bound to
11 sulfate ions (Ni et al., 2019). Chen et al. (2019) also found that Pb in both eutrophic and non-eutrophic
12 lakes was commonly complexed as $\text{Pb}(\text{HS})_2$, PbCO_3 and to a lesser extent: Pb^{2+} , PbOH^+ , $\text{Pb}(\text{OH})_2$,
13 PbSO_4 , $\text{Pb}(\text{CO}_2)_2^-$, and PbHCO_3^+ .

14 Temperature and seasonality also influence Pb adsorption and transport in freshwater systems. Pb
15 showed higher concentrations during the spring than summer in river samples (Zhang et al., 2016a).
16 Zhang et al. (2016a) also found that in the spring, the majority of Pb is found in the inert form (not
17 reactive with NH_4^+ or OH^- ion exchange resins) and only ~1 to 5 % of Pb found in the organic or labile
18 forms. But in the summer, there were higher percentages of labile Pb ranging from ~10 to 60% in the
19 rivers. The organic fraction was the same in both seasons, while the labile fraction increased (on average)
20 from 6.75 to 19.95 % between spring and summer. On average, the labile Pb fraction increased in all the
21 rivers during summer. The increase in labile concentrations might be attributed to human activities,
22 leading to increase potential toxicity in these rivers. In winter months, Sun et al. (2018) found that Pb^{2+}
23 has a low binding energy in ice compared with other cations ($\text{Fe} > \text{Cu} > \text{Mn} > \text{Zn} > \text{Cd} > \text{Hg} > \text{Pb}$). Lombardi et
24 al. (2014) found that total, dissolved, complexed, and labile Pb species were all higher in the winter, while
25 Pb was present more in the particulate form in the summer. Chen et al. (2019) observed the highest
26 dissolved Pb concentrations in July for a eutrophic lake, while the highest dissolved Pb concentrations
27 were in January for a lake covered by floating and submersed macrophytes. The greatest increase in Pb
28 complexation with DOM occurred in the eutrophic lake in July, while it occurred in the non-eutrophic
29 lake in April. However, the degree of Pb complexation to DOM was significantly larger in the non-
30 eutrophic lake in all seasons. The mobility of Pb in sediments showed significant seasonal variations,
31 reflected by a high release of Pb during the spring and summer in the algae-dominated region and during
32 the autumn and winter in the macrophyte-dominated region. A possible mechanism is that in the algae-
33 dominated regions of the lakes, increased bacterial abundance in the sediments during the spring
34 promoted microbial reduction of Fe/Mn oxides, which likely released Pb from sediments.

35 Pb was found to be higher during periods of extreme flooding (Milačić et al., 2017), where Pb
36 inputs are primarily derived from heavy industry activities or mining and metallurgy activities (depending
37 on the site). Valencia-Avellan et al. (2017) also found that Pb concentrations increased with peak flow in
38 an ephemeral tributary. This is likely because Pb is strongly associated with both particulate and colloidal

1 Fe and Al oxides, and also cerussite (PbCO_3), SO_4^- , and DOC, which can all increase during high flood
2 periods which are associated with the resuspension of sediments into water.

3 Light also has an influence on the dynamics of Pb in freshwater systems. Within an acid mine
4 drainage impacted wetland, it was found that increased light levels caused a reduction in ferrous iron and
5 this was associated with an increase in Pb concentration ([Duren and McKnight, 2013](#)). The mechanism
6 for this process is the formation of superoxide radicals (O_2^-) and H_2O_2 from the photoreduction of DOM
7 in the wetlands, where H_2O_2 reacts with Fe_2^+ and converts it to Fe_3^+ (reducing the amount of Fe_2^+ during
8 the day). During photolysis, [Drozdova et al. \(2020\)](#) observe two simultaneously occurring processes: 1)
9 the degradation of high molecular weight organo-mineral colloids and the formation of low molecular
10 weight organic molecules and Pb complexes, and 2) the formation of the >0.22 μm particulate aggregates
11 of Pb and OM. The DOM degradation produces both CO_2 and HCO_3^- whereas Pb which is initially
12 associated with organo-ferric colloids are subjected to coprecipitation with newly formed Fe(III)
13 oxy(hydr)oxides. Also, photolysis caused a decrease in Pb by 48% in solution and this may be because Pb
14 is correlated to changes in concentration of Fe, DOC, and humic substances. For instance, Fe-OH and
15 organic ligands can form ternary surface complexes with Pb. An alternative mechanism of metals removal
16 could be their precipitation in the form of individual metal hydroxides that occurs after photo-degradation
17 of metal-ligand (Me-L) complexes.

1.3.3.1.2. Saltwater Biogeochemical Influences

18 The transport of Pb through saltwater systems is influenced by a variety of biogeochemical
19 factors such as salinity, organic matter content, redox, alkalinity, and seasonality. Since the 2013 ISA
20 ([U.S. EPA, 2013](#)), new information was found on how Pb concentrations in solution increases with
21 increasing salinity and temperature, but decreases in the presence of dissolved organic carbon, Fe(III) and
22 Mn(IV/III) (hydr)oxides, which provide important binding sites for heavy metals under high dissolved
23 oxygen (oxic) conditions. There is also more information on the role of sulfide in estuarine systems and
24 on whether Pb comes from anthropogenic or natural sources.

25 Salinity of estuarine and coastal waters can have a strong influence on Pb fate and transport. ([Yao
26 et al., 2016](#)) found that the concentration of Pb adsorbed to PM decreases with increasing salinity in the
27 medium-low salinity of the estuary near the river mouth, indicating the release of Pb during early mixing
28 stages in the estuary. The metal release resulted from a balance between two opposite processes: (1) metal
29 mobilization due to ionic exchange or degradation of organic complexes and (2) metal re-adsorption onto
30 an existing or newly formed solid phase. Basically, with increasing salinity, cations such as Na^+ , Ca^{2+} ,
31 and Mg^{2+} compete for the adsorption sites on particle surfaces, thereby decreasing adsorption and
32 enhancing the release of sorbed Pb from the particle surfaces. Similarly, [Zhao et al. \(2013\)](#) observed that
33 as salinity in the Yangtze Estuary increased, Pb was released from the sediments, but it was minimal
34 (0.004–0.017%), due to preferential retention in Fe-Mn oxides and organic content. Pb cations seemed to
35 be sorbed more specifically to sites with high dissociation constants (and high sorption energies), making

1 them less vulnerable to leaching. ([Karbassi et al., 2014](#)) also observed greater Pb flocculation at lower
2 salinities (0.5‰) and constant pH of 8. However, another study found the opposite pattern; while the
3 partition coefficient K_d (L/kg), which is the relationship between the sorbed state to the dissolved state of
4 a metal, generally decreases with salinity due to higher ionic strength and competition for sorption sites,
5 [Alkhatib et al. \(2015\)](#) found the K_d for Pb increased with salinity (with K_d values at 234 L/kg in
6 freshwater, 575 L/kg in brackish water, and 1341 L/kg in seawater) and this is mainly attributed to
7 formation of insoluble metal species, like $PbSO_4$, which led to higher K_d values with the increase of
8 salinity. Another study with this pattern found that for PbS , freshwater exhibited the highest Pb release
9 followed by seawater and estuary water ([Chou et al., 2018](#)). Clearly, other factors like the types of
10 inorganic species and metals and other conditions must influence the impact that salinity has on the
11 transport of Pb in saltwaters.

12 The presence of different minerals in estuarine and seawaters can influence the transport of Pb
13 between sediments, saltwater, and the atmosphere. For example, [Shelley et al. \(2018\)](#) observed that Pb
14 and Al was significantly correlated ($r^2 = 0.478$) in saltwater and that Pb solubility was greater in saltwater
15 than in ultra-high purity water used as a control, though Pb solubilities decrease as aerosol loading
16 increased. Fe(III) and Mn(IV/III) (hydr)oxides provide important binding sites for heavy metals under
17 oxic conditions, and sulfide provides important binding sites for Pb under anoxic conditions ([Wang et al.,
18 2013](#)). Consequently, the reductive dissolution of Fe(III) and Mn (IV/III) (hydr)oxides could encourage
19 the release of Pb into solution. But dissolved levels of Pb became undetectable within 10 days suggesting
20 that it can be almost completely sequestered in the metal sulfide phases under sulfate-reducing conditions
21 (during bacterial sulfate reduction activity). [Morgan et al. \(2012\)](#) found acid volatile sulfide to have a
22 strong relationship with reactive Pb in estuaries and a strong relationship between FeS and Pb in
23 sediments, where Pb sulfates are more likely to precipitate than FeS due to lower solubility. Thus, FeS is
24 likely to retain Pb in estuarine sediments. [Tovar-Sánchez et al. \(2019\)](#) observed that both Pb and Fe were
25 abundant in the sea surface microlayer. And [Keene et al. \(2014\)](#) saw a strong correlation between total Pb
26 and reactive Fe in interfacial sediments of an estuary, with 50% of Pb being associated with reactive or
27 “acid extractable” phases in the sediment. [Ebling and Landing \(2015\)](#) studied the Pb levels in the sea
28 surface of the open ocean (“microlayer” – the thin layer at the boundary between the ocean and the
29 atmosphere) and measured dissolved, labile particulate, and refractory particulate trace element
30 concentrations of the sea surface microlayer. They found dissolved Pb to increase in the microlayer by a
31 factor of 2–3 over time, coinciding with an increase in Fe, which may have come from precipitation. At
32 the same time, the refractory particulate Pb increased by a factor of 23 in the microlayer. Pb in the
33 microlayer had retention time of about 1–2 days. The enrichment factor (EF) for Pb was >1 demonstrating
34 enrichment in the microlayer. [Cánovas et al. \(2020\)](#) observed Pb was abundant in PM (37–59% in
35 dissolved fraction) and that 66% of Pb was found forming Cl^- complexes, ~20% as CO_3^- complex, 5% as
36 Free Pb, and 5% as Pb hydroxide. Pb showed a balanced speciation between the uncharged and positively
37 charged species.

1 Particulate matter has been found to strongly influence Pb transport and fractionation in seawater.
2 [Angel et al. \(2016\)](#) reported that the amount of dissolved Pb concentrations in seawater was dependent on
3 the concentration of precipitate present, decreasing as the precipitate concentration increased. The
4 composition of the precipitate formed is likely to be a metastable Pb chlorocarbonate. [Feng et al. \(2017\)](#)
5 observed how the partitioning coefficient (K_p), for the amount of Pb sorbed to SPM was highest for Pb
6 compared with other trace metals (Ni, Cr, Cu, Hg, Zn, Cd, and As). And the K_p for Pb is higher in the
7 SPM than for the sediment-water interface. This is because SPM has a smaller particle size, and higher
8 specific surface area and OM content, and thus can adsorb more heavy metals. The exchangeable and
9 carbonate fractions of Pb also had significant positive correlations with the K_p for Pb in SPM and the
10 exchangeable, carbonate, and residual fractions of Pb had significant positive correlations with K_p for Pb
11 in sediments. Thus, adsorption is likely to be the dominant partition process of Pb. [Burton et al. \(2019\)](#)
12 found through a review study that the removal efficiencies (of the metal from the water column to SPM)
13 for 7 of the 12 estuaries were at or greater than approximately 75% for Pb. And metal removal efficiency
14 was greater for Pb than Cd and Zn, consistent with the metal's partition coefficient. Pb accumulates more
15 in the finer fractions of clay (<8 μm) and fine silt (8–16 μm) ([Yao et al., 2016](#)). Pb concentrations in the
16 bulk SPM varied from 25 to 38 mg Pb/kg, with an average of 32 mg Pb/kg. Pb had an average enrichment
17 factor (EF) value of 0.81 and were all <1.5. This indicates that the Pb concentrations originated from
18 natural weathering processes. The EF of an element is defined as the ratio of that element to a
19 conservative element in a sample divided by the ratio of that element to the same conservative element in
20 a background reference sample. An EF value between 0.5 and 1.5 suggests that the trace metals may be
21 entirely from crustal materials or from natural weathering processes, while an EF value >1.5 suggests that
22 a significant portion of trace metals is delivered from non-crustal materials, or nonnatural weathering
23 processes, like anthropogenic activities ($\text{EF} = [\text{Pb}]/[\text{Fe}]_{\text{sample}} / [\text{Pb}]/[\text{Fe}]$) ([Yao et al., 2016](#)). Another
24 study, however, observed that the EF for Pb was approximately 600, indicating it primarily originated
25 from anthropogenic contributions ([Xing et al., 2017](#)). One study by [Holmes et al. \(2014\)](#) examined the
26 role of estuaries in modifying the adsorptive properties of new and aged plastics towards trace metals and
27 found the absorption capacity of Pb on plastic surfaces to decrease from river water to seawater and with
28 decreasing pH due greater competition with other cations.

29 Temperature, oxic conditions, and organic content influence Pb transport in saltwaters. Within
30 estuarine waters, temperature was positively associated with free Pb concentration, with a 1°C increase
31 corresponding to approximately a 7% increase in free Pb concentration ([Dong et al., 2016](#)). Dissolved
32 oxygen (DO) was also found to be dominant factor that controlled the release of Pb from coastal
33 sediments, with increased hypoxia causing increased Pb in overlying waters compared with sediments
34 ([Liu et al., 2019](#)). Similarly, [Banks et al. \(2012\)](#) observed greater dissolved Pb concentration in porewater
35 estuarine sediments at lower DO levels, where the ratio of dissolved Pb concentration to metal
36 concentration was 1.2 for 5% DO, 1.1 for 20% DO, and 0.9 for 75% DO. In anoxic conditions the
37 presence of wetland plants, like *S. alterniflora*, could lead to higher concentrations of Pb in the sediments,
38 via pumping oxygen into the rhizosphere, which can cause the release of Pb to sulfates ([Wang et al.,](#)
39 [2013](#)). Similar to freshwater systems, within estuarine waters, DOC and free Pb concentration had a

1 negative relationship, indicating organic ligands in the water column were more important binding agents
2 for free Pb ions relative to particulate organic ligands ([Dong et al., 2016](#)). Also, the presence of HA
3 showed inhibition effects on its metal release of Pb from PbS ([Chou et al., 2018](#)). Carbon dioxide (CO₂)
4 also influences Pb; a model predicted that under scenarios of increasing CO₂, free Pb could increase from
5 9–97% and organically bound could increase by 5–43% ([Stockdale et al., 2016](#)).

