

NITROGEN DEPOSITION IN CALIFORNIA FORESTS: A REVIEW

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Abstract

Atmospheric concentrations and deposition of the major nitrogenous (N) compounds and their biological effects in California forests are reviewed. Climatic characteristics of California are summarized in light of their effects on pollutant accumulation and transport. Over large areas of the state dry deposition is of greater magnitude than wet deposition due to the arid climate. However, fog deposition can also be significant in areas where seasonal fogs and N pollution sources coincide. The dominance of dry deposition is magnified in airsheds with frequent temperature inversions such as occur in the Los Angeles Air Basin. Most of the deposition in such areas occurs in summer as a result of surface deposition of nitric acid vapor (HNO_3) as well as particulate nitrate (NO_3^-) and ammonium (NH_4^+). Internal uptake of gaseous N pollutants such as nitrogen dioxide (NO_2), nitric oxide (NO), HNO_3 , peroxyacetyl nitrate (PAN), ammonia (NH_3), and others provides additional N to forests. However, summer drought and subsequent lower stomatal conductance of plants tend to limit plant utilization of gaseous N. Nitrogen deposition is much greater than S deposition in California. In locations close to photochemical smog source areas, concentrations of oxidized forms of N (NO_2 , HNO_3 , PAN) dominate, while in areas near agricultural activities the importance of reduced N forms (NH_3 , NH_4^+) significantly increases. Little data from California forests are available for most of the gaseous N pollutants. Total inorganic N deposition in the most highly-exposed forests in the Los Angeles Air Basin may be as high as 25–45 kg ha^{-1} year $^{-1}$. Nitrogen deposition in these highly-exposed areas has led to N saturation of chaparral and mixed conifer stands. In N saturated forests high concentrations of NO_3^- are found in streamwater, soil solution, and in foliage. Nitric oxide emissions from soil and foliar N:P ratios are also high in N saturated sites. Further research is needed to determine the ecological effects of chronic N deposition, and to develop appropriate management options for protecting water quality and managing plant nutrient resources in ecosystems which no longer retain excess N. Published by Elsevier Science Ltd.

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INTRODUCTION

Elevated nitrogen deposition to forest ecosystems, in both dry and wet forms, has resulted in N saturation of forests in many parts of Europe (Dise & Wright, 1995), including the Netherlands (Boxman *et al.*, 1995; Duyzer *et al.*, 1992; Van Breemen *et al.*, 1987; Weyers *et al.*, 1992), Great Britain (ApSimon *et al.*, 1987; Emmett *et al.*, 1993), Scandinavia (Andersen *et al.*, 1993), and the Czech Republic (Adema *et al.*, 1993). Nitrogen saturation has also been described for some forests in the eastern United States (Aber *et al.*, 1989; Driscoll & Van Dreason, 1993; Foster *et al.*, 1989; Johnson & Fernandez, 1992; Johnson *et al.*, 1991b, 1993; Stoddard, 1994). Although atmospheric deposition of N has significantly increased in the United States and both aquatic and terrestrial ecosystems have been affected (NAPAP, 1990), little is known about N deposition and its potential impact on forests in the western states. This is despite the importance of Western forests as a major component of our natural resources, important for the preservation of species biodiversity, the national economy, and for recreational and aesthetic purposes (Raven, 1977). Based on the current limited information, the worst-case scenarios of N deposition effects on forests in the western US are in California, although forests downwind of other major urban centers in the West could also be significantly affected by N deposition (Böhm, 1992). Positive effects, such as enhanced tree growth, may result when N deposition is not excessive for a given ecosystem. However, chronic N inputs can lead to various negative effects, including nutrient deficiencies, soil acidification, altered species composition, decreased mycorrhizal root symbiosis, increased susceptibility to environmental stresses (including air pollutants), and elevated concentrations of nitrate in soil, ground water and streams (Skeffington & Wilson, 1988; Van Breemen & Van Dijk, 1988).

This paper summarizes and synthesizes current information for California forests on:

- (1) concentrations of the major gaseous, particulate, and wet-deposited N pollutants (Table 1);
- (2) N deposition to forest canopies;

Table 1. Nitrogen air pollutants discussed in the paper

Pollutant	Chemical formula	Molecular weight	Conversion factor for gaseous species ^a
Nitric oxide	NO	30	1 $\mu\text{g m}^{-3}$ = 8.419×10^{-4} ppm 1 ppm = 1227 $\mu\text{g m}^{-3}$
Nitrogen dioxide	NO ₂	46	1 $\mu\text{g m}^{-3}$ = 5.316×10^{-4} ppm 1 ppm = 1881 $\mu\text{g m}^{-3}$
Nitrous acid	HNO ₂	47	1 $\mu\text{g m}^{-3}$ = 5.202×10^{-4} ppm 1 ppm = 1922 $\mu\text{g m}^{-3}$
Nitric acid	HNO ₃	63	1 $\mu\text{g m}^{-3}$ = 3.880×10^{-4} ppm 1 ppm = 2577 $\mu\text{g m}^{-3}$
Peroxyacetyl nitrate (PAN)	CH ₃ C(O)OONO ₂	121	1 $\mu\text{g m}^{-3}$ = 2.020×10^{-4} ppm 1 ppm = 4949 $\mu\text{g m}^{-3}$
Peroxypropionyl nitrate (PPN)	C ₂ H ₅ C(O)OONO ₂	135	1 $\mu\text{g m}^{-3}$ = 1.811×10^{-4} ppm 1 ppm = 5521 $\mu\text{g m}^{-3}$
Nitrate (particulate and wet-deposited)	NO ₃ ⁻	62	
Ammonia	NH ₃	17	1 $\mu\text{g m}^{-3}$ = 1.439×10^{-3} ppm 1 ppm = 695 $\mu\text{g m}^{-3}$
Ammonium (particulate and wet-deposited)	NH ₄ ⁺	18	

^aAt 25°C and 0.101 MPa (1 atm.).

