Air Pollution Management in the 20th Century

Introduction

Air pollution has been a scientific and regulatory concern since before 1900. The 20th Century saw a dramatic evolution of air pollution management from particulates, to chemicals in those particulates, to vapors, and to reactive atmospheric chemistry. Much understanding of these issues has been gained and many control tools were developed by 2000, so that science and government are now left with the ultimate challenge for air pollution - greenhouse gas effects on climate change. That is not to say, however, that air pollution is solved. Air pollution problems are extremely complex, stretching even our current measurement, modeling, and control capabilities. This paper chronicles the evolution of air pollution and its management in the 20th Century, using lead as an example because this element plays such a representative role generally in environmental health as well as specifically in air pollution.

Identifying and solving air pollution problems in the United States has been a slow and arduous process, driven by several landmark events and paradigm changes. Similar to the Shifrin (2005) analysis of the scientific understanding and historical management of water and land pollution, the evolution of air pollution science occurred on a timescale of decades. While some historians suggest that particular events, such as the Donora, Pennsylvania air quality event, single-handedly set the wheels of change into overdrive, it will be shown that progress in our understanding of air pollution and our approaches to mitigating air pollution problems began well before that, and were the product of a series of landmark events (Table 1) with many years of research and scientific debate.

Prior to the 20th century and continuing until approximately the 1950s, air pollution control in the United States focused mainly on gross effects: visible black smoke and other solids (*e.g.*, fly ash) and gaseous by-products of incomplete coal combustion such as sulfur dioxides (). This narrow interest in air pollution was pervasive despite early recognition of other air pollutants and sources, including carbon monoxide and lead from automobile emissions (Bloomfield and Isbell, 1933; Green, 1931) as well as fine dust from sources other than fuel combustion, such as industrial and building operations (Monnett, 1923, as quoted in Briggs, 1941; Green, 1931). Industrial and residential combustion of coal and coke was widely considered to be the primary source of air pollution in urban and industrialized areas in the first half of the 20th century.

By the early 1900s, smoke had become an issue for both science and government (Monnett, 1923, as quoted in Briggs, 1941; Meetham, 1952; Faith, 1959; Swain, 1949). For example, smelters caused complaints about crop and livestock damage in Utah, Montana, and California (Baskerville, 1910; Ebaugh, 1907; 1910; Westby, 1912; Selby Smelter Commission, 1915; Wells, 1917).[1] Smoke was a nuisance due to the soiling of merchandise, buildings, and clothes; injuring vegetation and animals; impacting visibility and loss of sunlight (PHS, 1936; Meller, 1931). The relationship between smoke and health was not established in the early 1900s (PHS, 1936). Drinker (1939) noted that air pollution was viewed more as a nuisance than a health hazard to the "average man."

The US Public Health Service emerged as the federal research authority on air pollution in the 1920s and 1930s, conducting such important work as a 1927 study quantifying loss of daylight due to smoke in New York City and the 1931-1933 atmospheric pollution study of 14 of the largest US cities (PHS, 1936). In addition to these large-scale studies, a number of cities, including Pittsburgh, Chicago, Cleveland, and St. Louis, sponsored air pollution measurement studies prior to 1930 to investigate their smoke problems (PHS, 1936; Meller, 1931). The air pollutants of interest in these early air pollution measurement studies included total airborne suspended particles, total dustfall, and sulfur dioxide (Schueneman, 1955; PHS, 1936).

A new form of air pollution emerged in the Los Angeles area: smog. In the summer of 1943, smog clouds descended on the city, reducing visibility to 3 blocks and eliciting reports of smarting eyes, respiratory discomfort, nausea, vomiting, and vegetative damage (SCAQMD, 1997; CARB, 2004). A local butadiene plant was originally blamed for this "gas attack" but, following the plant shutdown, these extreme air pollution events continued. The repeated occurrence of smog events prompted both heightened scientific and regulatory air pollution activity in the Los Angeles basin in the 1940s. In 1947, the nation's first air pollution district was formed in Los Angeles County. In 1950, Dr. Arie Haagen-Smit of the California Institute of Technology proposed a photochemical mechanism for smog formation, identifying nitrogen oxides and hydrocarbons from automobiles and the petroleum industry to be key causative agents (Haagen-Smit, 1950). This was a significant breakthrough with sweeping ramifications for the emerging air pollution field, as it highlighted air pollutants other than smoke as threats to public health and welfare.

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[1] McKee (1969) noted that few damage claims or reports of problematic smelter emissions have become a matter of public record since approximately 1940. McKee (1969) attributed this to the successful responses of smelter companies to address these early air quality problems, both through engineering measures (*e.g.*, use of tall stacks and stringent pollution control equipment) and through purchase of adjacent lands.

Approximately 1950 thus represents the emergence of the modern air pollution field and its focus on potential health implications and the levels and effects of specific air pollution constituents Clayton, 1949; McCabe, 1954). Air pollution was redefined, with air pollutants such as oxidants, hydrocarbons, and sulfur dioxide becoming the subject of increased attention and research activity (Lagarias, 1982). Nevertheless, many scientists at this time viewed air pollution incidents as providing only limited evidence for the acute health effects of major air pollution events, which they relegated to an unusual set of meteorological and geographical conditions (Clayton, 1949; Nord, 1951; Foulger, 1952a). In fact, Nord (1951) concluded: "In view of the fact that tobacco smoking- which is a highly intense form of personalized air pollution- is not regarded as harmful for most people, it seems doubtful whether the general contaminants in ordinary city air have any perceptible effect on the health of urban dwellers."

Evolution of Air Pollution Science

By the early 1900s, both gases and solids were recognized as forms of air pollution. The use of poison gases in World War I, acute occupational exposures such as in the "dusty trades," and early smoke-fog events in cities such as Glasgow and London (Chambers, 1968; Foulger, 1952b) are examples of this early recognition. Green (1931), Drinker (1939), Meller (1931) and Lagarias (1982) described sources of both point and fugitive airborne particulates, including from chimneys, metallurgy, vehicles (brakes, fuels, tires, and roads), earthworks, and factories. Smelters were noted not only to be sources of visible smoke but also of other air pollutants such as sulfur dioxide, sulfuric acid, metal fumes and metal dust (Baskerville, 1910; Ebaugh, 1910; Westby, 1912; Selby Smelter Commission, 1915; Wells, 1917; Swain, 1939).

Automobile emissions were recognized soon after the advent of leaded gasoline to be sources of airborne lead, although there was controversy regarding the health effects of auto emissions (Lewis, 1985; Hernberg, 2000). In fact, shortly after the introduction of lead as an anti-knock additive in gasoline in the 1920s, the US Surgeon General held a meeting among industrial representatives and public health experts in 1925 to debate the safety of tetraethyl lead as a gasoline additive (Lippmann, 1990; Needleman, 1998; Hernberg, 2000). Although several leading scientists including Dr. Alice Hamilton of Harvard University and Dr. Yandall Henderson of Yale University were very outspoken regarding the potential for widespread health risks associated with lead in automobile emissions, the meeting ended in disarray without any conclusions (Lippmann, 1990; Hernberg, 2000). A committee convened by the Surgeon

General concluded in 1926 that there was a lack of evidence to recommend a ban for tetraethyl lead. The large-scale use of tetraethyl lead in gasoline commenced.

Drinker and Hatch (1936) summarized the state of the knowledge regarding the various forms of particulate air pollution, defining dust, fumes, smoke, and mist or fogs based on such characteristics as their sources and mechanism of formation, particle size, and behavior. Early air pollution studies such as the 1931-1933 study of 14 of the largest US cities quantified amounts of carbon, silica, iron, and sulfur compounds to characterize the nature of the smoke problem (PHS, 1936). These studies also reported particle size distributions for suspended dust samples using microscopic analyses that identified particles with diameters down to approximately 0.2 microns.

By the 1930s scientists recognized particle size as a fundamental property affecting the behavior, effects, and control of airborne particles. Drinker and Hatch (1936) recognized that particle size not only dictated the settling properties of airborne particles, but also influenced their physiological effects. Munger (1952) understood that particle size was a factor determining the method of sampling and analysis of airborne particles, the equipment for separating particles from air, and the method for disposal. Both Munger (1952) and Clayton (1949) highlighted that particle size was a key factor influencing how long particles remain in the air and the distance they travel prior to deposition on the ground. Clayton (1949) distinguished between larger particles settling near their emission source under normal weather conditions and smaller particles (*i.e.*, less than a micron in size) remaining suspended in the atmosphere for distant transport.

The critical relationship between air pollution and meteorological conditions was also examined in the early 1900s. Wells (1917) identified 4 critical weather conditions associated with high ground-level SO_2 concentrations and plant damage at the American Smelting and Refining Co. smelter in Murray, Utah: (1) temperature above 40 degrees Fahrenheit; (2) relative humidity above 70%; (3) daylight; and (4) prevalent winds of 3 hours or more. The first 3 factors were related to stomata opening and uptake of gases by plants, while the fourth factor affected the duration of exposure. He concluded it was "essential" to have a complete installation of standard weather instruments at smelter sites.

Air transport concepts were recognized to provide rudimentary predictive theory and simple analytical techniques by the 1930s. Wells (1917) studied plume rise and atmospheric dispersion, observing that the greater the difference in temperature between the stack gases and the outside air, the greater the distance at which gases met the ground and the less the ground-level concentrations. This was the genesis of air modeling, which, in the 1930s and 1940s, was captured in simple mathematical expressions for predicting air concentrations resulting from pollutant sources (Bosanquet and Pearson, 1936; Sutton, 1947).

Some of the foundations of today's principles of air pollution were thus established by the 1940s, although technological limitations, prevailing beliefs, and a lack of validating data and quantitative theories were key barriers to the level of progress. At that time, air pollution studies were generally conducted for empirical investigation of the magnitude of smoke problems in cities through the collection of large amounts of data, rather than data collection for the development and testing of theories. Data analyses were typically simple in nature, such as ranking cities according to air pollutant levels and identifying qualitative relationships between different factors (*i.e.*, PHS (1936) reported that atmospheric pollution was less on Sundays than on weekdays, indicating the importance of industrial sources). Overall, there was a practical understanding of the relationship between air pollution and various factors, such as particle size and weather, but a general lack of quantitative theories, confirmatory models, and validating data analyses.

Source Inventories and Emissions Factors: 1950s and 1960s

Following the recognition that the air pollution problem was more complex than just a smoke problem, the 1950s saw the advent of pollutant emission inventories and source characterization (Schueneman, 1963; Rupp, 1956). Prior to this, air pollution studies generally focused on development of sampling methods and measurement of contaminants (Larson *et al*., 1953).

Some of the earliest emission inventories were performed in the Los Angeles area (Southerland, 2005). By using source contributions and relating them to air sampling data, Larson *et al*. (1953) developed emissions inventories for the base year 1951 in Los Angeles County and used adjustment factors to obtain estimates for 1940 and 1948. They used field testing data from more than 350 tests on pollution sources in the Los Angeles area. New York-New Jersey, Washington, and St. Louis inventories followed in the 1960s (NAPCA, 1969a).

Standardized emission factors for industrial air pollutant sources were developed by the US Public Health Service in the 1960s. This effort culminated in a May 1965 compilation that was expanded to cover additional sources and re-released as the "Compilation of Air Pollutant Emission Factors" in 1968 (PHS, 1968). As discussed in PHS (1968), the purpose of these early compilations was to provide source emission factors for use in conducting air pollutant emission inventories. PHS (1968) noted that emission factors may be based upon tests at a single facility or just a few facilities, and were thus intended for making estimations rather than exact calculations of emissions. Primary lead smelters were among the source types covered in PHS (1968), although these early smelter emission factors were limited to particulates and sulfur compounds from 3 types of lead furnaces (cupola, pot, and reverberatory/sweating) but no other smelting processes. Lead or fugitive emission sources were not addressed in the early emission factor compilations.[2]

Fugitive Emissions: 1970s to 1980s

Given their clear and obvious presence, stacks and other industrial point sources typically were the focus of early pollution control measures up through the preparation of the earliest state implementation plans for TSP (Total Suspended Particulates) in the early 1970s (US EPA, 1977a). Fugitive emission sources were known to exist prior to the 1970s, but due in part to difficulties in their measurement and estimation of air quality impacts, they were not emphasized (US EPA, 1977a). A renewed interest in fugitive emission sources only developed in the mid-1970s when they were identified as potential barriers affecting attainment of the TSP National Ambient Air Quality Standards (NAAQS) in some parts of the country (Lillis and Young, 1975; US EPA, 1977a; 1982).

The increased attention did not immediately translate into reliable emission factors or improved, accurate technologies for quantifying air quality impacts. Our ability to predict fugitive source impacts remained inadequate, and to the present, is limited and uncertain. Twenty years after initiating a series of focused fugitive emission studies, US EPA (1993) concluded that it was still then not possible to draw any firm quantitative conclusions about fugitive emissions measurement accuracy due to the general lack of available validating studies. US EPA then sponsored more research on fugitive emissions. In their seminal article, "EPA Looks At Fugitive Emissions," Lillis and Young (1975) distinguished between 2 categories of fugitive emissions – industrial (*i.e.*, process-related) fugitives and fugitive dust. Industrial fugitive emissions were defined as "both gaseous and particulate emissions that result from industrial

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[2] Section 4 discusses in greater detail the evolution of the emission factors compilations, which became the responsibility of the US EPA in the early 1970s and are now known as the AP-42 documents. As discussed in this section, air lead emission factors for a number of industrial sources, including primary lead smelters, were provided for the first time in 1979. The 1986 Supplement A to the fourth version of AP-42 provided, for the first time, lead emission factors for fugitive sources at primary lead smelters as well as particle size distribution data for fugitive sources (US EPA, 1986b).

related operations and which escape to the atmosphere through windows, doors, vents, *etc.*, but not through a primary exhaust system, such as a stack, flue, or control system" (Lillis and Young, 1975). Fugitive dust emissions were defined as natural or anthropogenic dusts (particulates only) made airborne by wind, human activity, or both (Lillis and Young, 1975). In a later guidance document, US EPA (1977a) defined industrial process fugitive particulate emissions (IPFPE) to include fugitive emissions (industrial) and fugitive dust emissions that originated from within industrial facilities (IPFPE is hereafter used in this section to refer to any fugitive emissions arising from within the boundaries of an industrial facility).

Lillis and Young (1975) and US EPA (1977a) noted that fugitive emissions could have a greater effect on air quality in close proximity to the source than stack emissions, in part because they tend to occur at or near ground level and remain there due to a lack of plume rise. They cited a handful of studies to support this idea, including a 1975 study of fugitive lead emissions at the Silver Valley/Bunker Hill primary lead smelter in Idaho, where preliminary results indicated that fugitive lead emissions were approximately 2 to 3 times the magnitude of process (stack) emissions (Lillis and Young, 1975). The air quality significance of IPFPEs was a subject of debate at the US EPA-sponsored "First Symposium on Fugitive Emissions," held May 17-19, 1976, in Hartford, CT. For example, Yocum (1976) agreed with their significance, but argued that fugitive emissions were site-specific (and thus defied unified prediction. On the other hand, McCutchen (1976) argued that iron and steel mill fugitive emissions were largely "cosmetic" in nature.

Uncertainty and debate surrounded the potential air quality impacts of IPFPEs in the 1970s, mainly due to difficulties associated with their measurement. Conventional techniques were generally not appropriate for measurement of IPFPEs since they are not emitted from a definable point source such as a stack (Lillis and Young, 1975; US EPA, 1977a). Few reliable emissions data on IPFPEs were available to characterize even the relative magnitude of emissions (Lillis and Young, 1975; US EPA, 1977a; 1979a). Lillis and Young (1975) and US EPA (1977a) concluded that the lack of reliable data contributed to the general lack of inclusion of fugitive emission sources in source emission inventories prepared by states and local air pollution control agencies in support of the first SIPs for TSP.