6 Several studies also found that within saltwater systems, pH has a negative impact on free Pb
7 concentrations. [Vasyukova et al. \(2012\)](#) also found that % dissolved Pb decreases as pH increases, and Pb
8 is an element strongly associated with colloids and exhibit significant increases of relative proportion of
9 colloidal forms with pH increase. Another study found that the partition coefficient K_d (L/kg), which is
10 the relationship between the sorbed state to the dissolved state of a metal, was greatest for Pb at pH 7, at
11 16434 L/kg, indicating that more sorbed Pb was present at neutral pH ([Alkhatib et al., 2015](#)). When
12 assessing the precipitation of Pb from PbS, there was minimal release of Pb from PbS at pH 8,
13 intermediate Pb released at pH 7 (213 mg/L/m²) and even more Pb released at pH 5 (386 mg/L/m²), which
14 suggests that H⁺ plays a role in the oxidative dissolution of Pb sulfides ([Chou et al., 2018](#)).

15 Seasonality, rainfall, and tidal flows can influence Pb dynamics in estuaries and coastal waters. In
16 the study by [Hierro et al. \(2014\)](#), Pb was found primarily in PM (average of 825 µg/L). Particulate matter
17 Pb concentrations (per volume) were 2–3 times higher in PM carried by ebbing tide compared with the
18 rising tide, due to increased PM when there is a fall in sea level. Often, Pb sorbed/coprecipitated with Fe
19 hydroxides, and highest particulate concentrations coincided with the estuarine maximum turbidity zone.
20 In terms of rainfall, one study found that increased rainfall resulted in lower free Pb concentrations, likely
21 due to dilution ([Dong et al., 2016](#)), while another study found higher levels of Pb in the spring when the
22 rainfall amounts were larger compared with summer months ([Xing et al., 2017](#)). Additionally, this study
23 found strong correlations between crustal-derived elements (Al, Fe) and anthropogenic elements (Pb, Cd,
24 Zn) likely due to both being influenced by air deposition rainwater runoff ([Xing et al., 2017](#)). Also, during
25 seawater-freshwater interaction from seawater intrusion to an aquifer, it was observed that Pb exhibited
26 significant correlation with colloids and was thus sensitive to the flow of the colloidal fraction where
27 seawater and freshwater are interacting ([Tan et al., 2017](#)).

1.3.3.2. Transport into Water (including Runoff)

28 The 2006 Pb AQCD concluded Pb in runoff was mostly in the particulate fraction and identified
29 runoff as being dependent on storm intensity and time between rain events ([U.S. EPA, 2006a](#)). The 2013
30 Pb ISA provided information on Pb runoff from roadways, urban areas, and snow melt into watersheds
31 ([U.S. EPA, 2013](#)). New research provides additional support for both the 2006 AQCD and 2013 Pb ISA
32 conclusions with additional information on runoff following fire events and urban sources of Pb unique to
33 city history and planning.

1.3.3.2.1. Urban

1 Pb in runoff in urban areas is correlated with surrounding land use characteristics such as
2 impervious surface area and road density. In a study that examined Pb concentration and distribution in
3 bed sediments of the Palolo drainage basin in Hawaii, [Hotton and Sutherland \(2016\)](#) found of the three
4 streams that comprise the basin, Palolo had Pb concentrations of 134 mg Pb/kg compared with Pukele
5 (24 mg Pb/kg) and Waiomao (7 mg Pb/kg). Furthermore, Palolo had high concentrations along its entire
6 length whereas Pukele and Waiomao showed highest concentrations downstream. This high Pb
7 concentration along Palolo was correlated with urban land-use characteristics including street length,
8 storm drain length, the number of storm drain inlets and outlets, as well as vehicle counts and overall
9 population. Urban development around Palolo was higher than around the other two streams in the
10 drainage basin. In a similar study, sediment Pb concentration was measured in highly urbanized
11 watersheds across several U.S. cities ([Nowell et al., 2013](#)). Pb was positively correlated with local urban
12 factors and study area variables including population density, urban land cover, road density, and amount
13 of impervious surface area as well as total organic carbon. Boston had higher Pb concentrations than other
14 cities with comparable urbanization and sediment total organic carbon and the authors highlight this
15 higher Pb concentration in Boston likely reflects the city's long history of industrial activity and high-
16 density development. [McKenzie and Young \(2013\)](#) examined Pb water column fractions following storm
17 events in creeks draining different surrounding land-use areas. Highways and urban areas had higher
18 runoff loads compared with agricultural and natural sites. Agricultural storm loadings were similar to
19 those in natural systems and irrigation loadings were less than storm loadings. Pb was primarily
20 associated with suspended sediments so would have low mobility and bioavailability. Highway runoff, on
21 the other hand, had high levels of dissolved Pb.

22 Several recent studies highlight urban-specific sources of Pb to city waterways due to city design
23 and historical pollution. For example, a study by [Coxon et al. \(2016\)](#) examined and mapped the
24 contamination histories of the rivers that drain the lower western Chesapeake Bay basin. Sources of
25 contamination have changed over Virginia's history and reflect the development of the area. Western
26 mountain reaches have elevated Pb levels due to lithology and historical mining while agriculture and
27 urbanization contribute to Pb enrichment across the drainage basin. Norfolk naval base and shipyard is a
28 current and significant source of metal enrichment, as are incinerators, older office buildings with Pb
29 paint, and ordnance storage. Furthermore, changes in urban land-use management have led to legacy Pb
30 pools becoming a new source and Pb enrichment downstream of urban areas is high—with sediments
31 downstream of Richmond showing particularly high Pb enrichment levels. Overall, fuel combustion,
32 street dust, and highly contaminated urban soils are the contemporary suppliers of Pb to the Virginia
33 Chesapeake waterways. In the Gwynns Falls watershed area in Baltimore, Pb concentration in riparian
34 sediments decreased with increasing distance from the city center (when normalized for sediment surface
35 area). Also of note, three hotspots of contamination in the urban system occurred adjacent to areas that
36 had been identified in 1979 as artificially filled ([Bain et al., 2012](#)). A non-U.S. study found that despite
37 restoration efforts enacted 30 years prior, Pb sediment contamination in an urban lagoon in Sardinia

1 exceeded 100 mg Pb/kg at 22 of 34 monitoring locations ([Atzori et al., 2018](#)). High Pb concentrations
2 were not correlated with OM content and showed similar contamination patterns with mercury (Hg).
3 Instead, Pb (and Hg) peaks were in sites with proximity to a chlor-alkali plant and an airport. In a study
4 that examined trace metal export in relation to soil concentrations, soil and water properties, and
5 watershed land use across several New England watersheds, Pb export rates varied from 0.03 to 0.37 kg
6 Pb/year/km² ([Richardson, 2021](#)). Dissolved Pb concentration was not correlated with soil Pb
7 concentration but was positively correlated with aquatic Zn and dissolved organic carbon concentrations.
8 Furthermore, dissolved Pb export was positively correlated with watershed cover of wetlands and
9 negatively correlated with percent of forest cover. The author suggests the positive correlation with
10 wetland cover may be due to wetlands serving as a reservoir of historic pollution and the negative
11 correlation with forests is due to there being less development and therefore less pollution, as well as
12 potentially higher retention of trace metals (Cu, Pb, and Zn) by soil iron oxides. Another recent study
13 sought to assess the trace metal loading rates in the Great Lakes basin and estimated results found Pb
14 inputs to Lakes Superior and Michigan were primarily atmospheric while Lakes Erie and Ontario received
15 proportionally more Pb from tributary inputs and to a less degree, from connecting lake channels. Lake
16 Huron, being in the middle, unsurprisingly receives Pb from atmospheric deposition and tributaries in
17 relatively more equivalent contributions ([Bentley et al., 2022](#)). Lastly, another study examined the effects
18 of road salts and deicers on metal mobilization within the soil profile ([Schuler and Relyea, 2018](#)). Sodium
19 (Na) can displace Ca and magnesium (Mg) in the soil which can increase porosity of the soil structure
20 leading to mobilization of metals. Salts can also mobilize Pb by breaking down the organic-rich colloidal
21 structures that often bind Pb within the soil matrix. Salts can also displace metals, including Pb, from
22 binding with organic compounds because the binding affinity is higher for Na, Ca, and Mg than it is for
23 heavy metals. Although this study did not measure Pb concentration in water or stream sediments, it
24 highlights how salts and road deicers can increase Pb mobility into water systems through the physical
25 and chemical changes to the soil matrix—an interaction effect unique yet widespread in urban systems.

1.3.3.2.2. Non-Urban

26 As in urban environments, Pb in runoff in non-urban areas is primarily within sediments and
27 runoff into waterways is driven by storm events and overall precipitation patterns. A European study
28 examined trace metal budgets across 14 forested catchments over the period of 1997–2011 ([Bringmark et
29 al., 2013](#)). Due to high anthropogenic deposition for decades, Pb accumulation in catchment soils was
30 high. Pb is bound to soil OM in these soils leading to high retention of Pb in the system. At higher altitude
31 sites which experience greater precipitation, retention is lower due to greater runoff and transport of PM
32 out of the system. In a similar study in England, while Pb deposition has decreased in recent decades,
33 legacy Pb in peat catchments is a continuing source of Pb to waterways ([Rothwell et al., 2011](#)).
34 Atmospheric deposition was measured at 34 gPb/ha/year while fluvial outputs were 316 gPb/ha/year.
35 Following storm events, Pb runoff into waters occurs primarily as suspended particles (261 gPb/ha/year)
36 with a smaller aqueous portion (55 gPb/ha/year). In the Snowy Mountains of Southeast Australia, down-

1 catchment reservoirs with large catchment size showed comparable sediment metal enrichment values to
2 soil enrichment values indicating the soil surface which contains metal values reflective of anthropogenic
3 deposition is the source of sediment to reservoirs, not eroded subsoils (which is less contaminated)
4 ([Stromsoe et al., 2015](#)). However, Pb (and chromium) are depleted in reservoir sediments compared with
5 soils, indicating these more particle-reactive materials are bound up within the soil matrix and are not
6 being washed into down-catchment reservoirs.

7 In waterways draining the Alberta badlands, total Pb concentration frequently exceeded Alberta
8 guidelines for freshwater biota ([Kerr and Cooke, 2017](#)). However, Pb concentration was positively
9 correlated with TSS due to an increase in sediment mass, not due to increased sediment Pb concentration,
10 highlighting the importance of erosion and precipitation interactions in arid systems. In contrast, a study
11 in Brazil found metal fluxes, including Pb, were highest during the dry season compared with the wet
12 season and were due to suspended sediments ([Bezerra da Silva et al., 2015](#)). Unlike the studies measuring
13 flux in the badlands of Alberta, this increase was not due to bedload but due to a lack of dilution. The
14 watershed included agricultural and industrial sources as inputs and Pb was likely from coal combustion,
15 solid waste incineration, and legacy Pb from petroleum but sources could also include pesticide use,
16 sewage sludge runoff and agricultural wastewater.

17 Pb runoff and accumulation from soils into lakes is also influenced by snow and ice melt. Overall,
18 three catchments in the Pyrenees have greater Pb concentration in lake sediments than surrounding soil
19 and bedrock ([Bacardit et al., 2012](#)). Lakes within the two watersheds above 2000 meters a.s.l. had lower
20 Pb concentrations than lakes within the lowest watershed (1655 meters). The lower Pb accumulation in
21 these high elevation lakes may be due to snowmelt moving soil bound Pb further down this catchment
22 and bypassing the lakes altogether due to lake ice. [Kim et al. \(2015\)](#) measured dissolved Pb (and other
23 trace elements) from melting glaciers along the Antarctic coastline in Marian Cove and determined
24 glacier meltwater is a significant source of Pb to the cove. In ice, Pb was measured at values between 90–
25 920 pM and 88–550 pM in snow. Pb ranged from 30–120 pM in seawater and Pb concentration decreased
26 with increasing salinity in seawater samples.

27 As described in Section 1.2.4, fires are a large and increasing contributor to ambient air Pb
28 concentrations. A few recent studies have specifically examined Pb (and other metals) movement
29 following fires. Overall, metal mobility to waterways following a burn event is largely dependent upon
30 the first storm event following the fire. A review of metal mobilization following fire found fires can lead
31 to mobilization of Pb into waterways ([Abraham et al., 2017](#)). Pb bound within the soil matrix, in
32 vegetation, or other burned materials, is released following a burn and is readily washed into downstream
33 waterbodies with rainfall. [Burton et al. \(2016\)](#) found the total Pb (unfiltered) median water concentrations
34 downstream of the Station Fire in California was higher than concentrations outside the burn area and
35 higher than concentrations measured prior to storm events. Furthermore, these post-fire total Pb
36 concentrations exceeded the recommended aquatic use criteria (CCC) following storm events. The
37 authors suggest these higher Pb concentrations within burned areas was likely due to ash runoff because

1 Pb concentration in ash samples was higher than soil samples. Pb source in the ash was primarily
2 vegetative or biogenic in origin but Pb in ash collected from residential burn areas was higher. A 2011
3 global review that examined water quality in forested catchments following wildfire reported Pb exceeded
4 water quality guidelines in Australia and the United States and is associated with high suspended solids
5 concentrations following post-fire rainfall events ([Smith et al., 2011](#)).

