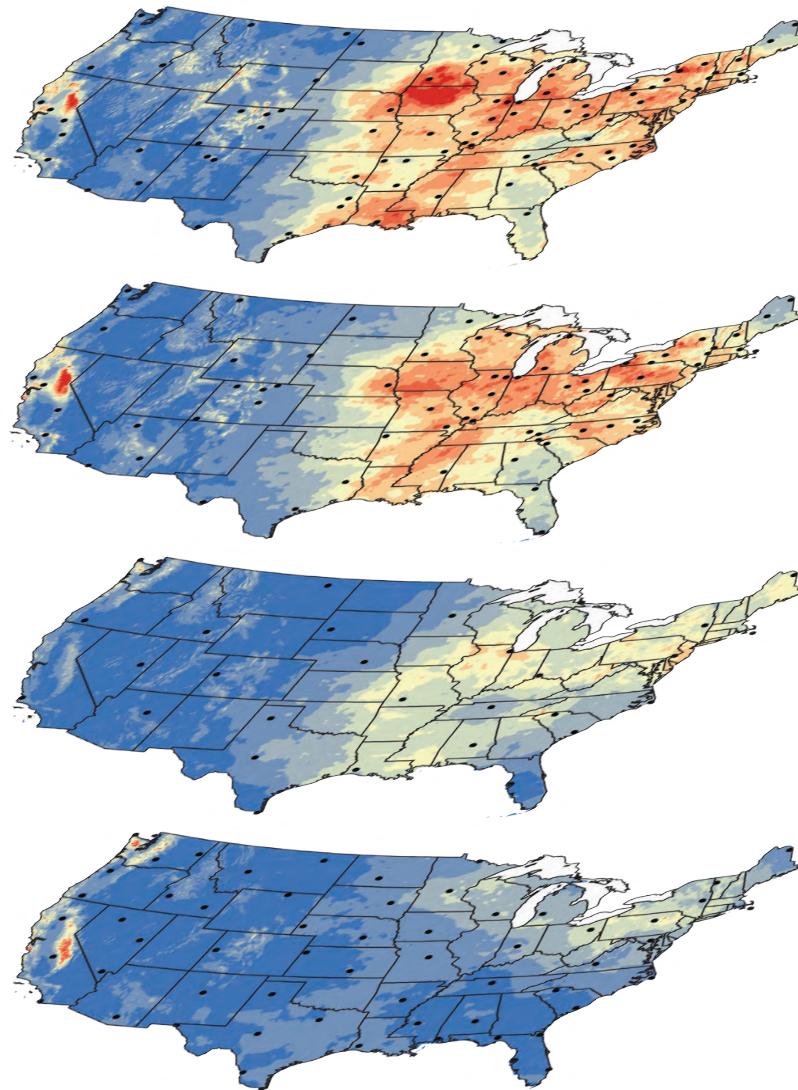


Estimates of Inorganic Nitrogen Wet Deposition from Precipitation for the Conterminous United States, 1955–84



Scientific Investigations Report 2014–5067

Front Cover. Top, Inorganic nitrogen deposition for 1983; Top middle, Inorganic nitrogen deposition for 1982; Bottom middle, Inorganic nitrogen deposition for 1964; Bottom, Inorganic nitrogen deposition July 1955–June 1956.

Back Cover. Kings River Experimental Watershed (National Atmospheric Deposition Program, National Trends Network, CA28) monitoring station, Sierra National Forest, California. Photograph taken by Eric Hebert.

Estimates of Inorganic Nitrogen Wet Deposition from Precipitation for the Conterminous United States, 1955–84

By Jo Ann M. Gronberg, Amy S. Lutzke, and Donna L. Knifong

National Water-Quality Assessment Program

Scientific Investigations Report 2014–5067

**U.S. Department of the Interior
U.S. Geological Survey**

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SALLY JEWELL, Secretary

U.S. Geological Survey
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Foreword

The U.S. Geological Survey (USGS) is committed to providing the Nation with reliable scientific information that helps to enhance and protect the overall quality of life and that facilitates effective management of water, biological, energy, and mineral resources (<http://www.usgs.gov/>). Information on the Nation's water resources is critical to ensuring long-term availability of water that is safe for drinking and recreation and is suitable for industry, irrigation, and fish and wildlife. Population growth and increasing demands for water make the availability of that water, measured in terms of quantity and quality, even more essential to the long-term sustainability of our communities and ecosystems.

The USGS implemented the National Water-Quality Assessment (NAWQA) Program in 1991 to support national, regional, State, and local information needs and decisions related to water-quality management and policy (<http://water.usgs.gov/nawqa>). The NAWQA Program is designed to answer: What is the quality of our Nation's streams and groundwater? How are conditions changing over time? How do natural features and human activities affect the quality of streams and groundwater, and where are those effects most pronounced? By combining information on water chemistry, physical characteristics, stream habitat, and aquatic life, the NAWQA Program aims to provide science-based insights for current and emerging water issues and priorities. From 1991 to 2001, the NAWQA Program completed interdisciplinary assessments and established a baseline understanding of water-quality conditions in 51 of the Nation's river basins and aquifers, referred to as Study Units (http://water.usgs.gov/nawqa/studies/study_units.html).

In the second decade of the Program (2001–2012), a major focus is on regional assessments of water-quality conditions and trends. These regional assessments are based on major river basins and principal aquifers, which encompass larger regions of the country than the Study Units. Regional assessments extend the findings in the Study Units by filling critical gaps in characterizing the quality of surface water and groundwater, and by determining water-quality status and trends at sites that have been consistently monitored for more than a decade. In addition, the regional assessments continue to build an understanding of how natural features and human activities affect water quality. Many of the regional assessments employ modeling and other scientific tools, developed on the basis of data collected at individual sites, to help extend knowledge of water quality to unmonitored, yet comparable areas within the regions. The models thereby enhance the value of our existing data and our understanding of the hydrologic system. In addition, the models are useful in evaluating various resource-management scenarios and in predicting how our actions, such as reducing or managing nonpoint and point sources of contamination, land conversion, and altering flow and (or) pumping regimes, are likely to affect water conditions within a region.

Other activities planned during the second decade include continuing national syntheses of information on pesticides, volatile organic compounds (VOCs), nutrients, trace elements, and aquatic ecology; and continuing national topical studies on the fate of agricultural chemicals, effects of urbanization on stream ecosystems, bioaccumulation of mercury in stream ecosystems, effects of nutrient enrichment on stream ecosystems, and transport of contaminants to public-supply wells.

The USGS aims to disseminate credible, timely, and relevant science information to address practical and effective water-resource management and strategies that protect and restore water quality. We hope this NAWQA publication will provide you with insights and information to meet your needs, and will foster increased citizen awareness and involvement in the protection and restoration of our Nation's waters.

The USGS recognizes that a national assessment by a single program cannot address all water-resource issues of interest. External coordination at all levels is critical for cost-effective management, regulation, and conservation of our Nation's water resources. The NAWQA Program, therefore, depends on advice and information from other agencies—Federal, State, regional, interstate, Tribal, and local—as well as nongovernmental organizations, industry, academia, and other stakeholder groups. Your assistance and suggestions are greatly appreciated.

William H. Werkheiser
USGS Associate Director for Water

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Conversion Factors

Inch/Pound to SI

Multiply	By	To obtain
Length		
inch (in.)	2.54	centimeter (cm)
inch (in.)	25.4	millimeter (mm)
Mass		
ton per day per square mile [(ton/d)/mi ²]	0.3503	megagram per day per square kilometer [(Mg/d)/km ²]

SI to Inch/Pound

Multiply	By	To obtain
Length		
centimeter (cm)	0.3937	inch (in.)
millimeter (mm)	0.03937	inch (in.)
meter (m)	3.281	foot (ft)
kilometer (km)	0.6214	mile (mi)
meter (m)	1.094	yard (yd)
Application rate		
kilograms per hectare per year (kg/ha)	0.8921	pounds per acre per year (lb/acre)
kilograms per hectare per year [(kg/ha)/yr]	0.8921	pounds per acre per year [(lb/acre)/yr]

Horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).

Concentrations of chemical constituents in water are given in milligrams per liter (mg/L).

Abbreviations

AirMoN	Atmospheric Integrated Research Monitoring Network
ASCII	American Standard Code for Information Interchange
EPA (USEPA)	U.S. Environmental Protection Agency
MAP3S	Multistate Atmospheric Power Production Pollution Study
NAD 83	North American Datum of 1983
NADP	National Atmospheric Deposition Program
NAWQA	National Water-Quality Assessment
NCDC	National Climatic Data Center
NH_4^+	ammonium
NO_3^-	nitrate
NOAA	National Oceanic and Atmospheric Administration
NTN	National Trends Network
NWS	National Weather Service
PCN	Precipitation Chemistry Network
PRISM	Parameter-elevation Regressions on Independent Slopes Model
USGS	U.S. Geological Survey
WMO	World Meteorological Organization

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Estimates of Inorganic Nitrogen Wet Deposition from Precipitation for the Conterminous United States, 1955–84

By Jo Ann M. Gronberg, Amy S. Lutke, and Donna L. Knifong

Abstract

The U.S. Geological Survey's National Water-Quality Assessment program requires nutrient input information for analysis of national and regional assessment of water quality. Historical data are needed to lengthen the data record for assessment of trends in water quality. This report provides estimates of inorganic nitrogen deposition from precipitation for the conterminous United States for 1955–56, 1961–65, and 1981–84. The estimates were derived from ammonium, nitrate, and inorganic nitrogen concentrations in atmospheric wet deposition and precipitation-depth data. This report documents the sources of these data and the methods that were used to estimate the inorganic nitrogen deposition. Tabular datasets, including the analytical results, precipitation depth, and calculated site-specific precipitation-weighted concentrations, and raster datasets of nitrogen from wet deposition are provided as appendixes in this report.

Introduction

In 1991, the U.S. Geological Survey (USGS) began implementation of the National Water-Quality Assessment (NAWQA) program to assess status and trends of surface-water and groundwater quality in the United States. Data development of nutrient inputs to the land surface of fertilizer, manure, and atmospheric deposition has been an essential part of these assessments. Atmospheric deposition was determined to be the largest nonpoint source of nitrogen input to undeveloped watersheds in the eastern part of the United States where the deposition rates are highest, in areas near the Great Lakes, and in the arid and mountainous West where human development is sparse (Dubrovsky and others, 2010).

Long-term changes (over several decades) in nitrogen deposition will be an important consideration in upcoming NAWQA trend assessments. Currently (2014), the longest national-scale record of nitrogen deposition is available from the National Atmospheric Deposition Program (NADP)

National Trends Network (NTN). The NADP NTN began in 1978, and national maps of inorganic nitrogen deposition are available for 1985 through 2012. Because of the importance of historical atmospheric deposition data in long-term trend assessment, carried out by NAWQA and other programs, there is a need to lengthen the period of record by adding nitrogen data that pre-dates the NADP NTN data. By expanding the record length, future trend studies can be more inclusive.

Purpose and Scope

The purpose of this report is to provide estimates of inorganic nitrogen from atmospheric wet deposition for the conterminous United States. These estimates are presented as maps and accompanying raster datasets of inorganic nitrogen deposition for 1955–56 (derived from Junge, 1958), 1961–65 (derived from Lodge and others, 1968), and for 1981–84 (derived from the National Atmospheric Deposition Program, 2013b) based on an interpolation of the various monitoring points throughout the United States. Estimates of nitrogen deposition provide information critical for characterizing the current and historical nutrient inputs to surface-water basins and groundwater recharge areas. These estimates consequently provide a method for explaining the occurrence and distribution of nutrients in surface water and groundwater for national and regional assessments of water quality and for evaluating management practices designed to reduce nitrogen in atmospheric deposition.

