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Methodology and Significance of Studies of Atmospheric Deposition in Highway Runoff

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A Contribution to the
NATIONAL HIGHWAY RUNOFF DATA AND METHODOLOGY SYNTHESIS

U.S. Department
of Transportation

USGS

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Methodology and Significance of Studies of Atmospheric Deposition in Highway Runoff

By JOHN A. COLMAN, KAREN C. RICE, and TIMOTHY C. WILLOUGHBY

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U.S. DEPARTMENT OF THE INTERIOR
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PREFACE

Knowledge of the characteristics of highway runoff (concentrations and loads of constituents and the physical and chemical processes which produce this runoff) is important for decision makers, planners, and highway engineers to assess and mitigate possible adverse impacts of highway runoff on the Nation's receiving waters. In October 1996, the Federal Highway Administration and the U.S. Geological Survey began the National Highway Runoff Data and Methodology Synthesis to provide a catalog of the pertinent information available; to define the necessary documentation to determine if data are valid (useful for intended purposes), current, and technically supportable; and to evaluate available sources in terms of current and foreseeable information needs. This paper is one contribution to the National Highway Runoff Data and Methodology Synthesis and is being made available as a U.S. Geological Survey Open-File Report pending its inclusion in a volume or series to be published by the Federal Highway Administration. More information about this project is available on the World Wide Web at http://ma.water.usgs.gov/fhwa/

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SI* (MODERN METRIC) CONVERSION FACTORS

APPROXIMATE CONVERSIONS TO SI UNITS

Symbol	When You Know	Multiply By	To Find	Symbol
LENGTH				
in	inches	25.4	millimeters	mm
ft	feet	0.305	meters	m
yd	yards	0.914	meters	m
mi	miles	1.61	kilometers	km
AREA				
in²	square inches	645.2	square millimeters	mm²
ft²	square feet	0.093	square meters	m²
yd²	square yards	0.836	square meters	m²
ac	acres	0.405	hectares	ha
mi²	square miles	2.59	square kilometers	km²
VOLUME				
fl oz	fluid ounces	29.57	milliliters	mL
gal	gallons	3.785	liters	L
ft³	cubic feet	0.028	cubic meters	m³
yd³	cubic yards	0.765	cubic meters	m³

NOTE: Volumes greater than 1000 l shall be shown in m³.

Symbol	When You Know	Multiply By	To Find	Symbol
MASS				
oz	ounces	28.35	grams	g
lb	pounds	0.454	kilograms	kg
T	short tons (2000 lb)	0.907	megagrams (or "metric ton")	Mg (or "t")
TEMPERATURE (exact)				
°F	Fahrenheit temperature	5(F-32)/9 or (F-32)/1.8	Celcius temperature	°C
ILLUMINATION				
fc	foot-candles	10.76	lux	lx
fl	foot-Lamberts	3.426	candela/m²	cd/m²
FORCE and PRESSURE or STRESS				
lbf	poundforce	4.45	newtons	N
lbf/in²	poundforce per square inch	6.89	kilopascals	kPa

APPROXIMATE CONVERSIONS FROM SI UNITS

Symbol	When You Know	Multiply By	To Find	Symbol
LENGTH				
mm	millimeters	0.039	inches	in
m	meters	3.28	feet	ft
m	meters	1.09	yards	yd
km	kilometers	0.621	miles	mi
AREA				
mm²	square millimeters	0.0016	square inches	in²
m²	square meters	10.764	square feet	ft²
m²	square meters	1.195	square yards	yd²
ha	hectares	2.47	acres	ac
km²	square kilometers	0.386	square miles	mi²
VOLUME				
mL	milliliters	0.034	fluid ounces	fl oz
L	liters	0.264	gallons	gal
m³	cubic meters	35.71	cubic feet	ft³
m³	cubic meters	1.307	cubic yards	yd³
MASS				
g	grams	0.035	ounces	oz
kg	kilograms	2.202	pounds	lb
Mg (or "t")	megagrams (or "metric ton")	1.103	short tons (2000 lb)	T
TEMPERATURE (exact)				
°C	Celcius temperature	1.8C + 32	Fahrenheit temperature	°F
ILLUMINATION				
lx	lux	0.0929	foot-candles	fc
cd/m²	candela/m²	0.2919	foot-Lamberts	fl
FORCE and PRESSURE or STRESS				
N	newtons	0.225	poundforce	lbf
kPa	kilopascals	0.145	poundforce per square inch	lbf/in²

* SI is the symbol for the International System of Units. Appropriate rounding should be made to comply with Section 4 of ASTM E380.

Methodology and Significance of Studies of Atmospheric Deposition in Highway Runoff

By John A. Colman, Karen C. Rice, *and* Timothy C. Willoughby

Abstract

Atmospheric deposition and the processes that are involved in causing and altering atmospheric deposition in relation to highway surfaces and runoff were evaluated nationwide. Wet deposition is more easily monitored than dry deposition, and data on wet deposition are available for major elements and water properties (constituents affecting acid deposition) from the inter-agency National Atmospheric Deposition Program/ National Trends Network (NADP/NTN). Many trace constituents (metals and organic compounds) of interest in highway runoff loads, however, are not included in the NADP/NTN. Dry deposition, which constitutes a large part of total atmospheric deposition for many constituents in highway runoff loads, is difficult to monitor accurately. Dry-deposition rates are not widely available.

Many of the highway-runoff investigations that have addressed atmospheric-deposition sources have had flawed investigative designs or problems with methodology. Some results may be incorrect because of reliance on time-aggregated data collected during a period of changing atmospheric emissions. None of the investigations used methods that could accurately quantify the part of highway runoff load that can be attributed to ambient atmospheric deposition. Lack of information about accurate ambient deposition rates and runoff loads was part of the problem. Samples collected to compute the rates and loads were collected without clean-sampling methods or sampler protocols, and without quality-assurance procedures that could validate the data. Mass-budget calculations comparing deposition and runoff did not consider loss of deposited material during on-highway processing. Loss of deposited particles from highway travel lanes could be large, as has been determined in labeled particle studies, because of resuspension caused by turbulence from passing traffic. Although a cause of resuspension of large particles, traffic turbulence may increase the rate of deposition for small particles and gases by impaction, especially during precipitation periods.

Ultimately, traffic and road maintenance may be determined to be the source of many constituents measured in highway runoff previously attributed to ambient atmospheric deposition. An investigative design using tracers of ambient deposition that are not present in highway traffic sources could determine conclusively what fraction of highway runoff load is contributed by ambient atmospheric deposition.

INTRODUCTION

The atmosphere is the pathway by which inorganic and organic compounds are transported from sources of air pollution to receptors on the ground. These compounds can be deposited locally around sources, when associated with large particles, or in areas far removed from the source, when associated with small particles or in vapor state (Majewski and Capel, 1995). Highway-travel surfaces are large, well-drained impervious areas whose efficiency as collectors of atmospheric deposition depends on the extent of traffic-induced particle resuspension. Deposited material that is not resuspended likely will enter highway runoff.

In addition to receiving constituents from the atmosphere, highways, and highway traffic contribute constituents to the atmosphere. Motor vehicles are recognized as a major source of pollutant emissions to the atmosphere and have a substantial effect on air and water quality (Moe and others, 1982; Ellis and others, 1987, and references therein). Vehicle emissions of gaseous oxides of nitrogen (N), carbon monoxide, trace metals, and organic compounds contribute substantially to the atmosphere (Harrison and Johnston, 1985). Whereas gaseous pollutants emitted from vehicles are rapidly and widely dispersed into the atmosphere, leaving no permanent record in the vicinity of the road, a portion of particulate emissions from vehicles is deposited locally, resulting in contamination of roadside vegetation, soils, surface waters (Harrison and Johnston, 1985; Ward, 1990), and in some cases, ground water. In addition, particulates can derive from the vehicle itself, such as from wear of brake linings, motorized parts, and tires (Shaheen and Boyd, 1975; Ward, 1990).

With the exception of eroded material from road cuts and road berms, most materials delivered to the road surface, and, therefore, subject to appearance in highway runoff, could be considered to be from ambient atmospheric deposition, from traffic, or from both sources. Yet, attempts to simulate loads of runoff constituents based on these causative factors have had little success (Driscoll and others, 1990a). The lack of simulation success limits attempts to mitigate causes of contamination from highway runoff or to estimate constituent loads in highway runoff from new construction. Unanswered are key questions such as whether variation in ambient deposition, from national or urban-to-rural gradients, are important to highway runoff quality in comparison to vehicle-related sources;

and the relative importance of highway as compared with other impervious surfaces (usually present in the urban environment) in determining runoff loads to receiving waters.

Complicating factors for modeling simulations may include complex on-highway processes, such as particle resuspension and deposition through turbulence impaction. Other difficulties are changes with time and among sampling programs in deposition-sampler construction materials, sampling procedures, and documentation of sample quality assurance. A final complicating factor is the substantial changes in atmospheric pollution loads that may have occurred during the data-collection period. Emissions of many types of airborne pollution have decreased since the passage of the Federal Clean Air Acts, beginning in 1970 (USEPA, 1999).

The U.S. Geological Survey (USGS) investigates all aspects of the hydrologic cycle including investigations on atmospheric deposition to highway surfaces in relation to constituent loads in runoff. The USGS maintains the database for the National Atmospheric Deposition Program (NADP). These activities account for the interest of the USGS in the topics reviewed in this chapter. The purpose of this report is to describe atmospheric deposition as it relates to highway runoff. Tools and techniques are reviewed, including data-quality assurance procedures that commonly are used for the assessment of atmospheric deposition when determining the occurrence, source, and load of constituents in highway runoff. In addition, this report describes sources of atmospheric-deposition data, reviews articles that discuss atmospheric sources to highway runoff (100 reports are listed in table form), and outlines possible strategies for continuing research within the context of information needed to simulate highway-runoff quality consistently on a regional or national basis.

FORMS OF DEPOSITION AND POST-DEPOSITIONAL TRANSPORT

Atmospheric deposition is defined as the solid, liquid, and gaseous materials deposited by atmospheric processes (Bricker and Rice, 1993). Atmospheric deposition can be divided into two categories—deposition that involves precipitation, called wet deposition (rain, sleet, and snow), and deposition that does not involve

precipitation, called dry deposition (settleable particulates and aerosols). Removal of contaminants from the atmosphere by fog, mist, and dew can be classified somewhere between wet and dry deposition but is more closely related to dry deposition (Majewski and Capel, 1995); these types of atmospheric deposition are sometimes termed occult deposition (Stensland and others, 1986).

Atmospheric-deposition samples can be collected in buckets as wet deposition, dry deposition, or bulk deposition (where a bucket is left open so that both wet and dry deposition are collected together). Dry deposition also can be evaluated by deposition onto standard surfaces or from deposition models that use concentrations in air as a function of particle size as inputs. The collection-bucket method for ground-based sampling of wet deposition is straightforward (Majewski and Capel, 1995), although the sampler design and means of sample collection can affect the results. In simplest form, bulk and dry-bucket collection of dry deposition also is straightforward. However, observed variability in multiple-sampler deployments and dependency of result on the bucket design or the collection surface has led to more complex collection devices (Lin and others, 1993; USEPA, 1997).

Wet Deposition

The two major processes that introduce atmospheric contaminants to wet deposition are rainout and washout. Rainout is a process that occurs in clouds and causes formation of water droplets on the pollutant by condensation, nucleation, or gas dissolution. The pollutants then are removed from the atmosphere by rainfall. Washout is the process in which falling precipitation scavenges airborne particulates and aerosols between the cloud base and the ground. In general, rainout is a more effective removal mechanism than washout. These processes can occur continuously during wet deposition because most storms produce convective air currents that add large masses of near-surface air to overlying clouds (Schroder and Hedley, 1986). The path of a storm can have a substantial effect on the chemistry of wet deposition delivered by the storm and may be a large source of variability in event-mean concentrations observed in highway-runoff studies (Driscoll and others, 1990a; Gay and Melching, 1995).

In general, much airborne material is removed from the atmosphere in the early stages of a precipitation event, and the rain becomes more dilute as the event proceeds. For computing annual depositional flux and for accurate comparison among sites, volume-weighting of constituents is necessary because of the concentration dependence on event size. Adjusting the concentrations of constituents by the amount of rainfall collected for the precipitation event allows for a fair comparison among sites. Example data collected from 1982 through 1993 at Catoctin Mountain, Maryland (Rice and Bricker, 1996) show that annual concentrations based on a straight average of samples generally are overestimated relative to volume-weighted annual averages (fig. 1). These data demonstrate the necessity of comparing volume-weighted rather than straight-average annual concentrations among sites.

Dry Deposition

Dry deposition provides a substantial mechanism for removing pollutants from the atmosphere and represents a larger deposition flux of many contaminants than does wet deposition (Kobriger, 1984; Harned, 1988). Solids, nutrients, trace metals, and organic compounds may be contributed to highway surfaces by dry deposition from background and pollution sources (Sartor and others, 1972; Gupta and others, 1981). Gupta and others (1981) reported that typical monthly dustfall loads in major cities across the United States ranged from 2,600 to 26,000 kg/km^2.

Atmospheric materials that contribute to dry deposition include settleable particulates, aerosols, gases, and vapors. Dry-deposition rates are dependent on the size, surface area, and mass of the particle (Legge, 1990). The largest airborne particles (greater than 2.5 µm) are derived from natural sources of mechanical abrasion and wind erosion, and are subject to gravitational settling. These particles are removed from the air mostly by dry deposition (Holsen and Noll, 1992). Smaller particles (from 0.1 to 2.5 µm) result from coagulation processes and primary aerosol emission, and are removed from the atmosphere by precipitation scavenging and by dry-particle deposition. The smallest particles found (less than 0.1 µm) are the result of secondary products of air-pollution emission and are removed mostly by particle coagulation (Holsen and Noll, 1992). Deposition of gases is part of dry deposition and occurs by adsorption on vegetation, soil, and other surfaces.
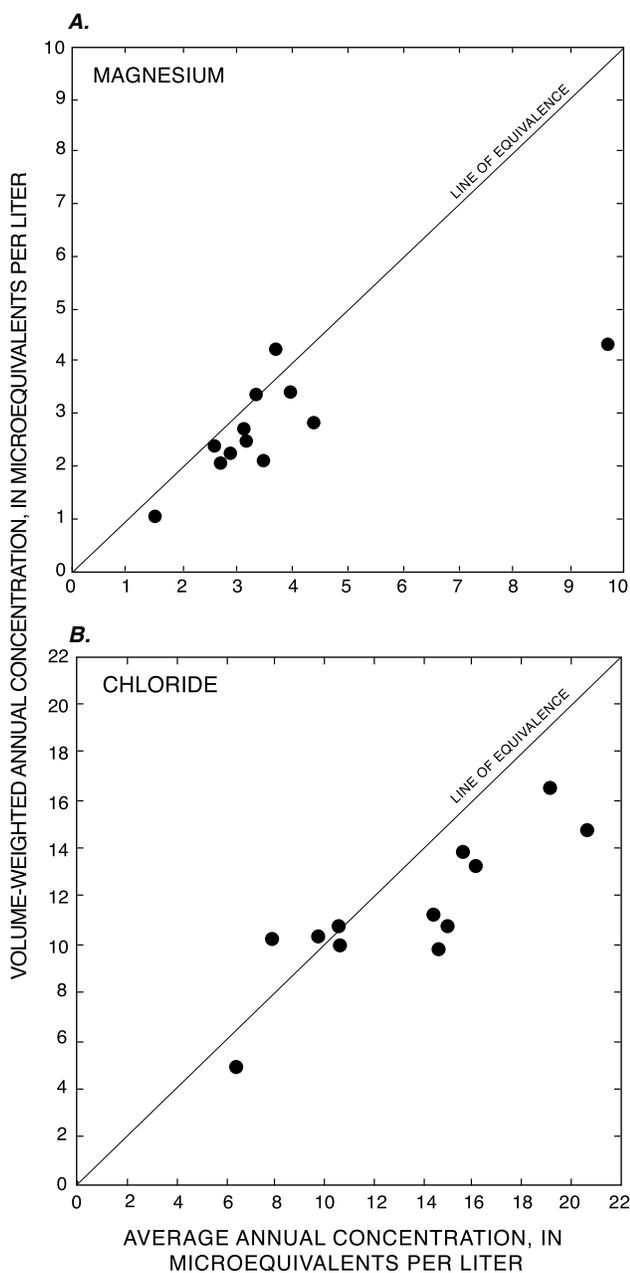
Figure 1. Comparison of average annual concentrations and volume-weighted annual concentrations from wet-deposition samples, 1982 through 1993 at Catoctin Mountain, Maryland, of (*A*) magnesium, and (*B*) chloride (Rice and Bricker, 1996).

The depositional transport of gases is increased by turbulence and roughness of the surface, which can move material through the boundary layer adjacent to deposition surfaces (Hicks and others, 1991). Dry deposition of gases onto highway surfaces has not been formally investigated.

Whereas the measurement of wet deposition by analysis of water collected in an appropriately constructed bucket is relatively straightforward, collection of dry deposition is complex because dry deposition depends on various physical characteristics of the collection surface used. Vandenberg and Knoerr (1985), for instance, measured the following variation for sulfate: 1.1, 3.2 and 5.8 mg $SO_4^{2-}/m^2/d$ for petri plate, interior surface of a bucket, and teflon filter surfaces, respectively. Deposition for N and sulfur (S) chemical species are particularly dependent on the type of surface, because the gas phases, nitric acid (HNO_3) and sulfur dioxide (SO_2), are important components of the total deposition (Hicks and others, 1991; Meyers and others, 1998).

Deposition rates determined by model simulation can be compared with deposition rates measured on a standard surface (Lin and others, 1993). In this report, deposition onto a standard surface refers to a polyvinyl chloride (PVC) plate with a sharply beveled (10 degrees) leading edge, on which are attached Mylar strips covered with a thin layer of Apezion L grease (Lin and others, 1993). The plate is directed into the wind with a wind vane. The device collects dry deposition on the grease-coated strips in a reproducible way with minimal turbulence. Model results have indicated that deposition velocities can be defined for all forms of dry deposition so that deposition can be modeled as a function of constituent concentration in air. For particulates, deposition velocities are functions of air turbulence, collection surface roughness, collection surface stickiness, and settling velocity, which differs by particle size. Particles in air can be separated by size using rotary-impactor samplers for large particles (6.5 to 36.5 μm) and conventional cascade-impactor samplers for small particles (0.43 to 10 μm) (Holsen

and Noll, 1992). Cascade-impactor samplers draw air through a series of differently sized jets arranged with sampling plates on which particles of successive size ranges are inertially impacted out of the air stream. For gases, deposition velocities can be modeled as functions of air turbulence and meteorological data (Hicks and others, 1991; Meyers and others, 1998).

Deposition to a standard surface has advantages over deposition to a dry-deposition bucket collector in terms of reproducibility of result and correspondence to total deposition where turbulence is at a minimum. Deposition to a standard surface still may need to be adjusted to apply to highway surfaces to account for on-highway processing because of highway-specific characteristics, such as traffic-induced turbulence and highway surface roughness. These adjustments have not been investigated but likely would depend on particle size of the deposited material. If particle-size data are necessary for an accurate estimate of highway loading, then air monitoring by impactor sampler as well as chemical analysis and estimated deposition velocity of each size fraction would be required.

Most mass-balance budget investigations, whether of highways, or urban or rural catchments, have relied on direct measurements of material collected in bulk or dust buckets to estimate atmospheric dry deposition. In perhaps the most exhaustive mass-balance investigation of a catchment, at Hubbard Brook, New Hampshire, deposition samples were collected in bulk collectors. Investigators at Hubbard Brook (Likens and Bormann, 1995) state that, although bulk collectors are "not very efficient at quantifying the deposition of very fine particles—smaller than 1µm— bulk collector results for calcium (Ca), magnesium (Mg), sodium (Na), potassium (K), and chloride (Cl) (dominated, presumably, by wet deposition and dry deposition of particles greater than 1µm), are fairly accurate." Bulk measurements of deposition for N and S, which have appreciable gas-phase deposition components, were adjusted upwards in the Hubbard Brook budgets (Likens and Bormann, 1995). If bulk collectors are assumed to measure large particle flux correctly,

they may be suitable for estimating constituent fluxes whose dry deposition is dominated by large-particle flux.

Saltation and Resuspension

In the geologic context, saltation refers to sediment transport in which particles are moved forward by bouncing along a surface, such as sand grains being pushed up a dune by the wind. In the highway context, saltation refers to deposition that bounces (as from a road salt application) or is splashed (particularly by moving traffic) and can result in material being moved off the road. Thus, saltation is a type of post-depositional transport. Vehicle spray has been shown to reintroduce contaminants to the atmosphere during periods of wet deposition (Irish and others, 1996) and clearly affects amounts of total precipitation and associated concentrations of constituents that are measured in runoff. Highway saltation measurements have been made with a collector that is slotted on the side to measure material that arrives at the collection location by way of a horizontal particle flight path (Harned, 1988; Kobriger and Geinopolos, 1984). Quality-assurance procedures for these saltation collectors, however, have not been documented. A blank could be measured by turning the collector away from the highway to determine how much of the total sample collected could have been from other than horizontal transport of resuspended material from the highway. Without associated quality-assurance measures, the saltation collectors only confirm that horizontal transport does occur, rather than providing a value for a budget term in the highway mass balance.

Resuspension also can be a sediment transport process, but it refers to finer particles that can be supported by the upward currents in turbulent eddies and moved greater distances than the sand-sized particles. Jones and Tinker (1984) found that the amounts of contaminants entrained by resuspension of wet deposition depend on traffic density, duration of the rainfall, wind speed, and wind direction. Likewise, dry deposition on

highways can be reintroduced into the atmosphere and transported to different locations by wind and anthropogenic activities, such as construction and automobile turbulence (Nicholson and Branson, 1990). Moe and others (1982) suggest that resuspended dust from roadways is preventing many cities from achieving national ambient air-quality standards for total-suspended particulates.

In a test with fluorescent marked particles, more than 70 percent of 12-μm silica particles were removed from the highway surface by four passes of a passenger vehicle at 40 miles per hour (Nicholson and Branson, 1990; fig. 2). Because kinetic energy increases with the square of velocity, a still greater percentage of particles would be removed by mixed traffic (cars and trucks) at more usual highway speeds of 60 to 70 miles per hour. Particle removal by vehicle turbulence was determined to be a function of particle size. Experiments were done with 4.2-, 9.5-, 12-, and 20-μm particles; only about 40 percent of particles were removed at 40 miles per hour when particle size was 4.2 μm (fig. 2).