1.3.3.3. Sedimentation, Transport, and Flux in Water and Sediment

6 As in the 2006 Pb AQCD and the 2013 Pb ISA ([U.S. EPA, 2013, 2006b](#)), chemical and physical
7 characteristics of the water such as pH, salinity, and flow rate as well as the chemical and physical
8 properties of the suspended sediments determine the fate of Pb and therefore influence rates of
9 sedimentation and flux as well as transport downstream. Recent publications provide additional support
10 regarding Pb adsorption onto organic-rich or small colloid particles as well as the importance of water
11 flowrate in settlement and downstream transport. Literature since the previous ISA provides new detail of
12 the effects of seasonality on Pb fate and transport in water. Seasonal patterns of precipitation can lead to
13 differences in runoff, flowrate, and turbidity, for example, which can subsequently alter sedimentation
14 rates, transport downstream, and flux from sediments.

1.3.3.3.1. Urban

15 Within urban environments, city infrastructure can lead to increased loading and movement into
16 downstream reaches. In an urban Baltimore area watershed, sediment concentrations of metals including
17 Pb increased with urbanization ([Bain et al., 2012](#)). Stormwater flow and well-drained soils with low OM
18 interact to increase runoff and downstream movement of Pb. In an arid California watershed, urban
19 infrastructure allows for the quick movement of water away from urban areas resulting in increased Pb
20 concentrations in receiving water bodies following rain events ([McKee and Gilbreath, 2015](#)). Pb
21 concentration was correlated with water turbidity due to Pb being primarily within the particulate fraction.
22 Another study examined the influence of runoff and other diffuse pollution sources on lake water and
23 sediment chemistry of Hough Park Lake in Central Missouri ([Ikem and Adisa, 2011](#)). The lake is
24 surrounded by a golf course with little natural buffering between the course and the lake. Average lake Pb
25 concentrations in sediments were 11.05 mg Pb/kg for littoral zone sediment and 0.79 mg Pb/kg in pelagic
26 zone sediment. Total Pb in the water column was 0.0004 mg/L in the spring. Pb content was primarily in
27 the residual phase (75%) and the authors suggest since all heavy metals were primarily within the residual
28 phase (lowest mobility phase), the source of heavy metals is likely due to erosion and runoff of parent
29 rock material.

30 Seasonal and local weather patterns interact with other factors such as soil and sediment physical
31 and chemical characteristics to transport Pb into and within water bodies. One study sampling water

1 column and surface sediment Pb concentrations in Lake Pontchartrain along I-10 shows seasonal
2 differences ([Zhang et al., 2016b](#)). Spring sediment concentrations ranged from 16.42–28.25 mg Pb/kg and
3 from 6.94–21.79 mg Pb/kg during the summer. Water column Pb concentrations in spring ranged from
4 4.65–7.4 µg Pb/L and from 4.7–10.4 µg Pb/L during the summer. The higher sediment concentrations in
5 spring and higher water column concentrations in summer may be due to warmer summer water
6 temperatures releasing more Pb from sediments. Differences between spring and summer could also be
7 due to less precipitation and sediment disturbance via turbidity during cooler months. Pb in sediments
8 was primarily in the stable residual fraction. In the San Juan River delta of Lake Powell, sediment loading
9 and associated Pb contamination in the downstream reaches reflect an interaction of seasonality and
10 precipitation ([Frederick et al., 2019](#)). During the spring, high sustained flow from snowmelt occurs at
11 high elevation tributaries with a history of mining. This increased flow in the spring contributes more Pb-
12 contaminated sediment to the downstream delta area. Tributaries that contribute greater sediment during
13 short rainfall events have lower Pb concentrations.

14 Transport and settlement patterns of Pb are also a function of sediment particle size. In a study
15 examining the downstream transport of heavy metals from the superfund site Iron Mountain into the
16 Sacramento River, Pb enters the Keswick Reservoir primarily in the dissolved form and is precipitated or
17 adsorbed into the particulate phase ([Taylor et al., 2012](#)). However, Pb does not settle out of the water
18 column and is instead transported far downstream due to particle association with the colloid phase. This
19 transport of fine contaminated particles occurs during both high and low flow conditions. In the Miami
20 River in Florida, Pb was negatively correlated with sediment particle size ([Tansel and Rafiuddin, 2016](#)).
21 As particle size decreased, Pb content increased with 900 mg Pb/kg found within the fine sediment
22 fraction. Due to Pb being bound to finer sediments, turbidity from boating, tidal action, and rain events
23 are of concern for resuspension and mobilization of Pb within the water column. Following a dam
24 removal on the Pawtuxet River in Rhode Island, fluxes of all metals including Pb increased in response to
25 river flow ([Katz et al., 2017](#)). As river flow increased, sediments were resuspended into water and this
26 particle-bound Pb moved downstream into Narragansett Bay. [Sebastiao et al. \(2017\)](#) examined Pb (and
27 other metals) in the river sediments at two paved river fords in suburban Philadelphia and across seasons.
28 Pb was found in higher concentrations in April, July, and in December, but these higher values were not
29 correlated with any rain event. Pb was positively correlated with organic content but only at the ford that
30 was less used. The authors suggest this relationship occurs due to less water movement from traffic which
31 in turn allows Pb to adhere to organic particles and settle out of the water column. Furthermore, since
32 sediment Pb concentration was still high during the winter when the fords are closed to traffic, Pb
33 persistence in this system is not seasonal.

1.3.3.3.2. Non-Urban

34 Seasonal effects such as snowmelt can impact Pb movement. Anthropogenic Pb per unit area in
35 high elevation lake sediments in the Pyrenees was lower than in the surrounding catchments, and this is

1 potentially due to Pb deposition largely accumulating in snowpack followed by melting and outwash into
2 lower elevation systems before the ice on the high elevation lakes can melt and incorporate this deposition
3 portion as happens at lower elevation lakes ([Bacardit et al., 2012](#)). In a study that measured Pb in surface
4 sediment at the river mouth of the Papaloapan River across a gradient of increasing water depth in the SW
5 Gulf of Mexico, Pb was positively correlated with organic carbon but only during the winter months
6 ([Rosales-Hoz et al., 2015](#)). During the summer, Pb was strongly positively correlated with Al₂O₃ (also
7 during the winter months though not as strongly). Pb concentration increased with water depth as did the
8 muddy proportion in sediments. Muddy sediment output from the river was greater during the summer.

9 As in urban aquatic environments, environmental and physical drivers of water flow patterns
10 largely govern the transport of Pb within non-urban aquatic environments with additional influence of
11 sediment and water chemistry. Pb water concentration at European bog and peaty riparian sites was
12 positively correlated with dissolved organic carbon (particularly at the bog location) ([Broder and Biester,
13 2017](#)). However, at this location, Pb concentrations in water were lowest during the spring snowmelt
14 likely due to a dilution effect whereas during high rainfall flow events, amounts exported increase while
15 water concentrations decrease. Pb was more likely mobilized due to OM decomposition than affinity for
16 forming Pb-OM complexes since all elements showed similar patterns regardless of OM affinity. Pb
17 concentrations in water were highest during the fall when dry periods were followed by high rain events.
18 High elevation peat mires, soils, and down-catchment reservoirs were sampled in the Snowy Mountains
19 in southeast Australia ([Stromsoe et al., 2015](#)). Pb input to peat mires is dominated by atmospheric
20 deposition and showed greater enrichment of Pb compared with down-catchment reservoirs. Down-
21 catchment reservoirs had depleted Pb levels in comparison due to a dilution effect of soil-bound Pb and
22 large catchment areas.

23 In an arctic peatland, Pb aqueous concentrations were 2–3 times higher in the spring compared
24 with the summer ([Stolpe et al., 2013](#)). Pb concentrations were correlated with DOM. Pb was also
25 primarily in the 0.5–4 nm colloid fraction. During the spring melt in an arctic peatland, pH and high
26 dissolved DOC occur due to erosion of acidic OM and fine particles while concurrently diluting the
27 contribution of the bicarbonate parental material to waterways. During the summer, alkalinity increases
28 while DOC decreases because water inputs shift to groundwater source. A European study examined trace
29 metal budgets across 14 forested catchments over the period of 1997–2011 ([Bringmark et al., 2013](#)). Due
30 to high anthropogenic deposition for decades, Pb accumulation in catchment soils was high. Pb is bound
31 to soil OM in these soils leading to high retention of Pb in the system. At higher altitude sites which
32 experience greater precipitation, retention is lower due to greater runoff and transport of PM out of the
33 system. In a similar study in England, while Pb deposition has decreased in recent decades, legacy Pb in
34 peat catchments is a continuing source of Pb to waterways ([Rothwell et al., 2011](#)). Atmospheric
35 deposition was measured at 34 gPb/ha/year while fluvial outputs were 316 gPb/ha/year. Following storm
36 events, Pb runoff into waters occurs primarily as suspended particles (261 gPb/ha/year) with a smaller
37 aqueous portion (55 gPb/ha/year).

1.3.3.4. Temporal Trends Documented in Sediments

1 Temporal trends of Pb deposition in sediment show distinct leaded gasoline peaks in the United
2 States.. These peaks are found globally, corresponding to the specific phase-out periods for multiple
3 countries. Patterns of increasing Pb concentration occurring from the mid-19th century through the mid-
4 20th century due to early industry as well as agriculture, weathering, and mining operations are
5 identifiable in North American lake and reservoir sediments. Following the peak deposition period in the
6 1960s due to leaded gasoline in North America, widespread decreases in Pb concentration in sediments
7 are seen over the following half century, but concentration values are still higher than background levels
8 showing continued deposition, non-point contamination, and/or legacy Pb runoff contributions.

9 Sediment dating of a mill pond in eastern Virginia shows local Pb sources (weathering and coal
10 combustion) were the primary inputs to Lake Matoaka during the years 1700–1775 ([Balascio et al., 2019](#)).
11 In 1780, Pb accumulation slightly decreased possibly due to a decline of industry which coincides with
12 the capital of Virginia moving from nearby Williamsburg to Richmond. Over the following two centuries,
13 Pb accumulation increased, and sources were from regional mining and Pb ore smelting activities. Pb
14 concentrations continue to increase during the 1900s to a peak maximum in 1975 followed by a sharp
15 decline. This rise and fall of Pb accumulation reflect the increase in coal combustion, smelting, and use of
16 leaded gasoline. Sediment records in Lake Anna in Virginia have higher Pb concentrations in the
17 downstream portion of the reservoir (often exceeding 50 mg Pb/kg) in comparison to the upper reaches
18 ([Odhiambo et al., 2013](#)). Furthermore, these higher concentrations in the downstream portions were
19 limited to the younger surface sediments. Lake Anna sits in a rural watershed and sediment cores do not
20 show the typical increase followed by sharp decrease indicative of leaded gasoline deposition. Instead,
21 sediment enrichment of Pb in addition with cadmium (Cd), Cu, and Zn point to mining runoff as the
22 source of Pb enrichment in sediments. An old sulfur (S) mine operated in the area until 1877, and a pyrite,
23 Cu, and Fe mine until 1920. All other mines ceased operation in the 1990s. A study that measured heavy
24 metal, polychlorinated biphenyl, and polycyclic aromatic hydrocarbon concentrations in dated sediments
25 of the lower Anacostia River in Washington D.C. found Pb concentration had two peaks: the first
26 occurred at corresponding depth of 1943 and the second in 1984 ([Velinsky et al., 2011](#)). The cause for the
27 early 20th century peak is not clear, as Pb sources in the area included both agriculture and industry. The
28 second peak in 1984 and subsequent decrease likely corresponds to the use and then phase-out of leaded
29 gasoline. A location near the Navy Yard and Government Services Administration showed an increase in
30 Pb concentration again over the years of 1989–2000, and the authors note this sampling location is close
31 to large storm water and sewer drainages.

32 In Horseshoe Lake near St. Louis, three main periods of variable Pb pollution were identified.
33 The first period was dated as pre-settlement, had low Pb concentration and the lowest $^{206}\text{Pb}/^{207}\text{Pb}$ ratio and
34 is representative of background parental material and deposition from flooding of the Mississippi River
35 basin ([Brugam et al., 2012](#)). The second period dated as post 1750 had increasing Pb concentration, and
36 the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio diverges from the ratio in the first period. The $^{206}\text{Pb}/^{207}\text{Pb}$ ratio increases and matches

1 Missouri ore samples, and this period coincides with the start of Pb mining in the region. The third period
2 dated from 1915 to the present also contained high Pb concentrations but a lower $^{206}\text{Pb}/^{207}\text{Pb}$ ratio than
3 period two. The source of Pb in this sediment layer is less clear because the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio is similar to
4 vehicle exhaust values of leaded gasoline but is also similar to a nearby old Pb smelter. The lower ratio
5 but high concentrations may also be reflecting erosion and settling of upstream agricultural runoff. The
6 parental material upstream under agriculture has a lower ratio than silt from the Mississippi River but a
7 higher Pb concentration due to agriculture. Lake Whittington, an oxbow lake, was created in 1937 by the
8 U.S. Army Corps of Engineers off the Mississippi River. Flooding of the Mississippi River is the main
9 source of sediment to Lake Whittington ([Van Metre and Horowitz, 2013](#)). Sediment analysis within the
10 lake shows Pb concentration increased from 1938 to the 1970s followed by a decrease. The increase is
11 due, in part, by greater sediment contribution from polluted up-river watersheds of the upper Mississippi
12 and Ohio rivers and less contribution from the cleaner Missouri River watershed which had extensive
13 damming in the 1950s. The concentration decrease post 1970s is explained by reduction in leaded
14 gasoline emissions.