An ultimate product of EPA's sponsorship was the 1976 US EPA *Technical Manual for Measurement of Fugitive Emissions: Volumes 1 through 3* (US EPA 1976a; 1976b; 1976c) that outlined 3 basic approaches to the measurement of atmospheric fugitive emissions that had been developed by this time: (1) quasi-stack,[3] (2) roof monitor,[4] and (3) upwind-downwind.[5] None of these techniques was widely accepted as an accurate measurement method (McCutchen, 1976) since each was burdened with technical limitations and uncertainties (US EPA, 1976a; 1976b; 1976c). The technical manual provided criteria for the selection of the most applicable of the 3 measurement methods for a given set of conditions, as well as detailed sampling procedures (US EPA, 1976a; 1976b; 1976c). However, the 1976 technical manual provided little quantitative information for assessing the measurement accuracy or emission factor reliability of these methods, as did a 1980 updated guideline protocol document (TRC, 1980).

Modeling was an alternative approach to measurement of air quality impacts from IPFPEs, but modeling was limited by the lack of reliable emission factors. The 1977 US EPA *Technical Guidance for Control of Industrial Process Fugitive Emissions* (US EPA, 1977a) noted the flaws of fugitive emission modeling due to several complicating factors, such as variable emission rates and the lack of detailed particle sizing data needed to model deposition.

EPA's IPFPE studies identified a number of possible fugitive emission sources at, for example, primary lead smelters, including: (1) unloading, material handling, and concentrate storage; (2) material handling, sinter crushing, particulate escaping sintering machine hoods; (3) fugitive emissions from blast furnaces; (4) fumes from molten metal handling during the refining process; (5) material handling operations; and (6) windblown dust resuspended from ground, storage piles, or open areas around smelter (US EPA, 1979a). Most notable was the 1977 US EPA study, *Sample Fugitive Lead Emissions from 2 Primary Lead Smelters* (US EPA, 1977b), which summarized measurements made at 2 primary lead smelters and provided a number of fugitive lead emission factors.

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[3] The quasi-stack sampling method refers to the temporary installation of a hood or enclosure over the source, with venting to an exhaust duct or stack of regular cross-sectional area. This method allows for the use of standard stack sampling techniques. The major limitations of this technique include its use for only those sources amenable to the installation of a hood or enclosure, and the possibility that the hood or enclosure may alter emissions.

[4] The roof-top sampling method involves the operation of samplers at building or other enclosure openings such as roof-top vents, doors, windows, *etc.* Limitations of this technique include having to sample very large openings and dealing with large variations in flow-through openings.

[5] The upwind-downwind sampling method involves the use of upwind and downwind ambient monitors to determine the differential air quality impact associated with IPFPEs. This technique is generally considered the least reliable of the three methods due to a variety of complicating factors, including background sources of the pollutant of interest, issues associated with monitor siting, *etc.* (US EPA, 1993a).

Improved Particle Characterization: 1980s to Present

Coinciding with improved analytical capabilities for lead and other trace metals (see Section 3), studies began to appear in the literature around 1980 on the chemical characterization industrial emissions (Eatough *et al.*, 1979; Foster and Lott, 1980; Harrison *et al.*, 1981; Harrison and Williams, 1983). In addition, better particle size distribution data were also collected.[6] At this time it was recognized that these data were not only important for characterizing pollutant sources, but also for assessing potential health effects (*e.g.*, by providing data on mobility and bioavailability) and for refining predictive air quality models (*e.g.*, by providing improved emission factors and inputs for deposition modeling

Air Sampling and Analysis

Particle Sampling

The development of air sampling was critical because control is difficult without measurement. Compared to other media (*e.g.*, soil, water), air sampling engineering has proven to be more difficult to develop. The main problem has been sample representativeness of particles and gases with precise characterization of their physical properties and chemical composition. The proper location of samplers and chemical analysis of collected samples has been a strong factor in this development.

Air particulate sampling remains imperfect today, but has evolved from sampling all particles by deposition (settling), to impingement of moderate air flows, to filtering of large volumes of air, to selective sampling of different-sized particles, to highly sensitive direct-reading instrumentation. Table 2 documents the development of various samplers for airborne particulates.

Dustfall collectors, consisting of a large conical funnel connected by tubing to a glass bottle, were among the rudimentary samplers used to collect dust and rain in a number of early air pollution studies conducted in US cities (PHS, 1936). Variations of the dustfall collector later evolved into a dustfall method codified in the 1950s and 1960s by the National Air Pollution Control Association (NAPCA) and

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[6] As discussed in Section 2.3, US EPA (1977c) is one of the few early studies reporting particle size distribution data for primary smelter emissions. In addition to this study, Dorn *et al.* (1976) reported particle sizing data for 1972 ambient air sampling conducted at a Missouri farm approximately 800 meters from the base of a lead smelter smokestack (smelter not named). For over 70 sampling days, Dorn *et al*. (1976) reported that approximately 66% of the collected Pb was associated with particles with diameters of less than 4.7 µm.

the American Society for Testing and Materials (Chow, 1995; Hendrickson, 1968). These samplers collected large particles (>5 μm) by gravitational settling into open containers over several weeks. They suffered many interferences from insects, fungi, bird droppings, and plant materials, and accuracy was affected by wind speed and direction. The uncertainty of the method was confirmed by highly variable results from nearby samplers (Chow, 1995). Smaller particles were poorly sampled due to their low settling velocities and wind effects.

Wet impingers, such as the Greenburg-Smith impinger and later the more portable midget impinger, were developed in the 1920s and soon thereafter gained widespread usage (Greenburg and Smith, 1922; Hendrickson, 1968; Marple, 2004). Impingers had a glass tube with a small orifice through which air was drawn at high velocity and sprayed onto a flat surface submerged in water or some other liquid. Good collection efficiencies were reported for particles with diameters down to about 1 μm for the Greenburg-Smith impinger (Giever, 1968). Impingers were used in 1920s and 1930s studies for sampling lead dust and fume (Bloomfield and Isbell, 1933; Littlefield *et al*., 1938; Case, 1941).

The Owens Jet Dust Counter was an early version of modern day impactor samplers (PHS, 1936; Marple, 2004). A pump plunger was used to draw a jet of air through a slit where water condensed on particles and the wetted particles impinged/adhered to a cover glass. Moisture was then evaporated and the cover glass was analyzed microscopically for counting and sizing of particles. This device was used in several prominent 1920s and 1930s air pollution studies, including the Kimball and Hand (1924; 1931) studies of the dust content of the atmosphere in Washington, D.C. and Chicago and the PHS (1936) atmospheric pollution study of 14 of the largest US cities. PHS (1936) reported that they were able to count and size particles down to a diameter of about 0.2 microns with the Owens Jet Dust Counter.

The method of collecting particles by drawing large volumes of air through a filter was first used in approximately 1885; however, the technique was not very accurate because of the lack of electrically operated pumps, timing mechanisms, and flow measurement devices (Chow, 1995). It was not until the late 1940s that this method was refined for measurement of airborne radioactivity from nuclear weapons testing (Chow, 1995). It was soon applied to a variety of particle types, including textile dusts, silica dust, lead fume, and tobacco smoke (Silverman and Viles, 1948). This early sampler, which relied on high volume air sampling and filter weighing, was the predecessor to today's Hi-Vol sampler.

Following its use in the 1953 Public Health Service's National Air Sampling Network (NASN, later renamed the National Air Surveillance Network), the Hi-Vol sampler quickly became the workhorse sampler for airborne particulates. The Hi-Vol became the Federal reference method in 1971 for the TSP National Ambient Air Quality Standard (NAAQS) (PHS, 1958; US EPA, 1971a), and up through the 1980s, thousands of TSP Hi-Vol monitors were operated throughout the United States. Although ambient particulate matter is no longer regulated as TSP and the Hi-Vol sampler is no longer commonly used to measure ambient particle mass concentrations,[7] the Hi-Vol sampler has remained part of the federal reference method for determination of lead in suspended particulate matter for compliance with the lead NAAQS since its promulgation in 1978 (US EPA, 1978a).

Although widely used over the years, the Hi-Vol sampler's measurement flaws have been extensively noted. Initially, lack of filter standardization (glass felt and polystyrene were often used) was a problem (NAPCA, 1969a), but the system was eventually standardized with flat glass fiber filters contained in a box with a peaked roof to prevent passive dust settling onto the filter (Chow, 1995). NAPCA's 1969 *Air Quality Criteria for Particulate Matter* study noted "… the filter material may contaminate the sample; different substances in the sample may react with each other; and losses may occur through volatilization of material." Greater use and study of the Hi-Vol revealed that collection efficiencies declined above particle sizes of 20 µm, while sampler orientation and wind direction/velocity also affected results (US EPA, 1986a; 1999a). Incidental dust settling was found to create positive biases of 10-15% despite the sloped roof of the units (Chow, 1995). Also, the glass fiber filters were shown to absorb sulfur dioxide and nitrogen oxides, potentially adding several $\mu g/m^3$ to the samples (Chow, 1995). US EPA (1977c) identified many issues associated with filter selection and sample preparation, including variable lead content of glass fiber filters. Scott *et al*. (1976) found no lead interference problems, however, thus leaving doubt. Lastly, carbon and copper from the pump were also identified as a source of contamination (Chow, 1995).

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[7] A re-evaluation of the TSP NAAQS beginning in 1978 resulted in the 1987 replacement of the TSP standards with standards for PM_{10}, defined to include those particles collected by a sampler with a specified penetration curve yielding an upper 50% cut-point of 10 µm aerodynamic diameter (US EPA, 1987a). This change in the NAAQS was motivated by the desire to regulate the inhalable fraction of airborne particles that can be deposited in the lower regions of the human respiratory tract. Although the first reference method for PM_{10} was essentially an updated version of the Hi-Vol sampler with size-selective inlets, a number of other sampler types were eventually designated as reference or equivalent methods for sampling PM_{10} and determining NAAQS compliance (Chow, 1995). In 1997, US EPA again changed the nature of the particulate matter NAAQS, retaining the PM_{10} standard but also adding new $PM_{2.5}$ standards to address fine particles. Although a variety of PM_{10} and $PM_{2.5}$ samplers are currently used to collect samples for lead determination, these samplers are not discussed in any detail in this section since the lead NAAQS continues to be for total suspended particulate (TSP), as collected by a Hi-Vol sampler. Later in this section, we discuss the development and use of size-selective samplers since, although they may not be required for regulatory lead sampling, they have provided important information characterizing lead emission sources and the fate and transport of lead particulates.

Despite heavy reliance on the Hi-Vol for total mass sampling, other particulate samplers also gained usage for providing other types of information not available from the Hi-Vol, such as near-continuous measures of airborne particle levels and particle size distributions. First developed in the late 1940s with widespread usage in the 1950s and 1960s (Longhurst, 2005; US EPA, 1977c), the AISI spot tape sampler was used for near-continuous particulate sampling over 2 to 4 hr periods. Although typically used as an indirect measure of particulate matter, commonly referred to as the soiling index or coefficient of haze, the AISI sampler was used for lead determination in some studies including a 1960s survey of airborne lead in Cincinnati, Philadelphia, and Los Angeles (Diggs *et al.*, 1963; Ludwig *et al.*, 1965). The AISI sampler was also used in the early 1970s to determine diurnal airborne lead concentrations in Chicago and Washington, although the study authors reported tape filter contamination with metals and low collection efficiencies for the paper tape filters relative to the glass fiber filters used in NASN Hi-Vol samplers (MacLeod and Lee, 1973). Near-continuous methods have continued to evolve, with several gaining federal equivalent status for PM_{10} in the early 1990s, including the Rupprecht & Patashnick Tapered Element Oscillating Microbalance (TEOM) PM_{10} Monitor, the Andersen Instruments PM_{10} Beta Attenuation Monitor, and the Wedding & Associates PM_{10} Beta Gauge (US EPA, 2005a).[8]

Size-selective samplers have been used since the 1970s to provide particle size distribution information for ambient particles, including by studies characterizing lead emission sources at primary lead smelters and the fate and transport of lead particulates. Modern size-selective samplers were developed in the 1940s with the earliest cascade impactor, but they did not gain widespread usage in ambient particulate matter studies until the 1960s and 1970s following the commercialization of the Andersen cascade impactor (Marple, 2004). Through its use in the NASN cascade impactor network in the early 1970s, the Andersen cascade impactor provided some of the earliest size distribution measurements (in 6 size ranges) for trace metal components including lead (Lee *et al.*, 1972). Early in this program, Lee and Goranson (1972) reported sample losses related to fitting the filter properly in the filter holder, flow rate variability, and system leaks. Although these sample loss problems were largely addressed as operators gained experience (Lee and Goranson, 1972), particle bounce and re-entrainment has been a persistent source of sample loss for these samplers since their development (US EPA, 1986a; Chow, 1995). In addition, some studies have shown a bias towards small particles due to fracturing of

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[8] Federal equivalent methods for criteria pollutants such as lead, PM_{10}, and $PM_{2.5}$ are established in accordance with Title 40, Part 53 of the Code of Federal Regulations (40 CFR Part 53). Federal equivalent methods must meet rigorous requirements for adequacy. US EPA maintains a listing of designated reference and equivalent methods (US EPA, 2005a).

larger particles at impaction (US EPA, 1982). Despite its limitations, the cascade impactor is still used today for collection of particle size distribution data.

The dichotomous sampler also gained usage as a size-selective sampler in the 1970s for collection of particles in 2 size ranges, with a typical particle diameter demarcation between 2.0 and 3.5 microns (US EPA, 1977c). Following research indicating that atmospheric particles commonly occur in 2 distinct modes, often referred to as "fine" and "coarse," the dichotomous sampler was developed in the 1970s using a "virtual" impaction technique that eliminated the particle bounce issue commonly experienced with cascade impactors (US EPA, 1977c; US EPA, 1986a). Dzubay and Stevens (1975) published one of the earliest studies where the dichotomous sampler was used to collect samples for lead determination, showing that at least 75% of the lead for ambient samples collected in St. Louis was contained within particles less than 2 μm in diameter.

Currently, state-of-the-art near-continuous instruments are available for measurement of particle size distributions (US EPA, 2004).

Chemical Analysis of Particles

Air particulates were originally analyzed by "wet chemistry" (Table 3), while today, AAS (Atomic Absorption Spectrophotometry) and ICP (Inductively Coupled Argon Plasma emission spectrometry) are accurate methods of choice (US EPA, 1998a).

For example, colorimetric analysis for lead was originally based on a reagent method using dithizone from the 1940s until the 1970s (Skogerboe *et al.*, 1977). This method was sensitive and accurate but was complex, costly, and required a skilled analyst (US EPA, 1977c; Skogerboe *et al.*, 1977). This method persisted to the 1970s (NRC,1972; Ludwig *et al*.1965).

Optical emission spectroscopy (OES) offered some improvements for lead in air particulates starting in the 1950s (PHS, 1958; US EPA, 1976d; 1977c; Lee *et al.*, 1972), and was a precursor to today's ICP spectroscopy,[9] relying on an electric arc or high-voltage flame as an emission source rather than a plasma flame. Prior to 1960, emission spectrometers were commonly found only in specialized

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[9] Robinson (1978) noted that the earliest uses of a plasma as the excitation source in atomic emission spectroscopy date back to the early 1960s, although detection limits at this time were extremely high.

trace-detection laboratories since they could be used for simultaneous analysis of a large number of elements (US EPA, 1977c).

Although OES was considered a rapid and practical technique for particulate lead determination, it had some key shortcomings, including lower precision and sensitivity than alternative techniques (US EPA, 1976d; Skogerboe *et al*., 1977). Scott *et al*. (1976) reported a method detection limit of 0.15 μg/m^3 for OES analysis of particulate lead samples, while US EPA (1976d) reported lower analytical discrimination values of approximately 0.02 μg/m^3 for OES analysis of NASN samples. US EPA (1976d) reported that the OES analysis of the 1970-1974 NASN samples was not "considered to be as sensitive or precise a method as a specific determination method for a single element, such as in the case of determination of lead by atomic absorption," useful for relative order but not accurate concentrations. In addition, OES required that samples be in solution, and prior to 1966, this was done using a high-temperature muffle furnace for NASN samples. Potential losses of the more volatile metals, including lead, as a result of the high-temperature ashing were later reported to be a problem for the analysis of NASN samples prior to 1966 (US EPA, 1976d; 1977c).