The resuspension results emphasize the complex effect that particle size may have on particle-associated pollutants. Dry deposition of gases and particles might be increased by vehicle turbulence, but resuspension by turbulence would differentially affect particles according to size. Redeposition of the resuspended large particles likely would occur before long transport, so pollutants associated in large particles could be swept from the traveled highway surface but be redeposited in substantial amounts nearby.

EXPLANATION
SUSPENDED PARTICLES – Particle size in micrometers (μm)

—·—·■—·—·	4.2 μm
·········▲·········	9.5 μm
—·—△—·—	12.0 μm
— — □ — —	20.0 μm
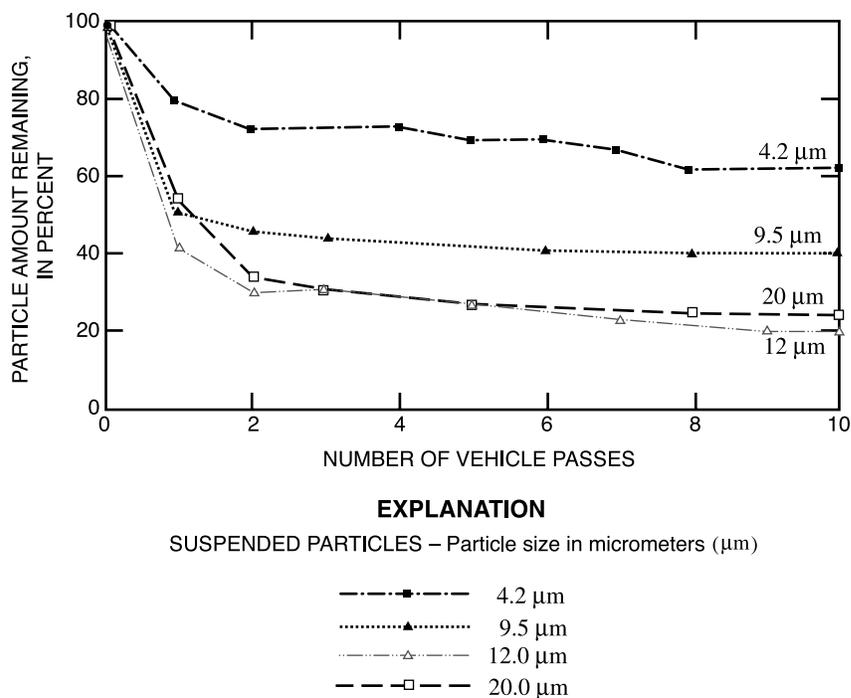
Figure 2. The resuspension of particles marked with fluorescent dye by turbulence from a medium-size vehicle traveling at 40 miles per hour, averaged for three separate experiments (Nicholson and Branson, 1990).

DATA FROM EXISTING MONITORING PROGRAMS

Atmospheric deposition has been studied intensively in the United States over the last 20 years, especially with regard to those constituents connected with acid deposition. Recently (1996) measurement of wet deposition of mercury (Hg) has begun to receive the same attention as the acid constituents, and wet deposition of other trace metals has been measured at some sites. Many of the scientific, technical, and logistic issues involved in creating a national network to characterize concentrations and loadings of pollutants from atmospheric deposition have been examined. The sources of information given here provide information on deposition of major elements, Hg, and precipitation, as well as conditions of meteorology and climate on a regional and national scale, as may be useful to highway-runoff investigations.

Wet deposition. The National Atmospheric Deposition Program (NADP), designed as a long-term program for chemical analysis of precipitation, was started in the United States in 1978 (Cowling, 1982). By late 1981, 93 sampling stations across the Nation had been established and were operational (Cowling, 1982). Now combined with the National Trends Network (NTN), the NADP/NTN cooperates with a variety of Federal, State, and private entities and currently operates more than 200

wet-deposition sampling sites spanning the continental United States, Alaska, Puerto Rico, and the Virgin Islands.

The NADP/NTN uses well-defined field methods and an extensive quality-assurance program to generate valid, defensible, and comparable results. The purpose of the network is to collect data on the chemistry of precipitation for monitoring geographical and temporal long-term trends. The precipitation at each station is collected weekly, and if it is of sufficient volume, analyzed for pH, conductivity, and the ions calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), ammonia (NH_3) and ammonium (NH_4^+), chloride (Cl^-), nitrate (NO_3^-), and sulfate (SO_4^{2-}). Data that can be obtained from the NADP/NTN are available on the Internet (NADP, 1998a). NADP/NTN data include weekly and daily precipitation chemistry; monthly, seasonal, and annual precipitation-weighted mean concentrations; annual and seasonal deposition totals; daily precipitation totals; and isopleth maps of constituent concentrations in precipitation and deposition rate [figs. 9*A-F* (at back of report); NADP, 2000a]. Data available from the NADP/NTN may be useful in establishing background wet-deposition constituent loadings for highway runoff studies.

The Atmospheric Integrated Research Monitoring Network (AIRMoN) is a subnetwork of the NADP/NTN run by the National Oceanographic and Atmospheric Administration (NOAA). In the AIRMoN program, wet-deposition samples are collected daily at a network of nine sites. Data are used for studying precipitation chemistry trends with a greater temporal resolution than the NADP/NTN (NADP, 1998b).

The NADP Mercury Deposition Network (NADP/MDN) is another subset monitoring program that measures Hg in precipitation. The program has been adding stations since startup in 1996, and currently (2000) includes more than 40 sites, most of which are in the eastern United States. Samples are collected weekly and collection and analysis are covered by a strong quality-assurance program. Results available through the Internet include annual volume-weighted concentrations and annual deposition (NADP, 2000b) plotted on a national map without isopleths (for example, fig. 3).

Dry deposition. Data from two dry-deposition measurement programs are available through the Internet. These programs are exclusively focussed on the problem of ozone and acid deposition. The goal of these two programs is to model dry deposition using measurements from reasonably non-complex collection of N and S chemical forms in atmospheric and meteorological data. The programs are the Atmospheric Integrated Research Monitoring Network (AIRMoN-dry program) run by NOAA Air Resources Laboratory (NOAA, 2000) and the Clean Air Status and Trends Network (CASTNET) run by the U.S. Environmental Protection Agency (USEPA) (USEPA, 2000a). Thirteen sites are available with dry-deposition estimates for SO_2, HNO_3, NO_3^-, and SO_4^{2-}. Because dry deposition of the measured constituents depends on deposition surface and local meteorological conditions, dry-deposition data users are cautioned against extrapolating to other conditions and locations (USEPA, 2000a).

Bulk deposition. The longest continuous record of precipitation chemistry in the United States is from the Hubbard Brook Experimental Forest in New Hampshire. Data have been collected at the Hubbard Brook site since 1963 and are discussed by Likens and Bormann (1995). Measurements have been limited to the concentrations of major ions and nutrients and water-quality properties such as pH. Estimates are made to correct bulk S and N deposition rates because of dry deposition of their gas phases, which are underestimated by the bulk collectors, as discussed previously.

Air pollution data. Although not directly a measure of deposition, air-quality information could be related to deposition. The USEPA's Toxic Release Inventory is a valuable source of information regarding the use, manufacture, treatment, transportation, or release of selected toxic chemicals into the environment. The Toxic Release Inventory can assist in locating point sources that might be affecting the atmospheric-deposition chemistry in an area (USEPA, 1998a). The States and cities collect data on air quality in a program administered by the USEPA (2000b). The data, which concern smog-producing constituents, NO_2, SO_2, O_3, and CO, particulates (PM_{10}) and lead (Pb), can be retrieved by monitoring site or by region (USEPA, 2000c).
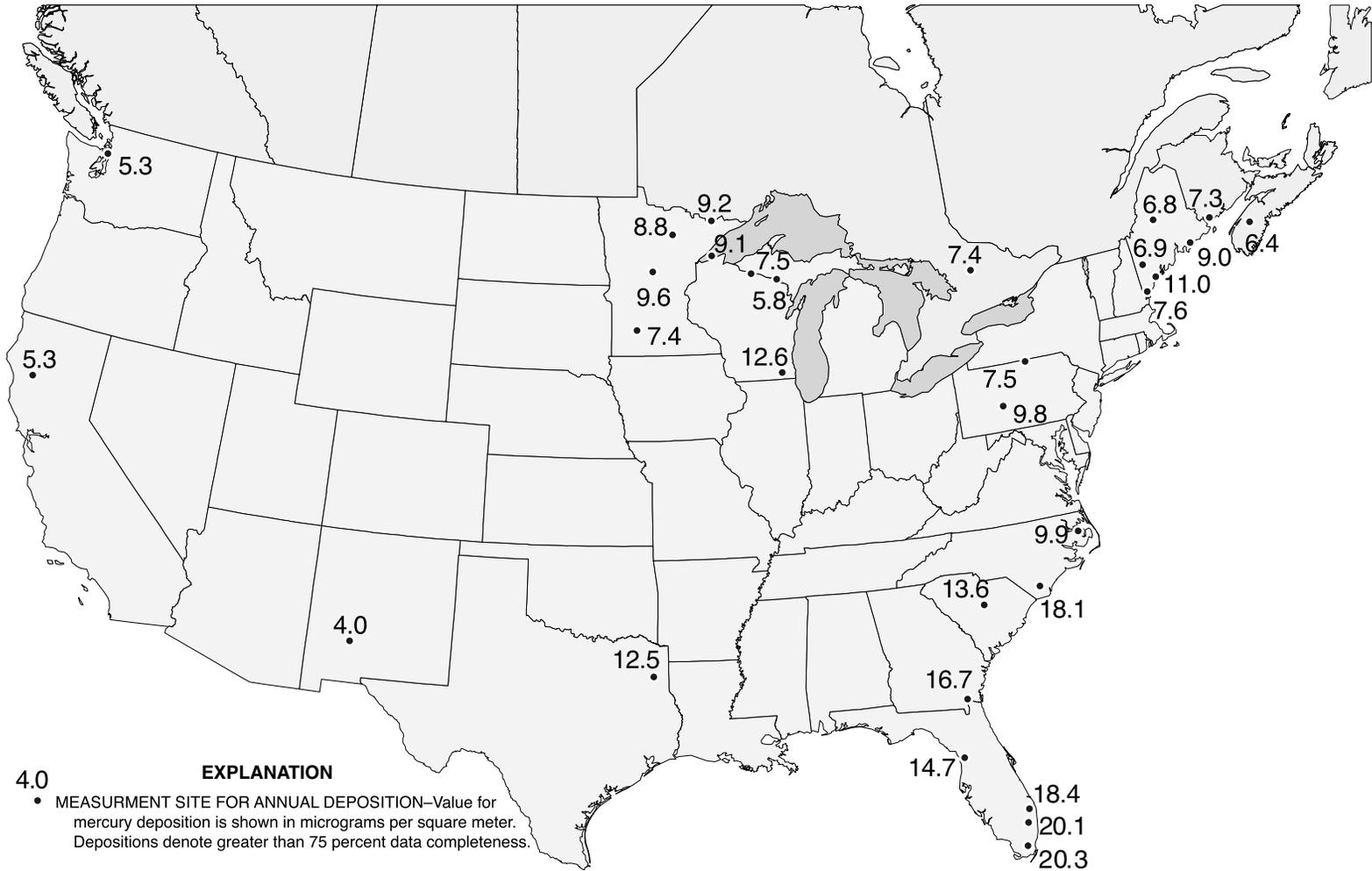
Figure 3. Annual wet-deposition rates of mercury during 1998 in the United States based on National Atmospheric Deposition Program/Mercury Deposition Network data (NADP, 2000c).

Meteorologic data. NOAA collects information concerning meteorological and climatological conditions throughout the United States. This information can be obtained from NOAA through their Regional Climate Centers. Information that may be useful in atmospheric-deposition studies includes daily and monthly high and low temperatures, precipitation amounts, wind speed and direction, and humidity (NOAA, 1998).

In summary, the available national network data are excellent for wet deposition and for acid-deposition constituents, but include only the one atypical trace metal, Hg. The dry-deposition data are focussed exclusively on acid-deposition constituents and are made available with a warning about use away from the local collection conditions.

MAJOR INORGANIC CONSTITUENTS AND WATER-QUALITY PROPERTIES

The major inorganic constituents and water-quality properties of atmospheric deposition are reviewed in this section. N and P species, often classified separately as "nutrients" in investigations of water, are included because of their relatively high atmospheric deposition.

Sources, Importance, and Chemistry

Common major inorganic constituents of highway runoff and their primary sources (Kobriger, 1984) are shown in table 1. Of the nine constituents, sources for four, bromide (Br⁻), N, P, S, are listed as atmospheric or partly atmospheric. In some locations, however, suspended solids likely could be added to the atmospheric source list, as could Ca^{2+}, Mg^{2+}, Na^+, and possibly Cl^- in warm areas, where these constituents are not applied in deicing salts.

Sediment transport is a major concern in highway runoff and certainly one source is atmospheric dust. Gupta and others (1981) reported (based on dust-bucket results) that typical monthly dustfall loads in major cities across the United States ranged from 86 to 860 mg/m²/d. Lin and others (1993) used modeled

Table 1. Common major inorganic constituents of highway runoff and their primary sources

[From Kobriger, 1984]

Constituent	Primary sources
Particulates	Pavement wear
Bromide	Automobile exhaust
Calcium	Deicing agents
Chloride	Deicing agents
Magnesium	Deicing agents
Nitrogen	Atmospheric deposition and roadside fertilizer application
Phosphorus	Atmospheric deposition and roadside fertilizer application
Sodium	Deicing agents
Sulfate	Roadway beds, fuel, deicing agents, atmospheric deposition

impactor-sampler data and deposition to a standard-plate collector to determine that total-particulate deposition at a Chicago, Illinois, site varied between 90 and 270 mg/m²/d, with 99.94 percent in the greater-than-2.5-µm fraction. Deposition of the greater-than-2.5-µm fraction is controlled by gravitational settling and may be subject to resuspension by traffic turbulence. A recent investigation comparing two sizes of road particulate matter (PM) concluded that the smaller $PM_{2.5}$ fraction, reflective of auto exhaust, exceeded the larger PM_{10} fraction, which is reflective of dustfall, at both urban and rural sites (Lamoree and Turner, 1999).

Major elements generally are not toxic and may be of less interest in highway-runoff investigations than are trace metals, many of which are toxic. Na, however, is important in drinking-water supplies because of its effect on the human circulatory system. Several highway-runoff projects have attempted to determine the extent to which Na contaminates aquifers and reservoirs near highways (Granato, 1996; Waldron, 1998). Also, the water-quality properties pH and specific conductance can affect chemical reactivity of atmospheric deposition on the highway surface (Bricker, 1999). Atmospheric deposition usually is acidic, particularly in the eastern United States (Rice and Bricker, 1992; Bricker, 1999), which could enhance the geochemical mobility of many highway contaminants. In contrast to typical trace-metal speciation in surface waters, a large percentage of trace metals in highway runoff (more than 50 percent) may be dissolved (Breault and Granato, 2000).

Character of Deposition Data

High-quality deposition data are available in greater quantity for the major constituents than for any other group because major inorganic constituents and water-quality properties (Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_3, Cl^-, NO_3^-, SO_4^{2-}, pH, and specific conductance) commonly are chosen for measurement in acid-deposition studies. Although accounting for only part of total deposition, the wet-deposition maps that are produced by the NADP/NTN enable observations about variability and relative deposition rates across the country. The degree of variability in the NADP/NTN wet deposition is small enough that data from the more-than-200 National collection sites can be contoured. In many areas, a reasonable approximation of the wet-deposition rate at a location between measured sites could be made by linear interpolation. Thus, highway mass-balance investigations could use the NADP data for a quality-assurance comparison check on ambient wet-deposition data collected on highway research sites. Because both wet and dry deposition originate from constituents in the atmosphere, wet deposition indicates, to a degree, the potential for constituent loads from dry and total deposition.

The deposition data maps indicate that there are lower deposition rates (order of magnitude less) for most of the major constituents in the west than in the midwest and eastern part of the country. There appears to be an eastern shift to deposition of constituents associated with combustion, such as SO_4^{2-}, (fig. 9A) and NO_3^- (fig. 9B) compared with those dominated by continental dust, such as Ca^{2+}, (fig. 9C) and those associated with agriculture, specifically the application of NH_3 in fertilizer (fig. 9D). Na^+ (fig. 9E) and Cl^- (fig. 9F) deposition rates are uniformly low across the country except in coastal regions where sea spray increases rates.

Trends analysis of the wet acid-deposition data indicate that acid deposition has decreased nationwide between 1980 and 1991 (Baier and Cohn, 1993). Statistically significant downward trends in SO_4^{2-} deposition occurred at 26 of 33 stations examined and at 3 stations for NO_3^-. Significant upward trends in pH were observed at nine stations, and a significant downward trend was observed at one station.

The dry-deposition networks (AIRMoN-dry and CASTNET) address only N, S, and ozone. At sites where both wet and dry deposition are measured, contributions from the two sources can be compared. Generally, the dry-deposition rates are much greater than the wet-deposition rates, and dry deposition of the gas forms (HNO_3, and SO_2) are substantial (10 times the particulate forms).

Reliable dry-deposition data for the other major elements are scarce. Ca dry deposition (7.1 mg/m^2/d) measured June through October 1991 in Chicago by standard surface deposition and by model simulation (Lin and others, 1993) was completely dominated by large-sized particles and more than 10 times the wet deposition measured by NADP in 1998 in the same area (0.69 mg/m^2/d). Measurements of dry deposition of six crustal elements—aluminum (Al), Ca, iron (Fe), Mg, silicon (Si), and titanium (Ti)—to a standard surface during July 1991 at three locations around southern Lake Michigan (at Chicago, on Lake Michigan, and at South Haven) varied in proportion to Ca deposition rates of 6.4, 1.5, and 3.0 mg/m^2/d, respectively (Holsen and others, 1993). This variation likely relates to prevailing wind direction and to relative proximity to a crustal source among the three sites.

Bulk collectors would represent a potential source of dry-deposition data if, as assumed by Likens and Bormann (1995), bulk collector results for Ca, Mg, Na, K, and Cl are accurate. An estimate of dry deposition could be made by comparison of their results with NADP wet deposition for northern New Hampshire. Replicates of bulk-collection results are variable, however (Kobriger and Geinopolos, 1984), so that the difference statistic, which represents dry deposition, likely would be inaccurate.

Phosphorus is not reported by NADP because concentrations in wet deposition generally are too low to be measured accurately. But dry deposition of phosphorus may be substantial and thus important in highway runoff that enters surface waters (Dudley and others, 1997). Analysis of year-long data collected in association with a lake budget in Walden Pond, Concord, Massachusetts, indicated that phosphorus deposition was dominated strongly by dry deposition (Colman and Friesz, 2001). Wet deposition was 0.010 mg/m^2/d, and dry deposition, measured in a bucket with water to simulate the lake-collector

surface, was 0.14 mg/m^2/d. The seasonal pattern of dry deposition indicated that pollen was a major component of the phosphorus load.

Uses in Highway-Runoff Investigations

Cl$^-$ and Na$^+$ are important major ions in the highway runoff context because of their use in road-salt applications and because of potential health effects of Na$^+$ in water supplies. Ambient Cl$^-$ and Na$^+$ deposition enter into road-salt runoff investigations as part of background loads. The NADP deposition maps (figs. 9E,F) show that Cl$^-$ in precipitation (less than 1 mg/L) never approaches the concentrations of Cl$^-$ in runoff associated with deicing in the north (100s to 1,000s of mg/L, Driscoll and others, 1990b). This result is the case even in northern near-marine areas (Massachusetts, Maine, Oregon, and Washington), where sea salt is incorporated into precipitation (Gay and Melching, 1995). If the wet-deposition rates are increased by a factor of 10 to account for dry deposition, resulting total deposition is still much less (10 times) in runoff at sites where road salt was applied.

The major elements may have a particular use in assessing how the highway processes of saltation and resuspension affect transport of ambient deposition to highway runoff. The major elements are more likely to be conservative in their chemistry and to be derived from ambient atmospheric deposition (rather than from engine and vehicle wear) than the trace metals. In addition, their measurements are less subject to quality-assurance problems during collection and analysis. Na$^+$ and Cl$^-$ might be used in warm areas where deicing agents are not used. Generally anions do not adsorb to solids, so the negatively charged chloride ion would be conservative during event-associated transport. Comparison of the Cl$^-$ runoff rate with Cl$^-$ deposition rate might indicate, for example, the net effect between the processes of loss from resuspension and that of increase because of turbulence impaction on the highway.

Although the major-element sources and chemistry are relatively simple, some considerations indicate that these likely tracers might give misleading results if the highway site is not carefully chosen. For example, concentrations may be greatly affected by bank erosion

and soil brought onto highways on tires and vehicle underbodies because aluminum (Al), Ca, Fe, Mg, manganese (Mn), and Si and suspended solids can be large components of these materials. Highway surfaces themselves can erode and contribute major ions, especially Mg and Ca, from concrete surfaces.

TRACE METALS AND CYANIDE

Trace metals and cyanide are reviewed in this section. Trace metals are deposited on highways from vehicles and the atmosphere. Cyanide is present in highway deicing salts.

Sources, Importance, and Chemistry

Trace metals and cyanide are important contaminants in highway runoff and may have substantial sources in ambient as well as vehicle-related atmospheric deposition. Trace metals are emitted to the atmosphere by both natural and anthropogenic sources. Natural processes, such as volcanic emissions and weathering of earth-surface materials (soils and bedrock), can contribute particulate-bound trace metals to the atmosphere. Anthropogenic activities are recognized as sources of trace metals to the atmosphere and include combustion of municipal solid waste (Campbell, 1976; Germani and Zoller, 1994), combustion of fossil fuels in coal- and oil-fired power plants (Campbell, 1976), releases from metal smelters (Crecelius and others, 1975; Ragaini and others, 1977), automobile emissions (Shaheen and Boyd, 1975; Ondov and others, 1982), and a large number of industrial stack emissions. Chemical analysis has indicated that fuels and exhaust contain many metals in measurable quantities, including Al, Ca, cobalt (Co), Fe, K, lithium (Li), Mg, Mn, Na, platinum (Pt), antimony (Sb), strontium (Sr), Ti, and V, in addition to the commonly studied trace metals cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), Pb, and zinc (Zn) (Hildemann and others, 1991; Hee, 1994). Vehicular emission of trace metals is well recognized as a source of contaminants to the roadside environment (Harrison and Johnston, 1985). Common trace metals in highway runoff and their primary sources are shown in table 2.