- (3) observed or predicted effects of N pollutants on vegetation, soils, nutrient cycling and water quality; and
- (4) possible interactions between N deposition and other air pollutants and environmental stresses.

VEGETATION TYPES AND EFFECTS OF GEOGRAPHIC AND CLIMATIC CHARACTERISTICS ON POLLUTION DISTRIBUTION IN CALIFORNIA

California has a wide variety of ecosystems including alpine, montane and subalpine forests, redwood forests, mixed evergreen forests, chaparral, coastal scrub, oak woodlands and desert scrub vegetation (Barbour & Major, 1977; Kricher, 1993).

In California, about 40% of the area is covered by forests (total area of forests equal to 16 249 000 ha). The most important forest types in the state are ponderosa pine, Douglas fir, fir-spruce, chaparral, and western hardwoods (Olson, 1992; Fig. 1).

Two aspects of the mean global circulation control the climate of California: light variable winds in summer associated with the subtropical anticyclone (Pacific High) located around latitudes 30°–40°, and frontal storms associated with cyclones embedded in the southwesterlies between latitudes 40°–60° in the winter (Rose *et al.*, 1992).

California may have the most Mediterranean climate in the world, based on winter predominance of precipitation and extreme summer drought. At nearly all the weather stations throughout the state more than 85% of the precipitation falls in the winter. Because of diversified topography however, variances within the Mediterranean climatic type occur repeatedly across

very short distances. Coastal regions have cool summers and almost completely frost-free winters. As little as 30 km inland a sheltered valley can have summers that are extremely hot with scores of days recording > 40°C, and sharp, though brief winter frosts. As is typical of Mediterranean areas, precipitation correlates positively with elevation and latitude. In the mountains of California most of the precipitation occurs as snow in winter, except for the coastal ranges, which receive mostly winter rainfall. Summer precipitation is scarce and occurs as short-lasting storms (Aschmann, 1973).

Geographic and climatic characteristics influence pollution distribution, exposures, and effects on forest ecosystems. The potential for severe air pollution episodes is determined by topographical and meteorological conditions conducive to the accumulation of atmospheric pollutants, and is a function of vertical mixing, horizontal dispersion, and magnitude of emissions (Böhm, 1992). For example, the distribution of air pollution within the Los Angeles Basin shows a persistent, low-level temperature inversion in conjunction with onshore flow and high pressure systems aloft, occurring during the warmer half of the year. Air pollutants injected into the marine layer are inhibited from vertical diffusion by the warm air layer above. Photochemical reactions together with a lack of pollutant dispersion causes air pollutants to accumulate beneath the inversion as the entire marine air mass moves inland. Surface heating weakens the inversion as the day progresses and the polluted air begins to mix vertically. The sea breeze and afternoon upslope flow advect pollutants to forests along the western slopes of the mountains located inland (Miller, 1992). Another example of air pollution dispersion in California and the neighboring states of Nevada and Arizona is the Los Angeles urban plume affecting visibility in the Grand Canyon

and Las Vegas areas in summer, when the westerly winds prevail (Hoffer *et al.*, 1981).

During the dry season, the polluted air masses accumulating over central California may be transported

into the Sierra Nevada by winds originating in the San Francisco Bay area and in the southern half of the Central Valley (Miller *et al.*, 1972; Stohlgren & Parsons, 1987). In winter, storms originating over the Pacific