Atomic absorption spectroscopy (AAS) eventually replaced OES and colorimetric dithizone for rapid, accurate, and sensitive particulate lead determination.[10] Although AAS originated in academic labs in 1955 (Skogerboe *et al*., 1977), it was not used for routine lead analysis until the late 1960s and early 1970s. Chakrabarti *et al*. (1966) is among the earliest studies employing AAS for lead determination in atmospheric particulate samples. Following this study, a growing number of air pollution studies employed AAS for lead determination, including Burnham *et al*. (1969), Kneip *et al*. (1970), Ranweiler and Moyers (1974), and Scott *et al*. (1976). This method was recommended as a reliable, sensitive, and interference-free method for airborne lead measurement in the National Academy of Sciences report on lead in 1972 (NRC, 1972), and in 1975 it was named a tentative ASTM method for testing of particulate lead in the atmosphere (US EPA, 1977c).

Although commonly used, AAS has inherent uncertainties including background absorption, chemical interferences, background light losses, and matrix effects (Skogerboe, 1974; US EPA, 1977c). In addition, some early studies showed that acid digests of filters, followed by AAS, yielded low results

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[10] Traditional AAS employs a flame to vaporize lead atoms in a sample. Nonflame (graphite furnace) AAS employs a graphite furnace rather than a flame to vaporize trace metal atoms. Nonflame AAS gained commercial usage in the 1970s (Skogerboe *et al*., 1977), and as discussed later in this section, is currently a federal equivalent method for lead determination in suspended particulate matter samples in support of the lead NAAQS.

due to incomplete digestion of particles (Skogerboe, 1974). Refinements to the AAS method were made in the 1970s to address some of these issues, and in a large mid-1970s study where approximately 1,000 NASN samples were analyzed using both AAS and OES, Scott *et al*. (1976) demonstrated the superior performance of AAS, including no discernible interference problems, a low method detection limit (0.1 μg/m^3), and low lead contamination of glass fiber filters. In the 1970s and 1980s, commercial AAS instruments became available and more widely used for rapid particulate lead determination, although US EPA (1986b) reported that there remained a persistent problem of sampling and laboratory contamination.

In 1978, AAS was promulgated as the federal reference method for lead determination in suspended particulate samples in support of the lead NAAQS (US EPA, 1978a). Detection limits for AAS are currently on the order of 0.002 μg/m^3 (US EPA, 1999a). AAS currently remains the federal reference method for particulate lead determination, although a number of other methods have achieved federal equivalent status. Federal equivalent methods for particulate lead determination currently include: nonflame GFAA spectrometry, X-ray fluorescence spectrometry (XRF), inductively coupled argon plasma-optical emission spectrometry (ICP-OES), inductively coupled argon plasma-atomic emission spectrometry (ICP-AES), and inductively coupled argon plasma-mass spectrometry (ICP/MS) (US EPA, 2005a).

These newer methods offer some unique advantages compared to AAS, but also some notable disadvantages (US EPA, 1999a). Some of these methods such as GFAA and ICP/MS have increased sensitivity for lead compared to AAS (detection limits for lead are approximately 0.00005 and 0.00001 μg/m^3 for GFAA and ICP/MS, respectively), but they both require greater operator skill (US EPA, 1999a). The ICP-based methods are the most expensive, and can be affected by interferences. XRF offers the advantage of being non-destructive and requiring minimal sample preparation compared to AAS; however, it is not very sensitive for single elements, and it can have problems with matrix offsets and background impurities. Overall, AAS remains a well-proven method for rapid analysis of particulate lead, but there are a number of additional methods currently available for rapid, accurate, and precise measurement of particulate lead.

Air Monitoring

Following the 1978 promulgation of the 1.5 μg/m^3 lead, EPA required the States to establish and operate monitoring networks for ambient lead. Despite this requirement, little guidance was available at the time regarding site selection criteria for monitors and how to set up a monitoring network. As noted in the 1977 *Air Quality Criteria for Lead* (US EPA, 1977c), "Much remains to be done toward establishing criteria for location of samplers." As discussed below, guidance and regulations regarding ambient air surveillance of lead were slow to develop, resulting in uncertainty regarding where to site monitors and what types of data to collect.

In the 1978 Federal Register notice promulgating the lead NAAQS, US EPA called for at least 2 permanent lead ambient air quality monitoring stations to be established in each urbanized area that: (1) had a 1970 population greater than 500,000; or that (2) had a lead air quality exceeding the 1.5 $\mu g/m^3$ quarterly average since 1974 (US EPA, 1978a). Further, it specified that at least one of the monitors be a roadway type monitor (located adjacent to a major roadway) and that at least one be a 'neighborhood site' monitor, in accordance with US EPA's *Supplemental Guidelines for Lead Implementation Plans* (US EPA, 1978b). This ruling did not specify specific sampling requirements in the vicinity of major lead point sources; instead it required SIPs to contain a "demonstration" that the standard would be met and maintained in areas in the vicinity of several specific point sources including primary lead smelters.

On May 10, 1979, a final ruling was published in the Federal Register that provided a more detailed set of requirements for air quality monitoring for all criteria pollutants, except for lead (US EPA, 1979b), which was still under development. This was defined as the State and Local Air Monitoring Stations (SLAMS) network. A National Air Monitoring Stations (NAMS) network was also defined in this ruling as a subset of the SLAMS network for US EPA's determination of national trends in air pollution.[11] The number of SLAMS in any Region was determined by the meteorology, population density, and emission density on a case-by-case basis, depending on SIP needs, while a minimum number of NAMS was defined.

US EPA's May 1979 ruling indicated that the agency had "reevaluated the need for specific regulations for lead monitoring around point sources" and concluded that the SLAMS network would satisfy the requirements (US EPA, 1979b). Furthermore, until lead monitoring regulations, siting criteria, and quality assurance requirements were issued, the ruling specified that States follow the draft guidelines for lead monitoring in the vicinity of point sources, which were later finalized until 1981 (US EPA,

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[11] Other types of stations were also defined. These stations included Special Purpose Monitors (SPMs), which the State could use to monitor around point sources or for special projects (without following the requirements set forth for SLAMS stations, unless they were used for support of SIPs); and the Prevention of Significant Deterioration stations or PSDs, generally operated for short periods of time by a source owner or operator (a SLAMS could also serve as a PSD).

1981a). States could also require point sources to conduct specific/targeted monitoring following the long-term monitoring guidelines (US EPA, 1979b).

On September 3, 1981, US EPA promulgated specific requirements for lead monitoring (US EPA, 1981b). The largest distinction between the lead guidelines and earlier guidelines for other criteria pollutants was the emphasis on mobile source monitoring for lead. Thus, 1 of the 2 stations required as a minimum in a NAMS lead network was to target roadway emissions. The second station was to aim at a larger geographical area or "neighborhood scale," typically located in a residential area with a high population and high traffic density.[12] This latter monitor would represent neighborhood scale impacts of point sources if they existed (US EPA, 1981b).

In a related action, US EPA finalized its "Guideline for Lead Monitoring in the Vicinity of Point Sources" in 1981 (US EPA, 1981a). This guideline required operator or State monitoring near point sources such as lead smelters, independent from NAMS or SLAMS urban site monitoring. Requirements included a minimum of 1 year of lead monitoring data to determine compliance with NAAQS, and as a secondary objective, to determine the contributions from fugitive emissions (US EPA, 1981a). One year of coincident meteorological data to provide information on the adequate placement of monitors was also required (US EPA, 1981a). The number of monitors would be determined by site-specific factors, *e.g.* meteorological conditions, terrain, and other nearby sources; dispersion modeling was allowed to help design a network based on emissions, wind speed, and wind direction (US EPA, 1981a). US EPA's minimum recommended network included 1 upwind background station, 2 monitors sited to capture the maximum concentration impacts of stack emissions, and 2 fugitive monitors (one close to the fenceline and 1 < 1 mile downwind).

Lead monitoring requirements were revised in November 1997 to account for the decline in leaded gasoline usage (US EPA, 1997a), but US EPA withdrew these revisions in December 1997 (US EPA, 1997b) due to technical criticisms. US EPA further revised and finalized the requirements in January 1999 (US EPA, 1999b), allowing the discontinuation of many lead SLAMS stations in urban areas along major roadways while maintaining a focus on stationary point sources.

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[12] Spatial scales for monitors were defined in this ruling and included a Microscale monitor, for street canyons and traffic corridors, where point sources may impact under "fumigation conditions;" a Middle scale monitor for areas several blocks in size, which was the typical scale for measuring point source impacts; a Neighborhood scale, an area with uniform land use patterns (*e.g.* residential area) from 0.5 to 4 km, often used for measuring impacts on children; an Urban scale monitor for areas 4 to 50 km to address city-wide air quality; and a Regional Scale monitor for areas greater than 50 km, typically used to measure background levels. A spatial scale representativeness was typically assigned to each monitoring station.

With US EPA's 1999 revisions, a minimum of 2 SLAMS stations (more in complex areas) was now required in areas exceeding the lead NAAQS in any of the most recent 8 quarters (US EPA, 1999b). With the discontinuation of many SLAMS lead monitoring stations, NAMS requirements were modified to require only 1 NAMS station at the maximum concentration site in 1 of the 2 most populated cities within each US EPA region, and 1 station in any city/county with NAAQS violations over the last 8 quarters (US EPA, 1999b). The focus of the revised NAMS network was on area-wide lead emissions rather than on stationary sources.

US EPA (1997c) established guidelines for States to establish monitoring networks around point sources to maximize the probability of capturing maximum lead concentrations, with the minimum number of monitors and at a minimum cost. The guidelines addressed source data collection, topography, land use, meteorology, and air modeling considerations. Requirements included a minimum of 2 monitors near the locations of maximum impact of point sources based on modeling and/or previous monitoring data (US EPA, 1997c). Furthermore, if both stack emissions and fugitive emission impacts were anticipated, 1 monitor was to be used for stack impacts and the other for fugitive emission impacts (US EPA, 1997c). A primary lead smelter case study was included in the guidance, as was emphasis on the use of site-specific information to determine both the number and the location of monitoring sites around any given stationary source to capture the maximum lead concentrations (US EPA, 1997c).

Air Modeling

Air quality simulation modeling predicts the geographic distribution of air concentrations caused by a specific emission source or combination of sources. Usually, the process of air modeling requires 2 major steps: (1) the estimation of the emission rate of a pollutant from a specific source (normally expressed in units of mass/time, *e.g.*, g/sec); and (2) the prediction of a time-averaged air concentration of a pollutant (normally expressed in units of mass/volume, *e.g.*, $\mu g/m^3$) resulting from the transport and dispersion of the pollutant emissions from the source to a given location. Figure 1 presents a timeline of important air modeling events since the 1940s that are discussed in this section.

There are many different types of air quality simulation models applicable for different distance scales and pollutants. For example, long-range transport models can track pollutants traveling for thousands of kilometers from a single source. Urban and regional scale photochemical models are used for ozone and other secondary pollutants (*e.g.*, sulfates and nitrates), and they incorporate hundreds of chemical reactions.

The Gaussian Plume Model

Atmospheric turbulence is difficult to simulate by a simple mathematical formulation because of the randomness of turbulent fluctuations in the atmosphere and the wide size range of turbulent eddies affecting dispersion. Nonetheless, driven partly by the use of mustard gas in World War I and the fear of further use of chemical warfare, a simple formulation, easily calculable by hand, was developed in the 1930s and 1940s to predict the air concentrations resulting from pollutant sources (Bosanquet and Pearson, 1936; Sutton, 1947). This mathematical technique is a relatively simple equation known as the Gaussian plume dispersion model, because the spread of a plume from a point source is approximated as a Gaussian (*i.e.*, bell-shaped) distribution in both the horizontal and vertical directions. Sutton (1947) described this equation almost exactly as used by current US EPA air dispersion models. Interestingly, even in the 1940s, a "factor of two" accuracy was claimed when comparing Gaussian plume predictions with limited experimental data (Sutton, 1947).

In the 1950s and 1960s, air modeling using a Gaussian plume formulation became more standardized through the use of dispersion coefficients based on experimental data. An important experimental program to study atmospheric dispersion was performed in 1956 over relatively flat terrain in Nebraska. This comprehensive program of over 70 tracer experiments (using SO_2 as a tracer gas) with detailed meteorological measurements became known as Project Prairie Grass, and the data from this program resulted in the first useful set of dispersion coefficients for air modeling purposes.

Although it was obvious from the Prairie Grass data that greater dispersion occurred during times of greater atmospheric turbulence, the problem was how to account for differences in turbulence or atmospheric stability in a practical manner that could be used in an air dispersion model. The key breakthrough has been credited to Pasquill (1961), who developed a set of discrete atmospheric stability classes based on 2 easily observable meteorological parameters: insolation and wind speed. The development of these discrete atmospheric stability classes, normally designated as a set of 6 classes (A through F, from very unstable to very stable), was a key factor in the practical and widespread use of Gaussian plume dispersion models.

Pasquill (1961) expressed dispersion coefficients based on the Prairie Grass data in the form of plume angular spread, as a function of discrete atmospheric stability class. Gifford (1961) converted Pasquill's angular spreads into a set of plume standard deviations in the horizontal and vertical directions (σ_y, σ_z) that could be directly input into the Gaussian plume equations. Turner (1961) expressed Pasquill's stability classes in terms of standard hourly airport observations, and published a classic document in 1967 called "Workbook of Atmospheric Dispersion Estimates" (Turner, 1967), which presented practical Gaussian plume equations and a set of dispersion coefficient graphs of σ_y and σ_z as a function of downwind distance for Pasquill's 6 atmospheric stability classes. These dispersion coefficient graphs have become known collectively either as the Pasquill-Gifford (PG) or the Pasquill, Gifford, Turner (PGT) dispersion curves, and other experimental dispersion programs conducted since the 1960s have verified the accuracy of these dispersion curves. The PG or PGT dispersion coefficients, based on the 1956 Prairie Grass experiments, have been used with only minor changes by most Gaussian plume models developed and recommended for use by US EPA from the 1970s to the present.[13]

EPA Regulatory Air Models

The history of US EPA regulatory air modeling began in 1970, and Irwin (2002) recently published an excellent reference document presenting a detailed evolution of US EPA air modeling. In 1970, 2 milestone regulatory developments occurred: (1) the US EPA was formed from parts of several other Federal agencies, and (2) the 1970 amendments to the Clean Air Act were passed, which specified the use of air dispersion modeling to predict the impact of new sources, and to verify that existing sources were not causing violations of the newly promulgated air pollution standards, known as the NAAQS, for criteria pollutants including total suspended particulates (TSP) and sulfur dioxide (SO_2).

The regulatory driving force of the 1970 Clean Air Act amendments created a new business enterprise -- air pollution consulting. During the 1970s, a number of small environmental consulting firms were formed to satisfy the new need for regulatory air modeling. The consultants were primarily meteorologists, who realized that their knowledge of atmospheric turbulence and dispersion theory could be put to practical use. Since US EPA funded a reasonable amount of academic research in atmospheric dispersion starting in the early 1970s, the first college courses devoted specifically to air pollution

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[13] In November 2005, US EPA recommended the AERMOD model to replace ISCST3 as a preferred short-range dispersion model (US EPA, 2005b). AERMOD does not use the discrete Pasquill stability classes, but calculates dispersion coefficients based on a continuous atmospheric stability formulation.

problems began in the mid-1970s, leading to the evolution of entire college programs in atmospheric science.

Since there were no standardized computer air models available in the early 1970s, air modeling at that time was usually done by hand calculations of Gaussian plume equations, which were generally based on the *Workbook of Atmospheric Dispersion Estimates* (Turner, 1967). A number of consultants developed proprietary dispersion models, which ran on the large mainframe computers of the time. Because there were no standard air models, and different consultants developed their own proprietary air models, US EPA and state regulatory agencies found it difficult to judge the accuracy and consistency of air modeling studies submitted to satisfy air regulations.

To partially alleviate this lack of standardization, US EPA developed a number of simple computer models in the early 1970s, based on the Gaussian plume model equations and the dispersion curves presented in Turner (1967). The equations for calculating the plume rise caused by the buoyancy and momentum of a hot point source (*e.g.*, a power plant stack) were based on the classic review of plume rise equations by Briggs (1969). These early models were designated as the UNAMAP (User's Network for Applied Modeling of Air Pollution) set of models, and were originally only available in an interactive mode through a teletype connection to US EPA in North Carolina. For more widespread use, the National Technical Information Service (NTIS) distributed the UNAMAP models on computer tapes to interested users in the 1970s and 1980s. With the advent of the Internet c. 1990, the US EPA established an air modeling web site, *www.epa.gov/ttn/scram*, which provides free downloads of recommended air models and meteorological data.