15 Sediments were sampled and dated in an oxbow lake in southwestern Pennsylvania to establish
16 historical contamination in an area of the country with a long pollution exposure history ([Ostrofsky and
17 Schworm, 2011](#)). Pb concentrations increased from 1915 to 1938 corresponding to the opening Donora
18 Zinc Works. Pb levels decrease during the 1940s corresponding depth layer which the authors suggest
19 reflects either a decrease in production or improvement in recovery methods. This downward trend
20 continues through the 1950s (Donora Zinc Works closed in 1957), but Pb concentration increases shortly
21 thereafter around the time a coal powerplant opened nearby. Pb concentration decreases again in the
22 1980s—perhaps in response to the cessation of leaded gasoline. However, As, Cd, and Zn concentrations
23 also decrease suggesting the Pb pollution patterns in this area during this time are instead linked to the
24 coal powerplant ([Rossi et al., 2017](#)). In Sandy Lake, Pennsylvania, Pb levels increase alongside Fe, Mn,
25 and S from approximately 1770 until the 2000s. The increase in concentrations seen in Pb and other
26 elements corresponds with the opening of a coal mine which directly contaminated Sandy Lake with acid
27 mine drainage in the late 1800s. The decrease in Pb levels in the 2000s likely reflects the decrease in
28 deposition from leaded gasoline. In another study, Pb sediment concentrations in Little Lake Bonnet in
29 Florida increased over the period of 1874 to 1920 with a peak of ~28 mg Pb/L ([Escobar et al., 2013](#)).
30 Concentrations increased further to ~38 mg Pb/L in 1949 with an overall peak in 1990 of ~72 mg Pb/L.
31 Pb concentration then declined after 2001 to ~60 mg Pb/L. Little Lake Jackson in Florida showed similar
32 patterns with a peak between mid-1970 and early 1980s. Isotope ratio analysis ties Pb peak patterns with
33 leaded gasoline. The general increasing concentrations in the lakes during the 20th century correspond
34 with broad regional industrialization during this period including pesticide and fertilizer use on golf
35 courses, Pb-arsenic insecticide use on nearby citrus groves, and proximity of coal power plants.

36 A detailed sediment analysis from Vermillion Lake in Sudbury, Ontario, Canada linked Pb
37 concentration to historical industry and leaded gasoline ([Schindler and Kamber, 2013](#); [Wiklund et al.,
38 2012](#)). Pb concentrations first started increasing in the late 19th century at a time when logging in the area

1 first started. Pb concentrations increased until a peak in the late 1960s into the 1970s. The subsequent
2 decrease in concentration corresponds to the phase-out of leaded gasoline in Canada in 1976 and union
3 strikes at nearby mines, resulting in low production. Pb ratios indicated multiple sources of Pb. The parent
4 rock and ores have a unique $^{206}\text{Pb}/^{207}\text{Pb}$ ratio and patterns in this ratio combined with increasing nickel
5 (Ni) concentrations over the period of 1905–1919 indicating Pb level was primarily due to mining
6 contamination. The continuing increase in Pb concentrations after this period would likely reflect greater
7 deposition from nearby smelters and refineries which has a similar ratio profile as leaded gasoline. [Child
8 et al. \(2018\)](#) examined sources and geographic extent of atmospheric metal deposition across eastern
9 Washington lakes within 50 km from the Trail smelter in British Columbia. Pb isotopes and deposition
10 profiles indicate the Trail smelter as a primary source of atmospheric trace metal deposition including a
11 lake outside the 50 km radius and upwind ([Louchouart et al., 2012](#)). A study by [Dunnington et al. \(2020\)](#)
12 examined Pb trends using sediment dating in multiple lakes across northeastern North America. Sampled
13 lakes included locations in the Adirondacks (northeast New York), lakes across Vermont, New
14 Hampshire, and Maine, as well as several lakes in Nova Scotia. In general, Pb concentrations decreased
15 from west to east. Sediment dating reveals anthropogenic Pb concentrations began increasing first in the
16 Adirondack region in 1859 followed by the VT-NH-ME region in 1874, and finally in Nova Scotia in
17 1901. Authors acknowledge early Pb emissions were likely due to coal combustion in the Adirondack and
18 New England lakes, but with the increase in Nova Scotia lakes after 1923, Pb from gasoline was likely an
19 important deposition source to all lakes in this study. Furthermore, looking across the whole sediment
20 core, Pb in youngest sediment is still higher than pre-industrial levels—an indication of continued
21 contamination and loading from legacy Pb in runoff.

22 In a review by [Marx et al. \(2016\)](#), global contamination records of Pb were examined using
23 sediment, peat, and ice cores from across North and South America, Europe, Asia, Australia, and both
24 polar regions. In North America, Pb contamination dating back to 6500 BCE was found and the authors
25 link this early contamination to pre-historic Cu use. Enrichment in this core increases between 1300 CE
26 and 1500 CE, where enrichment doubles, corresponding to the start of the industrial revolution. By the
27 1960s CE, enrichment peaks followed by a decline to 2002 (though enrichment is still well above the
28 1500 CE values). A peat and lake core in Canada record enrichment starting much later (1800s CE) and
29 linking it to coal mining. The peat core shows Pb enrichment peak in 1910 while the lake core peaks in
30 the 1970s. Overall, Europe and North America have higher enrichment values than Australia and
31 Antarctica. Globally, Pb enrichment starts in pre-historic era in Europe, North America, and east Asia
32 while in South America, enrichment starts during the Middle Ages. However, by the latter half of the
33 1800s, Pb enrichment is globally well above background levels, increasing until the 1970s when
34 enrichment declines in Europe and North America but continues to increase in east Asia and Australia. A
35 global study by [Zhou et al. \(2020a\)](#) sampled 168 rivers and 71 lakes from 1972 to 2017 to examine global
36 patterns in heavy metal pollution. Pb levels globally increased during the 1970s and 1980s to a peak level
37 of 257 $\mu\text{g}/\text{L}$ in the 1990s. Pb levels decreased slightly during the 2000s with a slight increase in the
38 2010s. Pb levels were highest in South American water bodies ($\sim 333 \mu\text{g Pb}/\text{L}$) and lowest in Europe
39 ($\sim 14 \mu\text{g Pb}/\text{L}$). Potential sources included rock weathering, fertilizer and pesticide application, mining

1 and manufacturing, and waste discharge. Across the decades the primary source of Pb was determined to
2 be from mining and manufacturing.

1.3.3.5. Sediment Pb Pools as Potential Sources to Surface Waters

3 The removal or breaching of worn-down dams, such as old mill dams, in the eastern United States
4 are a potential new source of legacy Pb for downstream waterways. One study by ([Niemitz et al., 2013](#))
5 found higher Pb sediment levels above a former mill dam draining former and current agricultural lands
6 compared with dammed sediment above a forested catchment within the upper reach of the Yellow
7 Breeches Creek watershed in Pennsylvania. Sediments at both locations, however, contained legacy Pb
8 from gasoline emissions. In another study, while Pb flux initially increased following the removal of
9 Pawtuxet River dam and was positively correlated with river flow, SPM concentration decreased and
10 remained lower than pre-removal levels for the duration of the study ([Katz et al., 2017](#)). This decrease in
11 SPM concentration, however, is potentially due to increased contribution of low-contamination level soils
12 previously above the flood zone downstream of the dam site, and sediment cores taken in multiple
13 locations upstream exceeded sediment quality criteria. Therefore, while removal of a dam may lead to a
14 dilution effect on downstream waters, pools of legacy Pb upstream of a dam will still move into
15 downstream waterways with the post-removal associated increase in water flow. Furthermore, these
16 contaminated sediments can transform to more biologically available forms as they contribute to
17 increased turbidity with increased water flow and salinity as with the Yellow Breeches and Pawtuxet
18 River's flow to the Atlantic (Chesapeake and Narraganset Bay, respectively). Interestingly, in a study
19 examining the effect of beaver dams on heavy metal retention and sediment contamination found Pb
20 aqueous concentration to be lower in the outflow because the dammed water acts as an oxidation pond
21 and results in sorption of Pb to iron oxides ([Shepherd and Naim, 2020](#)). In another study that examined
22 trace metals in surface sediments of a tidal tributary of the Chesapeake Bay, Pb concentrations were
23 above threshold effects levels at 44% of sites sampled. The Chester River watershed is a forested and
24 agricultural watershed and metal accumulation within riverine sediments is likely due to a combination of
25 multiple non-point source runoff as well as tidal exchange with the Chesapeake—highlighting metal
26 pollution transport can occur upstream in estuarine environments ([Krahforst et al., 2022](#)). Hurricane
27 Sandy associated storm surge resuspended Barnegat Bay sediments rich in metal contaminants and
28 transported them northward within the Bay ([Romanok et al., 2016](#)). The source of metal contaminants
29 within sediment silt comes from runoff from inundated urban lands, wetlands, estuaries, streams, and
30 from the resuspension into water of estuarine sediments that may have previously been considered pools
31 of legacy contaminants—including Pb from upstream deposits. Similarly, as discussed in Section
32 1.3.3.2.1, runoff and sediment concentrations have been linked to characteristics of urbanization
33 suggesting urban areas are a potential source of resuspended Pb in water from sediments following storm
34 or high-flow events.

1.3.3.6. Summary

1 In summary, literature since the 2013 Pb ISA supports previous conclusions regarding the
2 physicochemical drivers of Pb fate and transport in aquatic systems. Studies continue to report runoff
3 from urban or historically industrial areas contain higher Pb concentrations than non-urban areas with
4 new information highlighting relationships between street length and density, population density, and
5 land cover with runoff. Recent studies expand on the influence of seasonality and precipitation events on
6 runoff as well as transport and sedimentation. Timing of snow and ice melt can alter down-catchment
7 transport of Pb in high elevation watersheds, for example, while another study found water column
8 concentrations differed between summer and winter—possibly due to differences in precipitation patterns
9 influencing sedimentation and resuspension into water. A collection of recent studies linked Pb
10 concentration peaks in lacustrine and riverine sediment cores to national and global patterns of
11 industrialization in the late 19th and early 20th century, to increased vehicle abundance and associated
12 leaded gasoline in the mid-20th century, followed by a decline in Pb concentration coinciding with the
13 phase-out of leaded gasoline and stricter emissions regulations. Furthermore, new literature also
14 addressed the importance of turbidity and resuspension into water in relation to legacy Pb pools. While Pb
15 deposition has decreased in the last half century with the phase-out of leaded gasoline and stricter
16 regulation, Pb sediment pools in areas with a history of industry and urbanization are vulnerable to
17 resuspension into water and both down and upstream movement following a disturbance event. Dam
18 removal or other disturbances to streams in the eastern United States can lead to resuspension in water
19 and dissolution of Pb-contaminated sediment that was previously deposited. Lastly, with the predicted
20 increase in drought alongside less frequent but more severe precipitation patterns across most of the
21 United States, the potential for remobilization of legacy Pb is a growing area of concern and
22 consideration.

1.3.4. Fate and Transport in Urban Media

23 Additional media besides air, water, and soil are useful for understanding how Pb moves and
24 changes over time in the urban environment. These can include urban soil (Section 1.2.7), resuspended
25 airborne dust, road dust (Section 1.2.5), and house dust, between which Pb can be transported or cycled.
26 Pb concentrations are characteristically higher in urban soil than other soils. [Frank et al. \(2019\)](#) reported
27 the mean estimate of Pb concentration in urban residential soils was 3 times greater than for non-urban
28 soils in a meta-analysis of recent studies. [Obeng-Gyasi et al. \(2021\)](#) estimated that urban soil Pb
29 concentrations were approximately 7 times U.S. background levels (Section 11.1.3) through a comparison
30 of an analysis 84 studies of U.S. soil Pb with a U.S. Geological Survey study of U.S. background soil Pb
31 concentrations ([Datko-Williams et al., 2014](#); [Smith et al., 2013](#)). [Datko-Williams et al. \(2014\)](#) concluded
32 that there had been little change in urban U.S. soil Pb concentrations from 1970 to 2012. The highest
33 concentrations often occur in roadside soil and near buildings, reflecting the proximity to legacy Pb from
34 leaded gasoline and deteriorating paint, respectively ([Obeng-Gyasi et al., 2021](#)), although [Frank et al.](#)

1 [\(2019\)](#) reported lower soil Pb concentrations for roadside soils than for residential urban soils in their
2 meta-analysis of studies on soil Pb concentrations in the United States. The 2013 Pb ISA also reviewed
3 observations of decreasing soil Pb concentrations with distance from a road ([U.S. EPA, 2013](#)). There is
4 some evidence that the pattern of decreasing soil Pb concentrations with road distance are paralleled by
5 near-road air Pb concentrations (Section 1.2.6).

6 Pb has a very long residence time in soil, with models predicting more than a century until soil
7 concentrations return to steady-state levels ([Harris and Davidson, 2005](#)). This is consistent with the slow
8 transport rates typically observed for a range of conditions (Section 1.3.2) and facilitates the persistence
9 of long-term hot spots. For example, as described in Section 1.2.7, soil Pb in Durham NC ranged from 6–
10 8825 mg Pb/kg and soil Pb concentrations were higher around older homes ([Wade et al., 2021](#)). Home
11 age accounted for 40% of the variance in foundation soil Pb, with soil near painted houses containing
12 significantly higher soil Pb than for brick homes ([Wade et al., 2021](#)). Studies in East Chicago IL,
13 Greensboro NC, Brooklyn NY, and Philadelphia PA have explored the high spatial variability of urban
14 soil Pb concentrations, with hot spots related to income and racial disparities ([Caballero-Gómez et al.,](#)
15 [2022](#); [Pavilonis et al., 2022](#); [Haque et al., 2021](#); [Obeng-Gyasi et al., 2021](#)). Urban and neighborhood-scale
16 spatial variability of ambient air Pb concentrations have been observed in recent studies, but not directly
17 related to urban soil (Section 1.5.2).

18 Resuspension of Pb in contaminated soil and road dust by traffic, construction, and wind has been
19 described as a potential contributor to airborne Pb under some circumstances ([U.S. EPA, 2013](#)). The 2013
20 Pb ISA reported that contaminants associated with particles with diameters up to about 100 µm can
21 typically become resuspended into air, but particles larger than 10-20 µm typically do not remain airborne
22 long enough for appreciable transport ([U.S. EPA, 2013](#); [Nicholson, 1988](#); [Gillette et al., 1974](#)). However,
23 the extent of resuspension of contaminants in surface soil and dust particles into air depends strongly on
24 landscape, particle size and wind speed. The 2013 Pb ISA summarized factors influencing resuspension
25 into air in a complex urban landscape with heavy traffic, buildings, pavement and above- and below-
26 ground infrastructure ([U.S. EPA, 2013](#)). The critical diameter at which resuspension into air occurs is the
27 diameter at which the particle settling velocity becomes equal to the friction velocity of air needed to
28 move the particle at rest. Although this was estimated at roughly 20 µm in an open landscape ([U.S. EPA,](#)
29 [2013](#); [Gillette et al., 1974](#)), a higher friction velocity is expected for urban environments with traffic-
30 induced turbulence ([Britter and Hanna, 2003](#)). This could result in resuspension of somewhat larger
31 particles into air in an urban setting with heavy traffic ([Nicholson and Branson, 1990](#)). Near-road
32 observations indicate that the fraction of total particulate Pb associated with particles larger than 10 µm
33 can be 15% or more ([U.S. EPA, 2013](#)).