This report summarizes ammonium (NH_4^+), nitrate (NO_3^-), and inorganic nitrogen concentrations in atmospheric wet deposition, and their associated precipitation depths from 1955 through 1984, reconstructed from national historical datasets. The web address of the NADP NTN deposition data from 1985 to 2012 also is provided. Wet deposition removes gases and particles in the atmosphere and deposits them to the Earth's surface by way of rain, sleet, snow, and fog. Dry deposition is the process through which particles and gases are deposited in the absence of precipitation (Nilles, 2000). Dry and bulk (wet plus dry) deposition were not estimated

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in this report. Data files were compiled from each historical source and include station identifier, location, date, concentration, precipitation depth, and calculated annual precipitation-weighted mean concentrations (ammonium and nitrate, or inorganic nitrogen, depending on the data source). Precipitation-weighted mean concentrations are used to compensate for the different amounts of precipitation associated with the concentration values. Concentrations are weighted using the proportion of the associated precipitation to the total precipitation of the period (in this case, for the year) to calculate the mean. Methods for assembling the data, calculating site-level annual precipitation-weighted concentrations, and generating continuous precipitation-weighted mean concentration and inorganic nitrogen deposition across the conterminous United States, including the assumptions and restrictions applied, are described.

Data Sources

Site-specific concentration data and the associated precipitation-depth data (hereinafter referred to as “precipitation data”), are needed for calculating annual precipitation-weighted mean concentrations. Spatially continuous national raster datasets of these concentrations are interpolated from the site data and then used with the spatially continuous annual precipitation data to generate raster datasets of inorganic nitrogen deposition. The national station-level concentration datasets and the station-level and spatially continuous precipitation data needed for these calculations are described below. Regional concentration datasets also are described.

National Concentration Datasets

During the past 60 years, there have been four primary networks that have monitored wet deposition chemistry across the conterminous United States (Stensland and others, 1986). The first of these networks, herein referred to as “Junge,” was operated by the Air Force Cambridge Research Center from July 1955 through June 1956 (Junge and Gustafson, 1956). The second network, herein referred to as “Lodge,” was operated from 1960 to 1966 by the Public Health Service and the National Center for Atmospheric Research (Lodge and others, 1968). The third network was a joint effort operated by the National Oceanic and Atmospheric Administration (NOAA), U.S. Environmental Protection Agency (EPA), and World Meteorological Organization (WMO) from 1972 to 1982, herein referred to as the “NOAA–EPA–WMO network” (Artz and Miller, 1985). The fourth network, the NADP NTN (National Atmospheric Deposition Program, 2013c), began in 1978, overlapped the NOAA–EPA–WMO network, and is

still currently (2014) active. The NOAA–EPA–WMO network became part of the NADP NTN in 1980 and changed its sampling protocols to match the NADP NTN (Artz and Miller, 1985). A comparison of these national networks is provided in table 1, which summarizes the period of study, site locations, collection method, sampler type, preservative, reporting period, method reporting limit, and method of analysis. The time periods of these networks are summarized in figure 1.

Junge (July 1955–June 1956)

A network of 67 stations was established in the United States and over parts of the adjacent Atlantic Ocean to collect samples for chemical analysis (Junge and Gustafson, 1956). Three of the 67 stations were located outside the conterminous United States and three were ocean stations. Not all stations were shown in the Junge and Gustafson (1956) site map. Event-based, wet-deposition samples were collected from July 1955 through June 1956 and pooled together for average monthly concentrations of ammonium and nitrate, reported in milligrams per liter (mg/L). Event-based samples are defined as samples collected during a precipitation event. The monthly averages were then averaged again for 3-month periods (or quarters) from July 1955 through June 1956. The quarterly averages were presented in site maps (Junge, 1958); the original monthly averages were not in the publication. The site maps in Junge (1958) were labeled with dates July–September 1956, October–December 1956, January–March 1957, and April–June 1957. The map labels did not correspond to the report description of data collection from July 1955 through June 1956. Therefore, the authors assumed the map labels were in error and the data were treated as if the sample dates were from July 1955 through June 1956. Quarter 1 was defined as July–September 1955, quarter 2 as October–December 1955, quarter 3 as January–March 1956, and quarter 4 as April–June 1956. It is assumed that the quarterly averages are precipitation-weighted concentrations. The reporting limit for ammonium was 0.02 mg/L. When ammonium is below the detection limit, a value of 0.01 mg/L was reported on the ammonium maps (Junge, 1958). Although not explicitly stated in Junge (1958), the reporting limit for nitrate was assumed to be 0.02 mg/L, with a nondetect value of 0.01 mg/L. Annual concentration values for chloride, sodium, potassium, calcium, and sulfate were presented in a corresponding report (Junge and Werby, 1958), but are not used in this report. Station locations were shown with two-letter identifiers on the source report map (Junge and Gustafson, 1956) (fig. 2); however, latitude and longitude were not provided in either Junge and Gustafson (1956) or Junge (1958). Also, precipitation associated with the concentration data was not provided.

Table 1. Characteristics of the national chemistry networks.

[Abbreviations: Apr., April; Aug., August; Cu, copper; Dec., December; EPA, Environmental Protection Agency; Feb., February; Jan., January; Mar., March; mg/L, milligrams per liter; N, nitrogen; NA, not applicable; NADP, National Atmospheric Deposition Program; NH₄⁺, ammonium; NO₂⁻, nitrite; NO₃⁻, nitrate; NOAA, National Oceanic and Atmospheric Administration; Nov., November; NTN, National Trends Network; Oct., October; ppm, parts per million; Q, quarter; Sep., September; WMO, World Meteorological Organization; Zn, zinc]

Study	Junge ¹	Lodge and others ²	NOAA-EPA-WMO ³	NADP NTN ⁴
Period of study	July 1955–June 1956	Dec. 1960–Nov. 1966	1972–82	1978–present
Site location	National Weather Service facilities with 24-hour service, tended to be small airports	National Weather Service facilities, tended to be small airports	National Weather Service sites	National Atmospheric Deposition Program National Trends Network
Number of sites	67	33	11	⁵ 1979: 18–2012: 236
Collection method	Event based Manual exposure Pooled monthly samples	Event based Automated sampler Moisture-sensing grid Pooled monthly samples	Pre-1980: Automated sampler Pooled monthly samples Post-1980: Automated sampler, Moisture-sensing heated grid Weekly samples (no pooling)	Event based Automated sampler Moisture-sensing heated grid Weekly samples
Sampler	Wet-only sampler plexiglass funnel and polyethylene bottle in plywood box, heating device to melt snow.	Wet-only polyethylene bucket, transferred to polyethylene bottle	Pre-1980: Wet-only Misco collector, bucket poured into polyethylene bottle Post-1980: Wet-only Aerochem collector	Wet-only Aerochem collector, sample shipped out weekly for analysis
Preservative	Toluene	Toluene	Pre-1980: Chlorobenzene Post-1980: no preservative	None
Data reported	Quarterly average concentrations (mg/L)	Quarterly average concentrations (ppm) for a 6-year period	Pre-1980: monthly concentrations (mg/L) Post-1980: bi-weekly concentrations (mg/L)	Weekly
Quarters	Q1: July–Sep. 1956 Q2: Oct.–Dec. 1956 Q3: Jan.–Mar. 1957 Q4: Apr.–June 1957	Q1: Dec.–Feb. 1960–66 Q2: Mar.–May 1960–66 Q3: June–Aug. 1960–66 Q4: Sep.–Oct. 1960–66	NA	NA
NO₃⁻ reporting limit (mg/L)	⁶ 0.02	NA	0.02	1978–84: 0.02 1985–99: 0.03 1999–2012: 0.004–0.01
NH₄⁺ reporting limit (mg/L)	0.02	NA	0.02	1978–2004: 0.02 2005–12: 0.003–0.01
Inorganic nitrogen reporting limit (mg/L)	NA	⁷ 0.01	NA	NA
NO₃⁻ value reported when below detection limit (mg/L)	¹ 0.01	NA	NA	NA
NH₄⁺ value reported when below detection limit (mg/L)	¹ 0.01	NA	NA	NA
Inorganic nitrogen value reported when below detection limit (mg/L)	NA	NA	NA	NA
NO₃⁻ method of analysis	Colorimetric by chloroform solution of brucine alkaloid and sulfuric acid	NA	Colorimetric, reduction of NO ₃ ⁻ to NO ₂ ⁻ with sulfamilamide	1978–85: Colorimetric cadmium reduction 1985–2012: Ion chromatography
NH₄⁺ method of analysis	Colorimetric by Nessler's reagent after microdistillation to avoid interference	NA	Colorimetric, alkaline phenol and sodium hypochlorite	Colorimetric, phenate
Total inorganic N method of analysis	NA	Colorimetric, reduction of NO ₂ ⁻ and NO ₃ ⁻ by Cu-Zn alloy and an indophenol ammonia test	NA	NA

¹Junge (1958).²Lodge and others (1968).³Artz and Miller (1985).⁴National Atmospheric Deposition Program (2013b and 2013c).⁵Number of sites that met completeness criteria for the annual NADP NTN deposition maps, varies yearly.⁶Inferred from dataset, reporting level not defined in report.⁷Inferred from dataset, lowest concentration reported. Authors calculated method reporting limit from published values of six runs of lowest calibration standard as 0.012 mg/L.

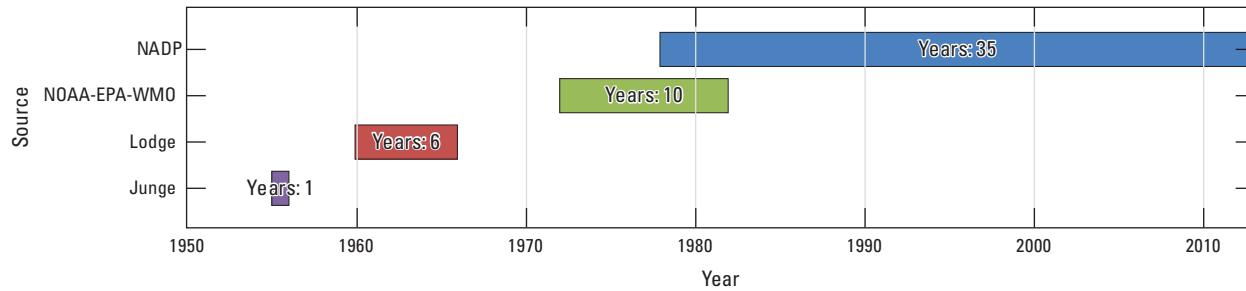


Figure 1. Time periods of national networks. EPA, U.S. Environmental Protection Agency; NADP, National Atmospheric Deposition Program; NOAA, National Oceanic and Atmospheric Administration; WMO, World Meteorological Organization.