Table 2. Common trace constituents in highway runoff and their primary sources

[From Kobriger, 1984]

Constituent	Primary sources
Cadmium	Tire wear and insecticide application
Chromium	Metal plating, moving engine parts, and break-lining wear
Copper	Metal plating, bearing and bushing wear, moving engine parts, break-lining wear, and fungicide and insecticide application
Iron	Autobody rust, moving engine parts, and steel highway structures
Lead	Automobile emission, tire wear, lubricating oil and grease, and break wear
Manganese	Moving engine parts
Nickel	Automobile emission, lubricating oil, metal plating, bushing wear, brake-lining wear, and asphalt paving
Zinc	Tire wear, motor oil, and grease
Cyanide	Deicing agents

By comparison with deposition rates of the major elements, the size of the atmospheric-deposition source for the trace metals is less well known. These metals are present in trace amounts in earth crustal materials, but vehicles have large percentages of these elements in their components, even pure sources such as copper wiring, cadmium plating, chromium trim, and substantial presence in gasoline. Although trace metals detected in highway runoff likely come from vehicles, quantifying that source by difference between deposition and runoff is difficult. Low concentrations (microgram per liter level) in deposition and runoff mean the load difference between the two is inaccurate unless great care is taken with precision in sampling methods (Breault and Granato, 2000).

Deicing agents such as rock salt (NaCl), calcium chloride (CaCl$_2$), and sand mixtures can contain impurities such as trace metals and cyanide, which is used as an anticaking agent. Measurable concentrations of Cd, Cr, Cu, Pb, Ni, Zn, and cyanide were detected in samples of deicing salts (Kobriger, 1984; Granato, 1996). Cd, Cu, Fe, Pb, Ni, Zn, and cyanide were found in deicing agents and sand mixtures that were used in Efland, N.C. (Harned, 1988).

The trace metals are important because of their toxicity in aquatic systems. In highway runoff, much of the trace-metal load may be in bioavailable, dissolved form (Bricker, 1999). Even the nondissolved load, however, which accumulates in sediment deposits on stream and lake beds, can be bioactive and is associated with tumors in fish and sediment toxicity (Buckler and Granato, 1999; Rice 1999). The chemistry of trace metals can be complex. In the aerobic aquatic environment, trace metals generally oxidize to cations and may become sorbed to solid phases or precipitate as metal oxides. The interaction between trace metals and organic materials is substantial and may lead to increased trace-metal mobility (Breault and others, 1996).

Character of Deposition Data

With the exception of wet deposition of Hg, deposition data for the trace metals is not available from a national network; therefore, information on rates and variability of trace-metal deposition data is much harder to assess than for the major elements. Another problem with gathering trace-metal deposition data is data quality for trace-metal concentrations, which typically are in the low or sub microgram-per-liter range in wet deposition (Vermette and others, 1995; Willoughby, 1995), or for Hg, in the low nanogram-per-liter range (NADP, 2000b).

Careful selection of collector construction materials and of protocols for cleaning, sample handling, and sample processing is necessary to avoid sample contamination of the trace metals. During the past 25 years, problems of contamination of trace-metal samples during sampling have been recognized. Contamination of Pb samples in oceanography studies was one of the first contamination problems to be recognized. Correction of the contamination problems resulted in estimates of seawater Pb concentrations orders of magnitude less than those previously measured (Patterson and Settle, 1976).

Similar downward adjustments in trace-metal concentration may be necessary for atmospheric-deposition estimates. Trace-constituent data on atmospheric deposition separated according to early studies (1987 and prior) without, and later studies (1993 and later) with, quality-assurance procedures are shown in table 3. The wet-deposition rates for trace metals in earlier studies (Chicago data of Lazrus and others, 1970) are much higher than in later investigations (Vermette and others, 1995; Willoughby, 1995), which may reflect changes in sampling, especially in sampler materials and in sample handling, since the earlier investigations. Changes in actual deposition rates, however, can not be ruled out as a cause for the much higher early deposition rates.

Table 3. Atmospheric deposition rates, for constituents of concern in highway runoff, measured by dry-, wet-, and bulk-deposition collectors and by dry deposition onto a standard grease-covered plate

[All rates are in milligrams per square meter per day. --, no data]

Constituent	Early investigations without quality assurance					Recent investigations with quality assurance			
	Dry deposition	Wet deposition	Bulk collector	Comments	Reference	Dry deposition to standard plate	Wet deposition	Comments	Reference
TRACE METALS									
Cadmium.............	--	--	0.0031	--	Hedges, 1987	0.0050	--	South Haven, Mich.	Holsen and others, 1993
			.0243	near rural highway	Harrison and Johnston, 1985	--	0.0002	Gary, Ind.	Willoughby, 1995
			.0014	far from rural highway	Harrison and Johnston, 1985	--	.0005	1 m precipitation assumed, average 3 sites	Vermette and others, 1995
	--	--	--	--	--	.0055	--	South Lake Michigan	Zufall and others, 1998
Copper.................	--	--	--	--	--	--	.0029	Gary, Ind.	Willoughby, 1995
		0.1800		Chicago, Ill.	Lazrus and others, 1970	--	.0027	1 m precipitation assumed, average 3 sites	Vermette and others, 1995
			.2143	near rural highway	Harrison and Johnston, 1985	.0600	--	Chicago, Ill.	Paode and others, 1998
			.0214	far from rural highway	Harrison and Johnston, 1985	.0100	--	South Lake Michigan	Paode and others, 1998
Chromium	--	--	--	--	--	--	.0115	Gary, Ind.	Willoughby, 1995
Nickel.................	--	.0267	--	Chicago, Ill.	Lazrus and others, 1970	.1270	--	Chicago, Ill.	Lin and others, 1993
						.0100	--	Lake Michigan	Holsen and others, 1993

Table 3. Atmospheric deposition rates, for constituents of concern in highway runoff, measured by dry-, wet-, and bulk-deposition collectors and by dry deposition onto a standard grease-covered plate—*Continued*

Constituent	Early investigations without quality assurance					Recent investigations with quality assurance			
	Dry deposi-tion	Wet deposi-tion	Bulk collector	Comments	Reference	Dry deposi-tion to standard plate	Wet deposi-tion	Comments	Reference
TRACE METALS—*Continued*									
Lead....................	--	--	--	--	--	--	0.0029	Gary, Ind.	Willoughby, 1995
	--	0.4600	--	Chicago, Ill.	Lazrus and others, 1970	0.0700	--	Chicago, Ill.	Paode and others, 1998
	--	--	1.6857	near rural highway	Harrison and Johnston, 1985	.0030	--	South Lake Michigan	Paode and others, 1999
	--	--	.0714	far from rural highway	Harrison and Johnston, 1985	--	.0060	1 m precipitation Assumed; average 3 sites	Vermette and others, 1995
Zinc	--	1.6533	--	Chicago, Ill.	Lazrus and others, 1970	--	.0066	Gary, Ind.	Willoughby, 1995
	2.8493	--	--	--	Barret and others, 1994				
ORGANIC COMPOUNDS									
PCBs	--	--	--	--	--	0.0028 to .0097	--	Chicago, Ill.	Holsen and others, 1991
						--	0.0026	1 m precipitation assumed; 50 km north of Minneapolis, Minn.	Franz and others, 1991
Total PAHs	--	--	--	--	--	--	.0528	1 m precipitation assumed; 50 km north of Minneapolis, Minn.	Franz and others, 1991
1,2,4,Trichloro-benzene............	--	--	--	--	--	--	.0088	1 m precipitation assumed; 50 km north of Minneapolis, Minn.	Franz and others, 1991

Part of the more recent trace-metal wet deposition data in table 3 were from a NADP pilot investigation of trace metals in precipitation (Vermette and others, 1995). The original data were volume-weighted concentrations for six metals in three sites (Illinois, Colorado, and Tennessee). Manganese varied the most among sites by a factor of 5. Cd and Cu varied by only 20 and 34 percent, respectively, but deposition rates for these constituents were very low. The deposition data in table 3 from this investigation were based on these volume-weighted concentrations and assumed annual rainfall of 100 cm (less would apply in Colorado; Smieszek and Granato, 2000). Other wet-deposition rates for trace metals from Gary, Indiana (near Chicago), are available in table 3 (Willoughby, 1995). These rates were measured with appropriate quality-assurance protocol in the heavily industrial Chicago area and may be considered to indicate high rates of wet deposition reliably. Note that even these methods might not prevent contamination for very low-concentration contaminants like Hg. The estimated maximum deposition rate was 0.00018 $mg/m^2/d$, which is three times the maximum rate measured in the 1998 MDN program (NADP, 2000b).

Ambient wet-deposition rates are better known for Hg than other trace metals because of the MDN national Hg deposition network of some 40 sites with excellent quality-assurance procedures. The network data indicate that ambient wet deposition of Hg ranges from 0.00001 $mg/m^2/d$ in Arizona to 0.00006 $mg/m^2/d$ in South Florida (fig. 3). These low Hg wet-deposition rates may represent less than half of total Hg deposition. Much of the two most common forms of Hg in the atmosphere, Hg^0 and Hg^{2+}, is transported to the earth through dry deposition (Lin and Pehkonen, 1999).

As is the case for the major elements, trace-metal deposition may be dominated by dry deposition. In the case of Pb, for instance, dry-deposition measurements made from June to October 1991 were 0.13 $mg/m^2/d$ in Chicago (Lin and others, 1993), whereas a Chicago-area average wet-deposition measurement in 1992–93 (Willoughby, 1995) was 0.0029 $mg/m^2/d$, 45 times less.

Pb is an example of an element whose release into the environment has changed because of environmental laws restricting its use, especially in gasoline (Callender and Rice, 2000; Callender and Van Metre, 1997). Average Pb concentrations in air (from 160 sites nationwide) decreased dramatically during 1978–88 (fig. 4, USEPA, 1998b). The comparisons in the pre-
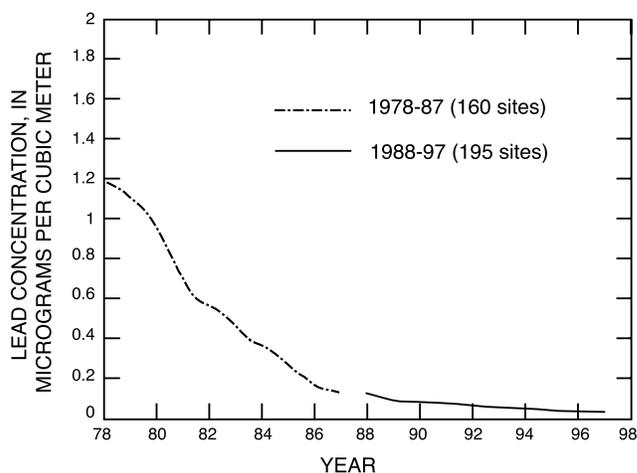
Figure 4. Long-term trend in ambient atmospheric lead concentration in the United States (U.S. Environmental Protection Agency, 1998b).

ceding paragraph, however, concern years (1992–93), during which Pb concentrations in the atmosphere were relatively stable.

Dry-deposition rates depend on particle size. Lin and others (1993) determined that although Pb concentrations were higher on smaller particles than on larger particles, particle flux for Pb was dominated by dry deposition of large-sized particles because deposition velocities for the large-size particles were much greater than deposition velocities for small-size particles. Investigation of dry deposition over Lake Michigan indicated that these relations may be seasonal. Zufall and others (1998) determined that deposition was dominated by large-sized particles (greater than 8 μm) during July 1994 but was dominated by smaller particles (4–8 μm) during January 1995.

Dry-deposition rates can vary dramatically from urban to rural areas and can be extremely low in rural areas. An investigation of deposition of three trace metals (Cu, Pb, and Zn) onto a standard surface in environments around Chicago (Paode and others, 1998) indicated that dry fluxes for all metals were greatest in Chicago, less in coastal South Haven, Michigan, and least over the water in Lake Michigan between Chicago and South Haven. Highest dry-deposition rates were 0.17 and 0.6 $mg/m^2/d$, for Cu and Zn, respectively; low rates were 0.003 and 0.005 $mg/m^2/d$, for Pb and Cd, respectively (table 3).

Finally, the extremely high trace-metal deposition rates associated with smelter operations should be noted. A recent investigation of dry deposition

downwind from a Zn-Pb smelter in Australia, using some quality-assurance procedures such as duplicate deployment, sample spikes, blanks, and reference materials, determined median rates of 19, 22, 12, 0.61, 0.40, and 0.052 mg/m^2/d for Pb, Zn, Fe, Cu, As, and Cd, respectively (van Alphen, 1999).

Uses in Highway-Runoff Investigations

Toxicity of the trace metals is the principal reason for investigating their deposition to highway surfaces. The wear of vehicle metal parts and tires is an important potential source of trace metals, and elevated concentrations (above background levels) of trace metals are measured in highway runoff. Trace metals, including isotopes, also may be investigated to help determine the relative contribution of ambient and vehicle sources to highway runoff. For these kinds of investigations, trace elements must be present in ambient-deposition sources, but not present in vehicle-related sources.

ORGANIC COMPOUNDS

Deposition of organic compounds is reviewed in this section. Organic compounds in deposition originate from vehicle and industrial sources.

Sources, Importance, and Chemistry

Vehicle emissions are a well-documented source of volatile, semivolatile, and other organic compounds to the atmosphere (Harrison and Johnston, 1985; Westerholm and others, 1988; Gardner and others, 1995). Organic compounds are common constituents in highway runoff (Lopes and Dionne, 1998). Volatile organic compounds tend to partition from the particulate and aqueous phases to the gaseous phase. Semivolatile organic compounds generally partition toward the particulate and aqueous phases. Semivolatile organic compounds, therefore, usually are deposited near the highway (Harrison and Johnston, 1985).

Polycyclic aromatic hydrocarbons (PAHs), which are semivolatile organic compounds, and their derivatives are among the organic compounds of most concern because of their effects on human health

(Harrison and Johnston, 1985). Harrison and Johnston (1985) determined that vehicle-generated PAHs were deposited primarily within 15 m of the highway. Elevated PAH concentrations (above background levels) at distances greater than 30 m from a highway usually were measured in winter and early spring, coinciding with the combustion of fossil fuels for domestic heating (Harrison and Johnston, 1985). Methyl *tert*-butyl ether (MTBE) is a volatile organic compound made from methanol and used as a gasoline additive to improve air quality in urban areas. The compound can have adverse human health effects, however (Squillace and others, 1995). The amount of MTBE released from motor vehicles is unknown but could be an important source of MTBE to the environment (Squillace and others, 1995).

Organic-compound pesticides have been recognized as potential air pollutants since 1946 (Daines, 1952). Before the 1960s, atmospheric deposition of pesticides generally was thought to be a local problem caused by spray drift (Majewski and Capel, 1995). Long-range movement of pesticides was thought to be minimal because their volatility and solubility in water are low. The detection of DDT (dichloro-diphenyl-trichloroethane) and other organochlorine compounds in fish and mammals in the Arctic and Antarctic proved that pesticides can be transported over long distances (Cade and others, 1968). Almost all pesticides investigated have been detected in air, rain, snow, or fog in the United States at various times of the year (Majewski and Capel, 1995). Pesticides, especially herbicides, can be applied to or near roadways. Harned (1988) measured elevated levels of the herbicide 2,4-D [(2,4-dichlorophenoxy)acetic acid] in a stream near a highway about 2 weeks after the herbicide was applied to weeds along the roadway.

Character of Deposition Data

Concentrations of organic pollutants in precipitation were measured in 1986 by Franz and others, (1991) at a site 50 km north of Minneapolis, Minn. If precipitation were 100 cm per year, polychlorinated biphenyl (PCB), phenanthrene, and *p,p′*dichloro-diphenyl-dichloroethylene (*p,p′*-DDE) wet-deposition rates would be 0.0026, 0.0086, and 0.00073 mg/m^2/yr, respectively. Results for other polyaromatic hydrocarbons (PAHs) and chlorinated compounds are in table 3.

The amount of PCB dry deposition to a standard surface has been investigated in Chicago. The estimated PCB flux varied between 0.0028 and 0.0097 mg/m^2/d, which was up to three orders of magnitude greater than that estimated to fall in nonurban areas (Holsen and others, 1991). Unlike the trace metals, these PCB dry-deposition values do not differ greatly from those for wet deposition.

Uses in Highway-Runoff Investigations

Excluding those organic compounds with roadway uses, such as the herbicide 2,4-D, investigations of some of the contaminant compounds might be useful for elucidating the on-highway processes that determine how much of what is measured as ambient deposition in collectors falls onto highways and moves into highway runoff. Although vehicle fluids and exhausts are sources of organic compounds to the atmosphere, some of the pesticides and PCBs are unlikely to be of vehicle origin. Unlike the major elements or the trace metals, these manmade compounds are not found in nature and their molecular structures are specific enough not to occur except through intentional synthesis. Manufacture and use of PCBs have been prohibited for over 25 years so that they are no longer part of vehicle materials or fluids. PCBs get into the atmosphere mostly in vapor form and may adhere to distinctly different particles and follow different atmospheric-transport processes than do the major elements or the trace metals.

REVIEW OF ARTICLES ON ATMOSPHERIC SOURCES FOR HIGHWAY RUNOFF

Atmospheric deposition has been a subject of many highway publications (table 5, at back of report). These include studies in which collectors were deployed transverse to the highway to determine the effect of traffic on atmospheric deposition and traffic-related processes of saltation and resuspension. In the highway-runoff context, however, atmospheric deposition is of interest chiefly as a source for constituents in runoff. Many highway-runoff studies result in questions about, or are designed to determine, which part of highway runoff comes from ambient atmospheric

deposition and which comes from traffic. These sources may covary because both traffic volume and ambient atmospheric deposition increase across the gradient from rural to urban land use.

Comparison of Traffic-Related and Ambient Atmospheric-Deposition Sources

Whether vehicles or ambient atmospheric deposition is the dominant source of runoff constituents is an important topic not yet resolved for many constituents measured in highway runoff. If traffic sources dominate, runoff loads would be similar for all highways, or proportional to traffic volume. If traffic contribution were small, runoff loads would depend on local atmospheric deposition, which can differ greatly along the gradient from rural to urban land use, particularly for dry deposition. For some constituents, both ambient atmospheric deposition and traffic sources may make large contributions, or their relative contributions might depend on land use. For instance, ambient air may be a problem for a given constituent in urban areas or near a smelter, but usually not in rural areas.

Deposition Rates and Highway Proximity

One way to determine the relative importance of traffic-related and ambient atmospheric-deposition sources is through examination of deposition rates at sites at different distances from the highway. Deposition fluxes of Cd, Cu, and Pb were determined to be highly elevated (6, 13, 33 times, respectively) in bulk samplers within 20 to 40 m of a highway in England in comparison with background levels measured 40 to 220 m from the highway (fig. 5) (Harrison and Johnston, 1985). Deposition rates for PAHs, which are a combustion product of fossil fuels, would be expected to decrease with distance from highways. The Harrison and Johnston (1985) investigation in England found that PAH deposition increased as the distances of measurement sites from their investigated highway decreased; otherwise, the highway had clean air blowing in off the Atlantic Ocean. Harrison and Johnston (1985) concluded that roadway-derived PAH was deposited primarily within 25 m of the road and was 100 times greater than background levels. The highest deposition rate recorded was 0.0010 mg/m^2/d. PAHs
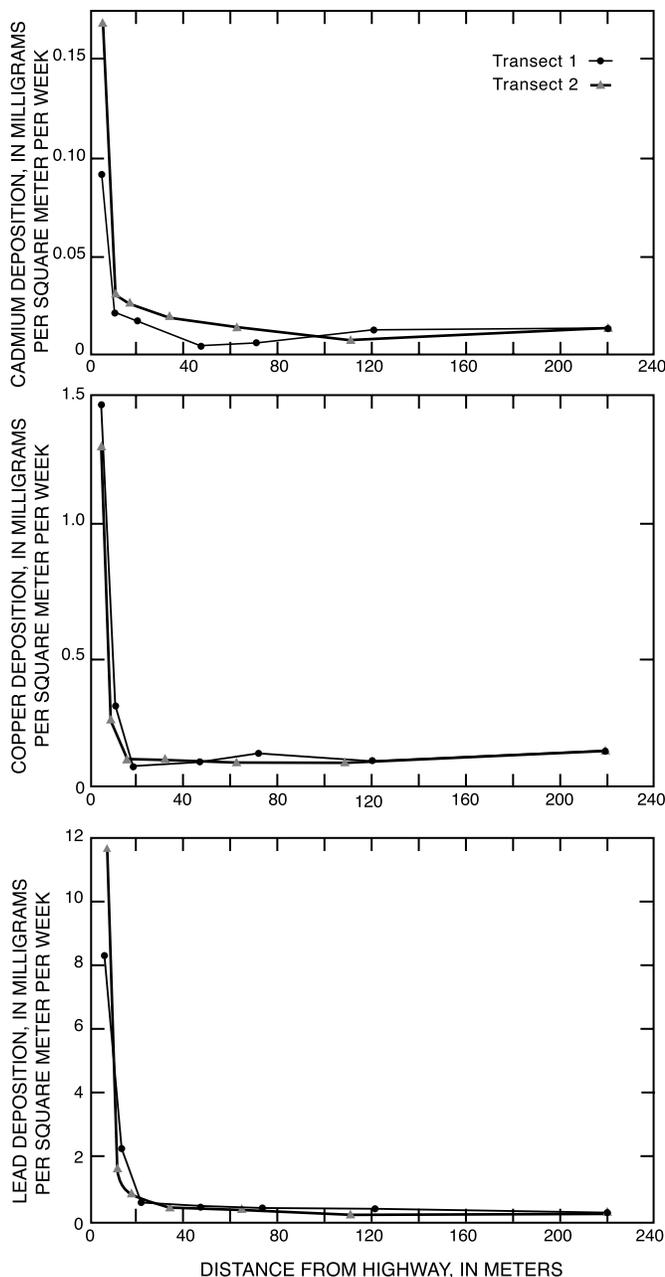
Figure 5. Average bulk deposition rate of trace metals for the period from October 14, 1981, to June 15, 1983, along two transects with distance from a rural highway in England (Harrison and Johnston, 1985).

are produced by non-vehicle sources of fossil-fuel burning as well, and the rate of deposition from urban air can be higher than the rural, near-highway rate (Harrison and Johnston, 1985).