34 Pb particles from soil and dust occur at sizes ranging from 0.1–10 µm, a size range with potential
35 for resuspension into air and inhalation ([O’Shea et al., 2021](#)). Laboratory studies and sampling in areas
36 with previous major emissions sources suggests the potential for resuspension of soil bound Pb to
37 contribute to airborne concentrations in those areas ([Pingitore et al., 2009](#); [U.S. EPA, 2006b](#)). A

1 previously reported modeling study estimated up to 90% of Pb emissions in Southern California are
2 attributed to Pb resuspension from soil and road dust into air ([Harris and Davidson, 2005](#)), though [Lankey](#)
3 [et al. \(1998\)](#) noted a smaller estimate of 43%. With estimated annual emissions of 54,000 kg airborne Pb
4 in 2001, modeled resuspension was identified as the largest source of airborne Pb in the Southern
5 California region ([Harris and Davidson, 2005](#)). Although Pb from contemporary sources can also be
6 resuspended into air, emissions from historical sources were considerably greater, and substantial Pb from
7 sources like leaded gasoline and historical industrial emissions or from non-atmospheric sources like
8 paint have accumulated in soil over many years, particularly in urban areas (Section 1.2.7).

9 Correlations between soil Pb concentrations and atmospheric Pb concentrations have been
10 observed in recent studies. [Laidlaw et al. \(2014\)](#) reported atmospheric Pb loadings increased by
11 $0.066 \mu\text{g}/\text{m}^2/28$ days for every mg Pb/kg increase in soil Pb. Another study found that a 1% increase in
12 estimated resuspended soil concentration in air corresponded to a 0.39% increase in atmospheric Pb
13 concentration (95% CI, 0.28 to 0.5%) ([Zahran et al., 2013](#)). The isotopic composition of Pb in airborne
14 particles is consistent with that of road dust and topsoil, with significant contributions (a binary mixing
15 model found from 32 ± 10 to $43 \pm 9\%$) of Pb from leaded gasoline ([Resongles et al., 2021](#)). Atmospheric
16 soil and Pb aerosols are 3.15 and 3.12 times higher, respectively, during weekdays and federal holidays
17 than weekends, suggesting traffic as a major driver of Pb resuspension into air ([Laidlaw et al., 2012](#)). In
18 London, 450-650 kg/year of Pb is emitted as resuspended dust, a similar magnitude as primary Pb air
19 emissions in urban locations ([Resongles et al., 2021](#)). Additionally, Pb isotopic composition was similar
20 for particles collected at ground-level and building height, suggesting Pb is well mixed throughout the
21 vertical column in urban environments ([Resongles et al., 2021](#)). Dust emissions are significant and
22 represent missing sources in the emission inventories ([Xu et al., 2019](#)). To reduce national-scale bias of
23 modeled Pb concentrations, a fivefold increase in anthropogenic emissions of Pb was necessary to
24 achieve agreement between simulated and observed ambient air Pb concentrations ([Xu et al., 2019](#)).
25 While these studies suggest that resuspension Pb from soil into air is a potentially important local source
26 of Pb in ambient air, it appears to be a much smaller contributor to current ambient air Pb concentrations
27 than leaded gasoline exhaust was in previous decades. Airborne Pb monitoring was originally required for
28 urban NCore multipollutant monitoring network sites (Section 1.4) but was discontinued in 2016 because
29 concentrations were consistently much lower than NAAQS levels (40 Code of Federal Regulations (CFR)
30 Part 58, Appendix A). Moreover, at five Pb monitoring sites near roads with heavy traffic, ambient air Pb
31 concentrations decreased from more than $1 \mu\text{g}/\text{m}^3$ in 1979 to less than $0.03 \mu\text{g}/\text{m}^3$ in 2010 ([U.S. EPA,](#)
32 [2013](#)).

33 Several studies have reported seasonal patterns of resuspension from soil into air, with highest
34 resuspension occurring in summer and autumn when soils are driest ([Resongles et al., 2021](#); [Mielke et al.,](#)
35 [2019](#); [Laidlaw et al., 2017](#); [Laidlaw et al., 2016](#); [Laidlaw et al., 2014](#)). In Detroit, atmospheric Pb is
36 44.8% higher in August than January (Figure 1-2) ([Zahran et al., 2013](#)). This seasonal pattern is also
37 observed in measurements of children's blood levels ([Laidlaw et al., 2017](#); [Mielke et al., 2017](#); [Laidlaw et](#)
38 [al., 2016](#); [Zahran et al., 2013](#)) and is discussed in detail in Section 2.4. National scale modeling of heavy

1 metal concentrations with the chemical transport model GEOS-Chem indicated that simulated heavy
2 metal concentrations in PM_{2.5} over continental North America were consistent with monitoring
3 observations in winter, but generally low in other seasons, suggesting that contributions of missing
4 sources of Pb follows a seasonal pattern similar to that observed for airborne soil components ([Xu et al.,
5 2019](#)).

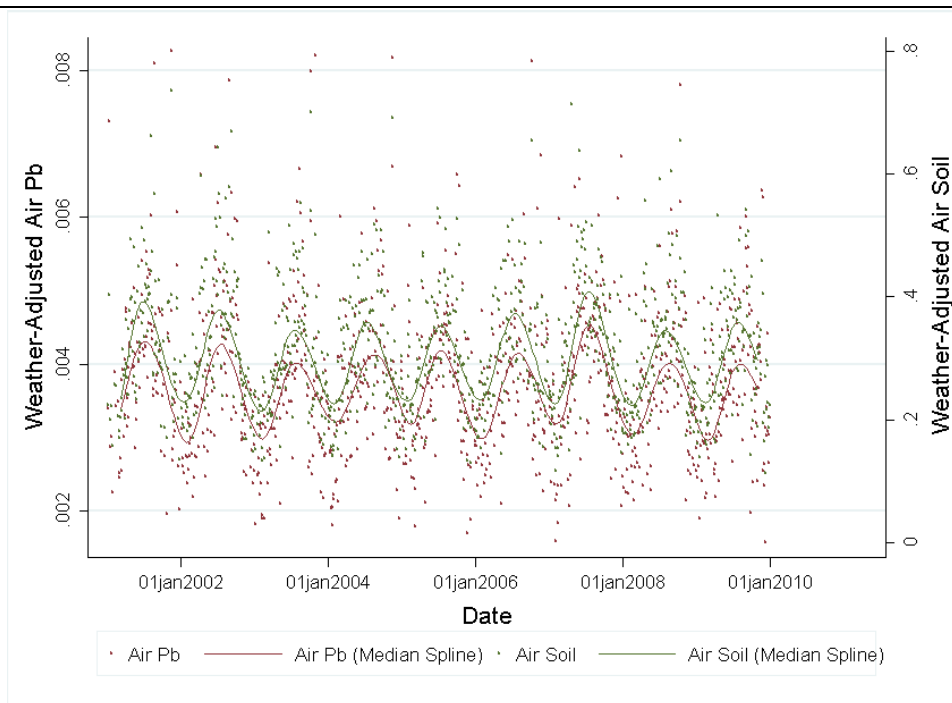


Figure 1-2 Ambient air Pb and air soil concentrations and median splines in $\mu\text{g}/\text{m}^3$ from Detroit MI. Air soil refers to the estimated ambient air concentration of soil-derived PM based on crustal element concentrations. Weather-adjusted concentrations are concentrations that have been adjusted for relative humidity, pressure, temperature, visibility, and wind speed using their known relationships with air Pb and air soil to determine their seasonality independent of short-term weather conditions. The median spline is a smoothing function based on a polynomial fit.

Source: Reprinted with permission from [Zahran et al. \(2013\)](#). Copyright 2013, American Chemical Society.

6 Though rare, extreme weather events can alter soil Pb concentrations drastically. Following
7 Hurricane Katrina in New Orleans, soil Pb decreased from 285 to 55 mg Pb/kg on public land and from
8 710 to 291 mg Pb/kg on private land ([Mielke et al., 2017](#)). These observed effects are likely due to result
9 in decreased resuspension into air following flooding, as well as transport of soil from outside the city

1 covering Pb-contaminated urban soil ([Mielke et al., 2017](#)). Recent Pb isotopic aerosol signatures show
2 origins from leaded gasoline, suggesting Pb sources have not changed substantially since the removal of
3 leaded gasoline ([Resongles et al., 2021](#)). Pb in PM₁₀ samples collected in London ranges from 3.9–
4 19.4 ng/m³, with deposition rates of 11,700–45,800 ng/m²/day for Pb associated with total suspended
5 particulate matter (TSP) ([Resongles et al., 2021](#)). 31% of PM₁₀ particles measured were attributed to
6 resuspended road dust in air ([Resongles et al., 2021](#)).

7 The larger size of resuspended dust particles compared with typical atmospheric particle size
8 distributions makes the atmospheric lifetimes and travel distances of the airborne dust Pb potentially
9 shorter than those expected for PM_{2.5} or PM₁₀ ([U.S. EPA, 2013](#)). As a result, resuspension of large
10 amounts of soil Pb into air does not appear to be an efficient process for Pb removal from a neighborhood.
11 However, resuspension followed by relatively rapid deposition provides a potential process for Pb to
12 translocate within neighborhoods, reducing high concentrations near busy roads while increasing it in
13 other areas. This pattern of evening out soil Pb concentrations in city centers over long time scales has
14 been described by [Laidlaw and Filippelli \(2008\)](#).

15 Association with airborne dust also provides Pb with a transport pathway indoors, where it
16 deposits as house dust. Along with urban soil, house dust has been a particular concern for accumulation
17 of Pb from deteriorating paint ([Lanphear et al., 1998](#)). The evidence for the link between atmospheric Pb
18 and house dust Pb near large industrial sources can be strong enough that urban-scale house dust Pb
19 concentrations have been used to effectively track changes in atmospheric deposition patterns caused by
20 the addition of a tall stack to a smelter ([Van Pelt et al., 2020](#)). Since there are also other processes for
21 transport of Pb between soil and house dust, an unknown portion of the Pb in house dust becomes
22 airborne after its release into the environment. However, there is potential for Pb resuspension into air to
23 serve as a source of both ambient air Pb and house dust Pb. In a recent study of childhood leukemia risk
24 from carpet dust, mean dust loadings were 24.5 and 15.3 µg/ft² in homes of children with and without
25 leukemia, respectively ([Whitehead et al., 2015](#)). These results compare to [Frank et al. \(2019\)](#), who
26 reported a mean of 13 µg/ft² for 535 floor samples and 214 µg/ft² for 380 windowsill samples in a meta-
27 analysis of studies published between 1999 and 2015. [Gillings et al. \(2022\)](#) observed that Pb in both
28 house dust and surface soil decreased with distance from mining areas. The decline with distance was
29 steeper for soil than for house dust ([Gillings et al., 2022](#)). Whether soil Pb was removed or buried deeper
30 under the surface was not discussed. There is also evidence for Pb-contaminated dust deposition onto
31 roofs, which could then undergo resuspension into the air during demolition ([Caballero-Gómez et al.,
32 2022](#)).

33 Recent research supports prior information on the influence of legacy Pb from leaded gasoline,
34 past industrial emissions, and deteriorating paint on soil Pb concentrations in some areas, with the highest
35 concentrations near roads and buildings. Within urban soil there appears to be gradual Pb transport away
36 from roads and buildings, and a slow reduction of soil Pb concentration gradients over time, potentially
37 due at least in part to cycles of resuspension into air, atmospheric transport, and deposition. There is also

1 transport between soil and other compartments, namely house dust, road dust, and air. Overall, transport
2 rates are extremely slow, and changes in concentration gradients near sources and hot spots is very
3 gradual, apparently remaining for decades without intervention or other external disturbances like
4 hurricanes or floods. Current research is taking place toward understanding rates of Pb transport and their
5 influencing factors either among compartments or among different locations in the same medium (i.e.,
6 soil, road dust). One salient observation is that the extremely slow movement of Pb through the urban
7 environment facilitates persistent hot spots of high soil Pb and house dust Pb concentrations. There is
8 little data on whether hot spots of high soil Pb and dust Pb concentrations also lead to pockets of high
9 ambient air concentrations, with the exception of near-road observations. Although not all urban Pb
10 transport involves air, there is evidence that resuspension of Pb in urban soil may contribute to airborne
11 Pb concentrations in some locations.

1.4 Monitoring of Pb in Ambient Air

12 This Section describes advances in development and evaluation of sampling and analytical
13 methods for monitoring and measurement of airborne Pb. Section 1.4.1 describes recent research to
14 evaluate the performance of the Federal Reference Method (FRM) for Pb in ambient air. Section 1.4.2
15 provides a summary of network monitoring challenges related to Pb airborne Pb sampling Section 1.4.3
16 describes recent advances in sampling and analysis of airborne Pb in monitoring and research.

1.4.1. Federal Reference Methods

17 Four national monitoring networks collect data on Pb concentrations in ambient air and report it
18 to the Air Quality System (AQS). Up-to-date network descriptions and monitor locations for the Pb
19 SLAMS, CSN, IMPROVE, and NCore networks are available in “Overview of Lead (Pb) Air Quality in
20 the United States” ([U.S. EPA, 2022](#)). In order to be used in regulatory decisions judging attainment of the
21 Pb NAAQS, ambient air Pb concentration data must be obtained through the FRM, or a Federal
22 Equivalent Method (FEM) defined for this purpose. Accordingly, for enforcement of the air quality
23 standards set forth under the Clean Air Act, EPA has established provisions in the Code of Federal
24 Regulations under which analytical methods can be designated as FRM or FEM. FRMs and FEMs for the
25 Pb NAAQS exist for both sample collection and sample analysis.