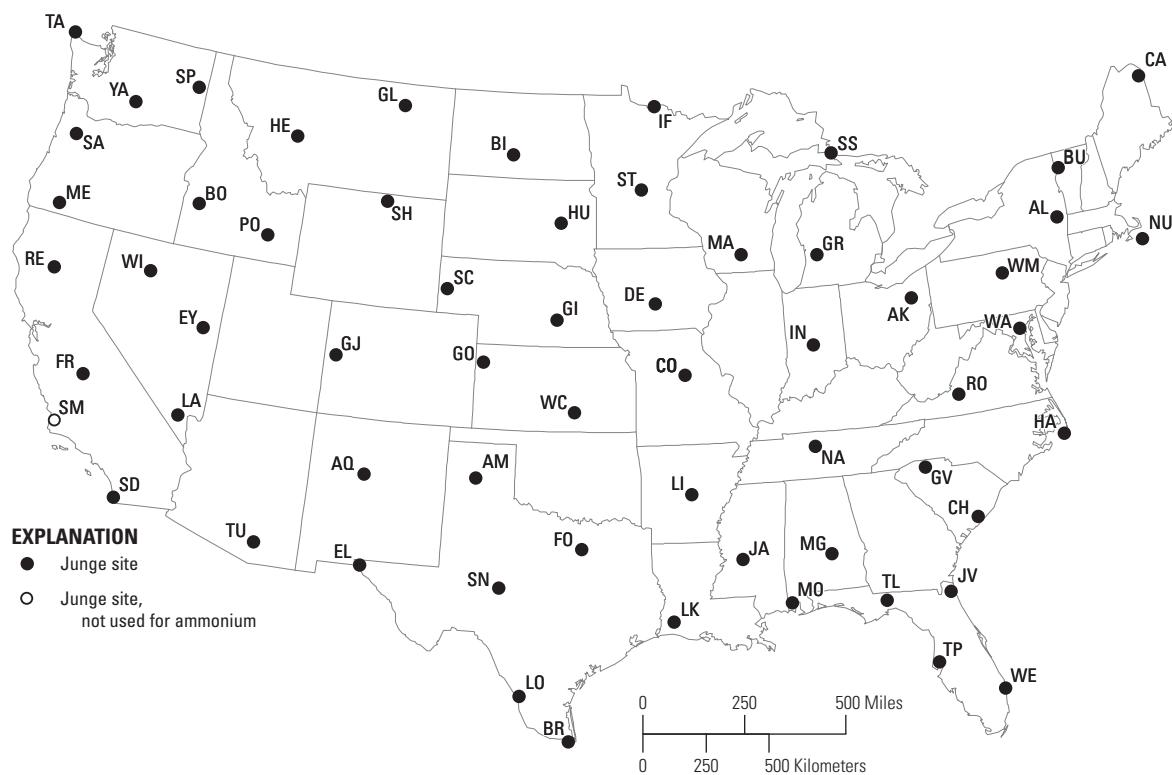


Figure 2. Location of Junge sites. See appendix 1 for more information on the sites.

Lodge (December 1960–November 1966)

The National Precipitation Sampling Network (also known as the Public Health Service/National Center for Atmospheric Research Network) was composed of 33 sites distributed throughout the conterminous United States (Lodge and others, 1968; fig. 3). Event-based, wet-deposition samples were collected from December 1960 through November 1966 and pooled together for monthly samples. Precipitation-weighted 6-year quarterly average concentrations of inorganic nitrogen (in milligrams per liter) were presented in concentration maps (Lodge and others, 1968). Data for each site consists of four values, with each value representing an average of the samples collected during the 6-year period for a given quarter. The quarters used by Lodge and others (1968) differ from those used by Junge (1958) and are grouped as follows: December–February, March–May, June–August, and September–November. The original sample values were not in the Lodge and others (1968) publication. The reporting limit for inorganic nitrogen was 0.01 mg/L (Lodge and others, 1968). Sulfate, chloride, sodium, potassium, calcium, and magnesium concentrations also were presented (Lodge and others, 1968), but were not used in this report. Station locations were shown on the source report map and listed by city and state (Lodge and others, 1968); however, latitude and longitude were not provided in the report. The L34 site was not listed in the Lodge site table; the Junge SM site location information was used for this site (fig. 2, fig. 3). Also, precipitation associated with the concentration data was not provided.

National Oceanic Atmospheric Administration, U.S. Environmental Protection Agency, World Meteorological Organization (1972–82)

The NOAA–EPA–WMO Regional Precipitation Chemistry Measurements consisted of data from 11 sites distributed throughout the United States (Artz and Miller, 1985; fig. 4). Samples were collected on a monthly basis from 1972 to 1980, and are referred to as the “pre-1980 dataset.” Artz and Miller (1985) did not discuss method reporting limits for the pre-1980 dataset; however, the lowest ammonium concentration value in the dataset was 0.05 mg/L and the lowest nitrate value was 0.02 mg/L. Calcium, magnesium, potassium, sodium, chloride, and sulfate concentrations also were collected, but were not used in this report. Latitude, longitude, elevation, and precipitation data were provided by Artz and Miller (1985). Beginning in 1980, the NOAA–EPA–WMO network joined with the NADP network and changed sampling equipment and sampling protocols. This “post-1980” part of the NOAA–EPA–WMO dataset is described in the NADP NTN section.

The NOAA–EPA–WMO pre-1980 dataset presented a challenge because of the sparse national coverage with only 11 sites and, for some sites, the limited number of values per year. Artz and Miller (1985) noted problems with the chemical

composition of the precipitation, determining that the problems were related to the collection of samples rather than analytical issues. Artz and Dayan (1986) noted higher deposition values for the NOAA–EPA–WMO study compared to the NADP dataset and attributed it to sample evaporation and poor sample collector efficiency. It was decided that this dataset would not be used to estimate nitrogen deposition.

National Atmospheric Deposition Program National Trends Network (1978–2012)

The NADP NTN has been monitoring atmospheric wet deposition since 1978 (National Atmospheric Deposition Program, 2013c). Wet-deposition samples are collected on a weekly basis. Chemistry data are available from 1978 to 2012, and raster datasets of concentrations and deposition for various analytes are available from 1985 through 2012 (National Atmospheric Deposition Program, 2013b). Annual precipitation-weighted ammonium and nitrate concentrations, date, latitude, longitude, elevation, and station precipitation data are part of the NADP NTN dataset. Site locations used in this report are shown on figure 5.

Regional Concentration Datasets

There were other atmospheric deposition studies taking place during these years: the USGS study from 1962 to 1963 (Gambell and Fisher, 1966), the USGS study from 1966 to 1968 (Pearson and Fisher, 1971), the Multistate Power Production Pollution Study Precipitation Chemistry Network (MAP3S/PCN) from 1976 to 1990 (Battelle Pacific Northwest Laboratories, 1977; and Butler and Likens, 1998), and the NADP Atmospheric Integrated Research Monitoring Network (AirMoN) from 1992 to present (National Atmospheric Deposition Program, 2013a). These studies fill some of the temporal gaps in the national datasets, but were all regional rather than national in scope, and in some cases, also were bulk deposition samples (which include dry and wet deposition), and therefore were excluded from the nitrogen estimations, herein. These datasets are described briefly.

A network of 27 sampling sites in eastern North Carolina and southeastern Virginia were sampled for major ions on a monthly basis from August 1962 through July 1963 (Gambell and Fisher, 1966). Sites were established at U.S. Weather Bureau cooperative-observer sites. Samplers were open continuously during the month and the data represent bulk deposition.

A network of 18 sites in New York, Pennsylvania, and New England were sampled for major ions from August 1966 through September 1968 (Pearson and Fisher, 1971). Not all sites were sampled during the entire period. Samplers were open continuously during the month and represent bulk deposition.

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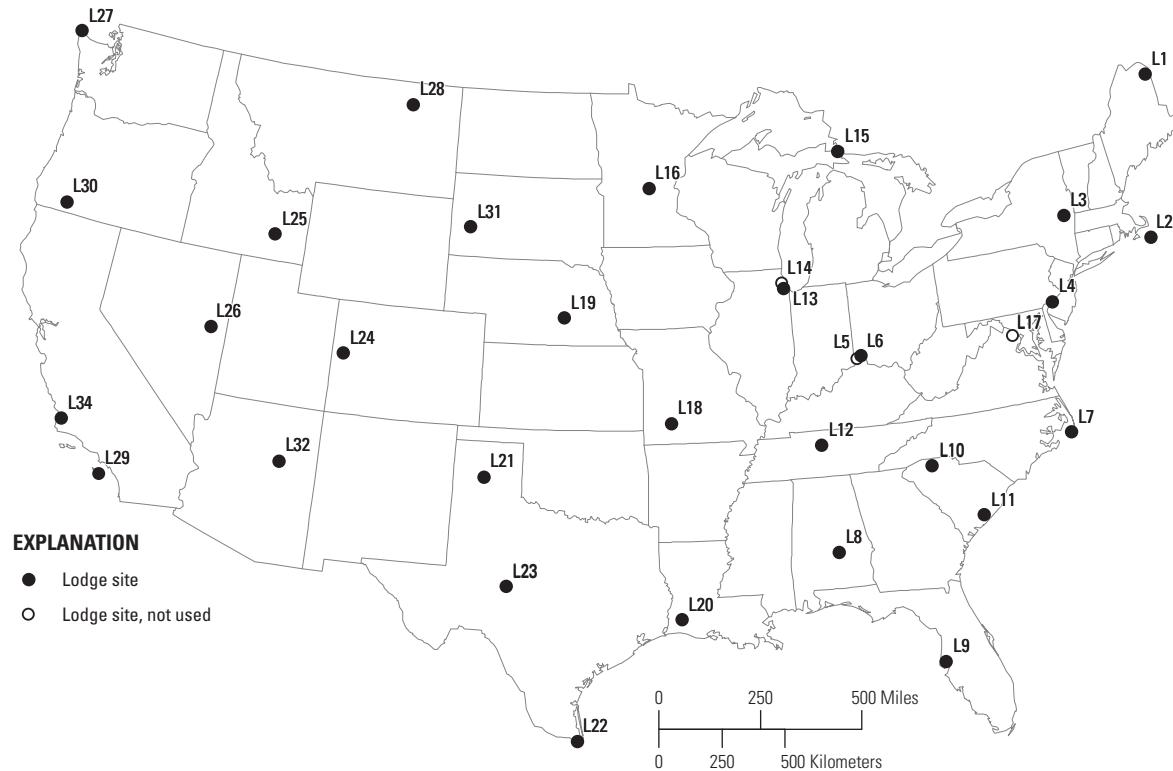


Figure 3. Location of Lodge sites. See appendix 2 for more information on the sites.



Figure 4. Location of NOAA-EPA-WMO sites. EPA, U.S. Environmental Protection Agency; NOAA, National Oceanic Atmospheric Administration; WMO, World Meteorological Organization. See appendix 3 for more information on the sites.