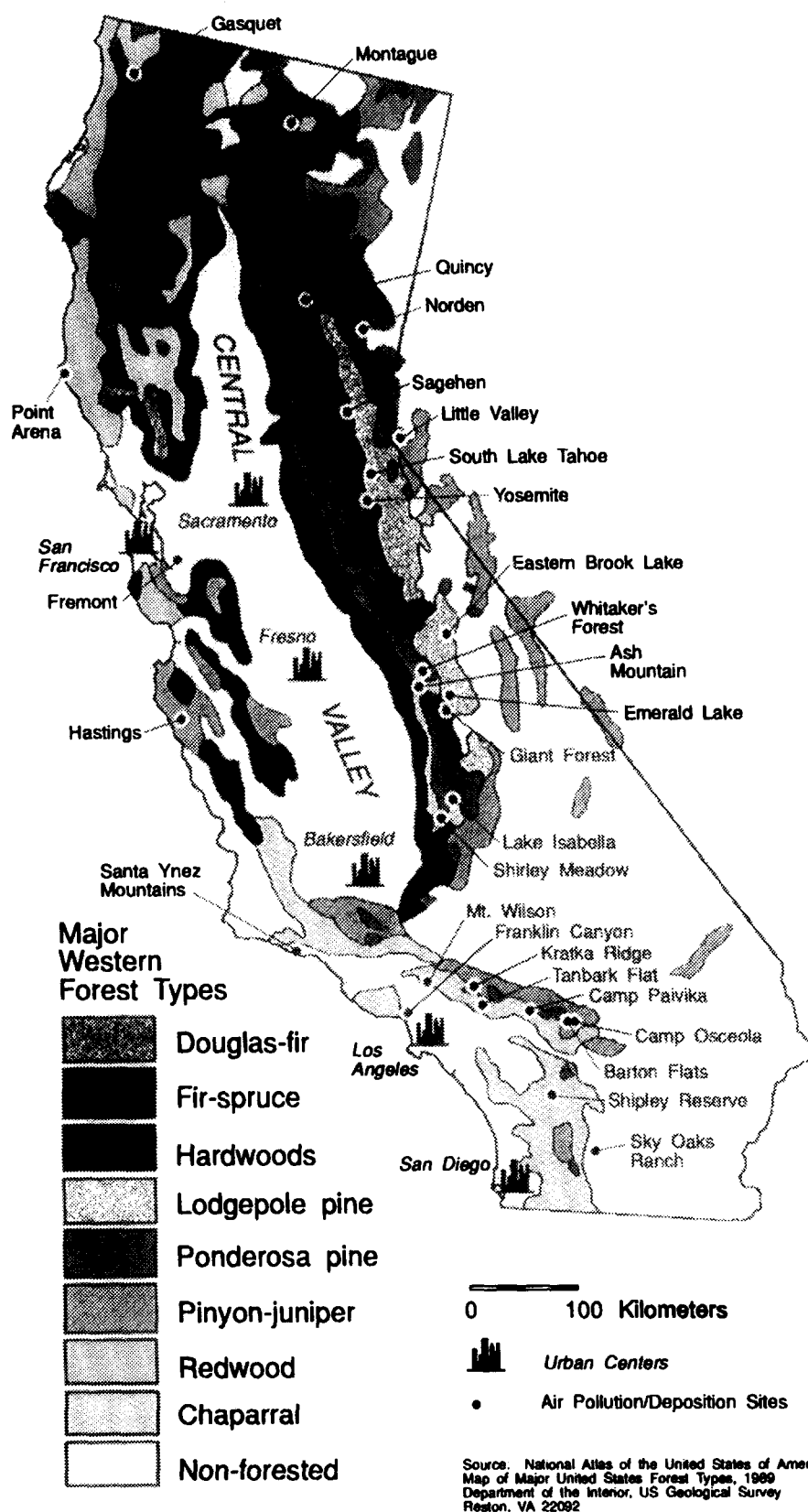


Fig. 1. Forest types in California and location of air pollution/deposition sites. Sites from Quincy in the north to Shirley Meadow in the south are Sierra Nevada locations.

Ocean tend to transport air masses north and northwest from the San Francisco Bay area into the northern Sierra Nevada and Coastal Range, while pollutants in the southern part of the Central Valley move into the southern Sierra Nevada and further north into the Valley (Stohlgren & Parsons, 1987). Air pollution exposure and transport in California and other western states has been reviewed by Böhm (1992).

The overriding influence of California's climate on atmospheric deposition is the relative dominance of dry deposition over wet deposition throughout large areas of the state (this may not be true in areas with high fog incidence, however). The greater magnitude of dry deposition is due to the temporal coincidence of high atmospheric pollutant levels and drought. The dominance of dry deposition is magnified in airsheds with frequent temperature inversions, such as occur in the Los Angeles Air Basin. In contrast, rainfall is more evenly distributed throughout the year in forests of the eastern US (Lindberg *et al.*, 1990), and wet and dry deposition are frequently of more equal importance (Lindberg *et al.*, 1986) in the overall atmospheric deposition budget. In forests of California, the timing and transport of ionic pollutants via throughfall into the forest floor and soil is governed by the cyclic pattern of long dry periods during which dry-deposited gaseous and particulate pollutants accumulate on canopy and forest floor surfaces followed by infrequent low-volume precipitation events which transport highly concentrated nutrient solutions to the forest floor. The extended summer dry period is probably also a major factor regulating the uptake rate of gaseous pollutants with a stomatal uptake mechanism. Therefore, potentially phytotoxic effects of high concentrations of pollutants, occurring when temperature and drought stress are highest, may be limited by reduced stomatal conductance (Krupa *et al.*, 1994). This situation is different from most of the forested areas in the eastern United States and central Europe where drought and high temperatures preventing air pollution toxicity rarely occur. Conversely, the situation described for California may apply to other ecosystems with a Mediterranean climate in different parts of the World.

NITROGENOUS AIR POLLUTANTS IN CALIFORNIA FORESTS

The potential for high N exposures and extensive distribution of N pollutants in California is strongly dependent on widespread vehicle use in large, developing urban centers with poorly established public transportation systems (Böhm, 1992). The air pollution emissions in California are generally characterized by a lower sulfur/nitrogen ratio than in the eastern United States or Europe (Young *et al.*, 1988) due to a relatively low number of stationary sources emitting sulfurous pollutants.

Although the effects of air pollution (photochemical smog) on forests in California have been recognized for

a long time, research efforts have focused almost exclusively on the phytotoxicity of ozone (Miller, 1993). In the early 1980s, it became evident that nitrogenous air pollutants, in the form of gases and aerosols (Grosjean, 1983; Russell *et al.*, 1985), or in wet precipitation, e.g. rain, snow, and fog (Jacob *et al.*, 1985; Munger *et al.*, 1983; Waldman *et al.*, 1985) were found in high concentrations near the urban centers of California and in adjacent forests (Bytnerowicz *et al.*, 1987; Grosjean & Bytnerowicz, 1993; Fenn & Bytnerowicz, 1993).

In the photochemical smog dominated areas, such as the Los Angeles Air Basin, oxidized N compounds, especially NO, NO₂, HNO₃, peroxyacetyl nitrate (PAN) and particulate NO₃⁻ are important in deposition processes (Russell *et al.*, 1985). In areas adjacent to agricultural activities, NH₃ and particulate NH₄⁺ may be more significant sources of deposited N (ApSimon *et al.*, 1987). In wet precipitation, NO₃⁻ and NH₄⁺ are the main sources of nitrogen. Other N air pollutants are also present in forests, although only limited information on their concentrations or deposition is available. The N trace gases, nitrous oxide (N₂O; an efficient and long-lived greenhouse gas; Rodhe, 1990) and NO can be released from forests soils as a result of microbial activity (Anderson & Poth, 1989). Recent data from California grasslands (Davidson, 1992), chaparral (Anderson *et al.*, 1988), and mixed conifer forests (Fenn *et al.*, in press) indicate that in the coarse-textured dry soils typical of these ecosystems, NO emissions are usually much greater than N₂O emissions. Another pollutant of interest is nitrous acid vapor (HNO₂) which is mainly formed at night in the polluted atmosphere. Through its photolysis it behaves as a powerful source of radicals in the early morning. HNO₂ is of special interest because of its presence in chemical reactions producing toxic substances, in particular carcinogens (Lammel & Perner, 1988). Ideally, all these major and minor N pollutants should be included in a budget of N compounds in forest stands.