The earliest US EPA models in the UNAMAP series in the early 1970s were simple FORTRAN computer programs that provided an alternative to hand calculations of Gaussian plume equations for a point source, and were called PTMAX (Point Source: Maximum), PTDIS (Point Source: Dispersion), and PTMTP (Point Source: Multiple Point Sources). Although relatively simple, these programs began a trend toward air model standardization. In 1977, US EPA developed a more sophisticated point source model called CRSTER[14] that included, for the first time, the effect of complex terrain interactions on an elevated point source.

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[14] Because US EPA wanted to quickly develop a sophisticated point source model, the model development effort became known as the "crash (CRS)" program for its tight deadlines (Irwin, 2002). When the model was adapted to include terrain, the model name was coined, CRSTER. Irwin (2002) provides a number of similar insights into how air models were named in his historical review.

To provide greater model standardization and to assure consistent application of air models, US EPA in 1978 published the first edition of "Guideline on Air Quality Models" (US EPA, 1978c), which specified "preferred" air models, like CRSTER, and provided guidance on meteorological and model input parameters. The "Guideline on Air Quality Models" has been updated frequently since 1978, and has resulted in widespread acceptance and standard use of US EPA's "preferred" air models, as well as the use of consistent meteorological and model input parameters, to provide a large measure of standardization in regulatory air modeling.

The Industrial Source Complex (ISC) Model

In 1979, US EPA added the simulation of aerodynamic building wake effects or "building downwash," based on experimental wind tunnel studies, to the CRSTER model, which became the first version of the ISC model (US EPA, 1979c). The ISC model quickly became popular because of its ability to handle terrain, building downwash, and any combination of multiple point, area, and volume sources. Because of computer limitations at the time, 2 ISC models were developed by US EPA: the detailed ISCST (Industrial Source Complex, Short Term) version, which used hourly meteorology to calculate short-term (1-hour, 3-hour, 8-hour, and 24-hour) averages of air concentrations; and the simpler ISCLT (Industrial Source Complex, Long Term) version, which used long-term averaged meteorology to calculate annual and quarterly averages.[15] These ISC models have been designated as "preferred" models in the various releases of US EPA's "Guideline on Air Quality Models" since 1979, and they have become the air models used for almost all regulatory air modeling of industrial sources over the last 25 years.

The original ISCST model in 1979 calculated deposition of particles by a simple gravitational settling velocity as a function of particle size, and used an empirical "reflection coefficient," which ranged from 0 to 1 depending on particle size, to estimate what fraction of the particles deposited when they contacted ground surface. This model formulation was designed for relatively large particles (greater than about 20 µm), which are affected primarily by gravitational settling. The area source algorithm used in ISCST was based on a finite line source technique, which was shown in a later study to predict unrealistic concentrations for receptors located within or near the area source (US EPA, 1989).

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[15] Although the ISCST model can also calculate annual averages, the long computer run times in the 1980s necessary to calculate hourly concentrations over an entire year (*i.e.*, 8760 hours) favored the use of the simpler and faster ISCLT model, which used an annual or quarterly frequency of wind speeds, directions, and stability classes (known as STAR data). With the advent of faster computers, the ISCLT model has become obsolete, and the ISCST model (as the ISCST3 version) has been used almost exclusively for all air modeling since 1996.

Because of inefficiencies in the FORTRAN programming of ISCST, a major overhaul of the programming in ISCST occurred in 1992, when the FORTRAN code was modified significantly to make the code more modular. The re-coded model was designated as ISCST2 (US EPA, 1992a). However, the basic deposition and area source algorithms were unchanged from the original ISCST model.

Because of the known deficiencies in the ISCST and ISCST2 area source calculations, an improved integrated area source algorithm was implemented into ISCST2, and released to the public in draft form as the AREA-ST model. Concurrently, development progressed on improved dry deposition algorithms that could more accurately model small particles (less than 10 μm), a new wet deposition technique, and an improved plume depletion technique that accounted for all particles that had deposited on surfaces. These techniques were independently implemented into ISCST2 and were released to the public in draft form as the DEPST model. The AREA-ST model and the DEPST model were then combined into a single draft model called ISC-COMPDEP, which in 1995 was released as the ISCST3 model (US EPA, 1995a).

Thus, it was not until 1995 that the ISC model was sufficiently improved to accurately predict air concentrations and surface deposition of fugitive area sources that release relatively small particles. Since 1995, only minor changes to the ISCST3 model have been made, including the addition of gas-phase deposition on surfaces in 1999. In November 2005, US EPA replaced the ISCST3 model with the AERMOD model as the preferred short-range air dispersion model, but allowed use of the ISCST3 model for regulatory air modeling until November 2006 (US EPA, 2005b).

Because of its widespread usage, a number of validation studies of the ISC model have been performed, in both rural and urban locations, over the last 25 years (*e.g.*, Bowers and Anderson, 1981; Schulman and Hanna, 1986; Riswadkar and Kumar, 1994; Kumar *et al*., 1999). The Kumar *et al*. (1999) study compared the ISCST3 model with air monitoring data for long-term predictions of SO_2 from 123 industrial stacks in Lucas County, Ohio. The results depended somewhat on the statistical measures used for comparison, but in general, the ISCST3 model consistently predicted within a "factor of two" of the observed values. Since Sutton (1947) claimed the same accuracy for the basic Gaussian plume model, it appears that a "factor of two" may be the limit of accuracy expected from the simplification of complex atmospheric turbulent processes by a Gaussian plume equation.

Emission Estimation

The overall accuracy of air concentrations predicted by a dispersion model is dependent on the accuracy of the emission rates input to that model. Since the air concentrations predicted by a Gaussian plume model are linearly proportional to the emission rate of a specific source, any errors in the source emission rate are linearly propagated to the predicted air concentrations. Similar to the evolution of air dispersion modeling techniques, source emission estimation techniques have improved with time. As a specific example, the evolution of lead emissions to the air from primary lead smelters is discussed in this section.

Like air quality dispersion models, emission estimation techniques for a large number of industrial sources have become standardized by US EPA over the years. The equivalent US EPA document to "Guideline on Air Quality Models" that describes standardized emission estimation techniques is entitled "Compilation of Air Pollutant Emission Factors," which has been published and continually updated by US EPA since 1972 (US EPA, 1972a). This document began in 2 earlier Public Health Service emission documents (PHS, 1965; PHS, 1968) published before the US EPA was formed in 1970. Because of its widespread use for recommended emission factors for a wide variety of industrial sources, this document has become known simply by its original Public Health Service document number, "AP-42."

The AP-42 document has evolved through 5 editions since 1972, with many improvements added to a specific edition either by replacement pages or new appendices. Figure 2 tracks the changes through the years of the AP-42 document specifically for particulate lead emissions to the air from primary lead smelters.

Since the first NAAQS air lead standard of 1.5 $\mu g/m^3$ for a quarterly average was not promulgated until 1978, the first 2 editions of AP-42 (US EPA, 1972a; 1973a) and the original third edition (US EPA, 1977d) did not include particulate lead emission rates to the air for any industrial sources. For primary lead smelters, only total particulates (TSP) and SO_2 emissions for point sources were provided in the first 2 editions and the original third edition of the AP-42 document.

In 1979, Supplement No. 9 of the third edition of AP-42 provided, for the first time, air lead emission factors for a number of industrial sources, including primary lead smelters (US EPA, 1979d).

Emission factors were presented, in terms of lb lead emitted to the air per ton of lead produced, for 4 process-related point sources at primary lead smelters, but no fugitive sources were mentioned. In 1980, Supplement No. 10 of the third edition of AP-42 provided 2 major additions relating to primary lead smelters: (1) limited particle size information was provided for point sources only; and (2) for the first time, particulate emission factors for 15 different types of fugitive sources at primary lead smelters were included (US EPA, 1980a). No air lead emission factors were provided for these fugitive sources, and the particulate emission factors were assigned the lowest certainty rating of E to reflect the poor quality and limited amount of underlying data used to derive the emission factor.

In 1986, Supplement A to the fourth version of AP-42 provided, for the first time, air lead emission factors for 5 fugitive emission sources as well as particle size distribution data for these fugitive sources (US EPA, 1986b). These air lead emission factors for fugitive sources were assigned the second lowest certainty rating of D, indicating their low reliability for specific sources within the source category population. Finally, the fifth version of AP-42 in 1995 combined and simplified many of the fugitive emission sources at primary lead smelters (US EPA, 1995b). In addition, their emission factor ratings were reduced to the lowest certainty rating of E. There have been no changes to the primary lead smelting emission section of AP-42 since 1995.

Control Technologies

Control technologies evolved concurrently with measurement and modeling developments. Early control efforts were motivated not only by pollution concerns but also by desires to improve byproduct recovery of useful byproducts (*e.g.*, rare metals, arsenic, sulfur dioxide) (Swain, 1939). Although the general principles underlying many of the primary particulate control technologies have been known for a century or more, technological advances and experience were required to improve the engineering of control efficiencies and to apply them to a larger variety of industrial processes.

Point Source Controls

The 4 primary particulate control devices developed in the 20th century (Table 4) are cyclones (centrifugal separators), electrostatic precipitators (ESPs), wet scrubbers, and baghouses (fabric filters) (NAPCA, 1969b; US EPA, 1977e; Cooper and Alley, 1994; EC/R, 1998). Each of these control technologies has been widely used at primary lead smelters, except for cyclones (US EPA, 1977e; 1980b), with ESPs and baghouses being the most prevalent (Stern, 1962; NAPCA, 1969b; US EPA, 1977e; 1980b; 1995c; Schupp, 2000).

Cyclones were used in industry for gas particulate cleaning by at least the 1890s, gaining commonplace usage during the 20th century (Zimmerman, 1933; Falk, 1955; Hardison, 1966; Cooper and Alley, 1994). In 1939, C.B. Shepherd and C.E. Lapple observed: "Centrifugal or cyclone collectors are widely used for the separation and recovery of industrial dusts from air or process gases. The usual type of cyclone is simple to construct and is very low in first costs compared with other types of dust collecting equipment" (Cooper and Alley, 1994). Falk (1955) described the use of multiple cyclones in series, while Hardison (1966) noted the use of cyclones as pre-cleaners ahead of other equipment. Although cyclones have not been common at primary lead smelters, US EPA (1980) reported the use of cyclones for dust control during milling and concentrating of lead ores prior to primary lead smelting.

In 1906, Dr. Frederick Cottrell invented ESPs for collection of sulfuric acid mists/fumes. Shortly following this invention, a large ESP for solids collection was installed in 1910 at the Anaconda, Montana copper smelter (Swain, 1949). ESPs gained widespread usage at industrial facilities by the 1930s (Green, 1931; Zimmerman, 1933; Swain, 1939), and revolutionized recovery of acid mists and metals at nonferrous smelters (Swain, 1939). NAPCA (1969b) stated: "The high-voltage electrostatic precipitator (ESP) is used at more large installations than any other type of high-efficiency particulate matter collector…High-voltage single-stage precipitators have been used successfully to collect both solid and liquid particulate matter from smelters, steel furnaces, petroleum refineries, cement kilns, acid plants, and many other operations" (NAPCA, 1969b, p. 81).

ESP improvements over the years have allowed increased collection efficiencies and wider application in a variety of industries, including primary lead smelting, particularly for treatment of sinter machine off-gases (Stern, 1962; NAPCA, 1969b; US EPA, 1977e; 1980b; 1995c; Schupp, 2000). ESP efficiencies approached 95% by the 1930s and 1940s (Cooper and Alley, 1994), 98% by the early 1950s (Anon., 1953), 99.5+% by the mid-1970s, and 99.9+% by the 1990s (Cooper and Alley, 1994). Although the ESP has been eclipsed by the baghouse as the dominant particulate control device at US primary lead smelters, it remained a widely used control technology at US primary lead smelters.

Wet scrubbers (wet collectors) in various forms have included simple spray chambers, centrifugal spray scrubbers, venturi scrubbers, impingement plate scrubbers, packed bed scrubbers, and mist eliminators (for removal of mists). Common use of wet scrubbers in industry was noted by the early 1930s (Zimmerman, 1933), and by the late 1940s, the development of the venturi scrubber was a key advance for efficient removal of fine dusts, mists, and fumes (Jones, 1949; Munger, 1951). With efficiencies of 98 to 99.8% reported for sulfuric acid mists in 1949 (Jones, 1949), 99.5% overall and 99.0% efficiency for 0-5 μm particles (Bond and Straub, 1972), venturi scrubbers are among the most efficient devices. Use of wet scrubbers at primary lead smelters was commonplace by the 1960s (Stern, 1962) in sulfuric acid "plants" for conversion of SO_2 to sulfuric acid (McKee, 1969; US EPA, 1980b; Wixson, 1982).

Baghouses (fabric filter bags) were used by industry since 1900.[16] Austin (1907) noted that "[a]t the American Zinc Works, near Canyon City, Colorado, the company has for years filtered all the gases from their roasters [sintering machines] and blast-furnaces…[after] having been filtered at the bag-house, there is no smoke visible. This is also true of the bag-house installed in connection with lead-smelting plants." Austin further noted that wool bags, not cotton, were required for filtering corrosive, sulfuric acid-containing roaster gases. Ebaugh (1910) observed that "gases coming from lead blast furnaces and from zinc plants had been filtered successfully for many years," and that a Utah lead smelter began baghouse treatment of roaster and blast furnace gases in 1908 in response to a 1906 court injunction. The US Bureau of Mines (1954) highlighted the successful use of a wool baghouse for blast furnace solids collection at the Selby lead smelter in California in 1908, while the Selby Smelter Commission (1915) observed that a baghouse was in use at the Selby lead smelter in 1914 for elimination of visible smoke from the roaster stack. Schupp (2000) noted the installation of baghouses at the Herculaneum, Missouri primary lead smelter in 1910. Stern (1962) stated that "filtration is the oldest and generally the most reliable of the many methods by which dusts, mists, and fumes may be removed from gases."

Although baghouses were already widely used in industry by the early 1930s (Green, 1931; Zimmerman, 1933), technological improvements such as the use of synthetic fiber bags with greater chemical and thermal resistance (Falk, 1955; NAPCA, 1969b) led to greatly expanded baghouse use in the 1950s and 1960s. Advances such as this were important to lead smelters because Wells (1917) noted that higher stack gas temperatures resulted in greater gas travel distances and lower ground-level gas concentrations. Despite these improvements, electrical precipitators competed with baghouses because they could withstand greater gas temperatures (Wells, 1917).

Through at least 1960, baghouses primarily used woven or felted natural fiber bags of cotton or wool, which required gas temperatures of below 250°F (Zimmerman, 1933; Falk, 1955; NAPCA, 1960) and offered only fair overall chemical resistance (NAPCA, 1969b). Bags constructed of synthetic fibers (*e.g.*, polypropylene, Nylon®, Dacron®, Teflon®), asbestos, or fiberglass first appeared at a secondary lead smelter by the late 1960s (Falk, 1955; Diamond Power International, Inc., 2001; NAPCA, 1969b). These materials offered greater chemical resistance (US EPA, 1991a) and allowed baghouse use at gas temperatures up to 550°F (NAPCA, 1969b; US EPA, 1977e). Fabrics developed by the 1990s, such as

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[16] Bags are by far the most common type of fabric filter; hence, the term "baghouse" is widely applied to fabric filtration (EC/R, 1998).

Gore-Tex®, Chem-Pro®, and ceramics, offer even greater chemical and/or thermal resistance (up to 1,000°F for ceramics) (US EPA, 1991a; EC/R, 1998). Slight increases in collection efficiencies, up to >99%, were also realized as a result of incremental improvements in baghouse technologies (Falk, 1955; NAPCA, 1960): 99.7% (including 99.5% of 0-5 μm particles) by 1965 (Bond and Straub, 1972), between 95-99.99% by the mid-1970s (US EPA, 1977e), and 99-99.9+% in the 1990s (EC/R, 1998).