26 There are two FRMs for Pb sampling in ambient air: (1) Reference Method for the Determination
27 of Lead in Suspended Particulate Matter Collected from Ambient Air (40 CFR part 50 Appendix G), and
28 (2) Reference Method for the Determination of Lead in Particulate Matter as PM₁₀ Collected from
29 Ambient Air (40 CFR part 50, Appendix G). The Pb-TSP FRM sample collection method measures Pb-
30 associated TSP and is required for all source-oriented NAAQS monitors, and the FRM for Pb- PM₁₀ is

1 accepted for Pb NAAQS monitoring at non-source-oriented monitors in specified situations ([U.S. EPA,](#)
2 [2013](#)).

3 The Pb-TSP FRM sample collection method specifies use of a high-volume TSP sampler that
4 meets specified design criteria (40 CFR part 50 Appendix B). Ambient air PM is collected on a glass fiber
5 filter for 24 hours using a high-volume air sampler. Variability in high-volume TSP sampler collection
6 efficiency associated with effects of wind speed and wind direction for particles larger than 10 μm has
7 been documented since the sampler was first implemented for TSP and Pb-TSP sampling and was a major
8 focus of the 2013 Pb ISA ([U.S. EPA, 2013](#)).

9 To provide informative background for addressing sampling issues associated with particles
10 larger than 10 μm , the 2013 Pb ISA reviewed historical research on large particle sampling as well as
11 recently developed low-volume samplers with manufacturer-designated TSP inlets. The 2013 Pb ISA
12 concluded that a high degree of variability had been observed between different models of manufacturer-
13 designated TSP samplers, that no alternative to the FRM TSP sampler had yet been identified, and that
14 there was still a need to assess the feasibility of a revised TSP sampling method for efficient collection
15 particles larger than 10 μm ([U.S. EPA, 2013](#)).

16 The low-volume Pb-PM₁₀ FRM sample collection method specifies use of a low-volume PM₁₀
17 sampler that meets specified design criteria (40 CFR part 50, Appendix Q). Ambient air PM is collected
18 on a polytetrafluoroethylene (PTFE) filter for 24 hours using active sampling at local conditions with a
19 low-volume PM₁₀ sampler and analyzed by XRF. Use of the FRM for Pb-PM₁₀ is allowed in certain
20 instances where the expected Pb concentration does not approach the NAAQS and in the absence of
21 nearby sources of Pb associated with particles greater than 10 μm diameter.

22 In addition to FRMs used in the SLAMS network, other methods are used in the CSN,
23 IMPROVE and National Air Toxics Trends Stations (NATTS) networks ([U.S. EPA, 2022](#)), as well as in
24 field studies unrelated to network applications. The most relevant of these methods are listed and
25 described in the 2013 Pb ISA ([U.S. EPA, 2013](#)). In the CSN and IMPROVE networks, Pb in PM_{2.5} is
26 collected on Teflon filters by low-volume samplers and analyzed by XRF ([U.S. EPA, 2013](#)). Other
27 sampling approaches, including cascade impactor sampling for Pb particle size distributions, saturation
28 samplers, and passive samplers to provide high-density coverage for evaluation of spatial variability were
29 reviewed in the 2013 Pb ISA ([U.S. EPA, 2013](#)), and recent applications to Pb spatial variability and size
30 distributions are reported in Section 1.5. Other analytical methods applied to Pb were also reviewed in the
31 2013 Pb ISA, including inductively coupled plasma atomic emissions spectroscopy, energy-dispersive X-
32 ray fluorescence (ED-XRF), proton induced X-ray emission spectroscopy, X-ray photoelectron
33 spectroscopy, Pb speciation methods including X-ray absorption fine structure as well as gas
34 chromatography and high performance liquid chromatography combined with inductively coupled plasma
35 mass spectrometry, Pb isotope ratio analysis, and continuous Pb monitoring methods ([U.S. EPA, 2013](#)).
36 Few new advances related to Pb analysis have been reported for these methods, but recent applications of
37 some are reported in Section 1.5.

1.4.2. Sampling Considerations

1 When the Pb NAAQS was revised in 2008, revision of the FRM from a method sampling total
2 suspended particulate matter (Pb-TSP) to a method sampling PM less than or equal to 10 µm in diameter
3 (Pb-PM₁₀) was also considered because of poor precision and variable collection efficiencies with wind
4 speed and direction for larger particle sizes associated with the Pb-TSP FRM. The Pb-TSP FRM was
5 developed in the 1950s for collection of Pb-TSP. It has been the FRM for airborne Pb since 1973, but
6 over the first decade after its designation as the Pb FRM, at least twelve studies reported variable sampler
7 performance depending on several factors, particularly particle size, wind speed, sampler orientation with
8 respect to wind direction, and extent of passive collection ([Krug et al., 2017](#)). Variations in Pb-TSP FRM
9 sampler performance could be due in part to broad acceptable performance ranges and the lack of a
10 strictly defined performance standard for evaluating TSP samplers ([Krug et al., 2017](#)).

11 The Pb-PM₁₀ FRM is not as vulnerable to sampling errors associated with the Pb-TSP FRM
12 because it is based on a strictly defined performance standard. Transition to a Pb-PM₁₀-based NAAQS
13 was not supported, however, because Pb associated with particles larger than 10 µm in diameter can be an
14 important contributor to airborne Pb exposure, in part because of potential resuspension of Pb in urban or
15 industrial soil or road dust into air in some locations (Section 1.3.4). Additionally, Pb-PM₁₀ can only be
16 used as a surrogate for Pb-TSP if the loss of particles larger than 10 µm in diameter can be compensated
17 by firmly establishing a quantitative adjustment factor based on concurrent Pb-TSP and Pb-PM₁₀
18 sampling that exhibits long-term stability over time ([Krug et al., 2017](#)). On the grounds of limited
19 comparisons of Pb-TSP and Pb-PM₁₀ available at the time, it was judged that more data sets were needed
20 before either national or site-specific relationships between Pb-TSP and Pb-PM₁₀ could be established.
21 Thus, the Pb-TSP sampling method was retained as the FRM because of the importance of including Pb
22 associated with particles larger than 10 µm in diameter and the lack of a quantitative adjustment factor.
23 However, a Pb-PM₁₀ FRM was developed and permitted for non-source-oriented monitoring sites, where
24 rolling 3-month average Pb-TSP concentrations were less than 0.1 µg/m³ and Pb associated with particles
25 larger than 10 µm was not anticipated.

26 Although the term TSP implies collection of airborne particles of all sizes, there is evidence that
27 the Pb-TSP sampler could still miss particles with diameters larger than certain upper limits for efficient
28 sampling. Although a TSP sampler collects more particulate mass than a PM₁₀ sampler, a substantial
29 fraction of airborne PM could be missed by both samplers if the size range of Pb-associated particles
30 extends beyond both of their practical size limits for efficient sampling. As described in the 2013 Pb ISA
31 ([U.S. EPA, 2013](#)), in previous observations the TSP sampling efficiency of 97% for 5 µm particles
32 dropped to 35% for 15 µm particles under the same conditions ([U.S. EPA, 2013](#); [Wedding et al., 1977](#)),
33 and the cut-point for 50% sampling efficiency was observed to decrease from 50 µm at a 2 km/hour wind
34 speed to 22 µm at 24 km/hour ([Rodes and Evans, 1985](#)). This suggests that at least under some
35 circumstances resuspended Pb at the high end of the Pb particle size distribution relevant for resuspension
36 into air might be collected with less than 50% efficiency. Insufficient data on size distributions of

1 airborne resuspended Pb beyond the size range efficiently collected by the Pb-TSP sampler makes it
2 difficult to assess either the fraction of Pb potentially missed by a Pb-TSP sampler or the contribution of
3 Pb from this size range to total airborne Pb. An additional challenge to measuring Pb over its entire PM
4 size range is that spatial variability is greater for coarse than for fine particles. As described in the 2019
5 PM ISA ([U.S. EPA, 2019](#)), PM_{2.5} concentrations on an urban scale are more uniform than for PM_{10-2.5}. By
6 extension, including a substantial amount of mass for particles larger than PM₁₀ is potentially
7 representative over a smaller area containing a smaller population.

1.4.3. Recent Advances in Sampling and Analysis

8 The 2013 Pb ISA described several new developments in sampling and analysis of airborne Pb,
9 including the addition of new FRMs and FEMs ([U.S. EPA, 2013](#)). Advances in the development of new
10 sampling and analytical methods described here stem in part from 2008 revisions of the Pb NAAQS.
11 Specifically, the 2008 revisions described conditions under which a Pb-PM₁₀ FRM could be used as an
12 alternative to the original Pb-TSP FRM. The trade-off between missing Pb from Pb-TSP from upper end
13 sampling bias and uncertainty versus the potential for missing a large fraction of airborne Pb using the
14 Pb-PM₁₀ FRM continues to be a driving force for further research. In addition, the lower NAAQS level
15 established by the 2008 revisions triggered a need for alternative measurement methods like ICP-MS with
16 better performance characterization at lower concentrations. Recent publications provide essential
17 characterization and comparisons of both old and new measurement methods. These include advanced
18 wind-tunnel studies of the original Pb-TSP FRM ([Vanderpool et al., 2018](#)) and other TSP samplers ([Krug
19 et al., 2017](#)), a detailed field comparison of collocated Pb-PM₁₀ versus Pb-TSP performance ([Ward et al.,
20 2019](#)), performance evaluation and interlaboratory comparison of the new ICP-MS FRM ([Harrington et
21 al., 2014](#)), and development of relevant reference materials in a suitable concentration range for XRF and
22 ICP-MS analysis of airborne Pb ([Yatkin et al., 2016](#)).

23 Following a surge of studies immediately after its designation as a FRM, little new work to
24 further evaluate Pb-TSP FRM sampler performance was carried out until [Krug et al. \(2017\)](#) summarized
25 and compared results from the previous evaluations of sampler performance conducted under various
26 conditions, and used up-to-date methods to quantify the effects of environmental and operational factors
27 affecting sampler performance in a controlled wind-tunnel setting using isokinetic samplers operating
28 alongside the sampler. [Krug et al. \(2017\)](#) reported that: 1) sampling effectiveness ranged from 42% to
29 92% based on particle diameter, across orientations and wind speeds, an effectiveness range that is
30 comparable to those reported in some previous studies and smaller than those reported in others; 2)
31 sampling effectiveness is a near monotonic decreasing function of aerodynamic particle size, as predicted
32 by sampling theory; 3) wind speed plays a significant role in sampling effectiveness; and 4) sampling
33 effectiveness varies with sampler orientation to the wind and the variability increases with wind speed,
34 but not to the extent observed in previous studies.

1 Several low-volume, portable commercial samplers have been widely referred to as TSP samplers
2 because of their lack of internal size fractionation. Described as samplers with manufacturer-designated
3 TSP inlets in the 2013 Pb ISA ([U.S. EPA, 2013](#)), these devices also have potential for airborne Pb
4 sampling, but have not been as extensively evaluated for collection efficiency as a function of particle
5 size as the Pb-TSP FRM. Some early efforts to characterize alternative low-volume TSP samplers were
6 summarized in the 2013 Pb ISA, and the value of testing them as potential replacements for high-volume
7 TSP sampling was discussed ([U.S. EPA, 2013](#)). This testing has now been completed by [Vanderpool et
8 al. \(2018\)](#), who conducted a study specifically to evaluate size-selective performance of six low-volume
9 manufacturer-designated TSP samplers at different wind speeds in a controlled wind-tunnel setting. Like
10 the Pb-TSP FRM, sampling performance generally decreased with both particle diameter and wind speed
11 for each of the inlets evaluated, and all sampling inlets exhibited some degree of measurement bias for
12 larger particles and at higher wind speeds ([Vanderpool et al., 2018](#)). On average over most wind speeds,
13 most samplers collected 75% to 95% of particulate mass expected for particle diameters ranging up to
14 30 μm ([Vanderpool et al., 2018](#)). However, these would still need to be evaluated more extensively before
15 approval as a FRM or FEM and use in the NAAQS monitoring network.

16 Other recent research includes a detailed assessment of collocated Pb-TSP and Pb-PM₁₀
17 monitoring results to expand on the limited available data for understanding the relationship between Pb-
18 TSP and Pb-PM₁₀ concentrations and to assess the suitability of a site-specific adjustment factor. Pb-TSP
19 and Pb-PM₁₀ data were collected every sixth day for more than three years from a monitor adjacent to the
20 Walkill secondary smelter in New York, which had been recently equipped with a wet electrostatic
21 precipitator to reduce emissions of larger particles. Data from the two samplers were strongly correlated
22 with an adjustment factor of 1.49, somewhat lower than previous observations for primary smelters from
23 a small number of samples ([Ward et al., 2019](#)). [Ward et al. \(2019\)](#) confirmed that implementation of a Pb-
24 PM₁₀ monitor at their source-oriented location would lead to underestimation of the total ambient air Pb
25 concentration without the application of an adjustment factor relating Pb-PM₁₀ to Pb-TSP. They also
26 suggested that development of a generic adjustment factor for all Pb monitoring locations was probably
27 not possible because of differences in particulate Pb characteristics between different locations and source
28 emissions. These results indicate that in addition to non-source-oriented sites where the Pb-PM₁₀ FRM is
29 currently used, there is also potential to obtain high quality data using Pb-PM₁₀ samplers at some source-
30 oriented sites. However, this approach also requires demonstration of a stable relationship between Pb-
31 PM₁₀ and Pb-TSP over time, which has yet to be evaluated.