Similar decreases (as PAH) in concentrations of trace metal as the distance from the roadway increases were observed in studies by Hedges and Wren (1987),

Warren and Birch (1987), and Hewitt and Rashed (1990). Atmospheric deposition of highway-generated total particulate matter (TPM) and associated constituents onto areas adjacent to roadway surfaces appeared to be related to average daily traffic, wind speed and direction, available surface load, and terrain and landscape features (Kobriger, 1984). Harned (1988) compared dustfall by dry deposition between a site on Interstate 85 in North Carolina and a site about 0.5 mi from the highway. Land use at the highway site was 100 percent transportation and utilities. Land use at the background site was predominantly mixed forest, cropland, pasture, and open space. Samples collected in dust buckets at the highway site were compared with samples collected in a wet-dry collector at the background site. By adding the wet and dry fractions collected at the background site, Harned (1988) concluded that the proportion of the total loading attributable to background conditions was unexpectedly high, ranging from 42 percent for Zn to 93 percent of the total particulate loading rate determined for the highway site. Because there were no other apparent sources for these constituents in the area, this high proportion of the total dustfall loading rate at the background site suggested that dust from highways can be carried considerable distances. Harned (1988) compared trace-element loadings in bulk-deposition samples collected at the background site with samples collected by dust buckets located near the highway and observed that median loadings computed from the background samples were smaller than loadings computed from highway samples. Harned (1988) also showed, by analyzing samples from soil and from dust buckets arranged perpendicular to the road, that the highway was a source of contaminants in local atmospheric deposition and that concentration levels of all the monitored trace metals decreased as distance from the road increased.

Near-highway transects of soils and deposition are important ways to determine relative importance of highway and atmospherically derived sources. For constituents collected using appropriate methods, deposition measurements may be better because they are not subject to leaching loss, as would be soil concentration measurements. The studies cited showing that deposition rate decreases with increasing distance from the highway indicate that highways are large sources of these contaminants relative to atmospheric deposition. Potential problems with these investigations may be present because measurements are taken from dust buckets or bulk collectors. These methods

of collection have uncertain quality-assurance procedures and uncertain collection efficiencies that may depend on turbulence conditions, which could be traffic-related.

Lower background dry deposition (Holsen and others, 1993) and wet-deposition values (Willoughby, 1995) have been measured in investigations with appropriate collector construction and quality-assurance programs in environments more urban than that of the Harrison and Johnston investigation (table 3). Thus, the 33-fold difference for Pb measured by Harrison and Johnston (1985) may be still greater if clean methods had been used to collect the background sample. The effect of uncertain collection efficiency might be to either over or under sample deposition and could, thus, increase or decrease the measured gradient.

Mass Balance

Mass balance is another approach used to separate vehicle and ambient-deposition contributions to highway runoff loads. Direct measurements of vehicle contributions to roadways are difficult to make because ambient and vehicle-related contributions at the roadway cannot be separated. Direct contributions from ambient deposition, however, can be quantified by use of an atmospheric-deposition collector removed a suitable distance from the road. Many investigators have used a mass-balance approach to compute the vehicle deposition by subtracting atmospheric deposition from runoff loads. The method has a major flaw if actual atmospheric deposition to the highways is different from that determined by the collector.

Barrett and others (1995) in their literature review on quantity and control of pollution from highway runoff and construction state that "atmospheric sources contribute a significant amount of the pollutant load in highway runoff." Cited were (1) Irwin and Losey (1978) with ambient bulk deposition versus runoff values for 15 runoff constituents for a highway bridge in Florida—contributions to bridge runoff from bulk collection ranged from 18 percent for Pb to 38 percent for Cu to 490 percent for Na; (2) Bellinger and others (1982) on precipitation loads to highway runoff for major ions (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, and SO_4^{2-}), which were actually small, averaging 2 percent, and total solids, which averaged about 10 percent; and (3) Gupta and others (1981) on dustfall, who reported deposition rates ranging from 130 to 900 mg/m^2/d at Federal highway monitoring sites. Barrett and others (1995) state that "if all the dustfall remained on the highway, dustfall load would approximately equal the load in the runoff."

Working with data from 31 urban catchments across the United States, Ebbert and Wagner (1987) determined percentage of runoff load that came from rainfall for 12 constituents. For example, 40 percent of N, 6 percent of Pb, and 2 percent of suspended solids measured in runoff was from rainfall input. Again, these values assume conservation during transport in the catchment. A complete mass-balance approach was taken by Kobriger and others (1982) for a rural highway site in North Carolina. Atmospheric fallout was 8 percent of the total material loading the highway surface, with vehicle deposition and highway maintenance (including sanding and salting) of 25 and 67 percent, respectively. Vehicular deposition was calculated as the difference between the highway-runoff load, and the salting-sanding and atmospheric load.

Problems may result with the mass-balance approach. Implicit in the mass-difference calculation to compute traffic contribution is the assumption that the atmospheric contribution is transferred to runoff with 100 percent conservation. But a contaminant may precipitate in the drain or catch basin, or be subject to resuspension and deposition off the highway. A large percentage of dry deposition, which composes a large proportion of trace-metal contaminants, apparently would be subject especially to resuspension (as reviewed under "Forms of Deposition and Post-Depositional Transport"). Likely, a large percentage of deposition from both vehicles and ambient atmospheric sources is moved off highways by resuspension, in which case comparisons of measured atmospheric loads with runoff loads leads to incorrect values for traffic loads.

Sample contamination during sample collection and processing could add to uncertainty in the mass-balance calculation. Much of the deposition data has been collected with methods that are known to cause contamination problems at the concentrations that might be expected in the samplers (Patterson and Settle, 1976; Windom and others, 1991). When samples for precipitation have been collected with clean techniques, concentrations have been lower than those without the clean-techniques procedures (table 3).

In another mass-balance approach to determining sources of highway runoff, the assumption has been made that ambient atmospheric loadings are the

difference between highway runoff loadings measured in urban and rural settings. Various investigators have implied that ambient-atmospheric inputs can be important even for those pollutants (trace metals) that would appear to be dominated by traffic. Driscoll and others (1990a) reported that the most important factor that affects the types and amounts of contaminants contributed to highway runoff is the surrounding land use. Constituent loadings in areas where the dominant land use is industrial have been reported to be substantially greater than in areas where the dominant land use is residential or commercial (Gupta and others, 1981; Schroder and others, 1989; Driscoll and others, 1990a). Results from highway runoff studies conducted in the early 1980s indicate that total particulate matter measured in bulk deposition collected at urban sites contained approximately four times greater loadings than total particulate matter collected at rural sites. Similarly, trace-element bulk deposition was greater at background urban sites than at background rural sites (Dannecker and Stechmann, 1990; Smith and Lord, 1990). Complicating this approach are traffic and atmospheric sources, both of which are less in rural areas than in urban areas.

Regression Models of Federal Highway Administration Data

Another means of attributing runoff constituents to sources is through regression equations. A fundamental mass-balance consideration in atmospheric sources to highway runoff concerns the precipitation/runoff water balance. Regressions of water balances of precipitation and by the Federal Highway Administration (Driscoll and others, 1990a) indicate that about 80 percent of the water that falls onto impervious sections of highways runs off. The relation relates runoff from a given surface area to precipitation on the surface area as a function of percent of surface area that is impervious and given as

Runoff = Precipitation [0.007 (Percent impervious surface area) +0.1] .

Of the 20 percent lost from impervious surfaces, presumably some water evaporates and some splashes off the highway. These processes, of unknown relative magnitude, would act with contrasting effects on wet-deposition constituents. Evaporation would concentrate nonvolatile constituents in the water, resulting in no constituent loss in runoff, whereas splash loss removes both water and constituents in the water from runoff.

Regressions of data from 10 years of investigation supported by the Federal Highway Administration (Driscoll and others, 1990a) indicated that simple models that simulate trace-metal pollutant concentrations in runoff were not possible. Event mean concentration (EMC) of each of the monitored constituents during a storm were regressed first against storm size and then against average daily traffic level (ADT). Lack of correlation against storm or runoff volume was taken to "eliminate any concern for possible bias because of differences in the size of monitored storms at different investigation sites." Initially, the analysis indicated that EMC was less for rural sites compared to urban sites for each of the pollutants monitored. ADT also was less at the rural sites and actually was used to distinguish between rural (less than 30,000 vehicles per day) and urban (greater than 30,000 vehicles per day) classifications. Subsequent regression of EMC and ADT was applied only to higher ADT-level urban data. Most of the latter regressions resulted in low degrees of correlation. Some of the regressions, such as for phosphate and nitrate EMC, had negative slopes leading to the counterintuitive results that more traffic results in less pollution. An exception was the regression result for Zn. With one outlier removed, the Zn correlation was positive with an r^2 of 0.70. The outlier, with an EMC almost three times the next highest value, was associated with a site downwind from a Zn smelter.

Driscoll and others (1990a) concluded that the explanatory variable database (that is ADT) was not sufficiently large for regression analysis to develop general relations that could be used to simulate pollutant loads in highway runoff. The data for the urban regressions came from some 24 highway sites throughout the continental United States. Given the degree of climate variation across the country, an argument for insufficient explanatory variables sounds plausible. Driscoll and others suggested explanatory variables should include climate variables, atmospheric deposition and removals, highway-site situations, operational situations, and surrounding land-use characteristics. The investigation emphasized that the "runoff quality differed significantly between highways in urban areas versus those in rural areas." Traffic densities are markedly different between these two categories of surrounding land use, but the lack of a clear correlation of trace-metal EMC with ADT within each grouping led to the conclusion that the general atmospheric-quality

differences between urban and rural areas were the most important effect. Cited also was an investigation in Germany that concluded ADT is not a dominant factor and that the characteristics of the surrounding area are a more important effect (Stotz, 1987).

Other causes of the lack of a correlation should be considered, especially that of questionable data. No amount of explanatory variables from an expanded data-collection program will improve the regressions between trace-metal concentrations and ADT if the runoff data are biased or not of comparable quality. For instance, variability in the Hg runoff data was extreme. EMC Hg values at the urban sites ranged from 1 to 1,500 ng/L. This variation was too large for vehicles to be the sole cause, because ADT varied only by a factor of three. Variation that occurs nationally in atmospheric deposition of Hg also is small. As measured by the MDN, wet-deposition Hg varies across the country by only a factor of two. Hg is difficult to sample even in relatively high-concentration environments because of sample contamination during the sampling and sample processing (Bloom, 1995). Unless extraordinary sampling methods are used, such as described by Colman and Breault (2000), much higher concentrations of Hg would be attributed to runoff than actually are present. Extraordinary trace-metal sampling methods, including strict quality-assurance procedures and sampling using a clean hands-dirty hands protocol, were not used in the highway-data collection.

Even without quality-assurance procedures, the EMC/ADT regression indicated Zn concentration in runoff was related to traffic volume. Zn is a component of tires and should provide a strong traffic-related signal in runoff. Also, Zn is the trace metal generally measured at highest concentration in runoff investigations. Zn sampling was subject to the same lack of quality assurance and clean technique as the other uncorrelated trace metals. Perhaps the Zn signal is strong enough in runoff sampling to be observed despite contributions from possible contamination.

Lack of quality assurance would obscure an EMC/ADT correlation actually present. Alternatively, concentrations in runoff actually may not be correlated to traffic volume. Factors contributing to a lack of correlation for trace metals might be that traffic is both a source of trace metals and of turbulence that potentially moves the metals off the road surface onto adjacent areas. The turbulence causes resuspension that can substantially remove large-sized particles. The large-size particles are not transported far before being redeposited, as reviewed above. Thus, an active highway

surface with no pollutant particles may be bounded by an enriched zone on the breakdown lane and (or) berm. This potential transport mechanism has been confirmed by various studies indicating that highways with curbs, which restrict horizontal transport of resuspended particulates, have higher pollutant loads in runoff than noncurbed sections of highway (Gupta and others, 1981; Kobriger and Geinopolos, 1984; Driscoll and others, 1990a). If included in the area drained, substantial runoff load could be generated by the adjacent off-road area. Data available in the investigation by Driscoll and others were not controlled for amount of the adjacent off-road area being drained. With high deposition rates expected in the off-road areas, the variable inclusion could greatly alter the correlation between traffic and runoff loads.

One final consideration concerns aggregation of data collected over long time periods. Emissions to the atmosphere of some constituents have changed in response to emissions control programs of the USEPA (USEPA, 1998b). There have been substantial changes in emissions of the criteria pollutants from vehicle exhausts during the past 27 years (fig. 6) (USEPA, 1999).
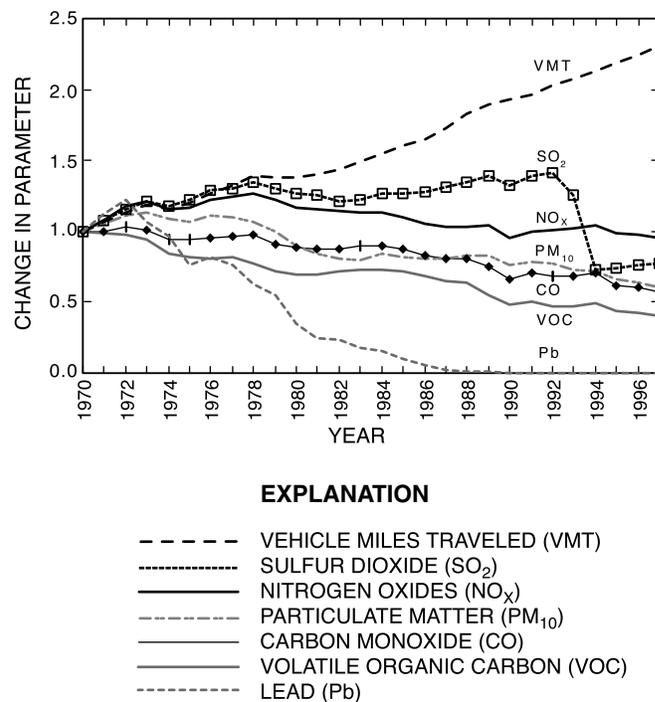

Figure 6. Change in criteria pollutant emissions from vehicles compared to vehicle travel, 1970–97 in the United States (USEPA, 1999).

On-Highway Processing

In general, little is known about on-highway processing of deposited materials. Greater knowledge of this area likely would lead to better simulations of constituent loads in highway runoff.

Two important on-highway processes, saltation and resuspension, were reviewed under "Forms of Deposition and Post-Depositional Transport." These processes relate to one prevalent on-highway processing theory: the number of dry days between large storms events (the antecedent dry period) can have a major effect on airborne particulates that build up on roadways. Associated with this theory are the concepts that light to moderate precipitation events of low intensity or intense rains of short duration do not clean the roadway completely, and that airborne particulate amounts increase at relatively constant rates for pollutants such as Pb, Br, and Ca (Moe and others, 1982). This buildup process, however, may be more associated with impervious surfaces in urban catchments than with active roadways. Resuspension of material deposited by traffic could greatly alter the effect of storm interval on runoff loads from travel lanes compared to nontravel lanes or other impervious surfaces. Investigations of runoff from travel lanes of a highway in Boston, Mass. (K.P. Smith, USGS, written commun., 2000), indicate no association of runoff of total-solids load with interval between storms. Results of the Boston investigation indicated that most material moves when there is heavy precipitation and soil adjacent to the highway erodes onto the impervious, drained, travel lane. (There are no breakdown lanes between the soil and the travel lanes at the site.)

The effect of turbulence on redistribution of highway sediment was described in Kobriger and Geinopolos (1984). As determined from sweeping and flushing studies, solids tend to accumulate in the break down and median lanes. Kobriger and Geinopolos (1984) conclude that runoff loadings come from these nontravel lanes rather than from travel lanes. The importance of on-highway processing was recognized by at least one investigator who has divided highway-runoff constituents into three categories: (1) constituents, such as suspended solids, that are affected by conditions during dry periods and that may be mitigated by dry-period activities; (2) constituents that are affected mostly by conditions during rainfall events and that may be mitigated only by use of runoff controls; and (3) constituents that are affected equally by dry and wet conditions (Irish and others, 1996).

The duration of a storm may interact with traffic turbulence and the deposition process of washout to affect highway-runoff quality substantially. In a Seattle, Washington investigation, constituent loads in highway runoff could be simulated by average traffic volume measured during the course of a rain event (Chui and others, 1982). Loads appeared to be more a function of kinetic energy provided by the moving vehicles than of rainfall. Washout by vehicle turbulence during wet weather might be particularly effective at trapping some of the gases exhausted from car tail pipes. Otherwise, as noted by Harrison and Johnston (1985), various major compounds that are present in vehicle exhaust, including oxides of nitrogen and carbon monoxide, are gases that rapidly and widely disperse, leaving no permanent record near the road.

Quality Assurance

Several of the investigative approaches described in this report would be compromised if concentrations measured at the low end of the concentration range were not accurate. If rates are being computed, a small absolute difference in concentration can lead to a large relative difference in deposition rate and in conclusions resulting from the application of the rate. The highway-runoff investigations completed in the 1970s and 1980s do not appear to have been conducted using clean methods to collect precipitation data or highway-runoff data. For example, no mention is made in the documentation regarding use of Teflon in sampler construction, cleaning of samplers before deployment, clean rooms or benches where samplers and samples are processed, or precautions used to transport samplers after cleaning. Nor is any verification generally given on sample integrity by reference to analysis of equipment blanks, trip blanks, standard reference materials, sample spikes or blind sample analysis, all usual quality-assurance procedures (Jones, 1999).

One quality-assurance procedure, use of multiple bulk samplers at a site to determine precision, was applied at a Milwaukee, Wisconsin, site in work

reported by Kobriger and others (1982). Groups of three samplers placed 0, 5, 15, 30, and 300 m from the highway were used to compute relative error (standard deviation divided by mean) for the measurements collected at each distance from the highway. The relative errors ranged from 2 to 93 percent. Mean relative error was about 20 percent. These results and other measures of quality assurance would be useful in future investigations to assess sources of variation in runoff data and to establish the quality and comparability of data collected at different sites.

STRATEGIES AND METHODS FOR FUTURE INVESTIGATIONS

Future investigation designs for atmospheric-deposition contributions to highway runoff should consider that currently little is known conclusively about the atmospheric source or how deposited material is transported from the highway to runoff. As discussed in the section "Review of Articles on Atmospheric Sources for Highway Runoff," uncertainties include measurements of atmospheric-deposition rates, especially in the highway investigation context, because virtually all wet and dry data have been collected with minimal quality-assurance procedures, and all dry data have been collected using bulk or dry-bucket samplers. In addition, the runoff database from projects sponsored by the Federal Highway Administration likely is not accurate for Hg and perhaps other trace metals because of lack of clean-sampling methods and application of quality-control procedures.

Even the correspondence between accurate dry-deposition rates, such as determined from deposition to standard surfaces, and net deposition to the highway that would be available for runoff is an area of uncertainty. Unlike most pavement surfaces, highway travel surfaces underlie a high-energy, turbulent environment because of traffic-induced air turbulence. The effect of air turbulence on highway-deposited materials generally is not considered in investigations of sources of highway runoff. Potential redistribution of particles by moving vehicles complicates determination of the part of a given constituent measured in highway runoff that can be attributed to atmospheric deposition. In

summary, on-highway processes that affect deposited material are poorly understood; these processes include effects from resuspension and evaporation. This poor understanding, in turn, has prevented accurate determinations of ambient atmospheric-deposition contributions to highway runoff loads in mass-balance calculations.

Given the effect that vehicles have on air quality in general, and on contributions to highway surfaces from vehicle wear, the contribution from vehicles to highway-runoff loads is likely to dominate over ambient atmospheric deposition for many pollutant constituents. For those constituents in runoff predominantly from vehicle sources, the details of ambient atmospheric deposition and processing may not be important, and investigations could be narrowed to vehicle-related sources. Much of what follows are strategies and techniques to determine whether ambient deposition sources are important for given constituents and to separate out the ambient deposition contribution to highway-runoff loads.

Particle Size and Sources

The potential importance of particle size for deposition and on-highway processing of deposited materials, and thus for connections between sources of deposition and highway runoff, already has been emphasized in this report. Multiple conceptual models for particle-dependent processing are possible. For some constituents, large-size particles that are subject to resuspension may dominate deposition. For other constituents, small particles that are removed by traffic turbulence or a combination of turbulence and precipitation may dominate. At present, it is not known which of these or other particle-dependent conceptual models may be dominant or acting in combination for each constituent.

Because of the complexity of processes operating on the highway surface, verification of a conceptual model may be difficult. Alternatively, tracers associated with one particle size or loading source could be used to indicate the net effects of on-highway processing of deposited materials.

Investigative Design

An initial investigative strategy could be to work out net highway processing for ambient-deposition constituents as a function of domination by large or small-sized particles. The fraction of total deposition of a constituent that is recovered in runoff could be related to the particle size that dominates deposition of the constituent, where total deposition is measured as the sum of wet deposition and dry deposition onto a standard surface removed from direct influence of the highway. Constituents would need to be chosen that were not contributed by highway maintenance procedures (including deicing) or vehicles. The data on relative deposition rates as a function of particle size would need to be obtained from literature values or modeled on the basis of deposition velocities, and site data on particle size and constituent concentration.

Once percent recovery as a function of particle size is known for constituents present only in ambient deposition, runoff loads from ambient deposition could be computed for other constituents with multiple (including vehicle) sources, such as Zn or Cu, from the equation

$$F_{rl} = (X_d Y_l)/(Y_d X_l),$$

where

F_{rl} is fraction of constituent X in the runoff load attributable to ambient deposition;

X_d (mg/m^2/d) is the ambient deposition rate of constituent X measured by wet deposition and standard surface collectors,

X_l (mg/m^2/d) is the load of constituent X in runoff from the highway;

Y_d (mg/m^2/d) is the deposition rate of ambient sole-source element measured by wet deposition and standard-surface collectors; and

Y_l (mg/m^2/d) is the load of ambient sole-source constituent in runoff from the highway.