Concentrations and biological effects of gases and aerosols

Nitric oxide (NO)

Most of the NO monitoring efforts have focused on urban areas, where NO concentrations are often high. During July 1993 the maximum hourly NO concentrations in California urban locations ranged between 6 and 70 µg m⁻³, in August between 11 and 125 µg m⁻³ and in September between 4 and 188 µg m⁻³ (Air Resources Board, 1993).

Very little is known of the concentrations of NO in forest locations in California. Concentrations of anthropogenically produced NO in forest receptor areas are generally low due to oxidation processes (including reactions with ozone). Generally, in the nonurban locations in the United States (California not included) the average concentrations of NO ranged between 1.2 and 12 µg m⁻³ (U.S. EPA, 1993). Monthly 24-h average concentrations of NO at Tanbark Flat in the San Gabriel

Mountains in the summer of 1985 ranged between 1.0 and 4.9 $\mu\text{g m}^{-3}$, while the maximum hourly average ranged between 4.1 and 30.0 $\mu\text{g m}^{-3}$ (Bytnerowicz *et al.*, 1987). At the same location during the 1989–1991 summer seasons the maximum hourly averages were <3.7 $\mu\text{g m}^{-3}$ (Grosjean & Bytnerowicz, 1993). At Shirley Meadow in the southwestern Sierra Nevada the monthly 24-h averages were <1.2 $\mu\text{g m}^{-3}$, while the maximum 1-h concentrations did not exceed 14.8 μg

m^{-3} (Temple & Bytnerowicz, 1993). NO concentrations occurring in most forested areas in California are not expected to cause phytotoxic effects (Wellburn, 1990).

Nitrogen dioxide (NO_2)

Most measurements of NO_2 have been done with the nitric oxide–ozone chemiluminescent method in which NO_2 is converted to NO. However, the method is not NO_2 specific and other oxidized N compounds (e.g.

Table 2. Concentrations of gaseous HNO_3 at various forested and other remote locations in California and elsewhere

Location (see Fig. 1)	Period of measurements	Concentrations ($\mu\text{g m}^{-3}$)	Method	Reference
<i>Northern and Central California</i>				
Gasquet	Summer 1989	Trace—daytime average Trace—nighttime average	Denuder difference	Ashbaugh <i>et al.</i> (1991)
Fremont	Summer 1989	0.8—daytime average 0—nighttime average	Denuder difference	Ashbaugh <i>et al.</i> (1991)
Sagehen	August 1994	0.64—24-h average	Annular denuder	Bytnerowicz, unpublished data
Yosemite National Park	Summer 1989	1.0—daytime average 0.3—nighttime average	Denuder difference	Ashbaugh <i>et al.</i> (1991)
Whitaker's Forest	Summers 1988–1990	1.1–1.8—daytime seasonal averages 0.3–0.7—nighttime seasonal averages	Annular denuder	Bytnerowicz and Riechers (1995)
Sequoia National Park	Summer 1989	0.7—daytime average 0.1—nighttime average	Denuder difference	Ashbaugh <i>et al.</i> (1991)
Eastern Brook Lake	Summer 1987 Winter 1987	0.36–24-h average 0.14–24-h average	Annular denuder/ filter pack	Miller and Walsh (1991)
Shirley Meadow	Summers 1989–1990	0.18–2.46—nighttime range 0.76–1.24—nighttime seasonal averages 0.28–4.82—daytime range 1.89–2.28—daytime seasonal averages	Annular denuder	Bytnerowicz and Miller (1991)
<i>Southern California</i>				
Tanbark Flat	Summers 1988–1991	3.0–27.3—daytime range 11.3–20.3—daytime seasonal averages 0.4–4.5—nighttime range 1.8–3.1—nighttime seasonal averages	Annular denuder	Grosjean and Bytnerowicz (1993)
Camp Paivika	September 1989	8.9–24-h average	Annular denuder	Fenn and Bytnerowicz (1993)
Barton Flats	Summers 1992–1994	0.45–9.69—24-h range 3.11–6.29—24-h seasonal averages	Annular denuder	Bytnerowicz <i>et al.</i> (1995)
Camp Osceola	September 1989	1.1–24-h average	Annular denuder	Fenn and Bytnerowicz (1993)
Shipley Reserve	Summer 1994	5.6–24-h average	Annular denuder	Bytnerowicz, unpublished data
<i>Other North American locations</i>				
Fortress Mtn, Alberta	1985–1987	0.0005–0.46 range of 48-h averages 0.31—mean of 48-h averages	Annular denuder	Legge and Krupa (1989)
Little Valley, Nevada	August 1994	2.02–24-h average	Annular denuder	Bytnerowicz, unpublished data
Mount Mitchell, North Carolina	Summer 1988	0.1–2.7—24-h average	Annular denuder	Aneja <i>et al.</i> (1991)
<i>Europe</i>				
Wolkerdorf, Austria	1990/1991	1.8–24-h annual average	Denuder difference	Puxbaum <i>et al.</i> (1993)
Black Forest, Germany	14 July 1987	2.1–24-h average	Laser-photolysis fragment-fluorescence	Papenbrock and Stuhl (1991)
North Sea coast		1.17–24-h average	Laser-photolysis fragment-fluorescence	Papenbrock and Stuhl (1991)
<i>Other remote locations</i>				
Mauna Loa, Hawaii	May–June 1988	0.33–24-h average	Filter pack	Ridley (1991)
Atlantic Ocean	September–October 1988	0.18–24-h average	Filter pack	Papenbrock <i>et al.</i> (1992)

PAN, peroxypropionyl nitrate, nitric acid, nitrate radical, dinitrogen pentoxide, nitrous acid and various organic compounds) are also converted to NO. Thus, much of the data on NO₂ concentrations entails a degree of uncertainty due to the inclusion of other N trace gases during analysis for NO. In rural and remote locations interferences from compounds other than NO₂ may be substantial (U.S. EPA, 1993).