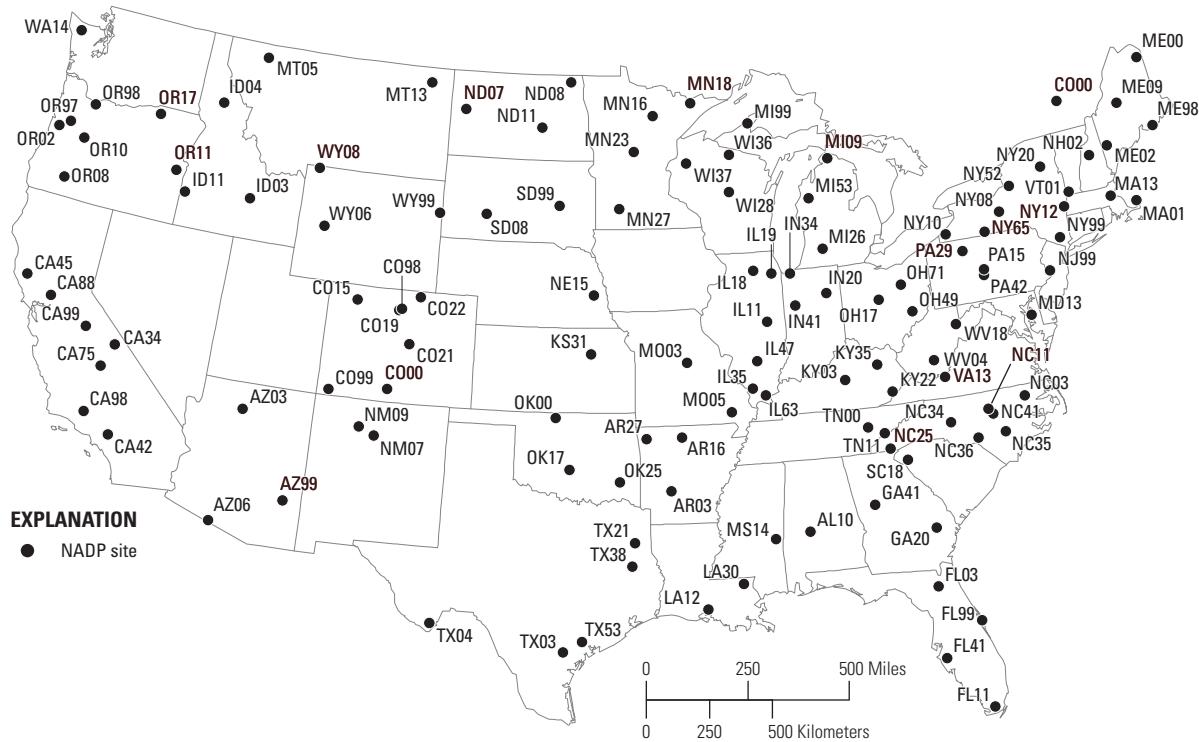


Figure 5. Location of NADP NTN sites, 1981–84. NADP, National Atmospheric Deposition Program; NTN, National Trends Network.

The MAP3S/PCN started sampling at four sites in September 1976 (Battelle Pacific Northwest Laboratories, 1977). Sampling continued until November 1990 at which time there were nine sites located in the northeastern United States (Butler and Likens, 1998). Samples were collected on an event basis and represent wet deposition.

The AirMoN (National Atmospheric Deposition Program, 2013a) started in 1992, using many of the sites from the MAP3S/PCN. The seven sites in the network are located in the northeastern United States. Samples are collected on a daily basis and represent wet deposition.

The locations of the sites in the national and regional concentration networks are shown in figure 6. In general, the sites in the national networks are well distributed throughout the conterminous United States, whereas sites in the regional networks are limited to the eastern one-half of the United States.

Precipitation Datasets

Precipitation data are needed, in combination with the concentration data, to estimate deposition. Precipitation data at National Weather Service (NWS) stations were available from the NOAA National Climatic Data Center (NCDC; <http://ncdc.noaa.gov/land-based-station-data/find-station>), and spatially continuous modeled precipitation data were available from the Oregon State University, Parameter-elevation Regression on Independent Slopes Model (PRISM) Climate Group (<http://prism.oregonstate.edu>).

National Oceanic and Atmospheric Administration National Climatic Data Center

Precipitation data, needed for the calculation of precipitation-weighted concentrations at the Junge and Lodge sampling sites, were not published in their respective reports. According to Junge and Gustafson (1956), the Junge sampling sites were located at the NOAA NWS stations, usually near small municipal airports; however, the specific NWS stations associated with each sampling site were not provided in the report. Although not specifically stated in Lodge and others (1968), the Lodge sampling sites also were assumed to be located at NWS stations. To determine the NWS stations most likely used for the Junge and Lodge studies, the National Oceanic and Atmospheric Agency National Climatic Data Center (2012) station information was screened based on station names and location. In some cases, the NWS station identified for a Lodge sampling site did not have precipitation data spanning the entire sampling period; in these cases, data from nearby NWS stations were substituted. These data substitutions are documented in the aggregated data files. An NWS station with precipitation data spanning the entire sampling period was not found for the site L17 in the Lodge data (fig. 3), so this site was excluded from further analysis. The precipitation depths and location information, including elevation (in meters), and latitude and longitude derived from the NCDC, were used in subsequent processing. Monthly precipitation from the NCDC was provided in tenths of millimeters and converted to centimeters.

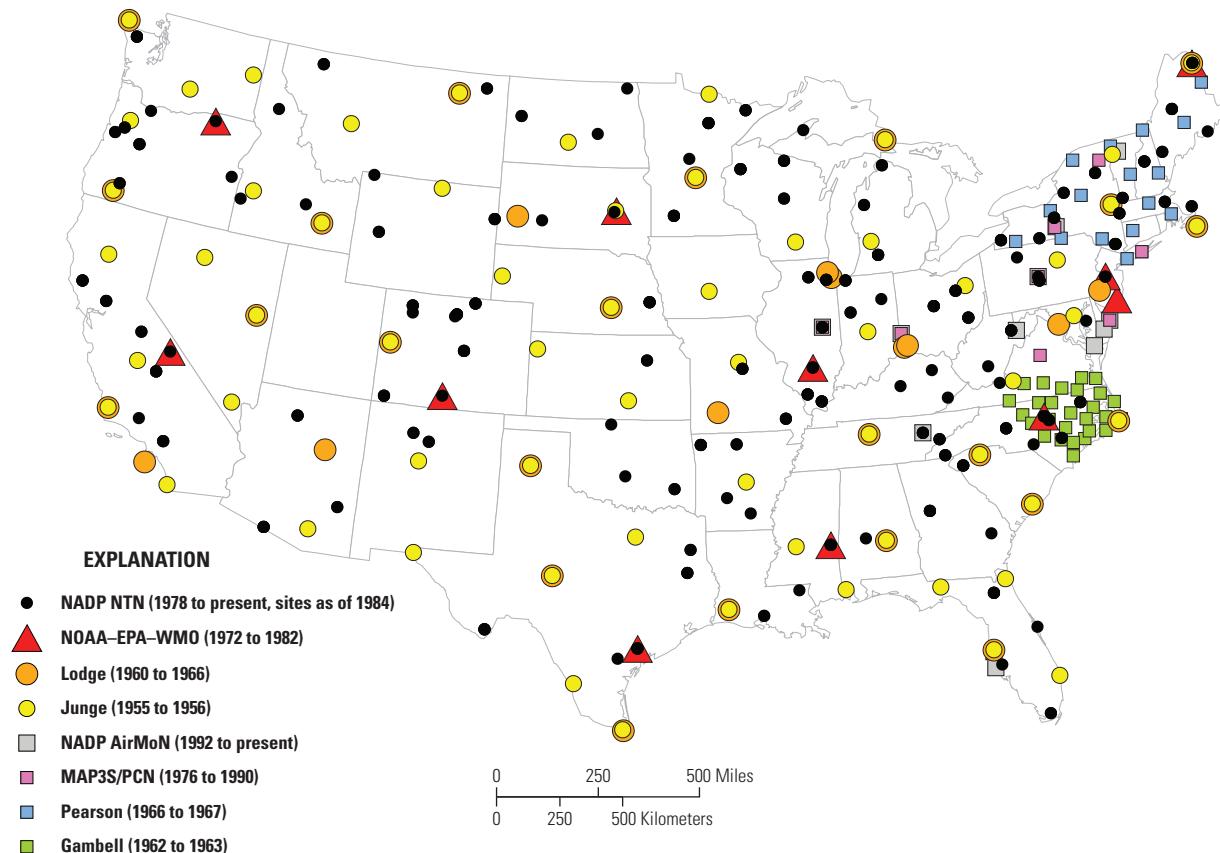


Figure 6. Location of network sites. AirMoN, Atmospheric Integrated Research Monitoring Network; EPA, U.S. Environmental Protection Agency; MAP3S/PCN, Multistate Power Production Pollution Study Precipitation Chemistry Network; NADP, National Atmospheric Deposition Program; NOAA, National Oceanic and Atmospheric Administration; NTN, National Trends Network; WMO, World Meteorological Organization.

Parameter-Elevation Regression on Independent Slopes Model Climate Group

Spatially continuous precipitation data for the conterminous United States are needed for the calculation of spatially continuous nitrogen deposition for national coverage. The PRISM American Standard Code for Information Interchange (ASCII) files of monthly and annual precipitation for the conterminous United States were retrieved from the Parameter-elevation Regression on Independent Slopes Model Climate Group, Oregon State University (2013). The PRISM ASCII files can be converted to digital grids of precipitation, which are estimated from many precipitation-monitoring networks, incorporating influences of topographical and geographical features. Monthly precipitation data were retrieved to correspond to the July 1955–June 1956 Junge dataset. Annual precipitation data were retrieved to correspond to the 1961–65 Lodge, and 1981–84 NADP datasets. Precipitation is provided by PRISM in millimeters times 100.

The PRISM ASCII files were converted to raster format, projected to a North American Albers projection, and resampled at 2,338.383 meter (m) by 2,338.383 m cell size (one-half the original cell resolution). The PRISM precipitation surface was modified by combining the station precipitation data (Junge, Lodge, and NADP) with the higher resolution PRISM data. A procedure was developed to blend the station data with the PRISM data to create a single precipitation surface (<http://nadp.sws.uiuc.edu/data/mapProcess.aspx>; Brian Kerchner, NADP, Illinois State Water Survey, written commun., April 15, 2013), using a modified distance weighting square interpolation within a 30-kilometer (km) radius of each station site. Essentially, the resulting precipitation surface reflects the values at the station location, and as the edge of the 30-km radius is approached, the values of the weighted raster cells approached that of the original higher resolution PRISM data. Outside the 30-km radius, the precipitation raster reflects the original PRISM data.

Data-Processing Methods

The following steps were followed to prepare the concentration and precipitation data and ultimately to create the raster datasets of inorganic nitrogen deposition. The data sources of ammonium, nitrate, and inorganic nitrogen concentration were checked for missing values. Annual precipitation-weighted concentrations were calculated for each site. National raster datasets (grids) of ammonium and nitrate, or inorganic nitrogen (depending on the data source) were created from the sites with calculated annual concentration values. These concentration raster datasets were coupled with national annual precipitation raster datasets to generate national deposition raster datasets.

Preprocessing Concentration Datasets

Most of the Junge (1955–56) sites (fig. 3) had quarterly average ammonium and nitrate concentration data for four quarters. The sites FR, LO, and SD were missing ammonium and nitrate concentration data for the July–September quarter. The sites GI, GL, GO, and LA were missing ammonium concentration data for one quarter; the SM site was missing ammonium data for two quarters and nitrate data for one quarter. For those sites with concentration data missing for only one quarter, precipitation amounts and the relation between precipitation and concentration during other quarters were investigated to gain insight into making a substitution for the missing values. On closer evaluation of the precipitation data from the NCDC and the PRISM, the missing data for FR and SM corresponded to zero or a trace amount of precipitation; therefore, no data substitutions were made for these sites. Although for some sites, concentration increased or decreased with increasing precipitation, it was difficult to reliably estimate a concentration value from a relation based on only three values per site. Therefore, the average value of the other three quarters was used for the missing quarters. The SM site was not used in generating the national ammonium concentration raster dataset.