Investigations of this type possibly could eliminate contributions of many constituents from ambient atmospheric deposition from further consideration. If, for example, more than 90 percent of a constituent in highway runoff is from nonambient deposition sources, further investigation of the ambient source may not be warranted, even in an urban environment.

Tracers of Atmospheric Deposition

Determination of the ambient-deposition fraction of runoff requires measurements of deposition and runoff of elements that are present in atmospheric deposition but not in vehicle or road-maintenance sources. Various isotopic tracers of atmospheric deposition (Pb-210 and Be-7) have been used in erosion and runoff investigations as a means to distinguish among sediment sources (Schroeder, 1995; Baskaran and others, 1993). Neither isotope would be likely to originate from nonatmospheric sources. Pb-emission sources to the atmosphere in the Straits of Dover have been identified using isotopic-ratio signatures (^{206}Pb/^{207}Pb, Deboudt and others, 1999). To use this ratio in tracing ambient deposition, knowledge of ^{206}Pb/^{207}Pb in highway or vehicle sources would be necessary.

Possible tracers in addition to the isotopes are the rare earth elements, major elements, or organic constituents such as pollen, or PCBs, which are not associated with traffic sources. The rare earth elements (such as the crustal element scandium) might act as good tracers of particles that dominate trace-metal ambient deposition. The presence of these elements in traffic sources has not yet been studied. Cost-effective analysis of their concentrations can be made using inductively coupled plasma-atomic emission spectroscopy (Fishman, 1993). Utility of a tracer would depend on local conditions. Use of the major elements, Na, Cl, Ca, K, would be appropriate only for road locations where deicing salt was not applied.

Appropriate Network for Data Collection

The general lack of accurate deposition information may indicate a need for a new investigative program to generate correct deposition data sets, especially for trace contaminants, measured across the country. The Federal highway system extends nationally, and deposition is likely to vary nationally, so only a national deposition program could provide the atmospheric source data needed. Until ambient deposition has been clearly established as a major source of pollutants in highway runoff, however, a national program modeled, for example, on the NADP network may not be warranted. An initial network that would determine

whether constituents were important in ambient deposition should be installed at highway sites investigated so that deposition and runoff can be compared as directly as possible.

Site Selection for Sampler Placement

Collection-site criteria established by the NADP for wet acidic deposition, as determined from the National Trends Network Site Operation Manual (National Atmospheric Deposition Program, 2000c), are appropriate for sites established to measure deposition to highways. These criteria include the requirement that sites be clear in all directions of any obstruction that extends upward more than 30 degrees from the horizon. Vegetation should be representative of the area and distance from atmospheric emission sources should be considered. Data-collection sites should be in the same direction with respect to prevailing winds from sources as are the highway sections in the investigations.

Duration and Frequency of Data Collection

In general, the duration of atmospheric-deposition studies conducted at a site should be a minimum of 1 year to detect seasonal variations in concentrations of the constituents of interest. For example, concentrations of pesticides in rain and air frequently are elevated (above background levels) during the warm-temperature months, which coincide

with application times (Majewski and Capel, 1995). Long-term sampling programs (longer than 1 year) are more desirable than short-term programs (less than 1 year) because interpretation of meteorological data for a single year can be misleading (Likens and Bormann, 1995). Long-term trends in deposition of specific constituents and the effects of climatic variations can be identified only in long-term data sets. Volume-weighted major-ion concentrations varied by as much as a factor of eight (for Na) among various major ions monitored in wet deposition at Catoctin Mountain, Maryland, for 9 years (Rice and Bricker, 1996, table 4). What may appear to be a seasonal pattern in a short-term data set may be insignificant in a long-term data set. Likewise, a seasonal effect may be minimized in the short-term data set and prove to be significant during a longer time period. Long-term data sets for a highway that has undergone an increase in traffic density also would be useful to determine the effect of the increased highway-runoff loads.

The sampling interval, which is the time between activation of the atmospheric-deposition sampler and retrieval of the sample, needs to be documented for all studies. Wet-deposition samples can be collected on an event basis, or, more commonly, at daily or weekly sampling intervals. Few studies have been conducted with sampling intervals longer than 1 week because of quality-control concerns (Likens and Bormann, 1995). Collection of dry-deposition samples requires short sampling intervals to reduce the amount of debris, such as leaves or bird droppings, that enters the collector.

Table 4. Annual volume-weighted mean concentrations of chemical constituents in precipitation collected from the U.S. Geological Survey precipitation-collection station at Catoctin Mountain, Maryland, 1982–91

[From Rice and others, 1993. All values in microequivalents per liter, except where noted]

Year	Precipitation (inches)	Calcium	Magnesium	Sodium	Potassium	Chloride	Nitrate	Sulfate
1982	32.86	8.51	2.10	9.95	2.68	11.20	23.51	45.30
1983	54.76	11.86	3.40	7.88	1.02	16.48	23.04	51.19
1984	56.65	13.17	2.39	11.01	1.88	10.35	21.68	58.79
1985	40.41	6.92	3.37	8.90	3.51	10.25	19.07	43.18
1986	36.72	10.73	2.22	6.01	1.85	10.80	24.64	54.78
1987	42.77	8.06	2.12	7.14	2.61	14.77	27.56	60.71
1988	36.28	8.67	2.51	5.24	2.28	9.99	25.30	61.73
1989	40.87	3.51	1.04	1.37	1.35	4.89	20.00	44.00
1990	49.65	7.33	2.71	6.93	2.35	10.75	23.24	49.55
1991	37.70	9.78	4.31	2.28	0.71	9.79	25.39	56.54

Ancillary Data Requirements

Recording the amount of precipitation is mandatory for making comparisons of concentrations of specific constituents nationwide. Types of rain gages include wedge and tube, weighing bucket, and tipping bucket. Other useful ancillary information for atmospheric-deposition studies includes the ambient wind speed and direction (from an anemometer) and the daily temperature fluctuation (from a thermistor). Electronic data loggers (such as the CR10 series of Campbell Scientific Instruments, Inc.) are useful for recording these types of ancillary information as well as digital output from recording rain gages (Church and others, 1999).

Collector Type and Installation

Collector types and installation are described in this section. Collectors of wet and dry deposition are considered.

Wet-Deposition Collectors

Several commercially manufactured wet-deposition collectors are available. Most of these collectors have a large collection area that drains into a bucket or through a funnel into a collection bottle or extraction cartridge (Majewski and Capel, 1995). Wet-deposition collectors are closed during dry periods to exclude dry deposition and open automatically when a sensor detects precipitation. If the collector is left open and is not cleaned just prior to a precipitation event, bulk deposition is collected (Majewski and Capel, 1995). Franz and others (1991) have reviewed the collection efficiencies of commercially available wet-deposition collectors. Modifications to commercially manufactured collectors may be necessary for the collection of specific constituents. For example, modifications were made to a standard Aerochem Metrics Model 301 wet/dry collector for the collection of major ions and trace elements in the heavily industrialized Gary, Indiana, area (fig. 7). Most of these modifications, which include the use of Teflon parts, were made to decrease possible trace-metal contamination of the sample from the sampler (Willoughby, 1995).

For collection of major inorganic ions in precipitation, the polyethylene bucket that comes with standard atmospheric-deposition collectors is sufficient (Robertson and Wilson, 1985). With the low detection limits used for trace metals and organic compounds, however, came the realization that for constituents other than major ions, the materials used to construct the sample container affected the chemistry of the sample. Constituents can sorb or desorb from the sample container. New materials such as Teflon started being used in the late 1970s to circumvent the sample-contamination problem. Teflon is the preferred material for the collection of trace elements (Fogg and Fitzgerald, 1979; Tramontano and others, 1987). Teflon, aluminum, and stainless steel are the preferred construction materials for the collection of pesticides in wet deposition (Majewski and Capel, 1995). These sampler materials are used for the respective constituents because they neither contribute to nor attenuate the constituents in significant amounts. Likewise, the need for clean sampling methods, such as the use of gloves for handling the sample container and the use of ultra-pure acids for the preservation of samples, became necessary as laboratory detection limits were lowered.

Dust Buckets and Bulk Collectors

Dry-deposition measurements are subject to many external effects, of which the principal one is the physical shape and dimensions of the sample collector. The airstream across the opening of the collector is subject to disturbances, and many collectors are not modified to minimize or eliminate these disturbances (Sanderson and others, 1963). Turbulence set up by air currents passing across the collector surfaces may cause resuspending of deposited material. Studies have indicated that dust volumes collected by an array of identical collectors at one site varied by as little as 15 percent (Sanderson and others, 1963). Conversely, dust volumes collected by an array of different types of collectors at a single site differed by as much as 100 percent (Sanderson and others, 1963).

Volumes of material deposited in bulk collectors could have a lack of reproducibility similar to that of the volumes deposited in the dust-bucket collectors. To the extent that constituent deposition is dominated by dry deposition of large particles, bulk collectors may be more appropriate to collect bulk-deposition samples than they would be if smaller particles or gases dominate deposition.

Wet-deposition collector

*Moving wet/dry deposition-collector lid (Teflon coated)

31-centimeter polyethylene wet-deposition funnel

*Teflon-coated lid arm

Dry-deposition collector

Sensor

Bucket housing

Bucket housing

Collector frame

13-liter polyethylene dry-deposition bucket

*Light bulb (to prevent freezing)

*1.3-centimeter Teflon tubing

Lid motor box

*Bottom enclosure

*5-liter Teflon wet-deposition-collection bottle

*Thermometer

*Modifications made to the standard Aerochem Metric 301 collector

Figure 7. Modified Aerochem Metrics Model 301 wet/dry collector that was used for the collection of trace elements in Gary, Indiana (Willoughby, 1995).

Impactor Samplers and Standard-Surface Collectors

For many of the constituents of interest, accurate assessment of dry deposition would require impactor or standard-surface collectors or both. For example, if particle size is required to assess on-highway processing, impactor samplers must be used to collect samples. Impactors appropriate for the range of particles that are important in atmospheric deposition are described in Lin and others (1993). Rotary impactor samplers are appropriate for large particles (6.5 to 36.5 μm) and conventional cascade impactor samplers for small particles (0.43 to 10 μm). Results from impactor collectors can be compared, using deposition velocities, to deposition rates measured using a standard-surface collector to determine whether modeled and experimentally determined deposition agree. Standard surface refers to a PVC plate with a sharply beveled (10 degrees) leading edge covered by Mylar strips coated with a thin layer of Apezion L grease (Lin and others, 1993). A wind vane directs the beveled edge into the wind. Dry deposition to this surface is a reproducible and accurate measure of deposition in places where surface roughness and turbulence is low. Processing the collectors consists of weighing the grease-coated Mylar film before and after collection, and then extracting the grease from the films for chemical analysis to determine constituent concentration.

Quality Assurance

Any analytical measurement contains a certain amount of error. Error can be introduced when samples are collected, transported, processed, or analyzed. Database errors can be introduced when data are stored on paper or electronically. To minimize all potential errors, atmospheric-deposition networks need a comprehensive, well-documented, quality-assurance plan. Without a quality-assurance plan, the error component of atmospheric-deposition data would be unknown and the usefulness of these data in environmental studies would be diminished. In addition, estimates of overall network data precision provide scientists who investigate precipitation chemistry with confidence in their findings. These estimates also provide a baseline against which to measure the performance of improved collectors and sample-collection procedures (Robertson and Wilson, 1985).

Field-Data Collection

To maintain data quality during collection, a training program for site operators is necessary, and data-collection procedures used by the site operator should be documented. Required documentation includes a description of the routine maintenance of the field equipment (wet or dry collector, rain gage, and other equipment); the procedures used to clean the sampling equipment (Willoughby, 1995); a description of the procedures for approaching the collector to remove the samples—for example, site operators should approach the collector from the downwind side to minimize windblown contaminants from entering the collector (Bigelow, 1982); the protocol for using "clean hands" procedures when working with the sample (Horowitz and others, 1994); and descriptions of procedures for removing the sample from the collector, measuring water-quality properties, and preserving the sample when necessary. During the site visit, the site operator should document the day and time the sample was collected and the time that the new sample-collection container was put in place.

If constituent concentrations or water-quality properties are measured in the field, it is necessary to determine the quality of field measurements. The NADP/NTN has an intersite comparison program to evaluate precision and accuracy of on-site measurements of pH and specific conductance (Willoughby and others, 1991; Nilles and others, 1992). For the intersite comparison program, identical samples with known pH and specific conductance are sent to site operators. Although the site operators know they are participating in an external quality-assurance program, they do not know the actual pH and specific conductance values of the solution. The results from each site operator are compared to all other site operators in the network (Willoughby and others, 1991). Results that are outside the acceptable limits are an indication of equipment problems or the need for additional site-operator training.

Other quality-control samples, such as field blanks, may be necessary at some sites. For example, an investigation of major ions and trace elements in wet deposition near Gary, Ind., used funnel rinses and system blanks to identify possible sources of contamination of the samples (Willoughby, 1995). In the funnel rinse, deionized water that had been adjusted to a pH similar to the wet deposition being collected was poured through the funnel immediately after the equipment was installed in the collector. The funnel rinse was processed by the same method as the wet-deposition samples to quantify possible contamination resulting from cleaning, shipping, and installing the funnel in the collector. The system blank was processed in the same manner as the funnel rinses and the wet-deposition samples, and was collected to evaluate possible contamination resulting from dry periods of 1 week. Acidified, deionized water was passed through the funnel at the end of the 1-week sampling period. The results were compared to the results from the acidified, deionized water-funnel rinses and the wet-deposition samples collected from October 17, 1995, through November 12, 1996, at the Gary Regional Airport (fig. 8). These data indicate that the quality-control steps successfully minimized sampling artifacts and show the difference between sampling artifacts and actual contaminant loadings during this sampling period.

The NADP/NTN also conducts a colocated sampler program. For the colocated sampler program, identical samplers are placed near each other at the same sampling site. Samples from the same precipitation event are collected in each collector and analyzed by the same laboratory using the same analytical methods. The results then are compared, and a variance is computed (Nilles and others, 1992). Peters and Reese (1995) compared results from colocated samplers and found that concentration in wet deposition generally was less variable and had lower concentrations of major inorganic ions and nutrients than did bulk deposition.
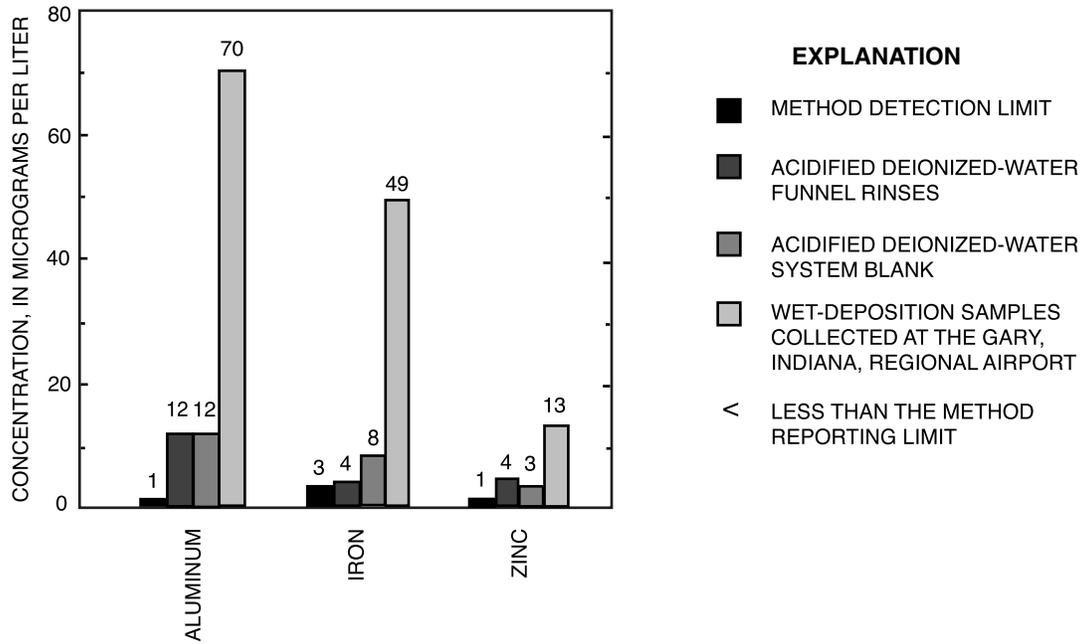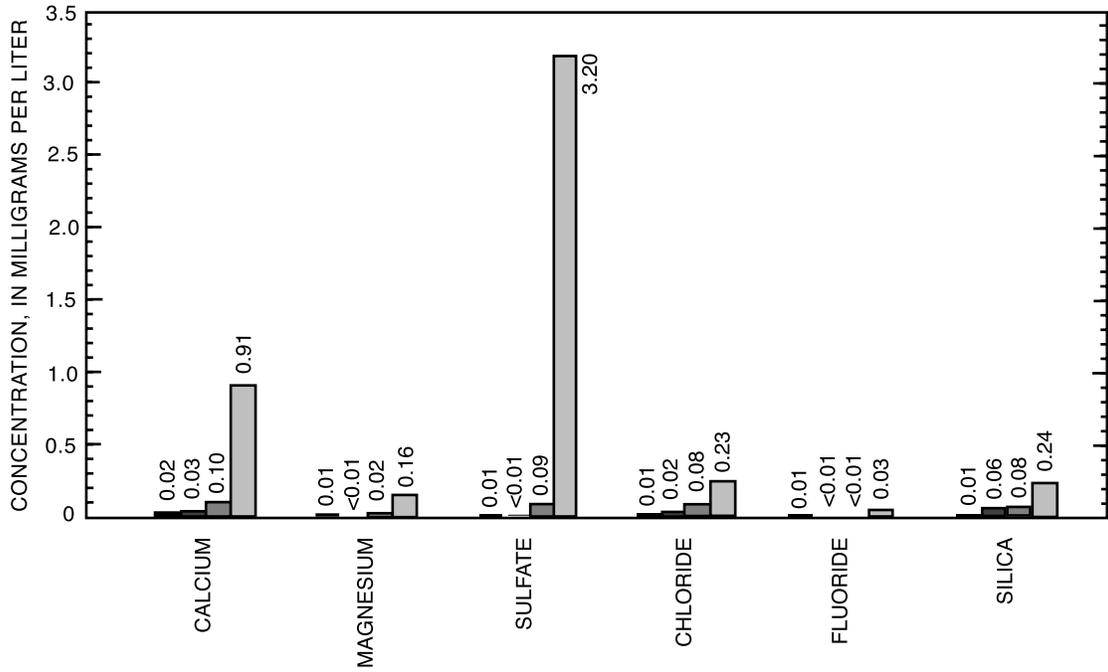
Figure 8. Median concentrations of chemical constituents measured in quality-control samples collected at the Gary, Indiana, Regional Airport from October 17, 1995, to November 12, 1996.

For studies in which contamination in highway runoff and in atmospheric deposition are being investigated, it may be necessary to establish a background site to determine constituent loadings that are not directly related to highway runoff. For background sites, all known emission point sources within a 50-km radius, as well as the constituents being emitted into the atmosphere by these sources and their combined annual emissions, should be recorded. It also is important to document land use and topography near the sampling site. A map of the sampling site should be included with the site-description documentation. For studies conducted near the highway, land use, topography, average daily traffic, and the number of days between storm events should be documented. Examples of sampling-site descriptions used by the NADP/NTN are provided in Robertson and Wojciechowski (1986).

Because the concentrations of atmospheric-deposition constituents generally are low, it is critical that data-collection efforts follow a quality-assurance/quality-control plan to ensure that the sampling program produces data of known quality. Quality-assurance (QA) programs include the precautionary actions used to prevent systematic bias. Examples of QA include using noncontaminating materials and sample containers, cleaning the sampling equipment, preserving samples soon after collection, and shipping samples overnight to the laboratory (Jones, 1999). Quality-control (QC) programs include the steps used to check that QA is effective and to evaluate variability in the data because of random error. Examples of QC include equipment-blank samples to determine if the sampling equipment is clean, replicates to assess sample variance and analytical precision, and samples spiked with analytes to evaluate analyte degradation and recovery (Jones, 1999). For small individual studies, the QA plan usually is described briefly along with investigation results. For large state, regional, or national studies, the QA plan should be published in conjunction with reports that document methods and results of these studies (Jones, 1999).

A technically defensible database used to assess the importance of atmospheric deposition to highway-runoff studies requires consistent national standards for data collection and processing. The Intergovernmental Task Force on Monitoring Water Quality (ITFM) is developing and implementing a national strategic plan to achieve effective collection, interpretation, and presentation of water-quality data and to improve the availability of information for decision making at all government levels and the private sector. The ITFM is composed of 10 Federal agencies, 9 State and interstate agencies, and 1 American Indian tribe. Products of the ITFM include a framework for monitoring programs; selection criteria for environmental indicators; and ecological, chemical, and physical criteria for evaluating water quality according to environmental management objectives (Intergovernmental Task Force on Monitoring Water Quality, 1995).

Laboratory Analysis

Most analytical laboratories have an internal quality-assurance program to evaluate the accuracy, precision, and bias of the measurements and to insure that appropriate protocols and control standards are being implemented. Examples of internal quality assurance include programs for data validation, analysis of method blanks, replicates, standard reference water samples, spike samples, and internal blind sampling.

Validation of sample data involves the comparison of data against acceptable limits, checking ion balances, and reanalysis of the sample, if necessary. Method blanks are prepared in the laboratory as an analyte-free matrix and are carried through the entire sample preparation and analytical procedure to identify contamination from the laboratory during sample preparation and analysis. A replicate sample is an environmental sample that is split into two or more aliquots and submitted to the laboratory for analysis (Bricker, 1999). Analysis of replicate samples provides information about the precision associated with sample handling (after splitting), shipping, storage, and laboratory procedures. Standard reference water samples are produced and certified by the U.S. National Institute of Standards and Technology and are used to assess performance and assess bias for each analysis, and to determine long-term estimates of method variability. A laboratory matrix spike is an environmental sample (that contains a known concentration of one or more of the analytes). The matrix spike is analyzed by the laboratory to assess method performance by recovery of analytes in a particular matrix (Jones, 1999)

Blind-sample programs can be internal or external and are used to quantify the bias caused by random laboratory contamination (Jones, 1999). The NADP/NTN uses an internal blind-sample program to assess the variation of analyte determinations caused by routine on-site and laboratory sample-handling

procedures. In the blind-sample program, samples with known concentrations are sent to the analytical laboratory for analysis. The samples are disguised so that the laboratory staff is not aware that the sample is a quality-control sample. Results reported for the sample then are compared to the known concentrations (Willoughby and others, 1991).