Limited information is available on concentrations of NO₂ in forest locations in California. Much more information is available for urban locations. In Pasadena and Upland of the Los Angeles Basin the 24-h average summer concentrations of NO₂ approached 75 $\mu\text{g m}^{-3}$ (SCAQMD, 1985). During July and August 1993 the maximum hourly concentrations of NO₂ throughout the urban areas of California ranged from 19 to 244 $\mu\text{g m}^{-3}$, and from 19 to 489 $\mu\text{g m}^{-3}$ in September (Air Resources Board, 1993).

NO₂ concentrations in U.S. nonurban monitoring locations usually exceed 1.9 $\mu\text{g m}^{-3}$ and exhibit short-term variability (U.S. EPA, 1993). In Tanbark Flat, a chaparral site in the San Gabriel Mountains in the Los Angeles Air Basin, the 1-h average concentrations of NO₂ (corrected for PAN and PPN) in the summers of 1989–1991 were in the range of 19–75 $\mu\text{g m}^{-3}$ (Grosjean & Bytnerowicz, 1993). At Shirley Meadow of the southeastern Sierra Nevada, the monthly NO₂ concentrations during the 1989 and 1990 summer seasons ranged between 1.9 and 9.4 $\mu\text{g m}^{-3}$, while the maximum hourly values ranged between 7.5 and 90 $\mu\text{g m}^{-3}$

(Temple & Bytnerowicz, 1993). Concentrations of NO₂ occurring in California forests are below levels which could cause visible injury on plants (Taylor & McLean, 1970; Wellburn, 1988, 1990). Long-term exposures to NO₂, however, could affect growth rates of trees. At low doses of NO₂ stimulation of growth can occur while at the high doses growth of trees can be reduced (Murray *et al.*, 1994). Short-term exposure of conifers to elevated ambient levels of NO₂ induced nitrate reductase activity and increased the organic N content in two different spruce species. Nutritional importance of the assimilated pollutant for the studied conifers was emphasized (Norby *et al.*, 1989; Nussbaum *et al.*, 1993). Even higher nutritional effects of NO₂ may be expected in California forests chronically exposed to photochemical smog.

Nitric acid (HNO₃)

Concentrations of gaseous nitric acid are higher in urban source areas than in forested and other remote sites (except some of the forest locations in southern California; Table 2). Ashbaugh *et al.* (1991) determined that concentrations of HNO₃ in urban locations of California (Sacramento, Bakersfield, Santa Barbara, Los Angeles) ranged between 0.1 and 0.3 $\mu\text{g m}^{-3}$ at nighttime, and between 1.0 and 6.8 $\mu\text{g m}^{-3}$ at daytime. Higher values (average concentrations of 2.8 $\mu\text{g m}^{-3}$ at nighttime and about 33 $\mu\text{g m}^{-3}$ in the daytime) were recorded during the 9-day period of measurements performed in Glendora, California, in August 1986. Similarly high values were determined in the Los

Table 3. Concentrations of HNO₂ at various forested and other remote locations in California and elsewhere measured with annular denuder systems

Location (see Fig. 1)	Period of measurements	Concentrations ($\mu\text{g m}^{-3}$)	Reference
<i>Central California</i>			
Sagehen	August 1994	0.005 24-h average	Bytnerowicz, unpublished data
Whitaker's Forest	Summers 1988–1990	0.00–0.52—daytime range 0.08–0.30—daytime seasonal averages 0.00–0.62—nighttime range 0.08–0.19—nighttime seasonal averages	Bytnerowicz and Riechers (1995)
Shirley Meadow	Summers 1989 and 1990	0.02–0.64—nighttime range 0.17–0.76—nighttime seasonal averages 0.00–1.94—daytime range 0.20–0.39—daytime seasonal averages	Bytnerowicz and Miller (1991)
<i>Southern California</i>			
Tanbark Flat	Summers 1988–1991	0.0–1.5—daytime range 0.2–0.5—daytime seasonal averages 0.0–2.0—nighttime range 0.8–0.9—nighttime seasonal averages	Grosjean and Bytnerowicz (1993)
Barton Flats	Summers 1992–1994	0.00–0.43—24-h range 0.03–0.21—24-h seasonal averages	Bytnerowicz, unpublished data
Camp Paivika	September 1989	0.09—24-h average	Fenn and Bytnerowicz (1993)
Camp Osceola	September 1989	0.00—24-h average	Fenn and Bytnerowicz (1993)
Shipley Reserve	Summer 1994	0.28—24-h average	Bytnerowicz, unpublished data
<i>Other locations</i>			
Little Valley, NV	August 1994	0.003—24-h average	Bytnerowicz, unpublished data
Fortress Mtn, Alberta	1985–1987	0.005–0.46—range of 48-h averages 0.05—mean of 48-h averages	Legge and Krupa (1989)

Angeles Basin during a September 1985 study by Solomon *et al.* (1988). By comparison, in Edmonton, Canada, the 24-h annual average in 1982/1983 was $0.30 \mu\text{g m}^{-3}$ with the highest daily average of $2.4 \mu\text{g m}^{-3}$ (Peake *et al.*, 1988).