For most Lodge sites (fig. 4), quarterly averages of inorganic nitrogen concentration data were available for each quarter (four values). Three sites were missing quarterly values: L15 and L31 were missing the September–November quarterly values, and L34 was missing the June–August quarterly value. The seasonal precipitation patterns and the relation between precipitation and concentration were investigated. For these years (1961–65), based on NCDC data, the only consistent seasonal pattern in precipitation was that for the L34 site—precipitation was always lowest during the June–August quarter and always less than 1 centimeter. Also, with the exception of the L31 site, where the highest concentrations were associated with the lowest precipitation, no strong relation between precipitation and concentration was observed. An average of the concentration values from the other three quarters was used here for the missing concentration values for these sites. The data from sites L5 and L14

were not used because values at those sites reflected local influences of major airports, and were not representative of national patterns, and therefore were not appropriate to use for later spatial interpolation of national patterns. The sites with fewer than 12 months of station precipitation data per year were not used for that year. These sites included L29 in 1961–62, and L20 in 1963–65. In addition, the reported value of zero at site L26 was set to 0.005 mg/L (one-half the reporting limit).

The NADP NTN data were prescreened by retrieving only data that met the completeness criteria used by the NADP program. The completeness criteria required that the data be restricted to sites where there are valid samples for at least 75 percent of the summary period, precipitation amounts are available either from the rain gage or from the sample volume for at least 90 percent of the summary period, and there are valid samples for at least 75 percent of the total measured precipitation; processing was limited to 1981–84. The 1978–80 data were not used because the data were limited in spatial coverage. Raster datasets for 1985–2012 already exist and are available directly from the NADP (National Atmospheric Deposition Program, 2013b).

Latitude and longitude were used to define the location of the sites. Horizontal datum was not specified for any of the data sources, but the latitudes and longitudes were treated as if the horizontal datum was North American Datum of 1983 (NAD 83). Points were created from the latitude and longitude, and then projected into a North American Albers projection.

Calculating Site-Specific Precipitation-Weighted Concentration

Quarterly precipitation values were calculated by summing NCDC monthly precipitation data for July–September 1955, October–December 1955, January–March 1956, and April–June 1956 for each Junge site. The Junge ammonium and nitrate quarterly concentrations were associated with the calculated quarterly NCDC precipitation data and processed by quarter to calculate the mean annual precipitation-weighted concentrations for July 1955–June 1956. The Lodge inorganic nitrogen concentrations were reported as 6-year mean quarterly precipitation-weighted concentrations from December 1960 through November 1966. The quarterly values were assumed to apply for each month in that quarter, meaning each month in a given quarter had the same value. In addition, the quarterly values were assumed to be the same for each of the 6 years, meaning each quarter (and thus month) had the same value from year to year. As a result, only the NCDC precipitation amounts varied from year to year when calculating the annual precipitation-weighted concentrations. The data were handled as monthly data instead of quarterly data so that they could be used together with monthly precipitation values to calculate annual precipitation-weighted concentrations for calendar years 1961–65.

For the Junge and Lodge data, mean annual precipitation-weighted concentration was calculated for each site and for each year using equation 1.

$$C_N = \frac{\sum_{x=1}^n P_x C_x}{\sum_{x=1}^n P_x} \quad (1)$$

where

for Junge, ammonium data

- C_N is the mean annual precipitation-weighted ammonium concentration, in milligrams per liter (NH_4^+);
- C_x is the precipitation-weighted ammonium concentration, in milligrams per liter, for quarter x ;
- P_x is the NCDC precipitation, in centimeters, during quarter x ; and
- n is the number of quarters.

for Junge, nitrate data

- C_N is the mean annual precipitation-weighted nitrate concentration, in milligrams per liter (NO_3^-);
- C_x is the precipitation-weighted nitrate concentration, in milligrams per liter, for quarter x ;
- P_x is the NCDC precipitation, in centimeters, during quarter x ; and
- n is the number of quarters.

for Lodge

- C_N is the mean annual precipitation-weighted inorganic nitrogen concentration, in milligrams per liter (N);
- C_x is the inorganic nitrogen concentration, in milligrams per liter, for month x ;
- P_x is the NCDC precipitation, in centimeters, during month x ; and
- n is the number of months.

The annual precipitation-weighted ammonium and nitrate concentrations were provided in the NADP NTN dataset.

Creating Raster Datasets of Nitrogen Deposition

Raster datasets of annual nitrogen deposition were generated by first creating raster datasets of the mean annual precipitation-weighted concentration data (ammonium and nitrate, or inorganic nitrogen) using selected sites for each dataset. Then the concentration raster datasets were multiplied by the modified PRISM precipitation raster and a unit correction factor. The approach used here is from Latysh and Wetherbee (2011).

Raster datasets of mean annual precipitation-weighted ammonium concentration (in milligrams per liter) and nitrate concentration (in milligrams per liter) for the Junge and NADP

data, and inorganic nitrogen concentration (in milligrams per liter) for the Lodge data were created using the Inverse Distance Weighting spatial interpolation tool in the ArcGIS software by Esri. Application of this tool varied depending on the spatial density of the monitoring points. For datasets with sufficient spatial density (sampling sites are no more than 1,000 km apart), interpolated values are based on direct measurements within a 500-km radius. For other datasets, interpolated values are based on the five closest direct measurements.

For Junge and NADP, the raster datasets of nitrogen deposition were created using the Raster Calculator ArcGIS tool to multiply the sum of the nitrogen equivalents of ammonium and nitrate precipitation-weighted concentrations on a pixel-by-pixel basis, by the adjusted PRISM precipitation, and a conversion factor (equation 2).

$$Ndep = ((NH_4^+ \times 0.7766) + (NO_3^- \times 0.2257)) \times ppt \times 0.001 \times 0.1 \quad (2)$$

where

- $Ndep$ is the nitrogen deposition, in kilograms per hectare;
- NH_4^+ is the mean annual precipitation-weighted ammonium, in milligrams per liter;
- NO_3^- is the mean annual precipitation-weighted nitrate, in milligrams per liter;
- ppt is the adjusted PRISM precipitation, in $100 \times$ millimeters;
- 0.7766 is the mole fraction of nitrogen in NH_4^+ ;
- 0.2257 is the mole fraction of nitrogen in NO_3^- ;
- 0.001 is the conversion factor to convert ppt in $100 \times$ millimeters to centimeters; and
- 0.1 is the conversion factor to convert milligrams per liter \times centimeters to kilograms per hectare.

For Lodge, the raster datasets of nitrogen deposition were created using a similar procedure except the deposition was calculated from the inorganic nitrogen concentration (equation 3).

$$Ndep = N \times ppt \times 0.001 \times 0.1 \quad (3)$$

where

- $Ndep$ is the nitrogen deposition, in kilograms per hectare;
- N is the mean annual precipitation-weighted inorganic nitrogen, in milligrams per liter;
- ppt is the adjusted PRISM precipitation, in $100 \times$ millimeters;
- 0.001 is the conversion factor to convert ppt in $100 \times$ millimeters to centimeters ; and
- 0.1 is the conversion factor to convert milligrams per liter \times centimeters to kilograms per hectare.

Description of Aggregated Datasets

The station data for Junge are in Junge_data.xlsx (appendix 1). The station data for Lodge are in Lodge_data.xlsx (appendix 2). The data files for Junge and Lodge contain site identifier; elevation; latitude; longitude; ammonium and nitrate, or inorganic nitrogen concentration (milligrams per liter); precipitation derived from NOAA NCDC (centimeters); and calculated mean annual precipitation-weighted concentration (milligrams per liter). Other chemical analytes from Junge and Werby (1958) and Lodge and others (1968) also are included in these data files. The NADP NTN data were not provided here, but can be accessed directly from NADP (<http://nadp.sws.uiuc.edu/>). The data from the national networks processed in this report span four decades, covering the 1950s (Junge), 1960s (Lodge), and 1980s (NADP). In general, the sites in the national networks are well distributed throughout the conterminous United States.

Although the data from NOAA–EPA–WMO (Artz and Miller, 1985) and Pearson and Fisher (1971) were not used to generate raster datasets of nitrogen deposition, the data tables were transcribed into digital format because currently (2014) the data are only available from hard copy reports

or online formats that are difficult to use directly. The data file for NOAA–EPA–WMO (NOAA_data.xlsx, appendix 3) contains site identifier and location information, precipitation (centimeters), and chemical constituents including ammonium and nitrate concentration (in milligrams per liter and milliequivalents per liter). The data file for Pearson (Pearson_data.xlsx, appendix 4) contains site identifier, and precipitation (inches) and analyte load (thousandths of tons per day per square mile), in the original units used in the report. Data from Gambell and Fisher (1966) were not transcribed because the sites were not uniquely identified.

Raster datasets of annual inorganic nitrogen deposition generated for 1955–56 (Junge, fig. 7A), 1961–65 (Lodge, fig. 7B–F), and 1981–84 (NADP; fig. 7G–J) are presented in 2,338.383-m by 2,338.383-m resolution, and the units are in kilograms per hectare, using the same resolution and units as NADP NTN raster datasets for easier comparison between sources. Analysis of the data is beyond the scope of this report; however, it is important to point out that some regions with high deposition likely are an artifact of the low station density. These raster datasets are available as appendixes 5–14. Raster datasets of the NADP data for 1985–2012 can be accessed directly from NADP (<http://nadp.sws.uiuc.edu/>).