In addition to internal laboratory quality assurance, networks should have an external quality-assurance program. The NADP/NTN conducts an interlaboratory comparison program to evaluate bias and analytical precision in measurements made by laboratories that measure atmospheric-deposition samples. For the interlaboratory comparison program, identical samples are sent to participating laboratories. The laboratories analyze the samples using the same procedures used to measure environmental samples. The results from the laboratories are compared, and analytical precision and bias are determined for each laboratory and for all the laboratories as a group (Willoughby and others, 1991).

Establishing, defining, and documenting detection limits is an important part of quality-assurance plans (Jones, 1999). The Minimum Reporting Level (MRL) is the lowest concentration that can be quantified accurately by a given analytical method. The Method Detection Limit (MDL) is the lowest concentration that the method can detect reliably. Investigation objectives define whether the MRL, the MDL, or both will be used and documented. It is difficult to combine and compare results from different studies, or within one investigation, when detection limits are variable or unknown (Majewski and Capel, 1995). Analytical and effective detection limits for many constituents have been improved during the past 5–10 years; therefore, the QA plan, and any data reports produced by an investigation, should document analytical and effective detection limits and any changes in those limits. When constituents are not detected at the MRL, the results from screening at the MDL should be recorded. Because many constituents in atmospheric deposition have relatively low concentrations (low micrograms per liter level), pre-concentration or extraction processes are often used to increase effective detection limits by proportionally increasing the concentration of constituents in the analyte matrix. Therefore, detection limits are often a function of the the available sample size used for the pre-concentration or extraction processes, as well as of the analytical method.

SUMMARY AND CONCLUSIONS

A general national model for simulation of pollutant constituents in highway runoff is not available at present because the processes leading to incorporation of chemical constituents in runoff are not understood, or because the data for modeling the chemical constituents in highway runoff are inadequate. A better understanding of atmospheric-deposition processes and deposition rates is necessary for improving the modeling capability. Little is known about deposition processes and rates that are specific to highway environments. For many of the pollutants that are important in highway runoff, dry deposition in nonhighway environments is greater than wet deposition. This result means that on-highway processing of the particles that make up dry deposition is important in determining the part of deposition that becomes runoff. In an investigation of these processes, labeled particles have been found to be efficiently removed from highway surfaces by traffic. Concentrations of particles in deposition collectors and knowledge of settling indicate that new deposition locations of removed particles would not be very distant from their original settling locations so that there may be a sharp gradient between the "swept clean" travel lanes and adjacent emergency-lane and berm surfaces. But depending on particle size, traffic could either sweep the travel lanes clean or cause more deposition by impacting particles (or gases) on the highway surface.

Runoff loads of many trace metals and organic constituents that are products of combustion would be expected to originate from traffic rather than from ambient deposition. If ambient deposition could be demonstrated to be insignificant compared to traffic sources, then predictive deposition models would be simplified. Many mass-balance budget investigations have indicated, however, that ambient deposition is a substantial part of the runoff load. Comparisons of deposition with runoff loads have been based on the assumption that on-highway transport of deposition is conservative. Because of resuspension from traffic-induced turbulence, however, transport is unlikely to be conservative.

Lack of quality-assurance programs or clean-hands techniques in collecting deposition and runoff samples has contributed to the problem, sometimes in subtle ways. For instance, sharp gradients have been measured in deposition rates of materials in a row of

collectors deployed on a transect transverse to highways. The finding that more deposition is measured near the highway than far from the highway indicates that traffic is a major source of deposited materials. Whereas detection of the gradient is an indication that collection methods are appropriate, comparison of the background results of the highway studies with deposition rates measured using clean methods and quality-assurance protocol shows that the former may be 10 times greater than actual background levels.

Programs that investigate atmospheric-deposition sources to highway runoff and which include analysis of tracers of ambient deposition may accomplish two types of objectives: determination of whether ambient deposition is important for pollutant constituents and determination of the character of on-highway processing of atmospheric deposition. Such programs may be logical precursors to any national investigation of atmospheric deposition that might be undertaken in support of highway-runoff projects.

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Figure 9

A. Estimated sulfate ion deposition, 1998 [Kilograms per hectare (kg/ha)]
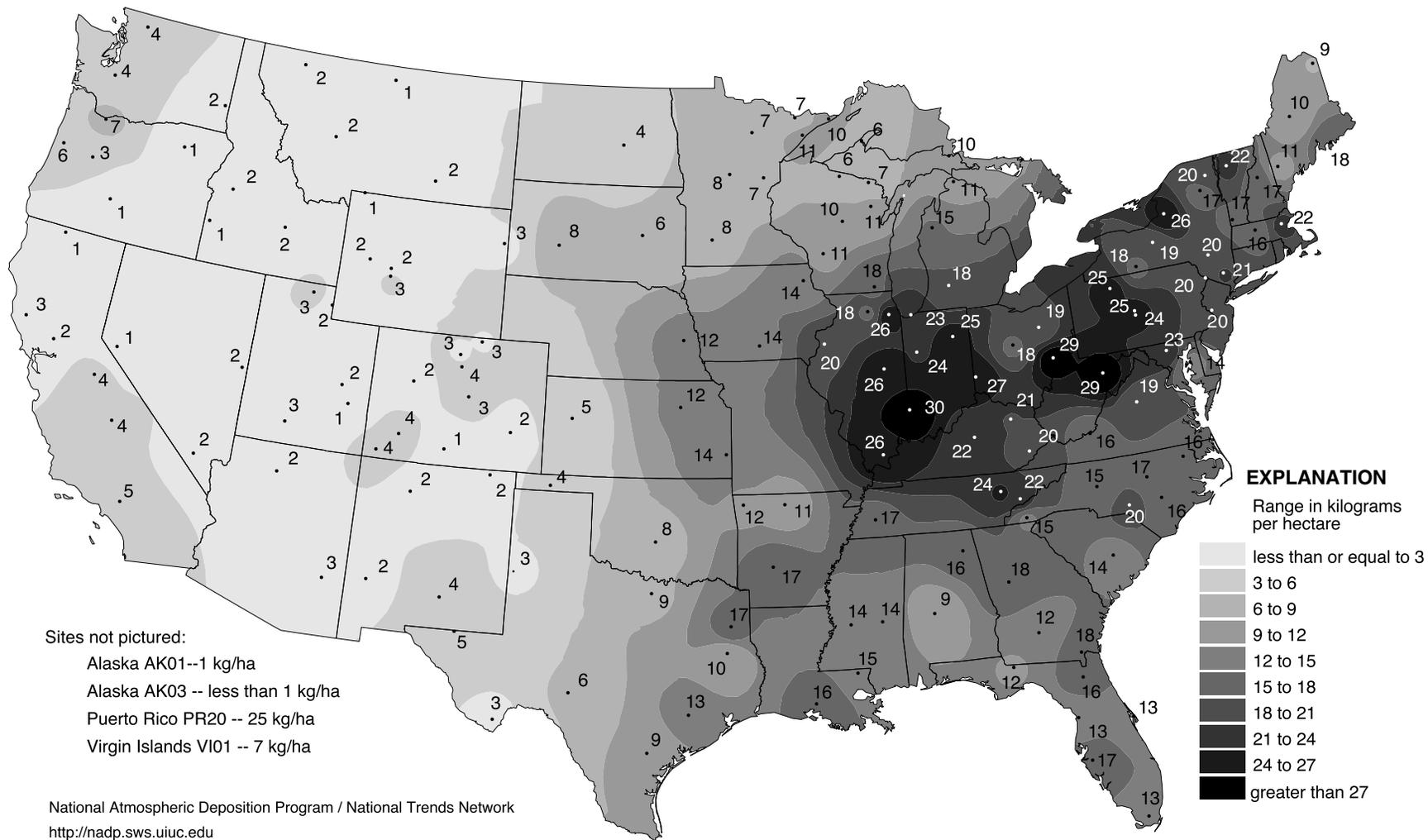
Sites not pictured:
 Alaska AK01--1 kg/ha
 Alaska AK03 -- less than 1 kg/ha
 Puerto Rico PR20 -- 25 kg/ha
 Virgin Islands VI01 -- 7 kg/ha

National Atmospheric Deposition Program / National Trends Network
http://nadp.sws.uiuc.edu

EXPLANATION

Range in kilograms
per hectare

less than or equal to 3
3 to 6
6 to 9
9 to 12
12 to 15
15 to 18
18 to 21
21 to 24
24 to 27
greater than 27

Figure 9. Annual wet-deposition rates mapped for 1998 for the United States based on National Atmospheric Deposition Program / National Trends Network (NADP/NTN) data for (*A*) sulfate, (*B*) nitrate, (*C*) calcium, (*D*) ammonium, (*E*) sodium, and (*F*) chloride.

Figure 9   44

B. Estimated nitrate ion deposition, 1998 [Kilograms per hectare (kg/ha)]
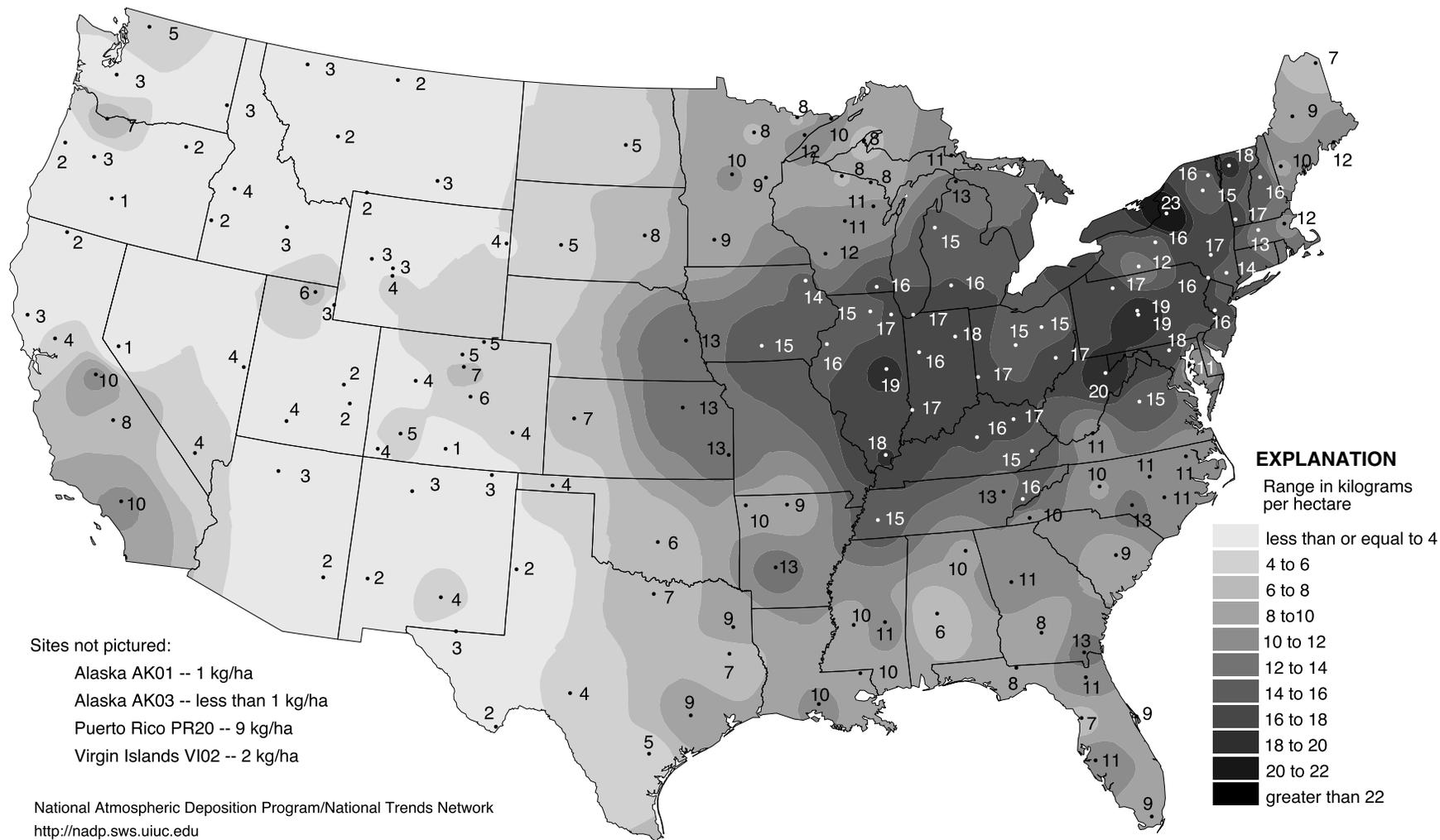
Sites not pictured:

Alaska AK01 -- 1 kg/ha

Alaska AK03 -- less than 1 kg/ha

Puerto Rico PR20 -- 9 kg/ha

Virgin Islands VI02 -- 2 kg/ha

National Atmospheric Deposition Program/National Trends Network
http://nadp.sws.uiuc.edu

EXPLANATION

Range in kilograms per hectare

- less than or equal to 4
- 4 to 6
- 6 to 8
- 8 to 10
- 10 to 12
- 12 to 14
- 14 to 16
- 16 to 18
- 18 to 20
- 20 to 22
- greater than 22

Figure 9. Annual wet-deposition rates mapped for 1998 for the United States based on NADP/NTN data for (*A*) sulfate, (*B*) nitrate, (*C*) calcium, (*D*) ammonium, (*E*) sodium, and (*F*) chloride—*Continued*.

C. Estimated calcium ion deposition, 1998 [Kilograms per hectare (kg/ha)]
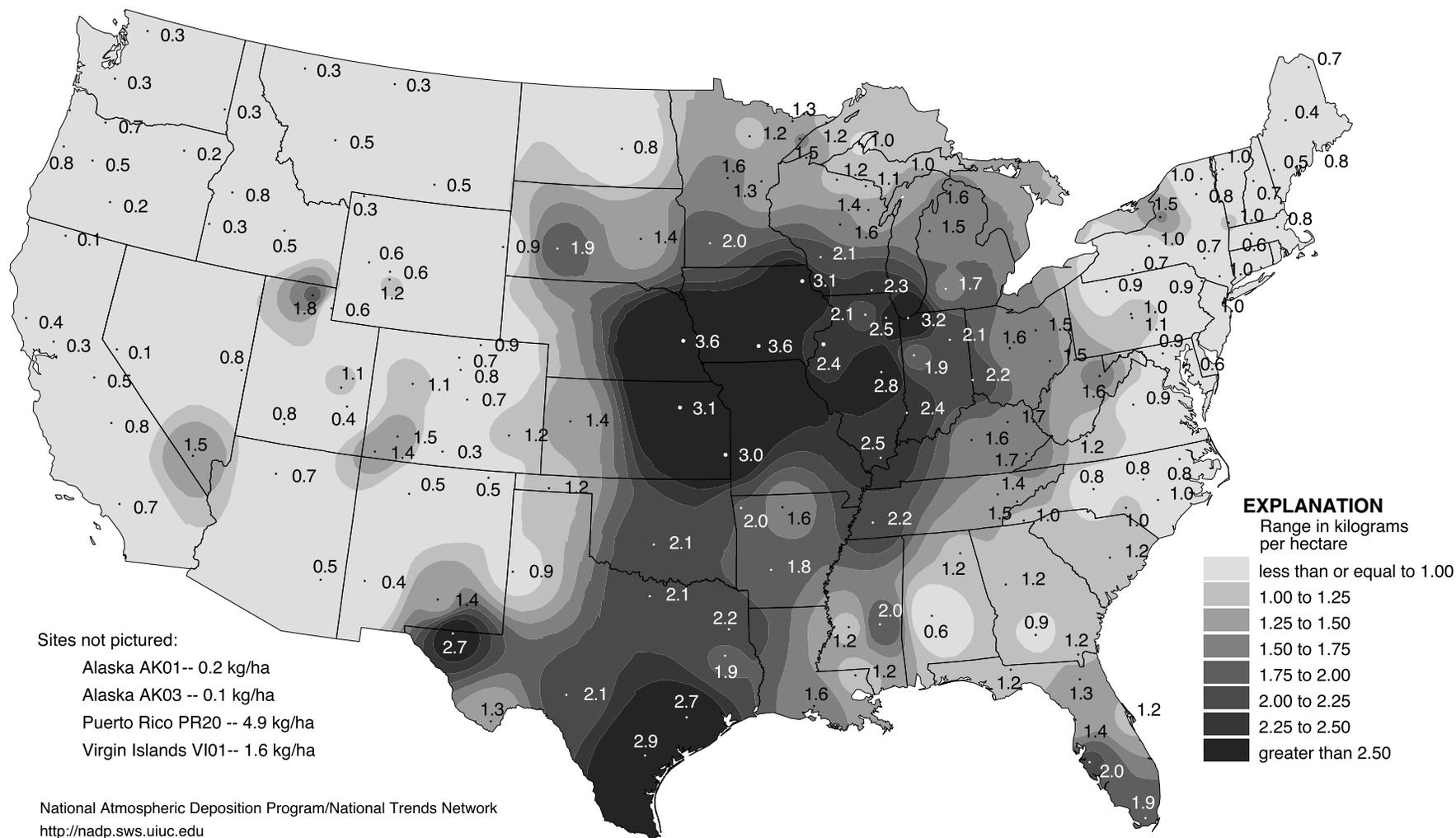
Sites not pictured:

Alaska AK01-- 0.2 kg/ha

Alaska AK03 -- 0.1 kg/ha

Puerto Rico PR20 -- 4.9 kg/ha

Virgin Islands VI01-- 1.6 kg/ha

National Atmospheric Deposition Program/National Trends Network

http://nadp.sws.uiuc.edu

EXPLANATION
Range in kilograms
per hectare

less than or equal to 1.00

1.00 to 1.25

1.25 to 1.50

1.50 to 1.75

1.75 to 2.00

2.00 to 2.25

2.25 to 2.50

greater than 2.50

Figure 9. Annual wet-deposition rates mapped for 1998 for the United States based on NADP/NTN data for (*A*) sulfate, (*B*) nitrate, (*C*) calcium, (*D*) ammonium, (*E*) sodium, and (*F*) chloride—*Continued.*

Figure 9   46

D. **Estimated ammonium ion deposition, 1998 [Kilograms per hectare (kg/ha)]**
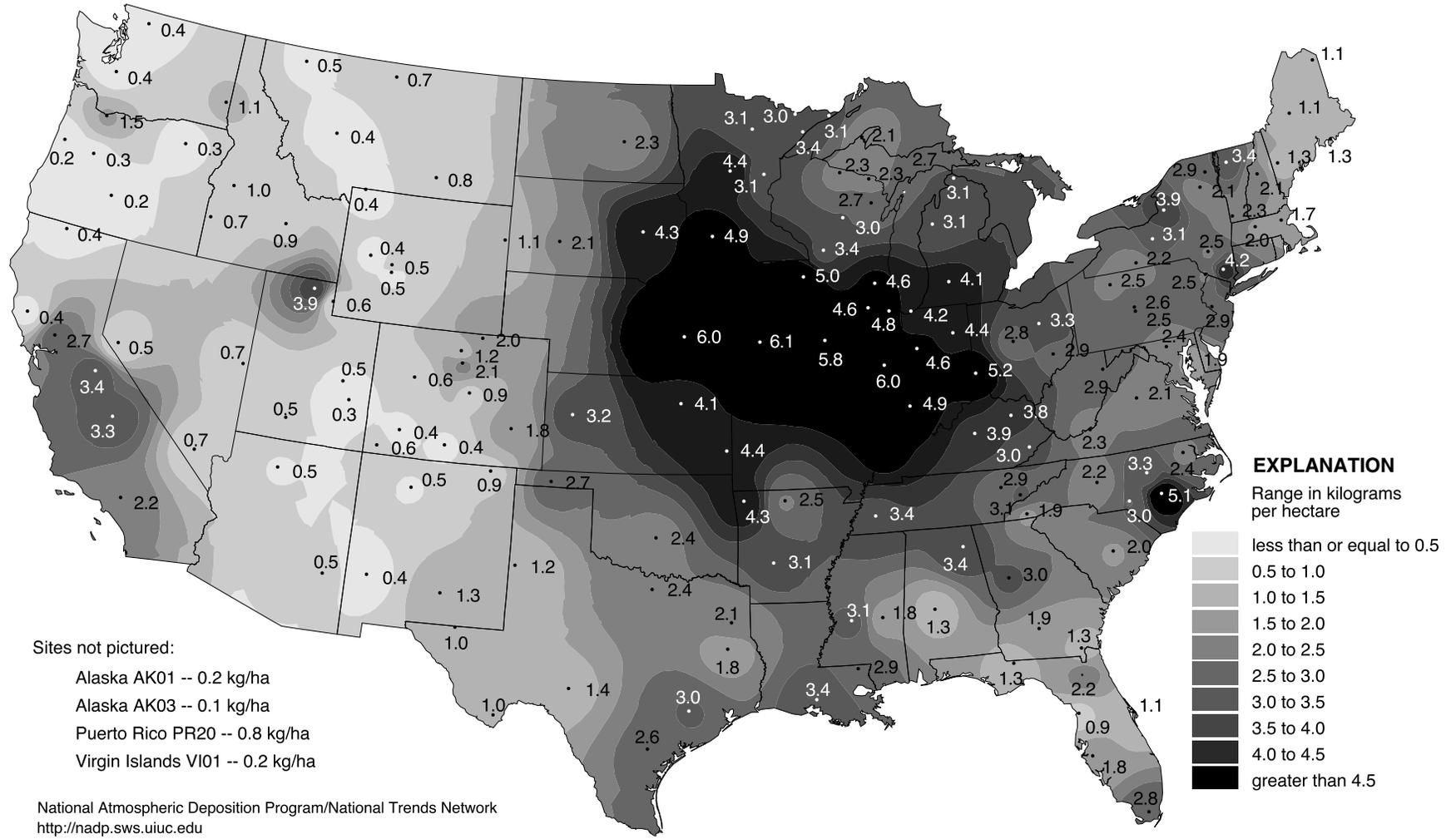
Sites not pictured:

Alaska AK01 -- 0.2 kg/ha

Alaska AK03 -- 0.1 kg/ha

Puerto Rico PR20 -- 0.8 kg/ha

Virgin Islands VI01 -- 0.2 kg/ha

National Atmospheric Deposition Program/National Trends Network
http://nadp.sws.uiuc.edu

EXPLANATION

Range in kilograms
per hectare

- less than or equal to 0.5
- 0.5 to 1.0
- 1.0 to 1.5
- 1.5 to 2.0
- 2.0 to 2.5
- 2.5 to 3.0
- 3.0 to 3.5
- 3.5 to 4.0
- 4.0 to 4.5
- greater than 4.5

Figure 9. Annual wet-deposition rates mapped for 1998 for the United States based on NADP/NTN data for (*A*) sulfate, (*B*) nitrate, (*C*) calcium, (*D*) ammonium, (*E*) sodium, and (*F*) chloride—*Continued*.