From the 1960s to the present, ESPs and baghouses have been the primary particulate control devices used at primary lead smelters (Stern, 1962; NAPCA, 1969b; US EPA, 1977e; US EPA, 1980b; US EPA, 1995c; Schupp, 2000), with baghouses becoming the more widely used technology (US EPA, 1998b). US EPA (1980b) noted that all 6 of the primary lead smelters in operation at the time used baghouses for particulates from blast and dross reverberatory furnace off-gases, and 3 of the 6 smelters used baghouses to collect particulates from the weak gas stream of sinter machines. Baghouses (or equivalents) were also required at primary lead smelters by federal regulation in 1999 as part of the final National Emission Standards for Hazardous Air Pollutants (NESHAP) rule (Section 9).

Fugitive Emission Controls

By the mid-1970s it became apparent that point source controls alone would not achieve the NAAQS for particulates. As discussed in Section 2.3, US EPA then turned its attention to fugitive (non-point source) emission control (Lillis and Young, 1975; US EPA, 1977a). In 1977, US EPA published *Technical Guidance for Control of Industrial Process Fugitive Particulate Emissions* (IPFPE), where IPFPE was defined as: "Particulate matter which escapes from a defined process flow stream due to leakage, materials charging/handling, inadequate operational control, lack of reasonably available control technology (RACT), transfer, or storage" (US EPA, 1977a). US EPA (1977a), as well as other concurrent US EPA reports (US EPA, 1977e; 1979a; 1979e), summarized the state of the knowledge on available control technologies for fugitive particulate emissions (both industrial process and open source). These documents addressed fugitive emissions at lead smelters, indicating that there was much to be learned about fugitive emissions and their control.

US EPA (1977a) discussed the following options for IPFPE control at smelters: building enclosures or hoods for the sintering and refining processes; proper operation and maintenance practices (*i.e.*, sealing of furnaces and ducts, prompt clean-up of spills); hoods for smelting and drossing processes; and enclosures, hoods, or wet suppression with water, foam, or chemicals for material storage, loading, and transfer areas. Building enclosures (typically ventilated to baghouses), however, presented worker health and safety challenges and were thus not immediately used at primary lead smelters (US EPA, 1979a). US EPA (1982) stated that use of localized hoods as opposed to building enclosures was "desirable from an economic and occupational exposure standpoint," but that building enclosures (though requiring large air flows) were being increasingly considered due to their ability to capture up to 95+% of fugitive emissions, as well as space constraints associated with the use of a number of localized hoods.

Retrofitting existing plants with fugitive emissions control systems was considered difficult due to space and operational limitations (US EPA, 1982), and there have been few major advancements in other types of techniques available for control of fugitive emissions. As noted by EC/R (1998): "The most widely used methods of controlling process fugitives are local ventilation [*i.e.,* hoods] and building enclosure/evacuation…paving of unpaved roads; eliminating, reducing, or managing truck transportation; and street cleaning are the most effective techniques to reduce fugitive dust emissions from roads" (EC/R, 1998)." These techniques are in essence the same as those identified by US EPA in its 1977 reports (US EPA, 1977a; 1977e). As discussed in Section 9, control of fugitive emission sources was addressed in US EPA's 1999 NESHAP regulations for primary lead smelters.

Air Pollution Regulations and Standards

Air pollution management efforts in the United States began in the late 19th century with simple technical approaches to readily apparent and straightforward problems. Early attempts were made to control visible smoke and airborne soot from emerging fossil fuel burning industrial facilities through municipal ordinances prohibiting emission of visible pollution and limiting the locations in which such smoke-producing activities could be conducted. In contrast, air pollution management today has one of the most complex regulatory structures in the United States, tying together air pollution control agencies at the local, regional, state, multi-state and federal levels of government (often within the shadow of international treaties). This apparatus seeks to manage air quality problems, which often present the following characteristics:

- Air quality problems are often difficult to define and measure;
- Many air quality problems present complex technological challenges to develop effective control strategies;
- Measurement of the effectiveness of the available control strategies in achieving stated objectives is often uncertain and imprecise;
- The premises, goals, and strategies underlying various aspects of air quality management are subject to broadly divergent degrees of acknowledgement and commitment; and
- The degree of control appropriate to address air quality problems deemed appropriate for management engenders intense scientifically- and policy-based debates.

Because of these characteristics, the process of developing, promulgating, complying with and enforcing air quality standards and regulations is quite complex, as are the air quality controls themselves. An examination of the development of the modern air quality control apparatus reveals how difficult it has been to develop consensus goals and policies, to translate them into laws and a coherent body of regulations governing polluting activities, and to have these goals and regulations translate into effective control of pollutants. This section provides an overview of the major stages of developing the current air quality control system in the United States and conveys some of the complexities underlying the current system for controlling pollution.

Early Regulation

The origins of early American efforts to control air quality are found in the smoky conditions of late 19th and early 20th century industrial cities. In cities such as Pittsburgh, St. Louis, Cincinnati and Chicago, it was commonplace for visible pollution resulting from fuel-burning industries to combine with fog and, during thermal inversions, to create conditions referred to as "Londoners" in which darkness during daylight hours caused work stoppages, school dismissals and accidents (Grinder, 1980, p. 84). Along with the visual impairment came dirt and filth and, ultimately, impaired health for the residents of these cities and a noticeably degraded urban ecology (Grinder, 1980, pp. 85-88).

These conditions gave rise to air quality control regulations in the form of local ordinances imposing emissions controls and land use controls, both growing out of legal bases in common law. Ultimately, these local ordinances were ineffective in the face of widespread industrialization and economic growth spurred on by the needs of 2 World Wars and by increasing complexities in the nature and reach of the causes of air pollution. The local programs were largely supplanted, beginning in the mid-1950s, by the current federal-state system of air pollution control.

Initial regulatory controls were carried out primarily at the municipal level (Grinder, 1980, p. 91; Reitze, 1991)[17] in response to these localized, though often citywide, conditions. These controls initially took the form of ordinances prohibiting or ordering the abatement of the smoke nuisance, *i.e.*, emission controls, as in Cincinnati, Cleveland, Pittsburgh, St. Louis and St. Paul (Reitze, 2001, p. 10). Many of these measures were the product of grass-roots organizations such as "Anti-Smoke Leagues" and, particularly, the efforts of women's clubs and organizations (Grinder, 1980, p. 88), typical of other Progressive Era social reform movements focused on the problems of industrialization. Development of these early ordinances was also the result of the failure of traditional common law legal principles such as nuisance, negligence and trespass to address air pollution problems effectively (Reitze, 1991, pp. 1555 – 1569; Kennedy and Porter, 1955).[18]

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[17] Municipal ordinances of this type sometimes ran afoul of state constitutions' allocations of authority, as in Missouri where an early smoke nuisance ordinance in St. Louis was held to be an unconstitutional exercise of the police power reserved to the state, leading to a petition drive for permission to enact the ordinance (Anon., 1897).

[18] For example, control of smoke pollution using nuisance law was frustrated by the fact that smoke was not considered to be a nuisance under the common law and that specific harm resulting from the smoke needed to be demonstrated for smoke to be established as a nuisance in a particular instance. The enactment of municipal ordinances overcame this limitation because municipalities or states were empowered to declare activities creating smoke pollution to be "public nuisances" or to regulate the activities under the police powers of the state, which authorize protection of the public health, safety and welfare.

Many municipalities adopted smoke abatement programs beginning with the earliest in Chicago in 1881 and continuing through the turn of the century (Reitze, 2001, p. 10). There are few examples from this period of air quality programs addressing air pollutants other than "smoke" (Reitze, 2001, p. 10). By 1912, most major United States cities had enacted smoke abatement programs.[19]

Early pollution control efforts acquired a technocratic aspect, as associations of engineers also became driving forces behind early municipal smoke nuisance ordinances (Reitze, 2001, p. 10). Engineers working on smoke abatement methods focused on optimizing combustion techniques as the "solution" to the smoke problem, which they viewed as an unnecessary waste of resources ancillary to the economic progress brought about by industrialization (Grinder, 1980, p. 89). However, along with this view that technology could be the solution to the smoke problem grew the contrary view that limitations to the technological ability to control or prevent smoke nuisance should be, and was adopted as, a defense to the enforcement of smoke abatement ordinances. In other words, in the interest of not inhibiting economic prosperity, there was the belief that required abatement measures should be limited to currently available technologies (Reitze, 1991, p. 1577).

Land use ordinances and regulations through the first half of the 20th century were generally limited in scope and jurisdiction and of questionable effectiveness at the citywide level, let alone against regional and trans-boundary impacts of air pollution. Zoning laws, the most common local land use control, were used initially to impose separation between industrial or commercial land uses and residential or recreational uses (Hadacheck, 1915, as cited in Reitze, 1991, p. 12). However, zoning laws and other land use laws and policies advancing this segregation have been blamed by many[20] for creating the conditions for one of the largest current air quality challenges: the land use patterns that necessitate widespread dependence upon automobiles for access to employment opportunities and affordable housing (Reitze, 2001, p. 12). At the risk of understatement, land use controls did not play a significant role in the control of air pollution during the first half of the 20th century.

Post-WWII, many cities established nuisance-based air regulations (Reitze, 2001, p. 11). These efforts often expanded the types of air pollution sought to be controlled beyond the usual smoke control provisions to address diverse local conditions. For example, Los Angeles initiated the regulation of smudge pots used to heat citrus groves at this time (Martineau and Novello, 2004, p. 5). Some programs also strived to address air pollution problems throughout an entire air quality basin, such as the Los

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[19] "By 1912, 23 of the 28 United States cities with populations in excess of 200,000 had smoke abatement programs" (Reitze, 1991, p. 1576).

[20] See Reitze, 2001, p. 12 and sources cited therein (describing high environmental costs of zoning-driven urban sprawl).

Angeles County Air Pollution Control District (Martineau and Novello, 2004, p. 5).[21] However, these programs were usually limited by minimal budgets (Reitze, 2001, p. 11; Schueneman, 1963).[22] State involvement was limited by the prevailing practice at the time of assigning authority to local governments for land use controls. Thus, with only a few exceptions, prior to the 1960s, air quality controls were largely limited to municipalities and local governments and had little regional or state-level presence (Reitze, 1991, p. 1580). The role of states in air quality control became much more pronounced due to the series of federal legislation that has led to the current system of air pollution control.

Federal Regulation

The first federal legislation that addressed air pollution specifically was enacted in 1955 (US Congress, 1959). This law authorized the expenditure of funds over a five-year period to conduct scientific and technical research on the causes of air pollution and to assist state efforts at research and training (US Congress, 1959). Although this act was reauthorized and extended for 4 years in 1959, the funding appropriated was always less than Congress authorized because "unlike water pollution, air pollution . . . is essentially a local problem" (Eisenhower administration Bureau of the Budget commentary on the air pollution legislation, as cited in Reitze, 2001, p. 14). Responsibility for administering these funds was vested with the Public Health Service in the Department of Health, Education and Welfare (DHEW), and, after its creation in 1960, in the Division of Air Pollution.

Congress passed the Clean Air Act of 1963, which was signed into law on December 17, 1963 (US Congress, 1963). This Act contained the first provision authorizing a federal agency to take enforcement action to abate air pollution. This provision established a mechanism by which a State, a State air pollution control agency or, with the permission of the State, a governing body of a municipality could request DHEW to convene a series of conferences and hearings to determine whether it was necessary to abate air pollution if alleged to endanger the health or welfare of persons outside the state or within the state (US Congress, 1963). The Secretary of DHEW was also authorized to take enforcement action and to invite the respective state and municipal authorities to participate (US Congress, 1963). Ultimately, if DHEW determined from these conferences and hearings that the alleged pollution was occurring and that effective progress toward abatement was not being made, the matter could be referred to the Attorney General to bring suit on behalf of the United States to secure abatement of the pollution

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[21] In 1947, the California Air Pollution Control Law authorized counties to establish air pollution control districts with the authority to issue permits, promulgate rules and regulations, and take steps to penalize violators (Kennedy and Porter, 1955, pp. 854,869-76).

[22] 17 states had air pollution programs expending $5,000 or more annually; total expenditures for 1961 were $2,000,000, 57 percent of which was by California.

(US Congress, 1963). This model of federal enforcement of air pollution provisions at the request of the states, in concert with the states or in place of state enforcement action, laid a cornerstone that has carried through to the present federal-state air pollution enforcement programs.

The 1963 Act also established a joint government-industry committee to evaluate progress in the development of automotive exhaust control devices and fuels and commissioned a report to Congress to recommend whether the discharge of pollutants from automotive exhausts should be subject to regulations (US Congress, 1963). This authority was expanded in amendments to the Act passed in 1965, namely the Motor Vehicle Air Pollution Control Act which authorized the Secretary of DHEW to issue emissions standards for new motor vehicles or motor vehicle engines (US Congress, 1965).

More key building blocks of the current air pollution control program were added in 1967 with enactment of the Air Quality Act of 1967 (US Congress, 1967). This Act directed the Secretary of DHEW to make grants to air pollution control agencies for a major portion (one-half or more) of the cost of planning, developing, improving, and maintaining programs for the prevention and control of air pollution and for the implementation of air quality standards to be developed under the Act (US Congress, 1967). The Act directed the Secretary to designate AQCRs, based on jurisdictional boundaries both interstate and intrastate, in which air quality control programs would be established to facilitate adequate implementation of air quality standards. The AQCRs would be defined on the basis of those aspects of climate, meteorology and topography which affect the interchange and diffusion of pollutants in the atmosphere (US Congress, 1967). The Secretary was further directed to develop and issue AQCs, consisting of summaries of the latest scientific information on the adverse effects of air pollution agents on man and the environment and the variable factors that may alter the effects on public health and welfare (US Congress, 1967).

Designation of the AQCRs and the AQCs was the centerpiece[23] of the Act, namely Section 108, which provided for development of air quality standards to protect the public health and welfare and outlined a programmatic means for the abatement of air pollution in any state or states which endangers the health or welfare of any persons (US Congress, 1967). Section 108(c) of the Act authorized states to adopt ambient air quality standards (AAQS) for any AQCRs within their respective state and to adopt a plan for the implementation, maintenance and enforcement of the AAQS. Under Section 108(c), if the

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[23] The Act also had provisions addressing motor vehicle emissions, including a provision directing the Secretary to prescribe standards applicable to the emission of any substance from any new motor vehicle or new motor vehicle engine which contributed to air pollution endangering the health or welfare of any persons and to prohibit the sale or importation of any new motor vehicle or motor vehicle engine unless it is in conformity with these emission standards (US Congress, 1967).

Secretary determined that a state's AAQS were consistent with the federal AQCs, that the plan would assure the achievement of the AAQS within a reasonable time, and that the state had adequate means of enforcement of the standards and the plan, then the state's standards and plan would serve as the air quality standards applicable to that state (US Congress, 1967). In the absence of state-enacted AAQS or implementation plans with respect to any AQCR or portion thereof, the Secretary was authorized to promulgate standards consistent with the purposes of the Act for that AQCR, after providing ample opportunity for the state to cure its nonperformance (US Congress, 1967). Once AAQS were developed for a state of an AQCR, Section 108 provided for enforcement of the AAQS either by the state or by the Secretary under some fairly arduous procedural limitations.[24]

These early measures to establish a coordinated federal and state government effort to control air pollution set the foundation for all subsequent air regulation. These measures established a federal-state relationship in which states were encouraged to take a leadership, hands-on role in establishing the front-line regulatory programs to deal with individual sources of pollution, while federal authorities could be exercised if states inadequately regulated the local pollution sources. Nevertheless, the federal government established the broad policies and criteria upon which the actual standards for air quality were to be based. In addition, it was largely the federal responsibility to provide funding, conduct research, set policies, and oversee state programs. Within this policy framework, the states were to develop implementation plans, which became the SIPs of today, detailing how the federal precepts were to be translated into regulations tailored to actual sources, and the states were to take the initiative in enforcing these state regulations, usually under state law.

As a result of this complex structure, the actual abatement of air pollution was attenuated under the 1967 Act and the earlier measures. It required the adoption of AQCRs and AQCs by DHEW, followed by the adoption of AAQS and an SIP by each state (or by the Secretary in the event of state default), promulgation of the necessary state laws and regulations to effectuate the SIP (often not until after federal approval of the plan[25]) then, finally, either enforcement action brought by the state under state law to enforce the implementation plan or the AAQS, or a tortuous enforcement proceeding brought

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[24] Section 2, subsection 108(c)(4) provides that, if the Secretary finds that the ambient air quality does not meet the AAQS in an AQCR or state because the state has failed to take reasonable action to enforce the standards, the Secretary may notify the relevant state of the failure and, if the failure does not cease within 180 days of notification, the Secretary may request the Attorney General to bring suit (in the case of interstate air quality standards violations) or, for intrastate violations, at the request of the Governor, may provide resources for the state to bring enforcement action or, again at the request of the Governor, request the Attorney General to bring suit to abate the air pollution (US Congress, 1967).