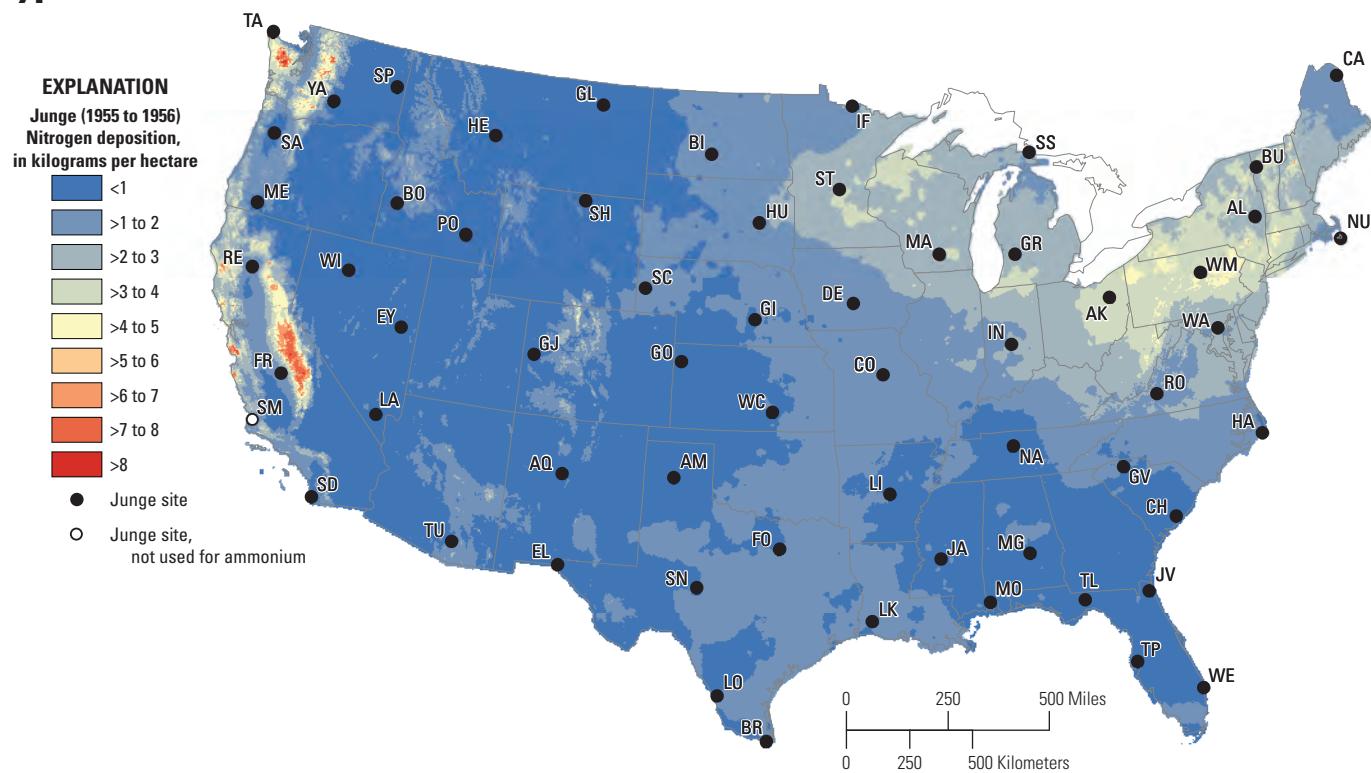
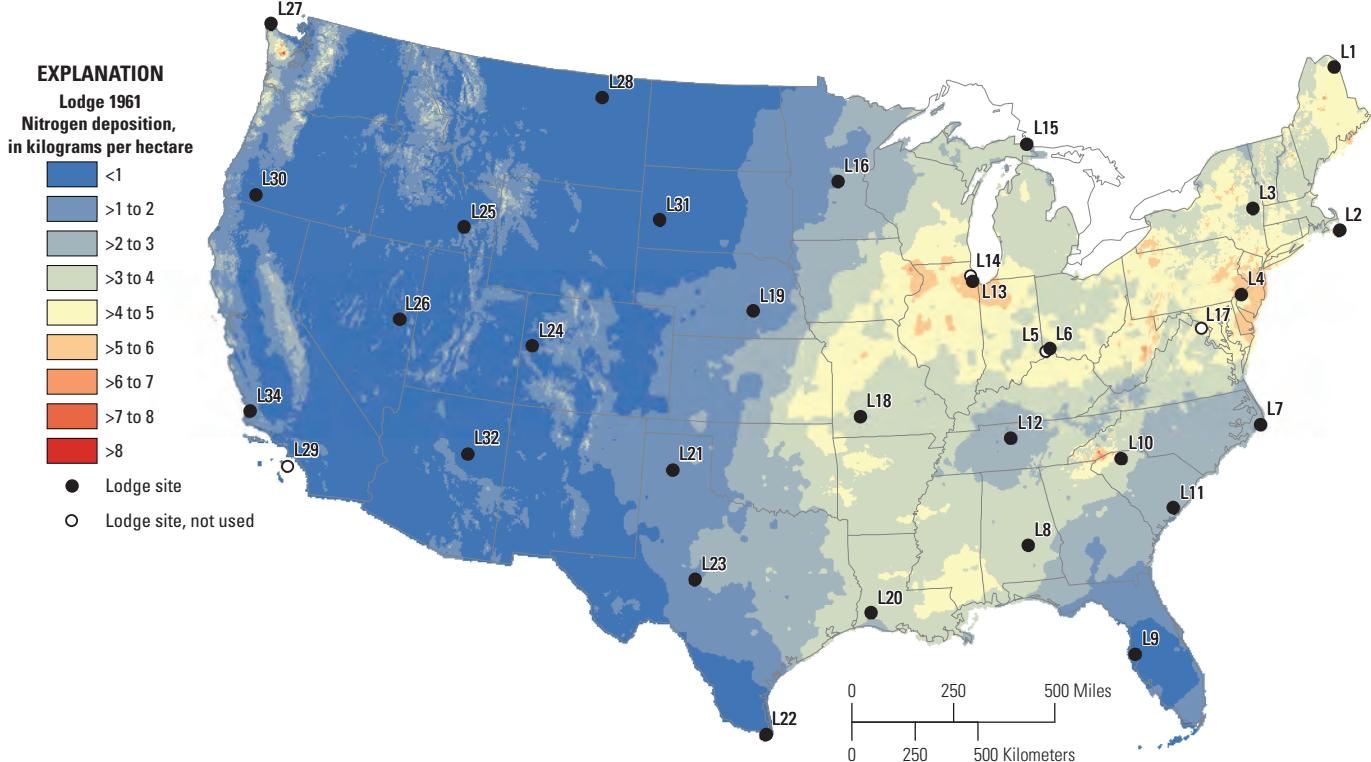
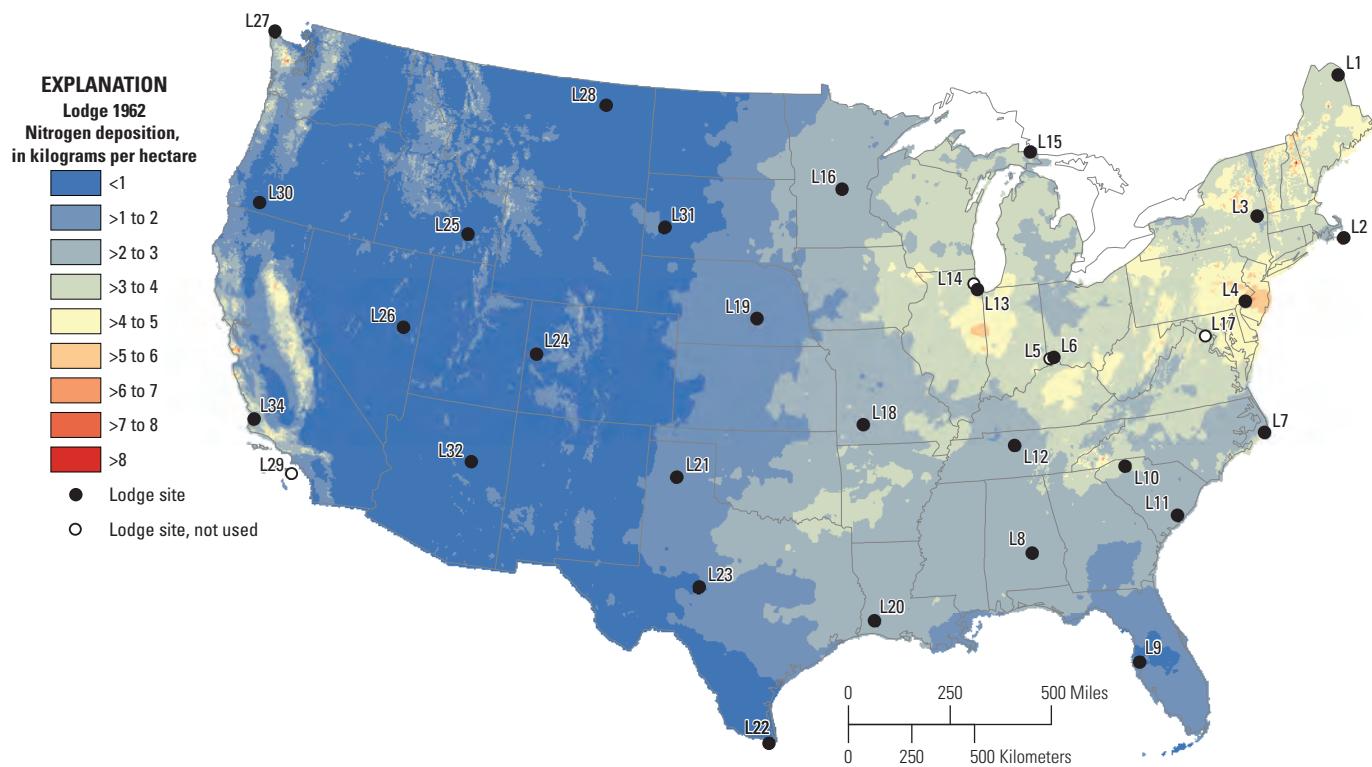
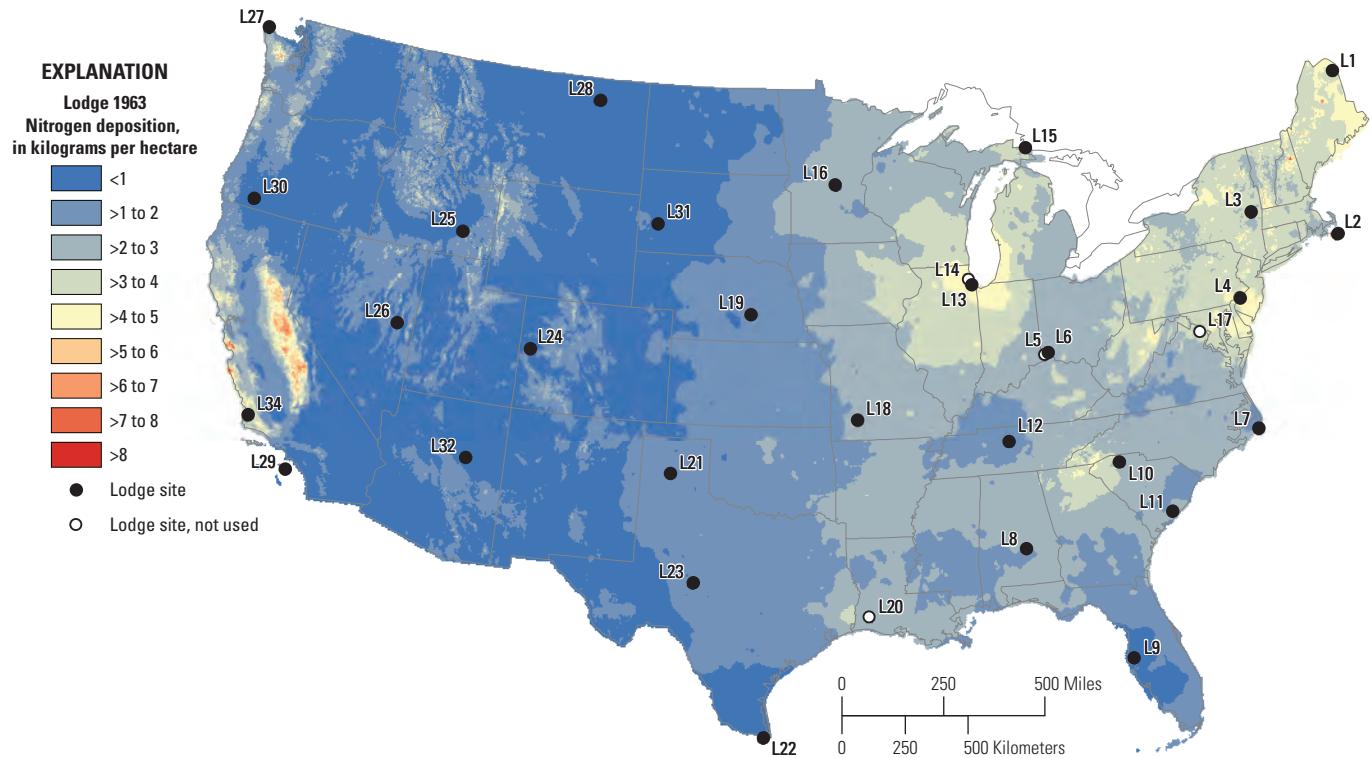
A**B**

Figure 7. Inorganic nitrogen deposition for *A*, July 1955–June 1956; *B*, 1961; *C*, 1962; *D*, 1963; *E*, 1964; *F*, 1965; *G*, 1981; *H*, 1982.; *I*, 1983.; *J*, 1984. NADP, National Atmospheric Deposition.