E. Estimated sodium ion deposition, 1998 [Kilograms per hectare (kg/ha)]
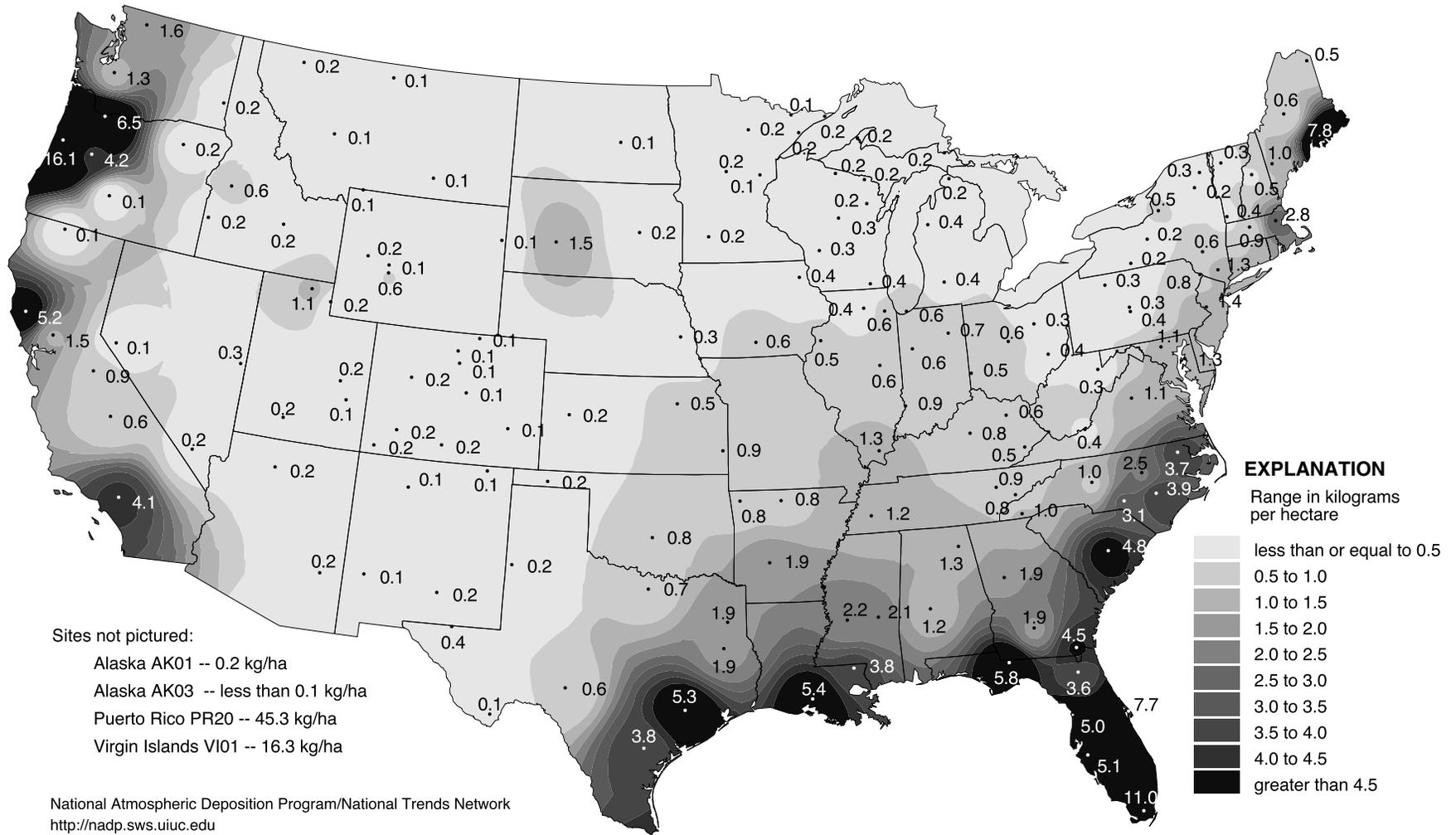
EXPLANATION

Range in kilograms per hectare

- less than or equal to 0.5
- 0.5 to 1.0
- 1.0 to 1.5
- 1.5 to 2.0
- 2.0 to 2.5
- 2.5 to 3.0
- 3.0 to 3.5
- 3.5 to 4.0
- 4.0 to 4.5
- greater than 4.5

Sites not pictured:

Alaska AK01 -- 0.2 kg/ha

Alaska AK03 -- less than 0.1 kg/ha

Puerto Rico PR20 -- 45.3 kg/ha

Virgin Islands VI01 -- 16.3 kg/ha

National Atmospheric Deposition Program/National Trends Network
http://nadp.sws.uiuc.edu

Figure 9. Annual wet-deposition rates mapped for 1998 for the United States based on NADP/NTN data for (*A*) sulfate, (*B*) nitrate, (*C*) calcium, (*D*) ammonium, (*E*) sodium, and (*F*) chloride—*Continued*.

Figure 9   48

F. Estimated chloride ion deposition, 1998 [Kilograms per hectare (kg/ha)]
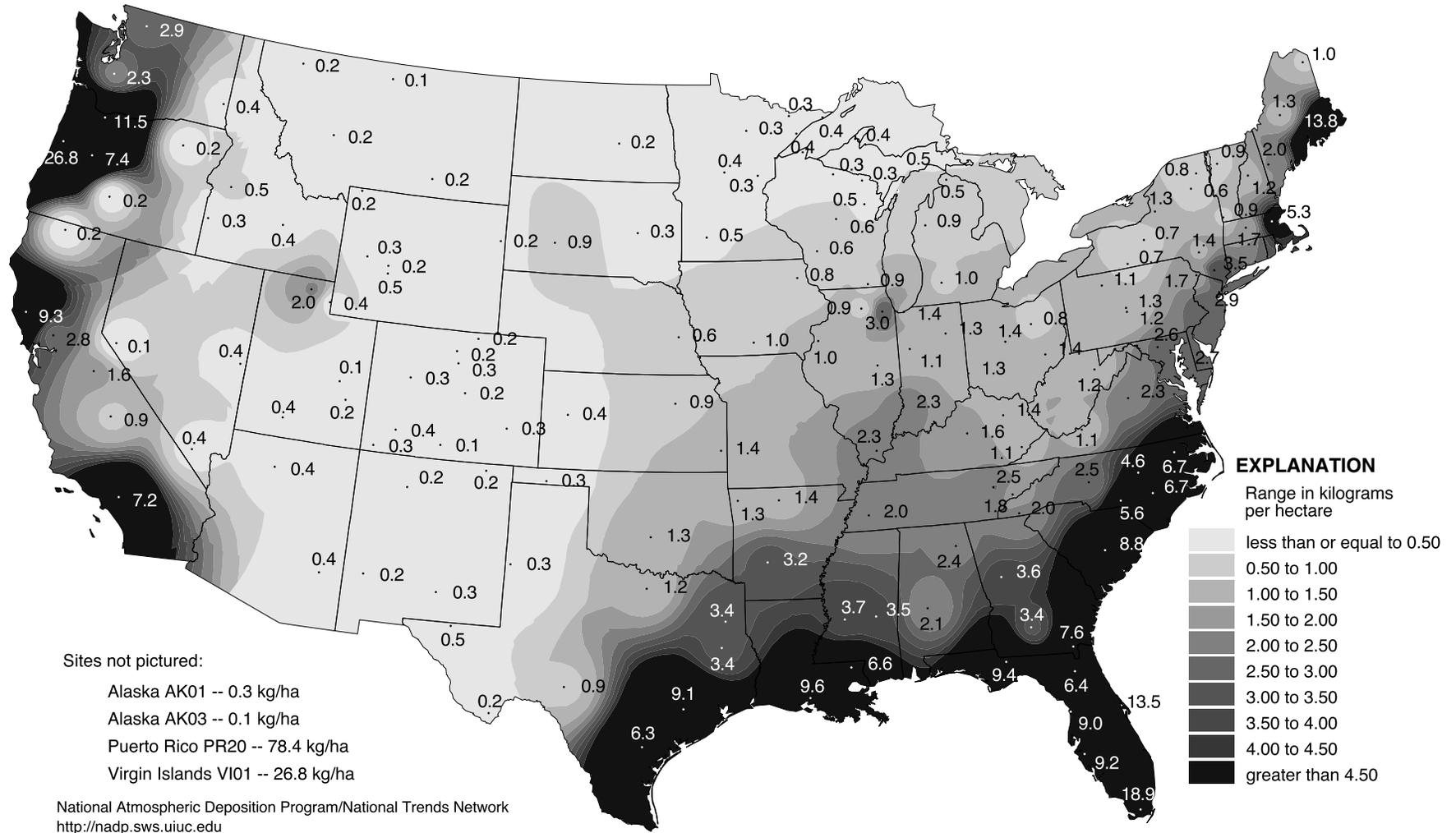
Sites not pictured:

Alaska AK01 -- 0.3 kg/ha

Alaska AK03 -- 0.1 kg/ha

Puerto Rico PR20 -- 78.4 kg/ha

Virgin Islands VI01 -- 26.8 kg/ha

National Atmospheric Deposition Program/National Trends Network
http://nadp.sws.uiuc.edu

EXPLANATION

Range in kilograms
per hectare

less than or equal to 0.50

0.50 to 1.00

1.00 to 1.50

1.50 to 2.00

2.00 to 2.50

2.50 to 3.00

3.00 to 3.50

3.50 to 4.00

4.00 to 4.50

greater than 4.50

**Figure 9.** Annual wet-deposition rates mapped for 1998 for the United States based on NADP/NTN data for (*A*) sulfate, (*B*) nitrate, (*C*) calcium, (*D*) ammonium, (*E*) sodium, and (*F*) chloride—*Continued*.

Table 5

Table 5. Reports that address highway-runoff constituent loads and atmospheric deposition sources

[**Report type:** D, data; D/I, data/interpretive; FHWA, Federal Highway Administration; MI, model/interpretive; R/S, Review/Summary. **Investigation type:** ND, not discussed; cm, centimeter; g/m²/d, gram per square meter per day; L, liter; μm, micrometer; %, percent; >, greater than]

Reference	Report type	Period of sampling	Type of sampling/method	Location/setting	Document method	
					Sample collection	Analytical
Adachi and Kobayashi (1992)	D/I	January–December 1990	Wet deposition/ND	Kobe, Japan/urban	ND	ND
Arimoto (1989)	R/S	ND	Bulk deposition	Great Lakes region/urban and rural	ND	ND
Asman and others (1982)	D/I	January 1978– December 1980	Wet deposition/precipitation collectors	Netherlands/rural	Yes	ND
Ball (1996)	D/I	ND	Dry deposition/vacuum cleaner	Sydney, Australia/urban	Yes	ND
Barkdoll and others (1977)	D/I	ND	Bulk deposition/continuous recorder, dustfall jar	Knoxville, Tenn./urban	Yes	ND
Beech (1980)	D/I	ND	Bulk deposition/collection from swimming pools	Miami, Fla./urban	Yes	Yes
Bidleman (1988)	R/S	ND	Bulk deposition/ND	ND	ND	ND
Bigelow (1982)	M/I	ND	Bulk deposition/wet-dry collector	ND	Yes	Yes
Brinkman (1985)	R/S	ND	ND	ND	ND	ND
Cadle (1985)	D/I	June 25, 1981– June 16, 1982	Bulk deposition/dual-filter method, denuder-difference method, impactors	Warren, Mich./urban	Yes	Yes
Cahill and Seiber (2000)	D/I	July 23–30, 1998; August 23–30, 1998	Water samples/1-L glass bottles	British Columbia, Yukon Territory, Canada, Alaska/ urban; Northern California/ urban	Yes	Yes
Capel and others (1998)	D/I	March –November 1988; March–April 1990	Wet deposition/wet-only rain sampler	St. Paul, Minn./urban	Yes	Yes
Clarke and Papapanayotou (1987)	D/I	October–December 1985	Vehicle emissions/filters	Leeds, U.K./urban	Yes	Yes
Dannecker and others (1990)	D/I	1986–87	Bulk deposition/wet only sampler, Bergerhoff instrument (dryfall)	Hamburg, Germany/urban	Yes	Yes
Dasch (1985)	D/I	August 1981– June 1982	Bulk deposition/wet-dry collector	Warren, Mich./urban; Lapeer, Mich./rural	Yes	Yes
Deboudt and others (1999)	D/I	March–October 1995	Dry deposition/cascade impactors	Straits of Dover, U.K./urban	Yes	Yes

A, atmospheric; NADP, National Atmospheric Deposition Program; PAHs, Polycyclic aromatic hydrocarbons, R, runoff; PCBs, polychlorinated biphenyl;

Reference	Investi- gation type	Chemical analyte	QA/QC	Comments
Adachi and Kobayashi (1992)	A	Major inorganic constituents, water-quality properties	ND	Ni^{2+} in rainwater can be used as indicator for air pollution by motor vehicle exhausts.
Arimoto (1989)	A	Trace elements, major inorganic constituents, organic compounds, water-quality properties	ND	Review of the atmospheric deposition of chemical contaminants to the Great Lakes Basin
Asman and others (1982)	A	Major inorganic constituents, organic compounds, water-quality properties	ND	Bird-droppings have an influence on the chemical composition of precipitation samples
Ball (1996)	A	Trace elements, major inorganic constituents	ND	Most of constituent load found to be sorbed onto sediment particles less than 70 μm in size
Barkdoll and others (1977)	A,R	Trace elements, major inorganic constituents, organic compounds, water-quality properties	ND	Discusses the effects of dustfall on urban stormwater quality
Beech (1980)	A	Trace elements, major inorganic constituents, water-quality properties	standard methods	Metal concentrations in swimming pools located along highways not appreciably different than other urban swimming pools.
Bidleman (1988)	A	Organic compounds	ND	Atmospheric processes of semivolatile organic compounds
Bigelow (1982)	A	Major inorganic constituents, organic compounds, water-quality properties	standards	NADP collection site operation manual
Brinkman (1985)	A,R	Trace elements, major inorganic constituents, organic compounds, water-quality properties	ND	Discusses sources and loadings of stormwater pollutants
Cadle (1985)	A	Major inorganic constituents, organic compounds	standards, blanks	Discusses factors affecting the seasonal variation
Cahill and Seiber (2000)	A,R	Organic compounds	blanks	Trifluoroacetate, breakdown product of hydrofluorocarbons and hydrochlorofluorocarbons, found at higher concentrations in surface waters near urban areas
Capel and others (1998)	A	Organic compounds, water-quality properties	standards	Discusses the wet atmospheric deposition of pesticides in Minnesota
Clarke and Papapanayotou (1987)	A	Major inorganic constituents, organic compounds	ND	Traffic-generated carbonaceous aerosols have a major effect on urban air pollution
Dannecker and others (1990)	A,R	Trace elements, organic compounds	ND	Pollutant concentrations in bulk deposition and runoff similar
Dasch (1985)	A	Major inorganic constituents, organic compounds	ND	Dry deposition strongly influenced by affinity of surface for gases and the retention characteristics of the surface for particles
Deboudt and others (1999)	A	Trace elements	blanks	Automobile source minor component of particulate lead in air

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Table 5. Reports that address highway-runoff constituent loads and atmospheric deposition sources—*Continued*

Reference	Report type	Period of sampling	Type of sampling/method	Location/setting	Document method Sample collection	Document method Analytical
deLuca (1991)	D/I	ND	Wet deposition/rainfall collector	Porto Alegre, Brazil/urban	Yes	ND
Diamond and others (2000)	D/I	ND	Bulk deposition/Kimwipes	Toronto, Ontario/urban	Yes	Yes
Dossett and Bowersox (1991)	M/I	ND	Bulk deposition/wet-dry collector	ND	ND	ND
Dupuis and Lord (1982)	R/S: FHWA	ND	Various	ND	Yes	Yes
Ebbert and Wagner (1987)	D/I	1977–83	Bulk deposition/rainfall collector, dryfall collector	31 catchments across United States/urban	ND	ND
Ellis and others (1987)	D/I	ND	ND	London region/urban	ND	ND
Endlich and others (1988)	D/I	1978–83	Wet deposition/automatic precipitation sampler, weighing bucket rain gage	Turners Falls, Mass./urban; Fort Wayne, Ind./urban; Lancaster, Kans./urban; Raleigh, N.C./ urban; Selma, Ala./urban; Zanesville, Ohio/urban	Yes	Yes
Fisher (1987)	R/S	ND	Highway related emissions	Europe/urban	ND	ND
Franz and others (1991)	D/I	May 8, 1986–October 13, 1986	Wet deposition/automatic wet deposition sampler	Minneapolis, Minn./urban	Yes	Yes
Fraser and others (1999)	D/I	September 8–9, 21, 1993	Vehicle emissions/low-volume particulate matter sampler, high volume dichotomous sampler	Los Angeles, Calif./urban; Long Beach, Los Angeles, Azusa, Claremont, Calif./urban	Yes	Yes
Garnaud and others (1999)	D/I	November 1994–May 1997	Bulk deposition/total deposition collector	Paris/urban	Yes	Yes
Gay and Melching (1995)	D/I	March 1983–April 1985	Wet deposition/precipitation collectors	Princeton, Mass./rural; Truro, Mass./rural	Yes	Yes
Germani and Zoller (1994)	D/I	November–December 1978	Dry deposition/cascade impactors	Alexandria, Va./urban	Yes	Yes
Grosjean (1983)	D/I	August–September 1981; February–March 1982	Air/filter samplers	Los Angeles region/urban	Yes	Yes
Gupta, Agnew, Gruber, and Kreutzberger (1981)	R/S: FHWA	March 1976–September 1977	Bulk deposition/precipitation gauge, dustfall bucket, weirs, flumes, level sensing instruments	Milwaukee, Wisc./urban; Harrisburg, Penn./rural; Nashville, Tenn./urban; Denver, Colo./urban	Yes	Yes
Habibi (1973)	D/I	ND	Vehicle emissions	ND	Yes	Yes
Halverson and others (1984)	D/I	July 1980–June 1981	Wet deposition/automatic precipitation collector	State College, Penn. region/ urban	Yes	Yes

Reference	Investi-gation type	Chemical analyte	QA/QC	Comments
deLuca (1991)	A,R	Trace elements, major inorganic constituents, organic compounds, water-quality properties	ND	Ammonia, chlorides, and sulphates from atmospheric deposition affect stormwater quality
Diamond and others (2000)	A	Organic compounds	blanks	Organic film containing semivolatile organic compuonds such as PCBs and PAHs can be seen in oily sheen found on roadways
Dossett and Bowersox (1991)	A	ND	ND	Sample contamination can occur when raindrops splash off of the sampler
Dupuis and Lord (1982)	A	ND	ND	Discusses instrumentation requirements for a comprehensive field-monitoring program
Ebbert and Wagner (1987)	A,R	Trace elements, major inorganic constituents, organic compounds	ND	Study indicates rainfalll is substantial source of some constituents, especially nitrogen species, in storm runoff from urban catchments
Ellis and others (1987)	R	Trace elements, organic compounds, water-quality properties	ND	Metal concentrations in stormwater runoff higher in high-way locations than in urban/residential and rural areas
Endlich and others(1988)	A	Trace elements, major inorganic constituents, organic compounds, water-quality properties	ND	Discusses chemical constituents in precipitation
Fisher (1987)	A	Major inorganic constituents	ND	Nitrogen oxides have direct affect on acid deposition
Franz and others (1991)	A	Organic compounds	blanks	All samplers collected at about 95% efficiency when periods of sampler malfunctioning were not considered
Fraser and others (1999)	A	Trace elements, major inorganic constituents, organic compounds	ND	Portion of fine organic particulate matter in the Los Angeles atmosphere attributable to direct particle emissions form vehicle exhaust is calculated to vary form 7.5 to 18.3%
Garnaud and others (1999)	A,R	Trace elements, major inorganic constituents, water-quality properties	standards, blanks	Particulate metal concentrations not appreciably varied in dry and wet fallout samples
Gay and Melching (1995)	A	Trace elements, major inorganic constituents, organic compounds, water-quality properties	standards	Precipitation samples collected to examine the quality of precipitation from storms
Germani and Zoller (1994)	A	Trace elements, major inorganic constituents	blanks	Mineral and ferrous metal particles constitute most of the material found on the first four stages of the cascade impactor samples
Grosjean (1983)	A	Organic compounds	blanks, controls	Approximately 85% of PAH levels would have been underestimated if sampling were limited to glass fiber filters
Gupta, Agnew, Gruber, and Kreutzberger (1981)	A,R	Trace elements, major inorganic constituents, organic compounds, water-quality properties	controls	Discussion of constituents of highway runoff, sources and migration of pollutants to receiving water, effects of pollutants and treatment methods
Habibi (1973)	A	Trace elements	replicates	Described methods used to assess lead emissions from vehicles
Halverson and others (1984)	A,R	Major inorganic constituents, organic compounds, water-quality properties	ND	Precipitation contributed most of nitrogen found in runoff, along with some sulfur and phosphorus

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Table 5. Reports that address highway-runoff constituent loads and atmospheric deposition sources—*Continued*