Very little is known of the direct effects of HNO_3 on plants. It has been established that HNO_3 has a very high deposition velocity compared to other pollutants and therefore it may be an important source of N for plants (Hanson & Lindberg, 1991). After 48 h of exposures at $230 \mu\text{g m}^{-3}$ HNO_3 , ponderosa pine seedlings showed significant changes in epicuticular waxes and increased nitrate reductase activity (Krywult *et al.*, in press). This finding as well as elevated concentrations of free amino acids in ponderosa pine needles (Pam Padgett, personal communication) indicate that HNO_3 may be a N source for that species. This is consistent with the results of HNO_3 fumigations for red spruce (Norby *et al.*, 1989). The observed changes in wax chemistry caused by HNO_3 and accompanying injury of the cuticle (Bytnerowicz *et al.*, unpublished data) may predispose plants to various environmental stresses (including drought and air pollutants) or pathogens. Because elevated concentrations of HNO_3 and ozone co-occur in the summer (Solomon *et al.*, 1992), interactions between these two pollutants are also possible.

Nitrous acid (HNO_2)

Information on concentrations of HNO_2 in forested areas and urban locations is scant (Table 3). The highest levels of HNO_2 occur just before sunrise and are commonly in the range of 3.8 – $7.6 \mu\text{g m}^{-3}$ in the Los Angeles area (U.S. EPA, 1993). According to Harris *et al.* (1982), HNO_2 concentrations in the Los Angeles area can reach about $15 \mu\text{g m}^{-3}$. By comparison, in Gothenburg, Sweden, in January–May HNO_2 concentrations ranged from 0.05 to $2.35 \mu\text{g m}^{-3}$ (Sjodin & Ferm, 1985). Practically nothing is known of the biological importance of HNO_2 . It is plausible that high concentrations of HNO_2 increase nitrite (NO_2^-) concentrations in cells to toxic levels. The highest potential for toxic effects could be

expected at low temperatures (Wellburn, 1990). However, at the presently observed levels in California forests, no toxic effects are expected to occur.

Peroxyacetyl nitrate (PAN) and peroxypropionyl nitrate (PPN)

Peroxyacetyl nitrate (PAN) and peroxypropionyl nitrate (PPN) are formed when organic compounds are oxidized in the presence of oxides of nitrogen (U.S. EPA, 1993). There is much more information regarding concentrations of PAN and PPN in urban locations than in forested and other remote locations (Table 4). For example, the 24-h average concentrations of PAN in February 1984 in Downey, CA, were $6.09 \mu\text{g m}^{-3}$; in March 1984 in Houston, TX, $3.71 \mu\text{g m}^{-3}$; and in March–April 1984 in Denver, CO, $3.20 \mu\text{g m}^{-3}$ (Singh & Salas, 1989). More information regarding ambient concentrations of PAN in various urban locations worldwide has been presented by Temple and Taylor (1983). Average 24-h concentrations of PPN in some of the urban locations were: in February 1984 in Downey, CA, $0.33 \mu\text{g m}^{-3}$; in March–April 1984 in Houston, TX, $0.25 \mu\text{g m}^{-3}$; and in March–April, 1984 in Denver, CO, $0.12 \mu\text{g m}^{-3}$ (Singh & Salas, 1989).

PAN is a phytotoxic agent which can cause characteristic necrotic lesions on foliage of sensitive agricultural and ornamental plants in southern California and in other North American locations (Taylor & McLean, 1970). The potential for serious PAN injury to plants is minimal in eastern North America and Europe but is much greater in California (Temple & Taylor, 1983). However, no information on PAN injury to forest plants has been reported.

NH_3

Ammonia is the only gaseous basic compound present in the atmosphere in appreciable concentrations. Both in the USA and in Europe, the production and use of animal manure contributes at least 70% to the total anthropogenic emission of NH_3 (Schuurkes *et al.*, 1988). Ammonia emissions from fertilizer production and as

Table 4. Concentrations of PAN and PPN at various forested and other remote locations

Location (see Fig. 1)	Period of measurements	Concentration ($\mu\text{g m}^{-3}$)		Reference
		PAN	PPN	
<i>California</i>				
Point Arena	January 1985	0.61—24-h average	0.03	Singh and Salas (1989)
	April 1984–March 1985	0.23—24-h average	0.02	
Tanbark Flat	Summers 1989–1991	0.45–51.0—24-h range	0.55–9.9	Grosjean and Bytnerowicz (1993)
Franklin Canyon	September 1991	0.50–34.7—24-h range	0.0–6.35	Grosjean <i>et al.</i> (1993)
<i>Other locations</i>				
Niwot Ridge, Colorado	July 1984	1.36—24-h average	0.09	Singh and Salas (1989)
Kananaskis, Alberta, Canada	Summer 1982	< 4.95—0.5-h averages		Peake <i>et al.</i> (1983)
Eastern Pacific	November–December 1981	< 0.59—1-h averages		Singh <i>et al.</i> (1986)

a result of inefficient application of N fertilizers (Fangmeier *et al.*, 1994), are also important sources of reduced N. Ammonium (NH_4^+) is not emitted in significant quantities and all of it originates from NH_3 (Asman *et al.*, 1989). In some high N deposition areas of the Netherlands affected by NH_3 emissions, throughfall deposition of NH_4^+ is over three times greater than deposition of NO_3^- (Van Breemen *et al.*, 1987). Large areas of forests have been affected by NH_3 near huge pig farms in eastern Germany (Fangmeier *et al.*, 1994). Very high concentrations of NH_3 (annual average of about $75 \mu\text{g m}^{-3}$) can cause direct damage to vegetation (Temple *et al.*, 1979; Van der Eerden, 1982), but NH_3 levels of this magnitude only occur near unusually strong point sources. Ammonia concentrations in forest locations in California are presented in Table 5. Con-

centrations in urban areas or locations close to major agricultural operations may be much higher than in the surrounding regions.