C**D****Figure 7.** —Continued

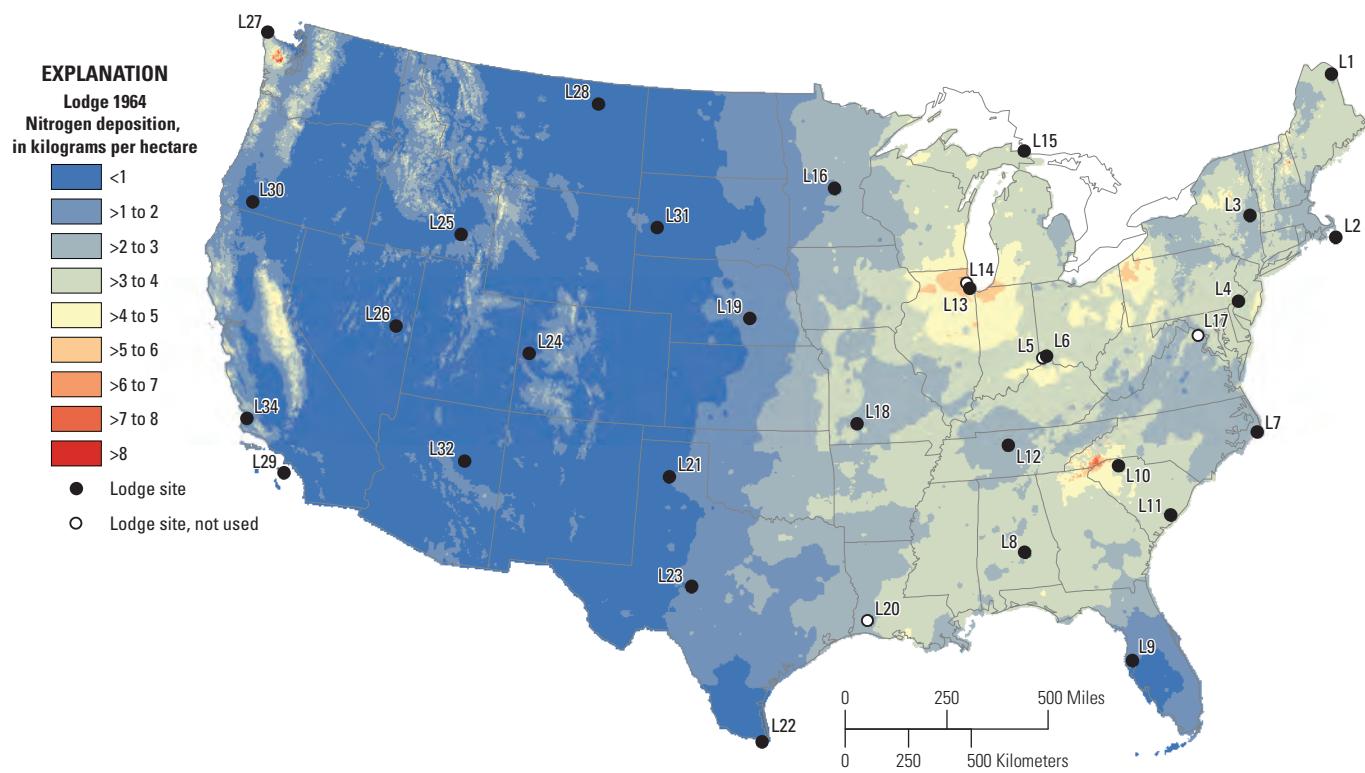
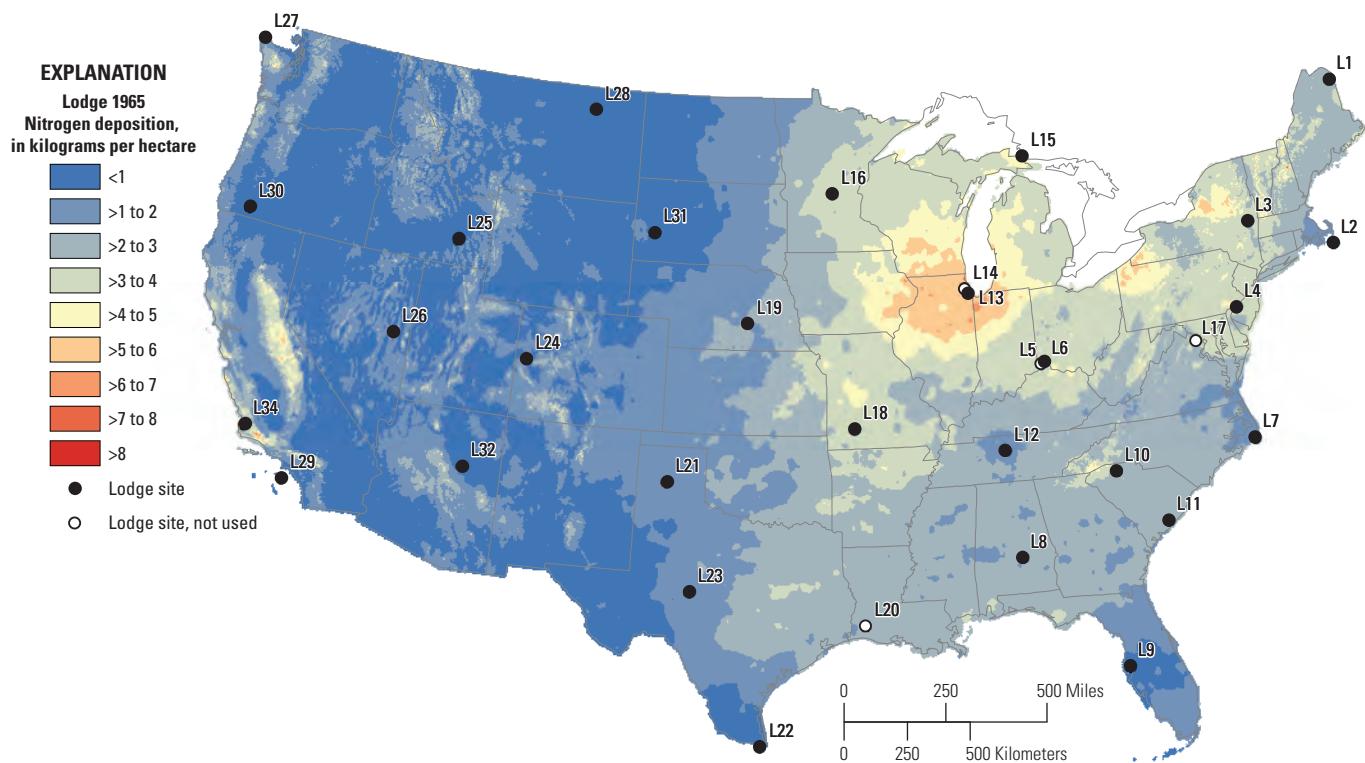
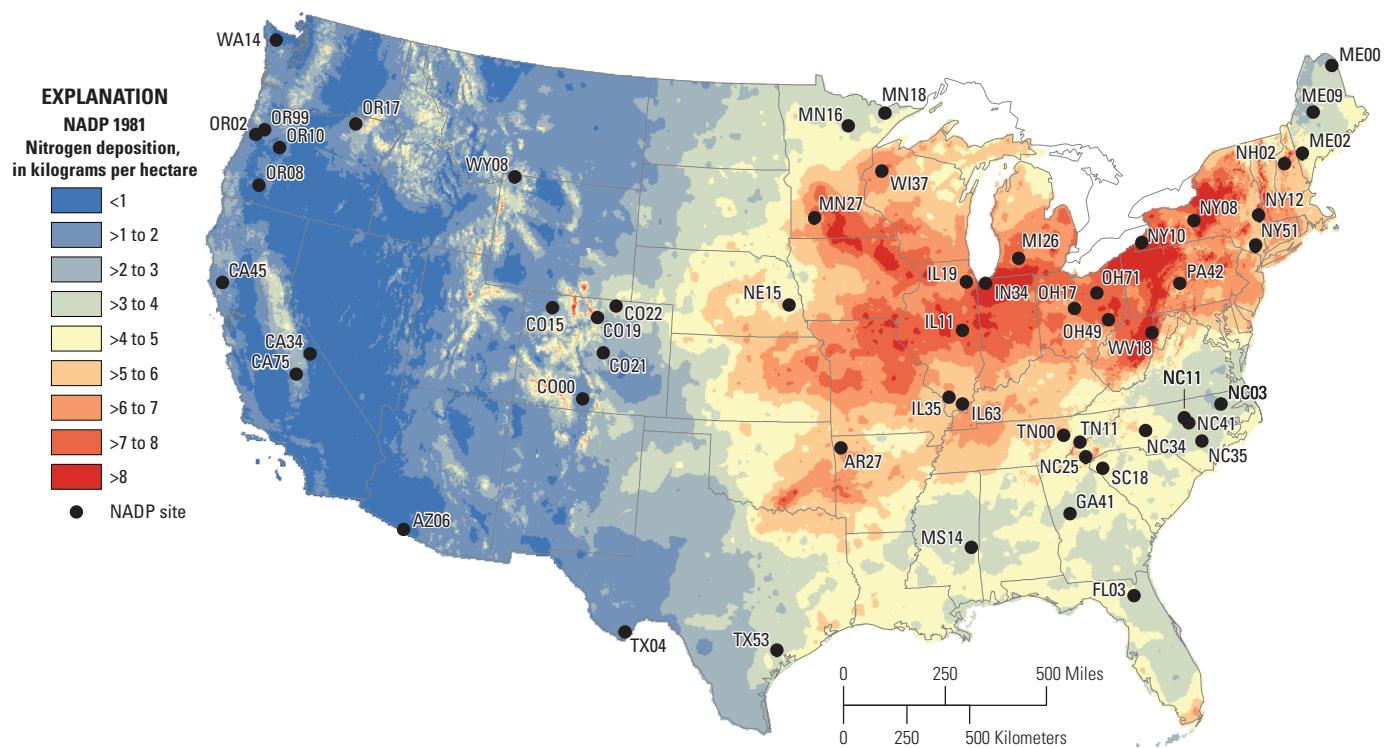
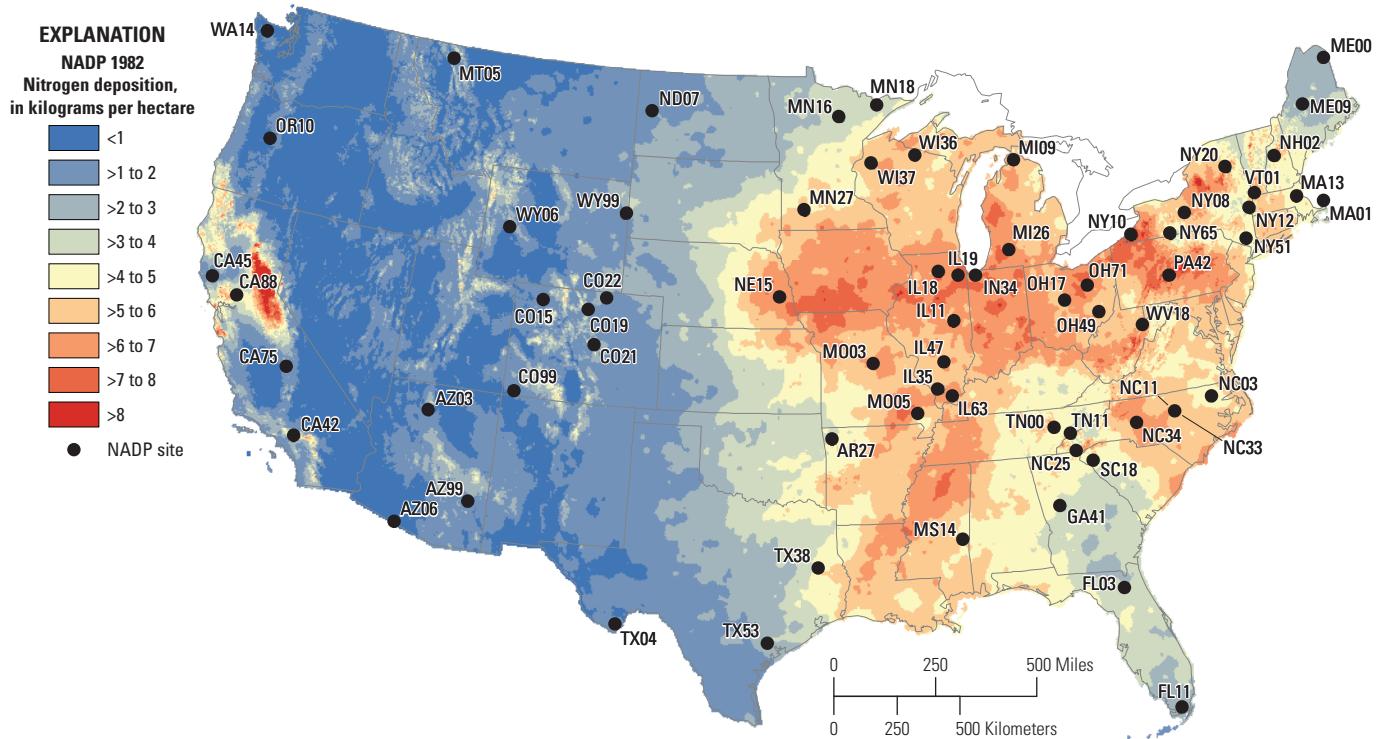
E**F**