[25] Following the 1994 decision in *Natural Resources Defense Council, Inc. v. EPA*, US EPA has taken the position that SIPs must be comprised of controls which are already in regulatory force, and presumably enforceable under state law at the time of submission of the SIP (Martineau and Novello, 2004, p. 45).

by the Attorney General on behalf of the state, in the event that the state had failed to take reasonable enforcement action on its own. Little air pollution control was achieved because the states proved to be less than enthusiastic in their embrace of these new authorities. Only 21 implementation plans were filed by the states between 1967 and 1970, none of which were approved by DHEW (Reitze, 1991, p. 1590). This problem of great delays between setting policy goals and actually controlling emissions is a product of the complex federal-state relationship built into the foundation by these early measures, which remains a central feature of air pollution control programs today.

In 1970, frustration with the slow progress of air pollution abatement and the generally heightened public environmental concerns combined to provide the necessary political support for the Clean Air Act Amendments of 1970, which asserted a major federal regulatory role[26] and strengthened enforcement authorities at the federal level. Authority to administer the federal role under the Clean Air Act and to conduct all of the federal air quality management responsibilities was given by the 1970 Amendments to the newly established US EPA (US Congress, 1970).

The 1970 Amendments significantly reconfigured the federal and state roles in the cornerstone programs in several significant ways. The Amendments required US EPA to develop a list of priority air pollutants (the "criteria pollutants") and to develop AQCs for these pollutants (US Congress, 1970). The Amendments removed the states' authority to establish AAQS and required US EPA to propose national standards, NAAQS, for each criteria pollutant. NAAQS were defined as levels of air quality of criteria pollutants, allowing an adequate margin of safety, that are requisite to protect public health (primary NAAQS) and to protect the public welfare from known or anticipated adverse effects of ambient air pollution by criteria pollutants (secondary NAAQS) (US Congress, 1970). Although states were assigned primary responsibility for assuring air quality in AQCRs, US EPA was authorized to establish any intrastate or interstate region as an air quality control region if necessary for the attainment of NAAQS (US Congress, 1970). States were required to submit for US EPA review an implementation plan that provided for the implementation, maintenance and enforcement of each primary and secondary NAAQS in each AQCR within the state in such a manner as to attain the NAAQS within stringent time limits (US Congress, 1970). If a state failed to submit an implementation plan that US EPA deemed approvable under standards set forth in the Act, US EPA was authorized to publish a regulation promulgating an implementation plan in place of the deficient state submission (US Congress, 1970). Moreover, if US EPA determined that a state was failing to enforce its implementation plan effectively, US EPA was

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[26] The 1970 Amendments expanded the federal regulation of mobile sources by imposing technology-based emission standards on motor vehicles and other mobile sources and authorizing land use and transportation controls in urban areas if necessary to achieve NAAQS (Reitze, 2001, p. 16).

authorized to notify the state and to enforce any requirements of the state's plan following the notification until the state satisfied US EPA that it would enforce its plan ("period of federally assumed enforcement") (US Congress, 1970).

The 1970 Amendments also directed US EPA to develop standards of performance for new stationary sources of air pollution (New Source Performance Standards or NSPS) under which new sources would be assigned to categories and regulations would be developed for each new source category to establish federal standards of performance (US Congress, 1970). A standard of performance was defined as "a standard of emissions which reflected the degree of emission limitation achievable through the application of the best system of emission reduction which (taking into account the cost of achieving such reduction) the Administrator determines has been adequately demonstrated" (US Congress, 1970). The NSPS program was also to be implemented through a federal-state partnership involving state submission of new source implementation plans, US EPA review and approval of the plans, and delegation of the authority to implement and enforce the NSPS program (US Congress, 1970).

The 1970 Amendments also required US EPA to develop a list of Hazardous Air Pollutants for which no AAQS is applicable which, in the judgment of the Administrator, may cause or contribute to an increase in mortality or an increase in serious, irreversible or incapacitating reversible illness (Hazardous Air Pollutants or HAPs) and publish emission standards for the listed pollutants (National Emission Standards for Hazardous Air Pollutants or NESHAPs) (US Congress, 1970). The states were again authorized to develop a plan for implementing and enforcing emission standards for HAPs acceptable to US EPA and thereby achieve delegation of the HAPs program (US Congress, 1970).

Thus, the 1970 Amendments established a federal-state partnership in which US EPA made basic regulatory and scientific judgments about the degree of control of criteria pollutants in ambient air necessary to protect public health and welfare, designated as NAAQS, and the states were required to develop a SIP under which the specific sources of criteria pollutants within their borders were to be abated to the extent necessary to achieve the NAAQS. If the state defaulted on its responsibility to develop and implement an effective SIP, US EPA was authorized to develop and implement a surrogate federal implementation plan (FIP) in order that air quality within the deficient state would attain NAAQS notwithstanding the state's default. Similarly, US EPA was directed to develop NSPS and emissions standards for HAPs, which could then be incorporated into a state's implementation plan, subject to US EPA's determination that the plan was adequate to achieve the goals of the Act.

Implementation of the 1970 Amendments at the federal level began briskly with the 1971 promulgation by US EPA of NAAQS for 6 criteria pollutants: sulfur dioxide, particulate matter, carbon monoxide, photochemical oxidants, hydrocarbons and nitrogen dioxide (US EPA, 1971b). Lead was not among these original pollutants. Among several significant policy judgments made by US EPA in promulgating these standards, US EPA determined that the development of NAAQS for criteria pollutants should not be limited by the feasibility of implementation of the standards. This determination was made based on the interpretation that the Clean Air Act does not permit any factors other than health to be taken into account in setting the primary NAAQS (US EPA, 1971b). No further criteria pollutants were named until 1976, when US EPA added lead to the list of criteria pollutants (US EPA, 1976e) in compliance with a federal court order in a lawsuit against US EPA brought by the Natural Resources Defense Council (NRDC) (US District Court, Southern District of New York, 1976). To date, no further pollutants have been listed and the initially listed hydrocarbon pollutants were dropped in 1983 (US EPA, 1983a).

Through the early 1970s, the states assembled the elements of their air quality control program envisioned in the 1970 Amendments. States upgraded their implementation plans to conform to the Amendments, enacted statutes and regulations which authorized the states to promulgate the specific emission controls necessary to seek attainment with NAAQS, and put in place the technical and bureaucratic infrastructure to pursue the goals of the Amendments (Martineau and Novello, 2004, p. 6). Despite, or perhaps because of, the fact that the states also responded briskly to the 1970 Amendments with SIPs, SIPs of uneven quality were submitted, which led US EPA to conclude that many were inadequate, resulting in FIPs and other US EPA requirements in response (Martineau and Novello, 2004, p. 6). Even the original 1970s SIPs approved by US EPA often failed to achieve attainment of the NAAQS within the statutory periods. This fact led directly to revisions of the Act in 1977 (US EPA, 1987b).

In 1977, a comprehensive package of amendments to the Clean Air Act was passed by Congress in response to frustration that the developing programs, and the submittal and review of SIPs had not met the goals of the 1970 Act. In particular, a disappointing number of the AQCRs across the country were meeting or making significant progress toward attaining the NAAQS (Martineau and Novello, 2004, p6).[27] As a result, the 1977 Amendments extended the deadlines to 1987 (US Congress, 1977) for reaching attainment with the primary NAAQS while imposing more stringent emissions standards on areas in nonattainment status (US Congress, 1977). A nonattainment area was defined to be "for any air

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[27] “When the [statutorily mandated] time for compliance [with primary NAAQS] arrived in 1975, many AQCRs failed to meet the NAAQS” (Reitze, 2001, p. 55).

pollutant an area which is shown by monitored data or which is calculated by air quality modeling (or other methods determined by the Administrator to be reliable) to exceed any national ambient air quality standard for such pollutant" (US Congress, 1977).

States were required to submit revised implementation plans for nonattainment areas to demonstrate how they would meet the revised deadlines and to obtain US EPA approval for the revised SIP by July 1979 (US Congress, 1977). In nonattainment areas, the revised SIPs were required to impose a higher technological standard of controls, RACT on existing major stationary sources, and to require permits for the construction and operation of major new stationary sources or major modifications to existing major stationary sources (the nonattainment area "New Source Review" program) (US Congress, 1977). These permits were required to impose emission controls to meet the lowest achievable emission rate (LAER) for any nonattainment criteria pollutant (US Congress, 1977). LAER was defined to be "the most stringent emission limitation which is contained in the implementation plan of any State for such class or category of source, unless the owner or operator of the proposed source demonstrates that such limitations are not achievable, or the most stringent emission limitation which is achieved in practice by such class or category of source, whichever is more stringent" (US Congress, 1977). Moreover, the Amendments required that new sources in nonattainment areas must compensate for their proposed increase in emissions of criteria pollutants in the area by obtaining a corresponding reduction in emissions of the criteria pollutant from their own sources or from other sources in the nonattainment area (US Congress, 1977).

Similar provisions applicable to new or significantly modified major stationary sources were imposed by the 1977 Amendments on AQCRs which were in compliance with the NAAQS ("attainment areas") in order to ensure that the nominally clean air in attainment areas was not degraded by increases in emissions. These provisions enacted into statute the "Prevention of Significant Deterioration" (PSD) program regulations which had been imposed upon US EPA as the result of litigation over the provisions of the 1970 amendments (US Court of Appeals, District of Columbia Circuit, 1972). This program required the issuance of permits for construction in attainment areas of new or modified "major emitting facilities" and required preconstruction review of the facility in order to determine that the facility would be consistent with the emissions limitations and implementation plan applicable to the attainment area (US Congress, 1977). These permits were required to impose the best available control technology (BACT), a stringent technology based standard, to prevent air quality deterioration.[28] The BACT

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[28] BACT was defined to be "an emission limitation based on the maximum degree of reduction of each pollutant subject to regulation under this Act emitted from or which results from any major emitting facility, which the permitting authority, on a

standard was also imposed by the 1977 Amendments on new sources in attainment areas under the NSPS program (US Congress, 1977).

The 1977 Amendments established an ambitious agenda and added considerable substantive law and complexity to the Clean Air Act. However, the heart of the amended Act remained the establishment, implementation and achievement of the NAAQS for criteria pollutants (Reitze, 2001, p19). The complexities of the NAAQS process and the attendant federal-state interactions in turn, resulted in the continuance of the slow speed of progress in developing and enforcing meaningful air quality standards nationwide. For example, to assure that the NAAQS were based on up-to-date technical data, the 1977 Amendments imposed the requirement that US EPA must, beginning in 1980 and every 5 years thereafter, conduct a thorough review of the AQCs and the air quality standards supporting and comprising the NAAQS and recommend any changes (US Congress, 1977). Whether because of the political climate of the 1980s or the inherent inertia of the Clean Air Act regulatory process, these periodic reviews proved to be time consuming and controversial (Belden, 2001, p. 19). The initial five-year review of the ozone standard commenced in the early 1980s and was not completed until 1993, based not on contemporaneous data but on 1986 data updated in 1989 (Belden, 2001, p. 19). The initial review of the lead criteria and NAAQS did not commence until 1984 and was not concluded until 1991 (Martineau and Novello, 2004, p. 35). Some commentators have observed that "litigation, chaos and uncertainty" in implementation of the 1977 Amendments resulted from the standoff between Congress and the White House beginning with the 1980 election, "even as the deadlines of the 1977 Amendments approached in the late 1980s and passed" (Martineau and Novello, 2004, p. 6).[29]

1990 Amendments and Current Air Pollution Regulation

The 1990 Amendments to the Clean Air Act (US Congress, 1990) built upon the programs and federal-state approach of the 1977 Amendments to refine most of the existing federal air programs, and enacted a number of significant new air pollution control programs.

Title I of the Amendments refined the provisions for attainment and maintenance of the NAAQS. This title defined more clearly the federal and state roles in designating AQCRs as being in attainment status or nonattainment status. Specific designations of nonattainment AQCRs were required for ozone,

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case-by-case basis, taking into account energy, environmental, and economic impacts and other costs, determines is achievable for such facility through application of production process and available methods, systems, and techniques, including fuel cleaning or treatment or innovative fuel combustion techniques for control of each such pollutant" (US Congress, 1977).

[29] These commentators note that, despite the chaos, air quality management programs matured at the state and local air quality regulatory level during the 1980s and undertook their own initiatives to deal with local issues (see also Reitze, 2001, pp. 19-21).

carbon monoxide and particulate matter criteria pollutants (US Congress, 1990). The designation of attainment or nonattainment carried with it increasing significance under the 1990 amendments as the respective designations resulted in varying degrees of control over existing and new sources within the designated AQCRs. In addition, nonattainment AQCRs for ozone and carbon monoxide were further classified based on the severity of their respective nonattainment conditions. The severity classifications for ozone and carbon monoxide (marginal, moderate, serious, severe and extreme) were then translated into the time periods within which an AQCR must come into compliance with the respective NAAQS and the rate of progress that must be demonstrated toward compliance.

Title I of the Amendments revised the manner in which nonattainment AQCRs are regulated, generally lengthening the periods allowed for coming into compliance with the NAAQS. Areas designated in nonattainment were required to submit revised SIPs that will "provide for the implementation of all reasonably available control measures as expeditiously as practicable (including such reductions in emissions from existing sources in the area as may be obtained through the adoption, at a minimum, of reasonably available control technology)" (US Congress, 1990), "require permits for the construction and operation of new or modified major stationary sources anywhere in the nonattainment area" (US Congress, 1990), and include enforceable emissions limitations, and such other control measures, means or techniques (including economic incentives such as fees, marketable permits, and auctions of emissions rights), as well as schedules and timetables for compliance, as may . . . provide for attainment of such standard in such area by the applicable attainment date" (US Congress, 1990). Title I also detailed specific programs for areas in nonattainment with the ozone, carbon monoxide and particulate matter NAAQS, with escalating requirements corresponding to increasing severity of the nonattainment (US Congress, 1990). The Amendments did not make significant changes to the nonattainment programs for sulfur oxides, nitrogen dioxide or lead, other than to modify the deadlines for coming into attainment (US Congress, 1990).

Title III significantly altered the Hazardous Air Pollutants program. Between 1970 and 1990, US EPA succeeded in listing only 8 Hazardous Air Pollutants under the initial Hazardous Air Pollutants program which was focused solely on the determination of adverse health effects of potential HAPs (Belden, 2001, p. 8). Rather than allowing US EPA to continue to develop a list of HAPs through a demonstrably slow regulatory process, the 1990 Amendments contained an initial list of 189 HAPs that would be subject to technology-based and health-based emissions standards and specified the procedures to modify or revise the list (US Congress, 1990). Title III also required US EPA to develop within 12 months a list of categories and subcategories of major sources[30] and area sources[31] of Hazardous Air Pollutants and to revise the list periodically thereafter, no less frequently than every 8 years.[32]

US EPA was directed to establish initial technology-based (Belden, 2001, p68) emission standards for each category and subcategory of major sources and area sources of listed Hazardous Air Pollutants (US Congress, 1990). A second phase of emission standards was to be developed if it was determined that the further regulation beyond the technology-based standards was necessary to achieve adequate protection of public health. [33] The promulgated emission standards[34] must require the maximum degree of reduction in emissions that the US EPA determines is achievable for new or existing sources in the category or subcategory of sources to which the standard is to apply, taking into consideration the cost of achieving such reduction and the health, environmental and energy impacts of doing so (the Maximum Achievable Control Technology, or MACT, standard) (US Congress, 1990). Title III retained a distinction between new and existing sources, requiring new sources to implement the emission control that is achieved in practice by the best controlled similar source, where existing sources could be regulated somewhat less stringently, but would still be bound to emission controls equivalent to those used by the top tier in emissions reduction effectiveness of similar sources (US Congress, 1990).[35]

Title III also established a program for prevention of accidental releases of Hazardous Air Pollutants (US Congress, 1990).[36]

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[30] A “major source” is defined to be “any stationary source or group of stationary sources located within a contiguous area and under common control that emits or has the potential to emit considering controls, in the aggregate, 10 tons per year or more of any hazardous air pollutant or 25 tons per year or more of any combination of hazardous air pollutants” (US Congress, 1990).