32 Revision of the Pb NAAQS in 2008 resulted in a NAAQS level that approached the limit of
33 quantitation for the Pb analysis FRM based on flame atomic absorption spectroscopy (FAAS).
34 Improvements in sensitivity, precision, throughput capability, and extraction efficiency since the
35 development of the original FRM provided additional motivation for development of a new FRM
36 ([Harrington et al., 2014](#)). To address these changes, a new Pb analysis FRM based on ICP-MS was
37 introduced in 2013 (40 CFR Part 50 Appendix G). The new FRM includes two extraction methods: hot
38 block HNO₃ extraction and ultrasonic extraction in a HNO₃/HCl mixture. The FRM was evaluated in a

1 multi-laboratory study that demonstrated acceptable sample stability after extraction; acceptable
2 equivalency with the FAAS FRM; acceptable intra- and interlaboratory precision; comparability across
3 relevant filter media; acceptable accuracy for analysis of botanical, geological, and industrial standard
4 reference materials; and method detection limits of less than 5% of the 2008 NAAQS levels ([Harrington
5 et al., 2014](#)). Considering these results, the ICP-MS-based FRM for Pb-TSP is considered more
6 appropriate for the 2008 NAAQS level than the previous FAAS-based FRM for Pb-TSP because of its
7 good performance at significantly lower ambient air Pb-TSP concentrations ([Harrington et al., 2014](#)).

8 Another useful recent advance for ambient air Pb measurement was the development of new
9 reference materials suitable for XRF analysis, which is used as an FEM for Pb NAAQS monitoring as
10 well as for quantifying Pb concentrations in the CSN and IMPROVE monitoring networks. The new
11 reference materials are useful for laboratory audits, federal equivalency method evaluation, calibration,
12 and quality control. They were generated by aerosol deposition over a range of Pb concentrations on
13 PTFE filters used for ambient air sampling, and good long-term stability was demonstrated ([Yatkin et al.,
14 2016](#)). The new reference materials fill a gap in commercially available reference materials because
15 previously available reference materials were not similar to filter media used for collection or the PM
16 matrix and contained Pb amounts that were not similar to typical ambient air Pb samples. [Yatkin et al.
17 \(2016\)](#) also used the new reference materials to conduct an interlaboratory comparison of XRF analysis
18 methods and to establish equivalence between XRF and ICP-MS Pb analysis methods.

19 Several advances have also recently taken place in research instrumentation to improve time
20 resolution. Performance was evaluated for an updated version of the Semi-continuous Elements in
21 Aerosol Sampler (SEAS-III). With this sampler, a high volume of sample is collected in a slurry for
22 analysis by ICP-MS. A collection efficiency of $87 \pm 16\%$ was reported and collocated precision was better
23 than 25% for 20 elements. For Pb, the collocated precision was 33% for sample concentrations averaging
24 4 ng/m^3 , or about 5 times the reported minimum detection limit of 0.79 ng/m^3 ([Pancras and Landis, 2011](#)).
25 A portable XRF monitor with subdaily time resolution has also been evaluated and applied to field
26 measurements of airborne PM ([Sofowote et al., 2019](#); [Tremper et al., 2018](#); [Furger et al., 2017](#)).
27 Extractive electrospray ionization combined with Time-of-flight mass spectrometry has also been
28 developed for real-time measurement of Pb in ambient air ([Giannoukos et al., 2020](#)).

1.5 Ambient Air Pb Concentration Trends

29 This Section summarizes ambient air Pb concentrations and trends. The 3-month average ambient
30 air Pb concentrations presented here were created using a simplified approach of the procedures detailed
31 in 40 CFR part 50 Appendix R and, as such, cannot be directly compared with the Pb NAAQS for
32 determination of compliance. Section 1.5.1 presents nationwide ambient air Pb concentration trends.
33 Section 1.5.2 summarizes results from numerous, mostly local studies on urban and neighborhood spatial

1 scale variability. Sections 1.5.3, 1.5.4, and 1.5.5 report the latest results on seasonal/diurnal trends, size
2 distributions, and background concentrations of Pb in ambient air PM from diverse locations.

1.5.1. National Scale Ambient Air Concentrations and Long-Term Trends

3 . For the period 2019-2021, concentrations of 3-month average ambient air Pb concentrations in
4 the United States were below $0.15 \mu\text{g}/\text{m}^3$ at most U.S. monitors and the median U.S. 3-month average Pb
5 concentration was $0.009 \mu\text{g}/\text{m}^3$ ([U.S. EPA, 2022](#)). Pb concentrations in ambient air in the U.S. have
6 decreased since the 1970s, mainly due to the phase-out of Pb in gasoline. In some cases, there has been a
7 more recent period of continued decline corresponding to reductions in Pb emissions from local and
8 regional industrial sources. A quantitative description of the trend based on monitoring network data is
9 problematic for two reasons. First, Pb concentration reporting requirements changed in 2010 from
10 measured Pb concentration at standard and temperature and pressure to Pb concentration measured
11 concentration under local conditions. As a result, daily concentration and design value data from before
12 2010 are not directly comparable to data from after 2010. Second, as numerous monitors have been
13 discontinued because of declining Pb concentrations, the proportion of monitors that is located near
14 sources has increased. The national median of maximum 3-month average Pb concentrations for the 74
15 Pb-TSP monitors in the U.S. that operated during the entire period declined by 89% from 1990 to 2010
16 ([U.S. EPA, 2013](#)). For a smaller population of 37 monitors with a higher proportion of source-oriented
17 monitors that operated continuously from 2010 to 2021, the national median of maximum 3-month
18 average Pb concentrations across monitoring sites decreased by 88% for monitors operating for that entire
19 period ([U.S. EPA, 2022](#)). This recent decrease was driven by the 2008 NAAQS revision and the steepest
20 declines were observed over the period from 2012 to 2015. Up-to-date graphics of annual maximum 3-
21 month average concentrations over the most recently available 3-year period and as well as ambient air Pb
22 concentration trends are available in “Overview of Lead (Pb) Air Quality in the United States” ([U.S.](#)
23 [EPA, 2022](#)).

24 These national trends of decreasing ambient air Pb concentrations are a continuation of longer-
25 term trends that can be traced back to the removal of Pb from gasoline in the 1970s and later reductions of
26 industrial use and processing of Pb in some areas and have been extensively documented in the 2013 Pb
27 ISA ([U.S. EPA, 2013](#)) and earlier assessments. The trend is generally reflected in long-term local and
28 regional observations of Pb concentrations in the U.S. and Canada for diverse locations and conditions,
29 including monitoring sites in New York state ([Buckley and Mitchell, 2011](#)), urban Baltimore ([Lin et al.,](#)
30 [2022](#)), Montreal ([Bagur and Widory, 2020](#)) and Edmonton ([Bari and Kindzierski, 2016](#)), and high traffic
31 areas of Cincinnati ([Grinshpun et al., 2014](#)). It is also reflected in long-term sediment records from
32 numerous locations in the United States, as described in Section 1.2.3. In general, there was a steep
33 decline in ambient air Pb concentrations in the 1970s and 1980s corresponding to the phase-out of Pb in
34 gasoline, and in some cases a more recent period of continued decline corresponding to reductions in Pb
35 emissions from local and regional industrial sources. In general, both AQS data and more detailed North

1 American field studies usually support continuing national and local long-term trends of decreasing
2 ambient air Pb concentrations.

1.5.2. Urban and Neighborhood Spatial Variability

3 The 2013 Pb ISA contains a comparison of Pb concentrations across six counties and past
4 literature on spatial distribution of airborne Pb in urban centers illustrating intra-urban variability ([U.S.
5 EPA, 2013](#)). These examples show that both point and non-point sources along with wind strength and
6 direction can play a role in distributing Pb across urban areas and create spatial variability. The 2006 Pb
7 AQCD ([U.S. EPA, 2006b](#)) contains additional information on Pb transport in the environment.
8 Ambient air Pb across urban and neighborhood scales may not be captured by national monitoring
9 networks because of contributions from more local emission sources ([Yu et al., 2011](#)). In particular, high-
10 emitting Pb sources may create local hotspots of elevated Pb in environmental media (Section 1.3.4).
11 Since the publication of the 2013 Pb ISA, additional studies have been published illustrating Pb
12 concentrations across urban centers. Recent studies on near-road spatial variability are discussed together
13 with traffic and road sources in Section 1.2.6. More general aspects of urban and neighborhood spatial
14 variability are described in this Section.

15 Spatial variability of ambient air Pb across New York City has been investigated in several
16 studies. A study of PM_{2.5} measurements across four boroughs of New York City found that spatial
17 patterns varied by season with the highest concentrations of Pb observed during the summer (7.94 ng/m³).
18 The sites within lower Manhattan had the highest concentrations Pb for both seasons. The authors
19 attributed Pb concentrations in lower Manhattan to waste incineration and traffic-related sources but
20 mention that Pb may be a poor tracer for incineration ([Peltier and Lippmann, 2011](#)). In a study of PM_{2.5}
21 samples used in a land-use regression model from 150 street-level sites across New York City, Pb was
22 found to have a mean value of 3.40 ng/m³ with a standard deviation of 1.52 ng/m³ and spatial coefficient
23 of variation of 0.45. Pb was attributed most strongly to boilers burning residual oil, which was correlated
24 with other elements consistent with emission factors data, results from combustion experiments, and
25 characterization of residual oil fly ash ([Ito et al., 2016](#)).

26 Other urban centers have also been investigated for spatial variability of ambient air Pb. Particles
27 between 2.5 and 10 μm were collected at ten sites in the greater Los Angeles area. The annual average Pb
28 concentration did not vary greatly between the Los Angeles (1.3 ng/m³), Long Beach (1.3 ng/m³),
29 Riverside (0.8 ng/m³), and Lancaster (0.5 ng/m³) sites. Principle component analysis revealed that Pb was
30 present most commonly with other elements that are tracers of abrasive vehicular emissions such as Sb,
31 Ba, Mo, Cu, Rh, and Fe ([Pakbin et al., 2011](#)). In another study spatial variations of trace elements in PM₁₀
32 around Paterson, NJ were investigated. Among 199 samples taken, there was an average Pb concentration
33 of 5.37 ng/m³ with a standard deviation of 5.07 ng/m³ ([Yu et al., 2011](#)). In a study of trace metals
34 concentrations across four sites in St. Louis the authors found that annual median air Pb concentrations in

1 PM₁₀ ranged from 6.01 ng/m³ (S.D. = 10.10 ng/m³) at a suburban site ~10 km away from the urban core
2 to 8.96 ng/m³ (S.D. = 16.00 ng/m³) at an urban site 3 km north of the urban core. Conditional probability
3 function plot graphs revealed that all sites were affected by Pb from a source to the south. A local
4 smelting plant was identified as the possible source ([Yadav and Turner, 2014](#)).

5 [Stevens et al. \(2014\)](#) investigated Pb-PM_{2.5} concentrations across six sites in Detroit, Michigan
6 that were part of the Detroit Exposure and Aerosol Research Study. The authors found that Pb
7 concentrations in PM_{2.5} were heterogeneous across sites, with the highest mean concentrations of Pb for
8 outdoor, indoor, and personal exposures occurring at two heavily industrial areas focusing on steel
9 manufacturing and automobiles. In another study spatial variability of PM_{2.5} metals in Massachusetts was
10 modeled. Pb was estimated to be most heavily concentrated in areas of high roadway density in
11 downtown Boston, similar to areas high in Al, Fe and Ti, indicating that the source of Pb was likely from
12 road dust and soil particles. The mean value for Pb among 62 sites was 4.5 ng/m³ with a predicted
13 coefficient of variation of 0.467 ([Requia et al., 2019](#)).

14 [Upadhyay et al. \(2011\)](#) investigated Pb in PM_{2.5} and PM₁₀ around three sites in Phoenix, Arizona.
15 Two sites were located south of Phoenix Sky Harbor International Airport representing a mix of urban
16 residential and industrial use while a control site was located east of the airport with no local point
17 sources present. The two sites closest to the airport had the highest mean air Pb concentrations among the
18 three sites, 4.6 ng/m³ and 4.7 ng/m³ versus 2.0 ng/m³ for Pb-PM_{2.5} and 6.3 ng/m³ and 5.6 ng/m³ versus
19 3.3 ng/m³ for Pb-PM₁₀, similar to other U.S. cities. Principle component analysis revealed that Pb in PM_{2.5}
20 was grouped with elements Cu and Zn, suggesting mobile sources and Pb in PM₁₀ was grouped with As
21 and Cr, suggesting combustion processes as a source. Pb concentrations also peaked on January 1st,
22 suggesting the influence of local fireworks.

23 Researchers may also use biological organisms as bioindicators for Pb concentrations in ambient
24 air. Pigeons were used in a New York City study. Samples of pigeons' blood revealed that the highest Pb
25 concentrations were found in the Soho/Greenwich Village neighborhood (mean = 23.121 µg Pb/dL) and
26 other neighborhoods of downtown Manhattan ([Cai and Calisi, 2016](#)). In another study, 26 lichen samples
27 and four atmospheric PM measurements were collected around Middletown, Ohio. Pb concentrations in
28 lichen samples reached a high of 151.27 ppm nearest the local steel plant but showed large heterogeneity
29 across samples with the lowest Pb concentration at 11.30 ppm found in a lichen sample outside the
30 general area, used as a background. Isotopic analysis of Pb species indicates that the Pb in these lichen
31 samples are a mix of coal fly ash and traffic-related PM, with some possible contribution from steel plant
32 emissions ([Kousehlar and Widom, 2020](#)).

33 [Jovan et al. \(2022\)](#) and [Kondo et al. \(2022\)](#) used local youth to collect moss samples around the
34 industrially adjacent South Park and Georgetown neighborhoods in Seattle, WA. [Jovan et al. \(2022\)](#)
35 found that the 79 samples collected by youth with minimal supervision had highly significant agreement
36 ($p = 0.001$) to 19 expert-collected samples. Pb concentrations peaked along the industrial core separating
37 the two communities. Among all samples there was a median value of 18.1 mg Pb/kg and minimum and

1 maximum values of 5.9 and 110.6 mg Pb/kg respectively. [Kondo et al. \(2022\)](#) assessed the spatial
 2 predictors of metal concentrations found in 61 of the moss samples. Traffic volume and block-group level
 3 percent people of color were found to be the spatial predictors significantly associated with higher Pb
 4 concentrations in moss.