Figure 7. —Continued

G**H****Figure 7.** —Continued

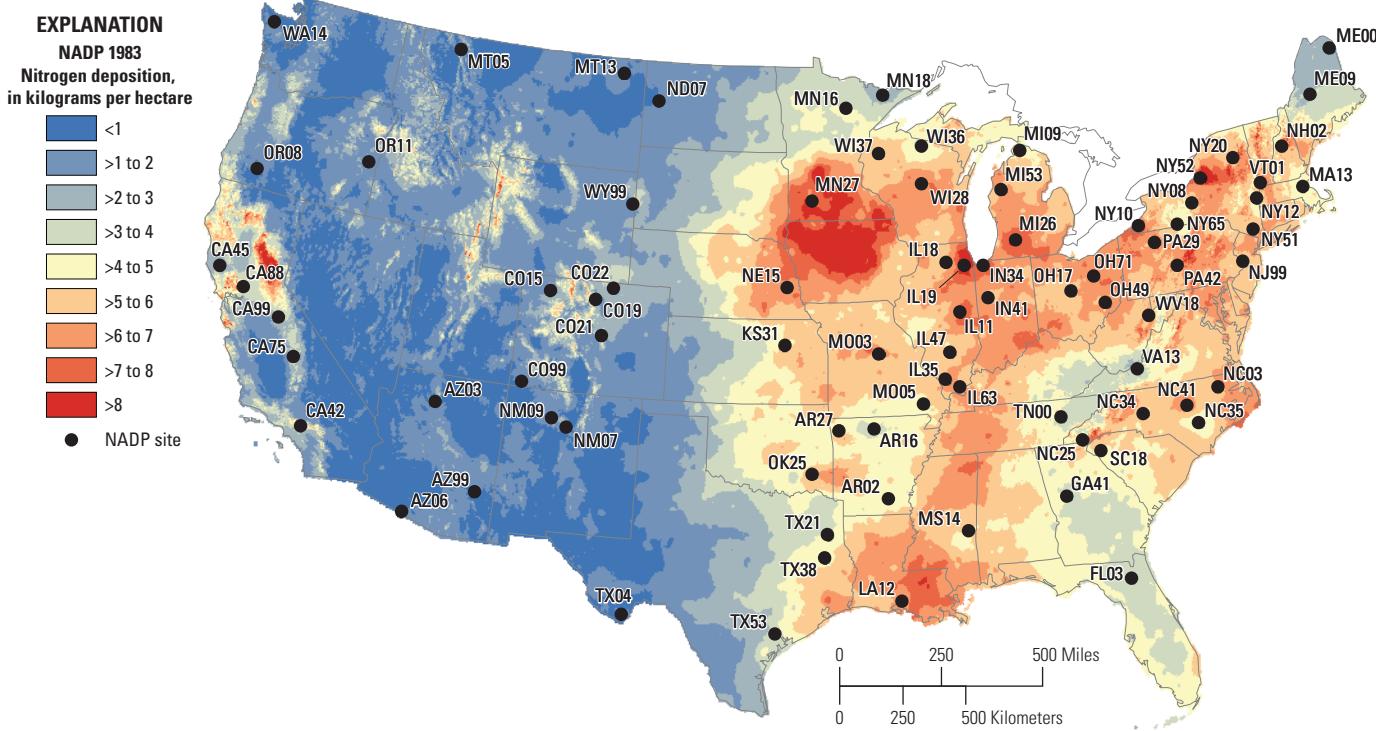
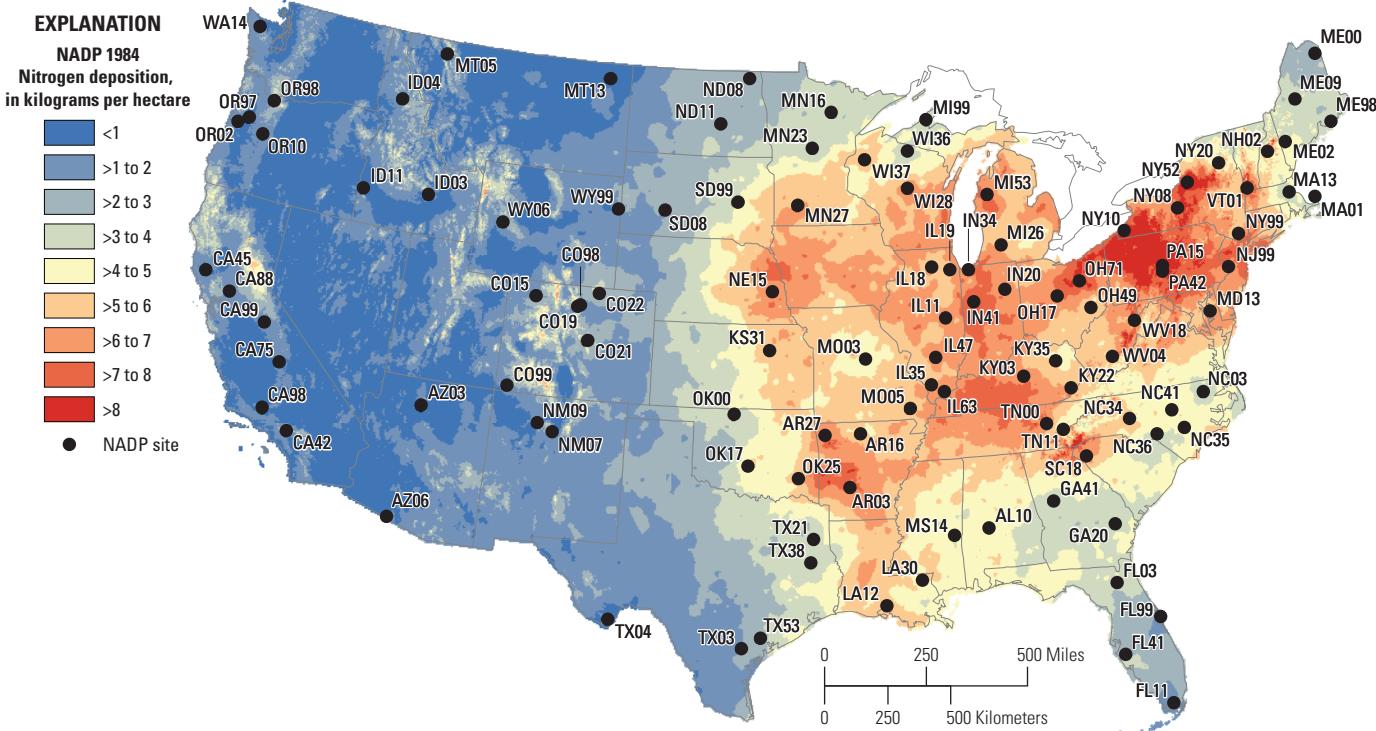
I**J**

Figure 7. —Continued

Summary

Inorganic nitrogen deposition was estimated for the conterminous United States for 1955–56, 1961–65, and 1981–84. These estimates were based on ammonium, nitrate, and inorganic nitrogen concentrations in wet deposition and on precipitation data, which were reconstructed from national historical data sources. The nitrogen deposition estimates, in kilograms per hectare, are presented as 2,338.383-meter by 2,338.383-meter resolution raster datasets. The units and resolution used here are the same as those used in the National Atmospheric Deposition Program National Trends Network raster datasets from 1985 to 2012, for easier comparison between these sources. The estimates presented here predate the already available National Atmospheric Deposition Program National Trends Network inorganic nitrogen deposition raster datasets, and effectively expand the length of nitrogen deposition records for use in characterizing nutrient inputs to surface-water basins and groundwater recharge areas.

The historic ammonium, nitrate, and inorganic nitrogen concentration and precipitation data, which provided the basis for the raster datasets, were reconstructed as tabular data files. This report documents the sources of these data and the methods that were used, first to calculate precipitation-weighted concentrations at the site level, and then to estimate the continuous precipitation-weighted concentrations and inorganic nitrogen deposition for the conterminous United States. Tabular datasets, including the analytical results, precipitation depth, and calculated precipitation-weighted concentration, and raster datasets of nitrogen deposition are provided as part of this report.

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Appendix

Appendix 1. Tabular Dataset of Junge Data

Download data here: http://pubs.usgs.gov/sir/2014/5067_downloads/sir2014-5067_Appendix_1_Junge_data.xlsx.

Appendix 2. Tabular Dataset of Lodge Data

Download data here: http://pubs.usgs.gov/sir/2014/5067_downloads/sir2014-5067_Appendix_2_Lodge_data.xlsx.

Appendix 3. Tabular Dataset of National Oceanic Atmospheric Administration, U.S. Environmental Protection Agency, World Meteorological Organization Data

Download data here: http://pubs.usgs.gov/sir/2014/5067_downloads/sir2014-5067_Appendix_3_noaa_data.xlsx.

Appendix 4. Tabular Dataset of Pearson Data

Download data here: http://pubs.usgs.gov/sir/2014/5067_downloads/sir2014-5067_Appendix_4_Pearson_data.xlsx.

Appendix 5. Raster Dataset of Inorganic Nitrogen Deposition, July 1955–June 1956

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1955.

Appendix 6. Raster Dataset of Inorganic Nitrogen Deposition, 1961

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1961.

Appendix 7. Raster Dataset of Inorganic Nitrogen Deposition, 1962

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1962.

Appendix 8. Raster Dataset of Inorganic Nitrogen Deposition, 1963

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1963.

Appendix 9. Raster Dataset of Inorganic Nitrogen Deposition, 1964

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1964.

Appendix 10. Raster Dataset of Inorganic Nitrogen Deposition, 1965

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1965.

Appendix 11. Raster Dataset of Inorganic Nitrogen Deposition, 1981

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1981.

Appendix 12. Raster Dataset of Inorganic Nitrogen Deposition, 1982

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1982.

Appendix 13. Raster Dataset of Inorganic Nitrogen Deposition, 1983

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1983.

Appendix 14. Raster Dataset of Inorganic Nitrogen Deposition, 1984

Download data here: http://water.usgs.gov/lookup/getspatial?sir2014-5067_atm_ndep_1984.

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