Particulate (aerosol) NO_3^- and NH_4^+

Particulate (aerosol) NO_3^- and NH_4^+ are secondary pollutants, formed as a result of chemical reactions of primary pollutants. According to Finlayson-Pitts and Pitts (1986) secondary pollutants usually have aerodynamic diameters $< 2.5 \mu\text{m}$, whereas naturally produced particles, such as dust are $> 2.5 \mu\text{m}$. No phytotoxic effects of particulate NO_3^- or NH_4^+ pollution have been reported. However, the potential importance of particulate pollution, especially particles $< 10 \mu\text{m}$ in diameter (PM 10), including various health effects has been recently recognized (Chow *et al.*, 1992, 1993).

Table 5. Concentrations of ambient NH_3 at various forested and other remote locations in California and elsewhere

Location (see Fig. 1)	Period of measurements	Concentrations ($\mu\text{g m}^{-3}$)	Method	Reference
<i>Central California</i>				
Sagehen	August 1994	0.35—24-h average		Bytnerowicz, unpublished data
Whitaker's Forest	Summer 1988–1990	0.51–2.28—nighttime range 1.11–1.56—nighttime seasonal averages 0.23–3.75—daytime range 1.92–2.48—daytime seasonal averages	Annular denuder	Bytnerowicz and Riechers (1995)
Shirley Meadow	Summers 1989–1990	0.31–4.65—nighttime range 1.76–1.95—nighttime seasonal averages 0.82–5.47—daytime range 2.06–2.31—daytime seasonal averages	Annular denuder	Bytnerowicz and Miller (1991)
Point Arena	April/May 1985	0.11–0.88—range 0.25—average	Tungsten oxide denuder tube	Roberts <i>et al.</i> (1988)
<i>Southern California</i>				
Tanbark Flat	Summer 1988–1991	0.3–4.7—daytime range 1.6–2.5—daytime seasonal averages 0.0–1.5—nighttime range 0.6–0.7—nighttime seasonal averages	Annular denuder	Grosjean and Bytnerowicz (1993)
Barton Flats	Summers 1992–1994	0.62–2.59—24-h range 1.42–1.49—24-h averages	Annular denuder	Bytnerowicz <i>et al.</i> (1995)
Shipley Reserve, CA	Summer 1994	2.28—24-h average	Annular denuder	Bytnerowicz, unpublished data
<i>Other locations</i>				
Little Valley, NV	August 1994	1.14—24-h average	Annular denuder	Bytnerowicz, unpublished data
Fortress Mtn, Alberta	1985–1987	0.04–1.30—range of 48-h averages 0.26—mean of 48-h averages	Annular denuder	Legge and Krupa (1989)
Niwot Ridge, CO	Oct/Nov 1984	0.04–0.48—range 0.13—average	Tungsten oxide denuder tube	Roberts <i>et al.</i> (1988)
Wolkendorff, NE Austria	1990/91	1.5—24-h average	Annular denuder	Puxbaum <i>et al.</i> (1993)

Table 6. Concentrations of particulate NO_3^- and NH_4^+ in forested and other remote locations in California and elsewhere determined with an annular denuder method

Location (see Fig. 1)	Period of measurement	Concentration ($\mu\text{g m}^{-3}$)		Reference
		NO_3^-	NH_4^+	
<i>Central California</i>				
Sagehen Valley	August 1994	0.15—24-h average	0.18—24-h average	Bytnerowicz, unpublished data
Whitaker's Forest	Summers 1988–1990	0.20–3.80—daytime range 0.30–1.40—daytime seasonal averages 0.15–2.20—nighttime range 0.44–1.05—nighttime seasonal averages	0.00–1.41—daytime range 0.39–0.41—daytime seasonal averages 0.00–1.40—nighttime range 0.11–0.33—nighttime seasonal averages	Bytnerowicz and Riechers (1995)
Shirley Meadow	Summers 1989–1990	0.03–0.94—daytime range 0.44–0.46—daytime seasonal averages 0.10–2.20 —nighttime range 0.68–0.74 —nighttime seasonal averages	0.00–0.26—daytime range 0.07–0.08—daytime seasonal averages 0.00–0.53—nighttime range 0.10–0.26—nighttime seasonal averages	Temple and Bytnerowicz (1993)
<i>Southern California</i>				
Tanbark Flat	Summers 1988–1991	0.1–21.8—daytimerange 3.0–5.1—daytime seasonal averages 0.1–2.7—nighttime range 0.5–0.7—nighttime seasonal averages	0.0–4.0—daytime range 0.9–1.3—daytime seasonal averages 0.0–2.7—nighttime range 0.4–0.7—nighttime seasonal averages	Grosjean and Bytnerowicz (1993)
Barton Flats	Summers 1992–1994	0.39–8.52—24-h range 2.22–3.28—24-h averages	0.07–1.99—24-h range 0.61–0.93—24-h averages	Bytnerowicz <i>et al.</i> (1995)
Camp Paivika	September 1989	5.1—24-h average	0.9—24-h average	Fenn and Bytnerowicz (1993)
Camp Osceola	September 1989	2.1—24-h average	0.1—24-h average	Fenn and Bytnerowicz (1993)
Shipley Reserve	Summer 1994	2.34—24-h average	0.91—24-h average	Bytnerowicz, unpublished data
<i>Other locations</i>				
Little Valley, Nevada	August 1994	0.51—24-h average	0.14—24-h average	Bytnerowicz, unpublished data
Fortress Mtn, Alberta	1985–1987	0.02–0.66—range of 48-h averages 0.13—mean of 48-h averages		Legge and Krupa (1989)

Practically nothing is known about the potential effects of elevated concentrations of particulate NO_3^- and NH_4^+ on plants. Probably the greatest effect of atmospheric deposition of NO_3^- and NH_4^+ on forests is to provide available N for plants and microbes, especially when these ions are solubilized and transported to the soil in rainfall, throughfall or stemflow. Concentrations of NO_3^- and NH_4^+ in selected forest locations in California are presented in Table 6.