1.5.3. Seasonal and Diurnal Trends

5 The 2013 Pb ISA ([U.S. EPA, 2013](#)) briefly illustrates that, depending on the measurement
 6 location, there can be seasonal variation for Pb in ambient air. Seasonal variability can depend heavily on
 7 local meteorological conditions including mixing height, wind direction, precipitation, and humidity ([U.S.
 8 EPA, 2006b](#)). [Levin et al. \(2020\)](#) discusses several additional factors that may contribute to local trends in
 9 ambient air Pb. As explored in Section 1.3.4, resuspension of Pb in soil can contribute Pb into air, with
 10 the highest contributions in dry summer months. Wildfires, which occur most often during the summer
 11 and into fall, can remobilize Pb deposited in the natural environment and Pb contained in man-made
 12 structures (Section 1.2.4). The 2013 Pb ISA ([U.S. EPA, 2006b](#)) includes past research that has identified
 13 seasonal variation of ambient air Pb in various locations. There was no research captured by the literature
 14 screening for this document that contained information on ambient air Pb diurnal trends.

15 Seasonal variation of Pb concentrations has been investigated in several studies of various
 16 locations since the 2013 Pb ISA ([U.S. EPA, 2013](#)). These studies have varied in their design with some
 17 measuring trends over a period of several years while others only measure a one-year period, and some
 18 have presented averages of Pb concentrations while others have presented more detailed monthly data of
 19 Pb concentrations. Table 1-1 below details study conditions and findings of seasonal variations from these
 20 studies.

Table 1-1 Seasonal variations in Pb concentration in Ambient Air

Study	Location	Time Period	Seasons (Months)	Source Attribution of Pb	Findings of Seasonal Variations
(Pakbin et al., 2011)	Greater Los Angeles Area	2008–2009	All Seasons	Abrasive vehicular emissions with some contributions from soil dust and vehicular catalytic converter wear	Average ambient air Pb concentrations highest in the winter for the more urban Los Angeles (1.3 ng/m ³ in winter versus 1.0 ng/m ³ in summer) and Long Beach (1.8 ng/m ³ in winter versus 0.8 ng/m ³ in summer) sites. Average ambient air Pb concentrations were higher in the summer for the semirural Riverside (0.5 ng/m ³ in winter versus 0.8 ng/m ³ in summer) and desert Lancaster (0.2 ng/m ³ in winter versus 0.7 ng/m ³ in summer) sites.

Study	Location	Time Period	Seasons (Months)	Source Attribution of Pb	Findings of Seasonal Variations
(Peltier and Lippmann, 2011)	New York City, NY	2008	Winter (January–March), Summer (May–July)	Incineration and Biomass Burning	Highest concentrations of Pb measured were found in lower Manhattan during the summer months, suggesting a highly localized source of Pb only present during this time. Pb concentrations were near 1 ng/m ³ with hot spot concentrations, on average, at 5 and 10 ng/m ³ during the winter and summer, respectively.
(Song and Gao, 2011)	Carlstadt, NJ	2007–2008	Winter (December–February), Summer (July)	Brake wear, direct vehicle emissions, other urban sources	Higher concentrations of Pb in winter for both coarse (2.04 ng/m ³ in winter versus 1.41 ng/m ³ in summer) and fine (2.82 ng/m ³ in winter vs. 1.27 ng/m ³ in summer) despite overall aerosol mass being lower in winter than summer.
(Yu et al., 2011)	Paterson, NJ	2005–2006	All Seasons	Traffic-related and industrial emissions	Pb concentrations highest in the winter, and second highest in the fall for mobile, industrial, and commercial sites. Pb concentrations were highest in winter and summer for the background site.
(Grinshpun et al., 2014)	Cincinnati, OH	2010–2011	Fall, Winter, Summer (months not specified)	Traffic-related emissions	Pb concentration was higher in the fall than winter and summer in a downtown Cincinnati location.
(Kundu and Stone, 2014)	Iowa	2009–2012	All Seasons	Diesel combustion, gasoline combustion, biomass burning, industry	There were no consistent seasonal patterns for Pb concentrations across sites measured.
(Prabhakar et al., 2014)	Southern Arizona	1988–2009	Summer (April–June), Fall (October–November), Monsoon (July–September), Winter (December–March)	Smelting operations and traffic-related emissions	Seasonal patterns were found to vary across sites, indicative of local conditions. Phoenix was impacted more heavily by traffic-related emissions, being an urban center, while Tonto National Monument was more affected by related smelting operations.
(Stevens et al., 2014)	Detroit, Michigan	2004–2007	Summer, Winter (months not specified)	Traffic-related and industrial emissions	Outdoor mean Pb concentrations were found to be slightly higher in summer for four sites. In contrast, site 5, a heavily industrial site had much a higher average Pb winter value (42 ng/m ³) than summer value (15 ng/m ³).

Study	Location	Time Period	Seasons (Months)	Source Attribution of Pb	Findings of Seasonal Variations
(Li and McDonald-Gillespie, 2020)	Tulsa, OK and Picher, OK	2010–2016	All Seasons	Atmospheric Pb blown from nearby chat piles	Picher showed strong seasonal Pb mass concentration peaks during the period of January–March and September, likely due to Pb blown from nearby chat piles. Tulsa did not show any strong seasonal variation over the measurement period.

NJ = New Jersey; NY = New York; OH = Ohio; OK = Oklahoma

1.5.4. Particle Size Characteristics

1 The size distribution of Pb-containing particles differs depending on source type and the
2 collection efficiency of Pb-TSP samplers. [Cho et al. \(2011\)](#) found that most studies included in their
3 review and published after 1986 indicated a shift in Pb particle size distribution from the fine fraction to
4 coarse fraction with the primary mode rising to 2.5–10 μm from a previous estimation of it being below
5 2.5 μm . Studies used to evaluate this shift ranged from sampling near roads, near industrial sources,
6 offshore in a lake environment, rural locations, and urban locations, within the United States and the
7 European Union. The elimination of the combustion of tetramethyl- and tetraethyl Pb in automobiles as
8 the dominant source of Pb-PM in the atmosphere, as indicated in Section 1.2, has led to larger Pb-
9 containing particles on average. However, coarse particles have higher settling velocities than fine
10 fraction or ultrafine fraction particles, resulting in measured concentrations of coarse or ultracoarse
11 (particles greater than 10 μm diameter) particles being spatially and temporally heterogeneous, as these
12 particles may drop out before they are collected by a TSP sampler. These topics have been discussed in
13 previous assessments ([U.S. EPA, 2013, 2006b](#)) as well as in Sections 1.3.1.1 and 1.4.2.

14 The literature search and screening process for the current iteration of the ISA did not find many
15 published studies containing information on Pb particle size distributions beyond what was included in
16 the 2013 Pb ISA ([U.S. EPA, 2013](#)). To briefly summarize information from the previous ISA, near-
17 roadway emissions may be the result of emissions directly from vehicular combustion or parts such as
18 brakes or wheel weights, sources near the roadway that are not related to traffic, or traffic-induced
19 turbulence causing resuspension of deposited Pb-bearing particles originating from wheel weights,
20 industrial emissions, or historic sources into air. [Sabin et al. \(2006\)](#) mentioned that freeways tend to be a
21 source of very large particles dispersed by turbulence from vehicular traffic and found a bimodal
22 distribution of particles at a near-road site in Los Angeles with a mode in the largest size fraction
23 sampled.

24 Other near-road studies in the 2013 Pb ISA ([U.S. EPA, 2013](#)) found a mix of size distributions at
25 near-road sites subject to different meteorological conditions and measurement techniques. [Hays et al.](#)
26 [\(2011\)](#) measured Pb in ambient air particles 20 meters north of a major interstate in Raleigh, NC. Pb was

1 found in PM_{10-2.5}, PM_{2.5-0.1} and PM_{0.1} size fractions, at 50 mg Pb/kg in each size fraction. Ambient air Pb
2 concentrations appeared unimodal and normally distributed over the accumulation mode with
3 0.4 ± 0.4 ng/m³, 1.4 ± 0.6 ng/m³, and 0.1 ± 0.02 ng/m³ for PM_{10-2.5}, PM_{2.5-0.1}, and PM_{0.1} size fractions,
4 respectively. Daily concentration changes were heavily correlated with traffic, including Pb-PM₁₀ samples
5 highly correlated with As samples, most likely resuspended from contemporary roadway sources. [Song
6 and Gao \(2011\)](#) collected measurements of ambient air PM using a sampling site approximately 5 meters
7 from the New Jersey Turnpike. The Pb mass size distribution had a bimodal concentration distribution in
8 summer and a trimodal distribution in winter with 47% and 58% of Pb mass measured in fine particles in
9 summer and winter, respectively (Pb mass concentration values mentioned in Table 1-1). Factor analysis
10 attributed the source of Pb to brake wear, fuel combustion, and urban pollution.

11 [Masri et al. \(2015\)](#) collected both fine-mode and coarse-mode ambient air PM at the Harvard
12 supersite in Boston, MA. This site is located atop a six-story building within one block of a four-lane
13 roadway and two major highways. Trace amounts of Pb were found to be exclusively associated with
14 fine-mode particles. Positive Matrix Factorization analysis was used to associate these particles across a
15 wide range of possible sources from with regional pollution, motor vehicles, crustal or road dust, oil
16 combustion and wood burning.

17 Size distributions have also been recorded at other sites as well, reflecting spatiotemporal
18 variability within and near cities. [Gonzalez et al. \(2021\)](#) analyzed data from five sites (Manila,
19 Philippines; Marina, CA; Tucson, AZ; Hayden, AZ; Mt. Lemmon, AZ) that measured both
20 submicrometer and supermicrometer particles (range 0.056 – 18.0 μm) which were then extracted for
21 further analysis. Pb mass concentrations within the submicrometer and supermicrometer ranges were
22 found to vary by site with Manila, Tucson, and Hayden (the location of an active metals smelter) having
23 higher mass concentrations in the submicrometer range while Marina and Mt. Lemmon had higher Pb
24 mass concentrations in the supermicrometer range. The Marina and Manila sites were also separated by
25 fire and non-fire-influenced datasets which showed the presence of a submicrometer mode for Pb mass
26 concentration in the fire-influenced data that was not present in the non-fire-influenced data. [Upadhyay et
27 al. \(2011\)](#) measured ambient air PM in Phoenix, Arizona both south and east of the Phoenix Sky Harbor
28 International Airport. The authors found Pb to be associated with both fine and coarse particles at three
29 sites in Arizona, with PM_{2.5}:PM₁₀ ratios between 0.5 and 0.7 for Pb at the three sites tested, indicating that
30 a substantial fraction of Pb was associated with both fine and coarse particles at each site. [Youn et al.
31 \(2016\)](#) performed chemical speciation on aerosol samples collected at an active smelting site in Hayden
32 AZ and an urban background site in Tuscon AZ. The particle size distribution of Pb mass found at the
33 active smelting site was bimodal with a large peak at 0.32 μm, and a smaller peak at 5.6 μm. The
34 background site was trimodal with peaks at 0.1 μm, 0.32 μm, and a smaller peak at 3.2 μm.
35 Submicrometer particles were attributed to the condensation and coagulation of smelting vapors, whereas
36 coarse particles were attributed to fugitive dust, including from mine tailings. Ambient air Pb
37 concentration values at the active smelting site were higher than in samples from the background site.

1.5.5. Background Concentrations

1 A small fraction of Pb in ambient air in the United States cannot be reduced by domestic emission
2 controls or domestic interventions within the United States. According to the 2013 Pb ISA, natural
3 sources of Pb to ambient air include suspension of surface soil containing natural Pb and wind erosion of
4 natural Pb-containing rocks. Previous assessments have suggested the evidence to indicate a plausible
5 range of natural background Pb concentration is 0.02 to 1 ng/m³ ([U.S. EPA, 2013](#); [Nrc, 1980](#)). As
6 described in the 2013 Pb ISA ([U.S. EPA, 2013](#)), average concentrations in this range have been measured
7 at remote monitoring sites such as Crater Lake, OR and Lassen Volcanic National Park, CA. In addition
8 to natural sources, intercontinental transport of Asian dust could also make a substantial contribution to
9 total atmospheric Pb, but generally less than 1 ng/m³ ([U.S. EPA, 2013](#)). The 2013 Pb ISA reviewed
10 evidence for intercontinental transport of Pb in African dust to the Southeastern United States and
11 described mixed results concerning whether intercontinental transport or natural sources contributed the
12 most to atmospheric Pb. The 2013 Pb ISA concluded that estimates of background Pb concentrations in
13 ambient air were well below current concentrations ([U.S. EPA, 2013](#)). No new studies on background Pb
14 concentrations in the U.S. since the last NAAQS review were identified in our literature search and
15 screening process.

1.6 Summary and Conclusions

16 Pb emissions and ambient air concentrations in the United States continue to steadily decline.
17 Major industrial sources have either reduced their emissions or closed, resulting in the emergence of
18 aviation gas as the dominant contemporary source. However, resuspension of soil containing Pb from
19 legacy sources is has the potential to contribute to atmospheric Pb in some locations, and high
20 concentrations of Pb associated with wildfires have been observed. Pb from these sources continues to
21 have potential health and ecological effects after atmospheric deposition to soil and water. A substantial
22 fraction of airborne Pb can be associated with PM larger than 10 μm in some locations. Bias and
23 uncertainty associated with sampling these large particles in Pb-TSP sampling are still an issue, although
24 it has become better understood and there have been several improvements in measurement tools,
25 including development of an ICP-MS-based FRM for Pb analysis, and introduction of reference materials
26 for analysis on filters. Overall, there have been substantial improvements in our understanding of and
27 research capabilities for airborne Pb.

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