[31] An “area source” is defined to be “any stationary source of hazardous air pollutants that is not a major source," not including motor vehicles (US Congress, 1990).

[32] US EPA was directed to make the list of categories of sources of hazardous air pollutants consistent with the list of categories of sources to be compiled for implementation of the new source performance standards pursuant to § 111 (US Congress, 1990).

[33] The health risk-based HAPs controls commenced with a requirement that US EPA investigate and report on, within six years of the 1990 Amendments, methods of calculating the risk to public health remaining or likely to remain from sources after application of the MACT emission standards, the public health significance of such remaining risk, the actual health effects experienced by neighbors in the vicinity of sources or experienced in response to background concentrations of HAPs, and recommendations concerning such remaining risk (US Congress, 1990). US EPA was required to promulgate second phase standards if the MACT standards do not reduce lifetime excess cancer risks to the individual most exposed to emissions of a source in the category or subcategory to less than one in a million (US Congress, 1990).

[34] US EPA was directed to establish emission standards for categories and subcategories of sources according to an aggressive schedule which required that emissions standards be promulgated for all categories and subcategories no later than 10 years after the date of enactment of the 1990 Amendments (US Congress, 1990).

[35] The top tier of similar sources was defined to be the average emission limitation achieved by the best performing 12 percent of the existing source in the category or subcategory for categories and subcategories with 30 or more sources or the best performing 5 sources for categories or subcategories with fewer than 5 sources (US Congress, 1990).

[36] This provision was a further legislative response to the Bhopal, India industrial disaster, which had previously been addressed in Emergency Planning and Community Right to Know provisions of the Superfund Amendments and Response Act of 1986 (US EPA, undated).

Other significant changes enacted in the 1990 Amendments include: Title II of the 1990 Amendments made widespread refinements and additions to the mobile source emissions control programs, including significantly strengthened emission standards for conventional motor vehicles and a program for regulation of mobile source-related air toxics (US Congress, 1990).[37] Title IV (Acid Deposition Control) established an emissions trading program to reduce emissions causing acid rain (US Congress, 1990). Title V (Permits) (US Congress, 1990) extended the Clean Air Act permit program to apply to existing sources and created a permitting process similar to those of other major federal environmental programs, *e.g.*, the National Pollutant Discharge Elimination System permitting program, whereby all requirements applicable to a permittee are captured by the permit, which is required to be renewed every 5 years (Reitze, 1991, p. 1611). Title VI (Stratospheric Ozone Protection) imposed stringent limitations on certain stratospheric ozone pollutants consistent with the provisions of the Montreal Protocol on Substances That Deplete the Ozone Layer (US Congress, 1990). Title VII (Provisions Relating to Enforcement) included more severe criminal enforcement consequences, including new authority for US EPA to undertake enforcement through administrative rather than judicial actions and to issue field citations for violations observed during inspections (US Congress, 1990).

Overall, the 1990 Amendments established an ambitious agenda for US EPA, the states, and the regulated community. It has been estimated that thousands of rulemakings by US EPA have been necessary to implement the 1990 Amendments, accompanied by thousands of guidance documents and even more interpretations (Martineau and Novello, 2004, p. 8). The states' responsibilities are multiples of US EPA's responsibilities, in that the states must apply the policies, regulations and guidance to individual sources of air emissions within their boundaries. And, of course, the regulated community must find a way to comprehend this complex body of regulations and laws, determine its impact on the affected businesses and organizations, and implement the required controls in a manner that is viable and effective.

The respective federal and state responsibilities were comprehensively set forth in US EPA's "General Preamble for the Implementation of Title I of the Clean Air Act Amendments of 1990" (US EPA, 1992b). This document focused primarily on the SIP submissions required for nonattainment areas by the Act. It also laid out US EPA's interpretation of the New Source Review provisions of the Act, which required that the states submit SIP revisions in 1992 and 1993 conforming their NSR programs for nonattainment areas to US EPA's interpretations. The Preamble identified 31 major deliverables

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[37] US EPA was instructed to conduct a study of the emission of then-unregulated toxic air pollutants from mobile sources and to develop regulations “containing reasonable requirements to control hazardous air pollutants from motor vehicles and motor vehicle fuels" (US Congress, 1990).

pertaining only to the ozone classification and CO classification portions of the Act which were due from the states within 4 years of enactment (US Congress, 1990). The Preamble sought to provide an overall picture of the activities necessary to complete the obligation that all necessary SIP revisions be made and approved by US EPA within 6 years of enactment (US Congress, 1990). The 6-year deadline was also the milestone by which ozone nonattainment areas in moderate or worse condition must achieve a 15 percent reduction in volatile organic compound (VOC) emissions. This timeframe illustrates the optimistic end of the range of expectations for the length of time it takes under the existing federal-state regulatory apparatus to determine specific air quality control policies and to actually see implementation of resultant improvements in air quality.

Air Quality Regulations for Lead

In 1961, the US Public Health Service, Division of Air Pollution, conducted a study of air lead concentrations in 3 major cities, as part of an evaluation of the public health aspects of a proposed increase in the allowable tetraethyl lead content of gasoline from 3 to 4 milliliters per gallon (Ludwig, *et al.*, 1965). The annual average air lead concentrations ranged from 1-2 $\mu g/m^3$ in Cincinnati, 1-3 $\mu g/m^3$ in Philadelphia, and 2-3 $\mu g/m^3$ in Los Angeles, with overall averages of 1.4, 1.6, and 2.5 $\mu g/m^3$, respectively. Averages in heavy traffic areas ranged from 14 to 44 $\mu g/m^3$. In a 1969 follow-up study, average (geometric mean) air lead concentrations in the urban portions of Philadelphia, Chicago, and New York were reported as 1.7 $\mu g/m^3$, 1.8 $\mu g/m^3$, and 2.1 $\mu g/m^3$, respectively (US EPA, 1972b). Air lead levels had declined slightly by 1979 with average concentrations of 1.1, 0.8, 0.9, 0.8, and 1.4 $\mu g/m^3$ reported for New York, Boston, Houston, Chicago, and Los Angeles, respectively (US EPA, 1986a).

Airborne lead concentrations decreased dramatically from about 1970 onward, largely due to the phase-out of leaded gasoline additives (US EPA, 2006a). From 1971 to 1980, the percent of urban stations reporting air lead concentrations of 1-1.9 $\mu g/m^3$ decreased from about 60% to less than 1% (US EPA, 1986a). Based on maximum quarterly average lead concentrations from 36 sites, the composite average concentrations decreased from about 1.3 $\mu g/m^3$ in 1977 to about 0.4 $\mu g/m^3$ in 1984 (US EPA, 1986a). Ambient air lead concentrations in the US decreased 94% from 1983 to 2002 (US EPA, 2003). Average air lead concentrations nationwide decreased from about 0.3 $\mu g/m^3$ in 1985 to about 0.04 $\mu g/m^3$ in 2000 (US EPA, 2006a).

National Ambient Air Quality Standards (NAAQS) for Lead

Lead was not among the original set of criteria pollutants for which the first NAAQS were adopted in the 1971.[38] As discussed in USBM (1986), US EPA relied upon the 1972 National Academy of Sciences publication "Lead: Airborne Lead in Perspective" in making a judgment that there was insufficient evidence at the time to identify an appropriate ambient air lead standard. In the early 1970s, US EPA's initial approach for control of airborne lead instead involved limiting lead emissions from automobiles, which were considered to be the principal source of lead in air (US EPA, 1978a).

Following a 1976 NRDC suit, US EPA was ordered to designate lead as a criteria pollutant and to develop a NAAQS for lead. US EPA added lead to the list of criteria pollutants in 1976 and established the first NAAQS for lead in October 1978.[39] Both primary and secondary NAAQS were set at 1.5 $\mu g/m^3$, averaged over a calendar quarter. These NAAQS of 1.5 $\mu g/m^3$ were retained in 1991 following an extensive review initiated in 1984. As discussed in US EPA (2007a), US EPA considered the latest lead health effects evidence, as reflected in such documents as the 1986 Air Quality Criteria for Lead (US EPA, 1986a), the 1990 Supplement to the criteria document (US EPA, 1990b), and the 1990 Staff Paper (US EPA, 1990c), in reaching its decision not to propose a revision to the lead NAAQS in 1991. The NAAQS are once again currently under review, with US EPA yet to propose revisions to the standards.[40]

The NAAQS for lead was established following a comprehensive scientific and technical policy assessment process that included the preparation of an "air quality criteria" document for lead. Following an extensive peer review process where opportunity for public comment was provided, the Air Quality Criteria for Lead was published by US EPA in December 1977 (US EPA, 1977c). US EPA (1977c) summarized the state of the science regarding what was known about lead in the environment and its potential for adverse health effects.

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[38] The first set of criteria pollutants included sulfur dioxide, particulate matter, carbon monoxide, photochemical oxidants, hydrocarbons, and nitrogen dioxide.

[39] The Lead Industries Association, Inc. brought a challenge against the lead NAAQS in 1979 in the federal Court of Appeals for the District of Columbia in which St. Joe Minerals Corp. intervened. This challenge was denied by the DC Circuit in 1980 and the US Supreme Court declined to review the denial of the challenge.

[40] As discussed in the December 17, 2007, Advance Notice of Proposed Rulemaking (ANPR) for the lead NAAQS (US EPA, 2007a), US EPA recently solicited comments on a range of alternative standard levels for the lead NAAQS, up to and including the current level of 1.5 $\mu g/m^3$. Staff within the US EPA Office of Air Quality Planning and Standards (OAQPS) previously made recommendations regarding ranges of alternative standard levels. As stated in the final Staff Paper for lead (US EPA, 2007b), OAQPS staff recommended that consideration be given to a range of alternative standard levels from as high as 0.1 to 0.2 $\mu g/m^3$, which represent current levels in many U.S. urban areas, down to the lowest levels of 0.02 to 0.05 $\mu g/m^3$ evaluated in the final Human Exposure and Health Risk Assessment for lead (US EPA, 2007c). OAQPS staff based their recommendations on a growing body of evidence on lead health effects, as well as on findings from the lead risk assessment of "estimated decrements in IQ for policy-relevant exposures associated with the current NAAQS that are clearly of a magnitude that might be reasonably be judged to be highly significant from a public health perspective." (US EPA, 2007b, p. 5-22)

Despite much progress in the understanding of the relationship between airborne lead and human health effects since publication of the 1972 NAS report, the Air Quality Criteria for Lead (US EPA, 1977c) and the final rulemaking for the Pb NAAQS (US EPA, 1978a) both acknowledged many remaining uncertainties. Among the key data gaps highlighted by US EPA (1977c; 1978a) were a variety of monitoring uncertainties including the lack of site selection criteria for locating airborne lead samplers, outstanding questions regarding interferences and collection efficiencies of various filter media, and uncertainties related to the extent to which a monitoring network is able to represent the actual exposure situations for young children. Significant data gaps also limited our understanding of airborne lead exposures and toxicity, as US EPA (1977c) highlighted the lack of quantitative data on the relative contributions of various sources of airborne lead to blood lead levels, the lack of information on the contribution of food to human exposure, and gaps in the knowledge of the effects of lead on the hepatic, cardiovascular, immunologic, and endocrine systems. Despite these and other key data gaps and uncertainties, the 1977 Air Quality Criteria served as the primary scientific reference underlying the first Pb NAAQS.

A variety of key issues associated with the technical and economic feasibility of the lead NAAQS was recognized by US EPA and others prior to its promulgation in October 1978. For example, US EPA had initially proposed NAAQS of 1.5 $\mu g/m^3$, averaged over 1 month, but changed to a less stringent quarterly averaging period following industry comments concerning the scientific and technical difficulties presented by the selection of the calendar month averaging period (US EPA, 1978a). US EPA also recognized that attainment of the standard may be problematic in areas with high background concentrations of airborne lead due to contributions from a variety of different sources (US EPA, 1978a). In fact, in promulgating the lead NAAQS, US EPA (1978a) noted that its own "economic analysis does indicate that there may be significant problems in attainment of the standard in the vicinity of nonferrous smelters and other large industrial sources of lead emissions." Previously, this idea that smelters may have great difficulties complying with the NAAQS was reflected in a September 29, 1978 press release on the lead NAAQS where Administrator Douglas M. Costle is quoted as stating that (US EPA, 1978d):

> Most primary smelters have their greatest impact in more sparsely populated areas. Our preliminary assessments indicate that some non-ferrous smelters may not be able to technically or economically achieve the standard. But our information on how bad the environmental problem is and how much it will cost to control these sources adequately is not good enough… During the next 6 to 9 months, we will work closely with the states and the affected industry to develop a plant-by-plant analysis of how serious the problems are, and what would be a reasonable compliance program for each smelter. We

> want to formulate a control strategy which will avoid significant disruption in the lead and smelting industries without compromising our goal of protecting public health. If our study of these impacts over the next 18 months indicates economic effects unwarranted by the health protection involved, we will consider a wide range of remedial action, including the possibility of seeking revisions to the Clean Air Act.

Because only generic emission factors and plant configurations were typically available, US EPA (1978a) encouraged state regulators and regulated entities to gather plant-specific air quality data, technical information and plant configurations in order to develop and "better consider approval of alternative approaches to emission control in the State plans. Recognizing the large uncertainties associated with the air quality impacts of fugitive emission sources from factories such as smelters, US EPA (1978a) concluded that "nonferrous smelters may have great difficulty in achieving lead air quality levels consistent with the proposed standard in areas immediately adjacent to the smelter complex."

It was thus well-recognized that few lead air quality data had been gathered around sources of fugitive emissions such as smelters, in part because there had never been any specific requirements in SIP regulations calling for the collection of such data around specific sources (US EPA, 1978a). To address this glaring data gap, as part of its promulgation of the NAAQS, US EPA (1978a) announced its intention to require installation of ambient air quality monitors in the vicinity of lead point sources. This proposed rulemaking was generally intended to initiate a process for the collection of facility-specific data needed to assess the nature and magnitude of the lead problem at major point sources, and to base further revisions of the SIPs consistent with the nature of the problem. It was specifically aimed at gathering information for characterizing the nature, extent, and impact of fugitive lead emissions from smelters. The final rulemaking for these proposed ambient air quality monitoring requirements at smelters, as well as the final version of the US EPA document "Guideline for Lead Monitoring in the Vicinity of Point Sources," was not published until nearly 3 years later in 1981 (US EPA, 1981a; 1981b).

As part of the October 1978 proposal involving ambient air quality monitoring in the vicinity of major point source, US EPA (1978a) announced its intention "to develop more accurate emission factors that relate the operation of a source to the amount of fugitive emissions the source generates." Although US EPA (1978a) acknowledged that there would be delays in the development of these factors such that they would not be available until some time after lead SIPs had been prepared and submitted by states, air lead emission factors for fugitive sources were not provided in the US EPA AP-42 emissions factor documents until 1986 and these emission factors were assigned the second lowest emission factor rating to D to indicate their low reliability (see Section 4.4).

Leaded Gasoline Phasedown

As discussed in Section 9.1, US EPA's rationale for not including lead among the original set of 6 criteria pollutants involved its belief that airborne lead could be controlled through its programs at the time for phasing out lead in gasoline (US EPA, 1978a). US EPA (1977c) cited data indicating that mobile-source emissions were responsible for an estimated 88 percent of total lead emissions in the United States. US EPA began its phasedown program for leaded gasoline in 1973, with its first reduction standards calling for a gradual phasedown of lead to 0.6 grams per total gallon by July 1, 1978 (US EPA, 1973b).[41] Implementation of this program was delayed by litigation until 1976. This program was tightened in 1982 and 1985, with the 1985 standard reducing the limit for lead content of gasoline down to 0.1 grams per gallon, effective January 1, 1986 (US EPA, 1985c). US EPA's 25-year program phasing out lead additives in gasoline was completed in 1996 when a direct final rule was published that banned the use of any lead additive in the production of unleaded gasoline (US EPA, 1996; ATSDR, 1999).