Reference	Report type	Period of sampling	Type of sampling/method	Location/setting	Document method Sample collection	Analytical
Harrison and Johnston (1985)	D/I	October 1981–June 1983	Bulk deposition/total deposition collectors	U.K./urban	Yes	Yes
Harrison and others (1985)	D/I	ND	Air/gas chromatography	Lancaster, U.K./urban; Hazelrigg, U.K./rural	Yes	Yes
Hedges and Wren (1987)	D/I	July 1983–July 1984	Bulk deposition/deposition canisters	Birmingham, U.K./urban	ND	ND
Herrick (1966)	M/I	ND	Dry deposition/dustfall collectors	ND	Yes	ND
Hewitt and Rashed (1990)	D/I	December 3, 1986–December 3, 1987	Bulk deposition/funnels covered with screens	Burton-in-Kendal, Cumbria, U.K./urban	ND	ND
Hicks and Matt (1988)	M/I	ND	Dry deposition/ND	ND	ND	ND
Hicks and others (1991)	M/I	ND	Dry deposition/filterpack sampler	ND	Yes	Yes
Hicks and others (1993)	R/S	ND	Bulk deposition/various	ND	Yes	ND
Ho and Tai (1988)	D/I	November 1979	Soil/trowel	Hong Kong/urban	Yes	Yes
Holdren and others (1993)	R/S	ND	Bulk deposition/ND	ND	ND	ND
Holsen and Noll (1992)	D/I	June 21,–September 29, 1991	Dry deposition/dry deposition collectors	Chicago, Ill./urban	Yes	Yes
Hopke (1991)	M/I	ND	Bulk deposition/ND	ND	ND	ND
Hvitved-Jacobson and Yousef (1991)	R/S	ND	Bulk deposition/ND	ND	ND	ND
James and Shivalingaiah (1985)	M/I	ND	Bulk deposition/ND	Hamilton, Ontarion, Canada/ urban	ND	ND
Jensen and Laxen (1987)	D/I	January 1985–April 1986	Air/'M' Type samplers	London region/urban	Yes	Yes
Jones and Tinker (1984)	D/I	ND	Wet deposition/rain gauge, weir	Preston, U.K./urban	Yes	ND
Klappenbach (1991)	D/I	1982–89	Wet deposition/precipitation collectors	Great Lakes region/urban and rural	Yes	Yes
Klaus (1985)	D/I	ND	Vehicle emissions/impactors	ND	Yes	Yes

Reference	Investigation type	Chemical analyte	QA/QC	Comments
Harrison and Johnston (1985)	A	Trace elements, organic compounds	blanks	Documents a substantial decrease in pollutant deposition fluxes with distance from the highway
Harrison and others (1985)	A	Trace elements	blanks	Organic lead associated with atmospheric aerosols only is a minor component in comparison with gas-phase alkyllead or particulate inorganic lead
Hedges and Wren (1987)	A	Trace elements, major inorganic constituents	ND	Wind direction and seasonal changes affect deposition rates of certain metals
Herrick (1966)	A	ND	blanks	A revision for Recommended Standard Method for Continuing Dustfall Survey
Hewitt and Rashed (1990)	A,R	Trace elements, organic compounds	ND	Approximately 90% of lead emissions from fast-moving vehicles dispersed away from road; PAH compounds have >95% of lower molecular weight species dispersed away from road
Hicks and Matt (1988)	A	ND	ND	Modeling and measurement of dry deposition
Hicks and others (1991)	A	Organic compounds	blanks	Filterpack used to estimate dry-deposition rates on a routine basis
Hicks and others (1993)	A	ND	ND	Separate dry- and wet-deposition measurements more accurately characterize total deposition than bulk-deposition measurements
Ho and Tai (1988)	A	Trace elements, major inorganic constituents	blanks, replicates	Soil and grass samples used to estimate the extent of aerial deposition of metals in the roadside
Holdren and others (1993)	A	ND	ND	Overview of the selection of models to represent ecosystem responses to specified leveles of deposition
Holsen and Noll (1992)	A	Trace elements, major inorganic constituents, organic compounds	blanks, replicates	Coarse particles and the compounds associated with them are responsible for the majority of dry deposition
Hopke (1991)	A	ND	ND	Discussion of receptor models
Hvitved-Jacobson and Yousef (1991)	A	ND	ND	Smith et al. reported dust fall loads in the USA approximately 0.23 $g/m^2/d$, 0.16–1.53 at the central region and 0.07–0.18 in the southern region and 0.06–0.16 at the eastern region
James and Shivalingaiah (1985)	A,R	ND	ND	Discussion of runoff models used to simulate quantity and quality of storm water runoff
Jensen and Laxen (1987)	A	Trace elements	blanks, standards	Concentrations of particulate-bound lead in air measured to see decrease after phase-down of lead content in gasoline
Jones and Tinker (1984)	A,R	Major inorganic constituents, organic compounds, water-quality properties	ND	Discusses transport of pollutants from road surface by spray
Klappenbach (1991)	A	Trace elements, major inorganic constituents	ND	Maximum volume weighted mean deposition rates for Pb, Fe, Zn, Cu, Na occur during winter months
Klaus (1985)	A	Trace elements, major inorganic constituents, organic compounds	ND	Composition of flue gas deposits from an Otto-cycle and a diesel engine investigated

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Table 5. Reports that address highway-runoff constituent loads and atmospheric deposition sources—*Continued*

Reference	Report type	Period of sampling	Type of sampling/method	Location/setting	Document method	
					Sample collection	Analytical
Kobriger and Geinopolos (1984)	R/S: FHWA	September 1978–October 1981	Bulk deposition/precipitation gauge, dustfall bucket, weirs, flumes, level sensing instruments	Milwaukee, Wisc./urban; Harrisburg, Penn./rural; Sacramento, Calif./urban; Efland, N.C./rural	Yes	Yes
Lamoree and Turner (1999)	D/I	February–April 1997	Vehicle emissions/MiniVol Portable Survey Samplers	St. Charles County, Mo./rural; St. Louis, Mo./urban	Yes	Yes
Lazrus and others (1970)	D/I	September 1966–March 1967	Bulk deposition/automated precipitation collector	32 stations across United States	Yes	Yes
Malmquist (1979)	D/I	April–October 1976	Bulk deposition/open beakers	Goteborg, Sweden/urban	ND	ND
Miguel and others (1998)	D/I	August 20–28, 1996	Vehicle emissions/stainless steel canisters	San Francisco Bay region, Calif./urban	Yes	Yes
Moe and others (1982)	D/I: FHWA	September 1977–September 1979	Bulk deposition/automatic recording rain gage, automatic water samplers, UC Davis stacked filter units	Dallas, Tex./urban	Yes	ND
Nader (1958)	D/I	ND	Dry deposition/dry deposition collectors	ND	Yes	Yes
Nikolaou and others (1997)	D/I	March 1993–March 1994	Air/CO sensors	Dallas, Lubbock, Houston, San Antonio, Tex./urban	Yes	Yes
Nilles and others (1992)	D/I	ND	Wet deposition/wet-dry deposition sampler, rain gage	United States/urban and rural	Yes	Yes
Nilles and others (1994)	R/S	1998–91	Wet deposition/wet-dry deposition sampler, rain gage	United States/urban and rural	Yes	Yes
Ondov and others (1982)	D/I	April 17–23, 1973; August 1973 December 1973–March 1974;	Vehicle emissions	ND	Yes	Yes
Pankow and others (1984)	D/I	March–April 1982; October–December 1982	Wet deposition/rain collector	Portland, Oreg./urban and semi-rural	Yes	Yes
Pankow and others (1998)	D/I	April–December 1997	Bulk deposition/multi-sorbent air-sampling cartridges	N.J./urban	Yes	Yes
Peters and Reese (1995)	D/I	April 14–June 30, 1992	Bulk deposition/wet-dry collector, dryfall collector, bulk collector	Lake Okeechobee, Clewiston, Fla./urban	Yes	Yes
Pirrone and Keeler (1993)	D/I	July 8–August 9, 1991	Bulk deposition/dichotomous sampler	Chicago, Ill./urban; Kankakee, Ill./rural	ND	ND

Reference	Investi-gation type	Chemical analyte	QA/QC	Comments
Kobriger and Geinopolos (1984)	A,R	Trace elements, major inorganic constituents, organic compounds, water-quality properties	controls	Discussion of constituents of highway runoff, sources and migration of pollutants to receiving water, effects of pollutants and treatment methods
Lamoree and turner (1999)	A	Organic compounds	blanks	Both PM20 and PM 2.5 the mass concentrations decreased with increasing distance downwind of the roadway
Lazrus and others (1970)	A	Trace elements, major inorganic constituents	ND	On basis of geographical distributions, lead, zinc, copper, iron, and manganese in atmospheric precipitation are derived primarily from human activity
Malmquist (1979)	A,R	Trace elements, major inorganic constituents, organic compounds	ND	Atmospheric fallout contributes substantially to the storm-water pollutants
Miguel and others (1998)	A	Organic compounds	ND	Gas- and particle-phase pollutant concentrations measured to determine PAH and black carbon emissions
Moe and others (1982)	A,R	Trace elements, major inorganic constituents, organic compounds, water-quality properties	ND	Particulate levels decreased sharply after rainshowers
Nader (1958)	A	ND	ND	No substantial differences measured among three different types of dustfall collectors
Nikolaou and others (1997)	A	Major inorganic constituents	ND	No evidence of increased carbon monoxide levels in vicinity of elevated or depressed freeways in comparison to at-grade level freeways
Nilles and others (1992)	A	Major inorganic constituents, water-quality properties	ND	Relative error increased for most analytes at lower concentrations; laboratory error is estimated to account for typically one-fifth of the overall collocated sampling error
Nilles and others (1994)	A	Major inorganic constituents, organic compounds, water-quality properties	duplicates	Colocation bias typically accounted for less than 25 percent of the overall measurement error for each site
Ondov and others (1982)	A	Trace elements, major inorganic constituents	ND	Lead, bromine, chloride, and barium can be used as markers of motor-vehicle contributions to pollution
Pankow and others (1984)	A	Organic compounds	blanks	Discussed design and use of a rain sampler
Pankow and others (1998)	A	Organic compounds	blanks, spikes, standards	Adsorption/thermal desorption with multi-sorbent air-sampling cartridges
Peters and Reese (1995)	A	Major inorganic constituents, organic compounds	blanks	Most of the Total-P in the dry deposition was Ortho-P, whereas only a small amount of the Total-P in the wet deposition was Ortho-P; inputs of insects to the bulk and dryfall-collectors factor affecting deposition estimates for P and NH3 + Org-N
Pirrone and Keeler (1993)	A,R	Trace elements, organic compounds	ND	Dry deposition of fine and coarse particles substantial source of trace metals discharged to sewer systems by storm water runoff

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Table 5. Reports that address highway-runoff constituent loads and atmospheric deposition sources—*Continued*

Reference	Report type	Period of sampling	Type of sampling/method	Location/setting	Document method Sample collection	Analytical
Randall and others (1981)	D/I	September 30, 1976– May 6, 1977	Bulk deposition/recording rain gages, automatic samplers, plastic funnels and containers	Virginia/urban	Yes	Yes
Revitt and others (1990)	D/I	July–September 1984	Bulk deposition/deposit gages, rain gage	Chilwell Gardens, Greater London/urban	ND	ND
Rice and Bricker (1996)	D/I	1991–93	Bulk deposition/wet-dry deposition collector	Catoctin Mt., Md./rural	Yes	Yes
Risley and Shanley (1994)	D/I	October 1986– December 1987	Wet deposition/wet-dry precipitation collector	Quabbin Reservoir region, Mass./rural	Yes	Yes
Rogge and others (1993)	D/I	May 1988	Vehicle emissions/vacuum sweeper truck	Los Angeles area/urban	Yes	Yes
Roorda-Knape and others (1999)	D/I	May 1995–August 1995	Vehicle emissions/ND	West of Netherlands/urban	ND	ND
Sanderson and others (1963)	D/I	November 1960– December 1961	Dry deposition/dustfall collectors	ND	ND	Yes
Sartor and others (1972)	R/S	ND	Street surface contaminants/ND	US/urban	ND	ND
Schroder and Hedley (1986)	D/I	May 11, 1982– May 13, 1982	Wet deposition/precipitation collectors	Denver, Colo./urban	Yes	Yes
Schroder and Malo (1987)	D	ND	Wet deposition/rain gage	ND	Yes	Yes
Schroder and Malo (1984)	D/I	ND	Bulk deposition/wet-dry collector	Raleigh, N.C./urban	Yes	Yes
Schroder and others (1989)	D/I	1985–88	Wet deposition/automatic wet-deposition sampler	Denver, Colo./urban	Yes	Yes
Schroeder (1995)	D/I	1986–87	Water samples/bailers	Fresno, Calif./urban	Yes	Yes
Schultz (1994)	D/I	June 15, 1988– August 31, 1989	Wet deposition/wet-dry automatic samplers	Cincinnati, Ohio/urban	Yes	Yes
Shahin and others (2000)	D/I	December 1993– October 1995	Dry deposition/automatic dry-deposition collectors	Lake Michigan region/urban	Yes	Yes

Reference	Investi-gation type	Chemical analyte	QA/QC	Comments
Randall and others (1981)	A,R	Trace elements, major inorganic constituents, organic compounds	ND	Washout of atmospheric contaminants occurred during the early stages of precipitation events
Revitt and others (1990)	A,R	Trace elements	ND	Contribution of local emissions to total deposition is considered and found to be important only for Zn; in rainfall metals are mainly particulate associated
Rice and Bricker (1996)	A	Trace elements, major inorganic constituents, water-quality properties	blanks, standards, duplicates	Contribution of large amounts of sulfate to watersheds by acidic deposition one of the underlying causes of acidification in watersheds
Risley and Shanley (1994)		Trace elements, major inorganic constituents, organic compounds, water-quality properties	ND	Discusses the effects of storm paths and variations within-storms on precipitation chemistry
Rogge and others (1993)	A	Organic compounds	ND	Fine particulate paved road dust is the third largest source of fine organic carbon particle emissions to urban atmosphere
Roorda-Knape and others (1999)	A	Major inorganic constituents, organic compounds	ND	Black smoke and NO_2 concentrations are higher close to motorways and decline with distance from the motorway
Sanderson and others (1963)	A	ND	duplicates	Comparison of dustfall collectors
Sartor and others (1972)	R	Trace elements, major inorganic constituents, organic compounds, water-quality properties	ND	Street surface contaminants contain high concentrations of materials known to cause problems of water pollution
Schroder and Hedley (1986)	A	Trace elements, major inorganic constituents, organic compounds, water-quality properties	blanks	During the beginning of a storm event, most of the inorganic constituents are at maximum concentration
Schroder and Malo (1987)	A	ND	blanks	Quality assurance program developed by the National Trends Network to collect long-term quality data
Schroder and Malo (1984)	A	Major inorganic constituents, water-quality properties	ND	Discusses the quality of wet deposition in the U.S.
Schroder and others (1989)	A	Trace elements, major inorganic constituents, organic compounds, water-quality properties	blanks	Precipitation acidity is associated with magnesium, fluoride, chloride, sodium, and calcium concentrations
Schroeder (1995)	A,R	Trace elements, major inorganic constituents, organic compounds, water-quality properties	standards	Wide variety of inorganic and organic contaminants generated in the catchment are removed by sorption within the top 4 cm of sediment in the recharge basin, and the contaminants have not degraded ground-water quality beneath the basin.
Schultz (1994)	A	Organic compounds, water-quality properties	ND	Urban wet deposition nitrate differs from non-urban deposition when compared on a monthly or seasonal basis, but not an annual basis
Shahin and others (2000)	A	Trace elements, major inorganic constituents	method detection limits, blanks, replicates	Automatic dry-deposition sampler developed that can make long term measurements

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Table 5. Reports that address highway-runoff constituent loads and atmospheric deposition sources—*Continued*

Reference	Report type	Period of sampling	Type of sampling/method	Location/setting	Document method Sample collection	Document method Analytical
Shi and others (1999)	D/I	November 1996–December 1997	Dry deposition/scanning mobility particle sizers, electrical low pressure impactor, condensation particle counter, thermophoretic precipitator	Birmingham, U.K./urban	Yes	ND
Shivalingaiah and James (1982)	M/I	ND	Bulk deposition/ND	Hamilton, Ontario, Canada/ urban	ND	ND
Shivalingaiah and James (1983)	M/I	ND	Bulk deposition/ND	Hamilton, Ontario, Canada/ urban	ND	ND
Simcik and others (1997)	D/I	November 1993–October 1995	Dry deposition/automatic dry deposition collectors	Lake Michigan region/urban	Yes	Yes
Simmons and Pocock (1987)	D/I	November–December 1982; February–March 1983	Dry deposition/ deposit canisters	Walsall, West Midlands, U.K./ urban	Yes	Yes
Sirois and Barrie (1988)	D/I	1979–82	Wet deposition/wet-only collector	Canada/urban	Yes	Yes
Stockham and others (1966)	D/I	February 1963–March 1964	Dry deposition/dustfall collectors	Chicago, Ill./urban	Yes	Yes
Tramontano and others (1987)	D/I	ND	Wet deposition/automatic collector	Lewes, Del./rural; Bermuda/ rural	Yes	Yes
Vandenberg and Knoerr (1985)	D/I	January 1981–April 1982	Dry deposition/deposition buckets, deposition plates, petri dishes, filter paper	Duke Forest, N.C./rural	Yes	Yes
Ward (1990)	D/I	ND	Dry deposition, soil/ND	U.K./urban	ND	Yes
Warren and Birch (1987)	D/I	ND	Dry deposition and soils/Warren Spring Laboratory 'M' type sampling system and dustpan and brush	East London/urban	Yes	Yes
Westerholm and others (1988)	D/I	ND	Vehicle emissions/dilution tunnel	Sweden/urban	Yes	Yes
Williams (1987)	R/S	ND	Vehicle emissions/	U.K./urban	ND	ND
Willoughby and others (1991)	R/S	1989	Wet deposition/wet-deposition collectors	US/urban and rural	Yes	Yes
Willoughby (1995)	D/I	June 1992–August 1993	Wet deposition/wet-dry collector	Gary, Ind./urban	Yes	Yes

Reference	Investi-gation type	Chemical analyte	QA/QC	Comments
Shi and others (1999)	A	Organic compounds	ND	Total particle number concentration higher at roadside; decrease in particle number concentration was greater than that of the mass concentration
Shivalingaiah and James (1982)	A,R	ND	ND	SWMM (Storm water management model) III runoff quality algorithms used to determine pollutant accumulation
Shivalingaiah and James (1983)	A,R	ND	ND	Addition of atmospheric-dustfall subroutine used to simulate pollutant accumulation on individual subcatchments NEWBLD to SWMM3-RUNOFF model improved the computed pollutographs
Simcik and others (1997)	A	Organic compounds	method detection limits, blanks	Particle mass and elemental fluxes higher in Chicago than in rural areas
Simmons and Pocock (1987)	A	Trace elements, major inorganic constituents	ND	Metal deposition rates in urban areas are highly spatially variable
Sirois and Barrie (1988)	A	Organic compounds, water-quality properties	ND	Dry deposition estimated from total deposition
Stockham and others (1966)	A	Organic compounds	duplicates	Discusses the variability of dustfall analysis because of the container and the collecting fluid
Tramontano and others (1987)	A	Trace elements	blanks	Discusses the results of the analysis of trace metals in precipitatio
Vandenberg and Knoerr (1985)	A	Major inorganic compounds	standards	Wide range of surface deposition rates are estimated form the variety of deposition surfaces used in this study shows uncertainty of the surrogate surface-measurement techniques
Ward (1990)	A	Trace elements, major inorganic constituents, water-quality properties	blanks, replicates	Elements related to motor vehicles are elevated in surface soils due to dry and wet deposition of emission or wear of components
Warren and Birch (1987)	A	Trace elements	ND	Highest overall contamination occurs at a site with highest traffic density
Westerholm and others (1988)	A	Organic compounds	ND	A significant fraction of emitted PAH is formed in the combustion process (>50%)
Williams (1987)	A	Organic compounds	ND	Summarization of the contribution of motor vehicles to the emissions and air quality in terms of the more important air pollutants in the U.K.
Willoughby and others (1991)	A	Major inorganic constituents, organic compounds, water-quality properties	duplicates	No significant difference among laboratory determinations for calcium, magnesium, sodium, potassium, ammonium, chloride, nitrate, sulfate, hydrogen ion, and specific conductance
Willoughby (1995)	A	Trace elements, major inorganic constituents, organic compounds, water-quality properties	controls, blanks	Chemical quality of wet deposition monitored; annual loadings could be used to estimate the contribution of wet deposition to the total annual constituent loadings

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Table 5. Reports that address highway-runoff constituent loads and atmospheric deposition sources—*Continued*

Reference	Report type	Period of sampling	Type of sampling/method	Location/setting	Document method	
					Sample collec-tion	Analyt-ical
Wu and others (1992)	D/I	June–July 1987; May and July 1988, May 1989	Dry deposition/deposition plates	Claremont, Calif./urban; Gettysburg, Penn./urban	Yes	Yes
Wu and others (1998)	D/I	August 1995–July 1996	Bulk deposition/automatic sampler, tipping bucket, nonrecording rain gage	Charlotte, N.C./urban and rural	Yes	Yes
Yu and others (1994)	D/I	June 5, 1990– July 9, 1991	Dry deposition/aerosol samplers, impactors	Chesapeake Bay region/rural	Yes	Yes
Zweidinger and others (1988)	D/I	May 1983	Vehicle emissions/roadside samplers	Raleigh, N.C. (U.S. Highway 70)/ urban	Yes	Yes

Reference	Investigation type	Chemical analyte	QA/QC	Comments
Wu and others (1992)	A	Trace elements, major inorganic constituents, organic compounds	blanks, duplicates	Surface resistance may be the rate-limiting factor in determining overall dry deposition
Wu and others (1998)	A,R	Trace elements, major inorganic constituents, organic compounds, water-quality properties	ND	Total suspended solids were higher at a bridge site than on typical urban highways
Yu and others (1994)	A	Trace elements, major inorganic constituents, organic compounds	blanks	Spatial and temporal differences of elemental concentrations examined
Zweidinger and others (1988)	A	Organic compounds	ND	Evaluated methods for estimating emission factors

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