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[41] US EPA (1996) states that the average lead content in gasoline in 1973 was 2-3 grams per gallon, or about 200,000 tons of lead per year.

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Table 1
Pre-Clean Air Act Landmark Air Pollution Events

Date	Event
1881	First US municipal smoke abatement legislation enacted in Chicago, IL.
1906	A Utah district court enjoined 4 Utah smelters not to operate in response to litigation launched by farmers alleging injury to crops and livestock.
1907	Municipal smoke inspectors formed the International Association for the Prevention of Smoke, later known as the Air Pollution Control Association and currently the Air & Waste Management Association.
1914	Selby Smelter Commission released its comprehensive report investigating conditions in and around the Selby Smelting and Lead Company smelter in Contra Costa County, California.
1919	US Bureau of Mines launched one of the most comprehensive air pollution studies to date in Salt Lake City, Utah, to identify the sources of the city's smoke problems.
1921	General Motors researchers identified tetraethyl lead as an effective anti-knock agent in gasoline.
1926	Surgeon General's committee affirmed the safety of tetraethyl lead for general use in gasoline, but recommended further federal study (studies never funded).
1927	US Public Health Service (PHS) quantified the loss of daylight due to smoke in New York City, reporting an average loss of 21.5 percent for yearlong measurements in Manhattan.
1930	Strong atmospheric inversion occurred between December 1 and 5 in Belgium's Meuse Valley, trapping industrial air emissions at ground level and contributing to more than 60 deaths and thousands of illnesses.
1943	First smog episodes recognized in Los Angeles, California.
1947	California adopted the Air Pollution Control Act, authorizing the formation of county air pollution control districts.
1948	Acute air pollution episode occurred in the small industrial town of Donora, PA, with an intense temperature inversion trapping particulates and other industrial emissions at ground level over a 4-day period, causing an estimated 20 deaths and between 5,000 and 7,000 illnesses among a population of approximately 14,000.
1950	First US Technical Conference on Air Pollution held in Washington, D.C., with over 750 people attending.
1950	Dr. Arie Haagen-Smit of the California Institute of Technology proposed a photochemical mechanism for smog formation, identifying nitrogen oxides and hydrocarbons from automobiles and the petroleum industry to be key causative agents.
1951	Oregon passed first state law providing statewide authority to a state air pollution control agency.
1952	London "killer fog" event occurred, with official reports blaming the thick sulfur and particulate-laden fog that occurred between December 5 and 9 for over 4,000 deaths over a 2-week period.
1953	New York City experienced its first major acute air pollution episode, with an estimated 160 to 220 deaths.
1953	The Public Health Service established the National Air Sampling Network (NASN), later renamed the National Air Surveillance Network.

1955 US Air Pollution Control Act of 1955 passed; major provisions provide for federal technical assistance to state air pollution control and funding for Public Health Service to conduct air pollution studies.

1955 National Air Pollution Control Administration (NAPCA) founded within the US Department of Health, Education, and Welfare (DHEW).

1962 Rachel Carson's *Silent Spring* is published, bringing to the public's attention the unforeseen harm that human actions can have on the environment.

1963 Air pollution inversion event resulted in over 400 deaths in New York City.

1963 First Federal Clean Air Act enacted, giving the federal government enforcement authority to combat both interstate and intrastate air pollution problems and providing grants for initiation and development of local or state air pollution control programs.

1965 Motor Vehicle Air Pollution Control Act enacted, authorizing the development of federal emissions standards for new motor vehicles.

1965 Dr. Clair Patterson of the California Institute of Technology published the seminal paper "Contaminated and Natural Lead Environments of Man," challenging the current paradigm that human exposures to lead were in large part natural in origin and that the body had adapted over time to handle chronic lead exposure.

1967 Federal Air Quality Act of 1967 enacted, with key provisions authorizing the federal government to define the atmospheric areas of the nation, designate air quality control regions (AQCRs), publish Air Quality Criteria (AQCs) and Control Technology Documents for use by the states in setting air quality standards, require the development of state implementation plans (SIPs), and expand ongoing air pollution control research.

1969 National Environmental Policy Act (NEPA) passed in Congress.

1970 US Environmental Protection Agency (US EPA) established.

Table 2
Key Dates in the Development and Application of Direct Sampling Methods Used to Collect Suspended Particulate Matter

Year	Sampling Method
1900s	• Dustfall collectors gained usage in early air pollution investigations
1920s	• Owens Jet Dust Counter developed • Greenburg-Smith Impinger developed
1930s	• Midget Impinger developed
1940s	• Precursor to modern Hi-Vol Sampler developed • First cascade impactor developed • American Iron and Steel Institute (AISI) Spot Tape Sampler developed
1950s	• Hi-Vol sampler selected for usage in Public Health Service's NASN
1960s	• Modified Andersen Cascade Impactor developed and applied to ambient suspended particulate matter
1970s	• Hi-Vol promulgated as federal reference method for TSP and Pb • Modified Andersen Cascade Impactor selected by US EPA for usage in NASN cascade impactor network • Dichotomous sampler gained usage as size-selective sampler
1980s	• PM_{10} replaced TSP as NAAQS, and PM_{10} federal reference methods first developed
1990s	• $PM_{2.5}$ NAAQS promulgated and $PM_{2.5}$ federal reference methods first developed

Table 3
Timeline of Analytical Methods Used to Determine Particulate Lead

Table	Analytical Method
1920s	• Chromate method (*i.e.*, modified Fairhall method) gains usage for lead determination, in particular for PHS investigations of tetraethyl lead
1940s	• Colorimetric dithizone gains acceptance as standard method for lead determination
1950s	• PHS first employs optical emission spectroscopy (OES) for trace metal analysis of NASN samples • Birth of modern AAS
1960s	• First applications of AAS for particulate lead determination • Commercial AAS instruments become available • Early applications of graphite furnace atomic absorption (GFAA) and ICP-AES for elemental analysis
1970s	• AAS promulgated as federal reference method for lead determination in suspended particulate matter • GFAA and ICP-AES become commercially available
1980s-present	• ICP-MS becomes commercially available • Methods such as GFAA, X-Ray Fluorescence Spectrometry (XRF), ICP-OES, ICP-AES, ICP/MS attain federal equivalent method status for particulate lead determination

Table 4
Development of Major Particulate Air Pollution Control Technologies

Year	Event
Baghouses (Fabric Filters)	
1900s-1910	First used at lead, copper, and zinc smelters in California, Colorado, Missouri, and Utah for roaster and blast furnace gases
1930s	Common industrial use (cotton or wool bags)
1955	Efficiency >99%; use limited to gas temperatures <250° F
1950s	Development of synthetic fiber filter bags
1965	Efficiency = 99.7% overall (99.5% for 0-5 µm particles)
late 1960s	First use of synthetic fiber (fiberglass) filter bags at a smelter
1969	Baghouses and ESPs - principal particulate control technologies used at primary lead smelters
1980	Efficiency between 95-99.9% at primary lead smelters
c. 1990	Development of ceramic fabric bags that withstand temperatures up to 900° F
1990s	Continued development of specialty fabric bags with greater chemical resistance and thermal resistance up to 1,000° F
1999	Baghouses (or equivalent) required as part of EPA's final NESHAP rule for primary lead smelting
Cyclones (Centrifugal Separators)	
c. 1890s	First used for industrial gas particulate cleaning
1955	Efficiency of 50-90% for 5-10 µm particles; cyclones used in series called multi-cyclones
1962	Usage noted at primary lead smelters
1966	Used as pre-cleaners ahead of ESPs
1969	Efficiency >95% for >40 µm particles; very low efficiency for <5 µm particles
1977, 1980	Cyclones were not noted by EPA to be used at any primary lead smelter
Electrostatic Precipitators (ESPs)	
1906	Dr. Frederick Cottrell first successfully electrostatically-precipitated sulfuric acid fumes
1907	First commercial ESP demonstration; first ESP installed at a gold smelter for collection of sulfuric acid fumes
1910	First large ESP used for particulate collection at the Anaconda, MT copper smelter
1930s	Common industrial use; efficiencies near 95%
1939	By this time, ESPs had revolutionized recovery of acid mists, lead and arsenic fumes, and precious metals at smelters
early 1950s	Efficiency up to 98-99%
1969	Baghouses and ESPs - principal particulate control technologies used at primary lead smelters
1969	Efficiencies of 90-98+% (dry applications) and up to 99+% (wet applications)
mid-1970s	Efficiency >99.5%
Wet Collectors (Wet Scrubbers)	
early 1930s	Common industrial use
1940s	Development of venturi scrubber (most efficient of wet collectors) for particulate removal
1962	Use of various wet collectors (including scrubbing and spray towers and venturi scrubbers) noted at primary lead smelters
1965	Efficiency = 99.5% overall (99.0% for 0-5 µm particles) for venturi scrubbers
1960s	Wide variety of wet collectors in industrial use, including: spray chambers, wetted filters, mist eliminators, and scrubbers (centrifugal, impingement plate, packed bed, venturi, etc.)

Table 5
Timeline of Ambient Lead Regulation

Year	Regulatory Activity
1970	Clean Air Act
1973	Leaded gasoline phasedown promulgated
1975	NRDC suit to make lead a criteria air pollutant
1976	Standards of performance promulgated for new and modified primary lead smelters
1976	Court order requiring designation of lead as criteria air pollutant
1977	First US EPA Air Quality Criteria document for lead issued
1978	Lead Primary and Secondary NAAQS promulgated
1986	US EPA issued revised Air Quality Criteria document for lead
1990	Clean Air Act Amendments
1990	US EPA issued supplement to 1986 Air Quality Criteria document for lead
1991	US EPA retained lead NAAQS of 1.5 $\mu g/m^3$ following completion of review process
1996	Leaded gasoline phasedown completed
1999	National Emission Standards for Hazardous Air Pollutants (NESHAP) adopted for primary lead smelters
2004	US EPA initiated review process for lead NAAQS
2006	US EPA issued final lead Air Quality Criteria document
2007	US EPA issued final lead Risk Assessment Report and final lead Staff Paper; US EPA issued Advance Notice of Proposed Rulemaking (ANPR) for lead NAAQS

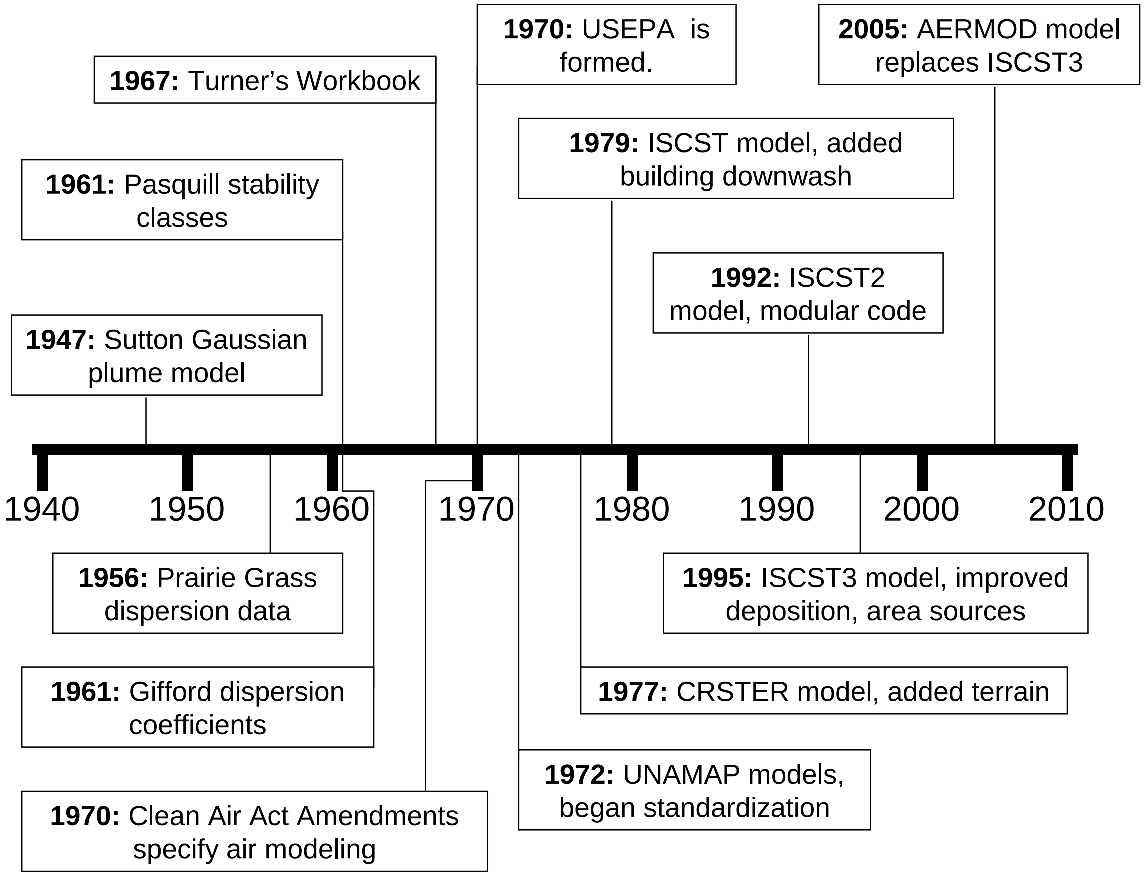

Figure 1
Timeline of Important Events in Air Dispersion Modeling

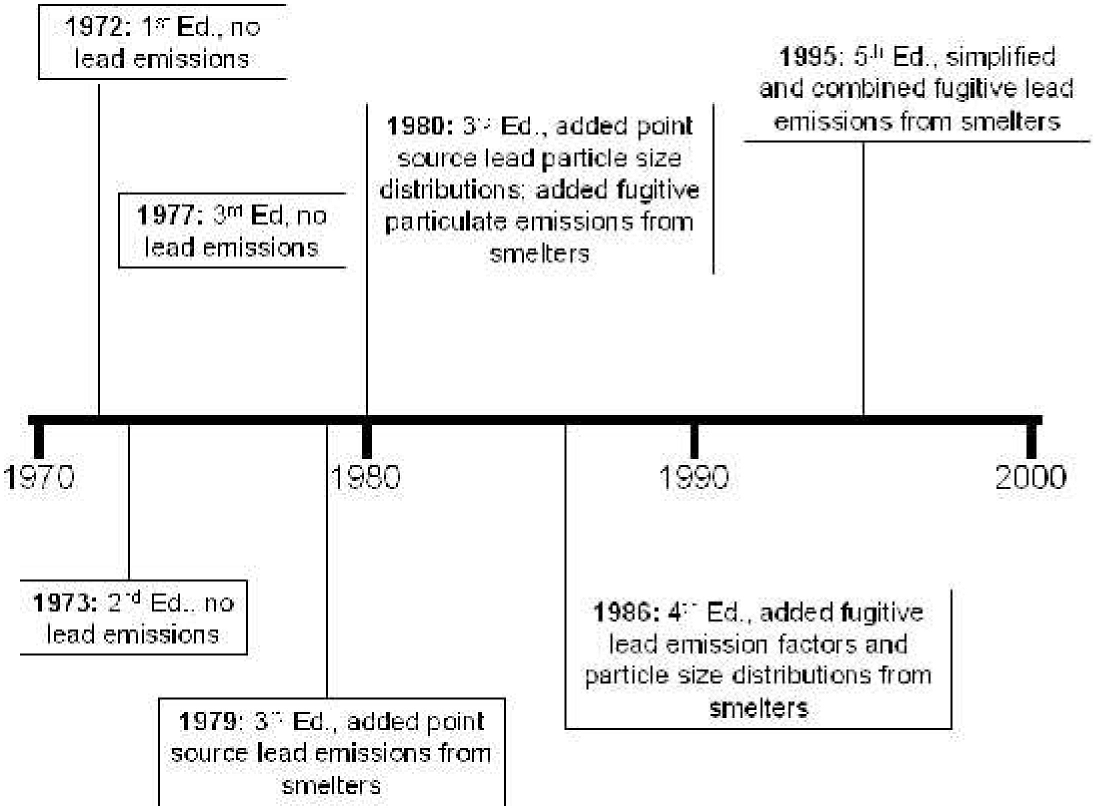

Figure 2
Timeline of AP-42 Changes for Lead Emissions from Primary Lead Smelters