Apportionment of gaseous and particulate N pollutants in selected locations in California forests

Information regarding concentrations of the individual N pollutants for a given area is needed to evaluate the potential for phytotoxicity and for calculating N deposition to a forest stand. The calculated apportionment of the various N pollutants can help establish the importance of individual pollutants, and may also indicate the relative contribution of stationary (industry, agriculture) and mobile (automobiles, aircraft) emission sources.

Tanbark Flat is in a highly-polluted area of the San Gabriel Mountains within the Los Angeles Air Basin (Bytnerowicz *et al.*, 1987), and is characterized by a high proportion of oxidized N compounds, mainly NO_2 , HNO_3 and PAN (Fig. 2(a) and (b)). Proportionally, NO_2 was a larger component of the total N pollution during the nighttime due to reduced HNO_3 , NO_3^- , and NH_3 components. This scenario is expected for a site located in direct proximity of a photochemical smog source area (Russell *et al.*, 1985).

Whitaker's Forest, in the western Sierra Nevada, is a receptor of air masses from the San Joaquin Valley—characterized by intensive agricultural activities and high photochemical smog of a local and remote origin (Miller *et al.*, 1972). The more heterogeneous pollution source types at Whitaker's Forest compared to Tanbark Flat is reflected in the apportionment of N pollutants (Fig. 2(c) and (d)). The proportion of oxidized N compounds at Whitaker's Forest is much lower, with > 60% of the N pollutants in reduced forms. Apportionments

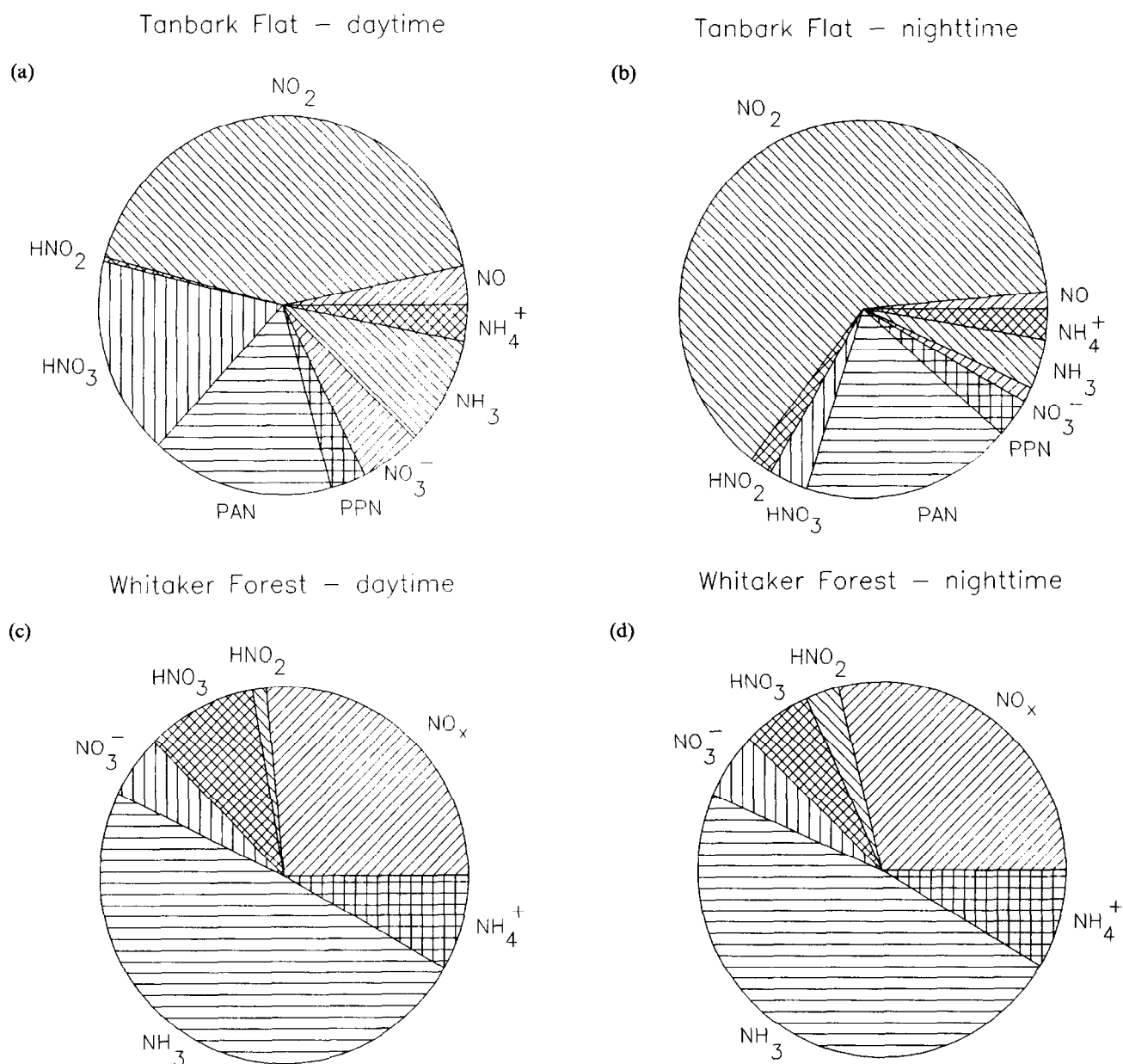


Fig. 2. Apportionment of N air pollutants (daytime and nighttime) in two forest locations in California in summer 1990. Modified data from Grosjean and Bytnerowicz (1993) were used for Tanbark Flat, and from Bytnerowicz and Riechers (1995) for Whitaker's Forest.

of individual pollutants at Whitaker's Forest were similar for daytime and nighttime. The situation described for Whitaker's Forest is probably typical for other forest sites in the western Sierra Nevada.

Concentrations of nitrogen ions in wet precipitation

In recent years much has been learned of the chemistry of wet precipitation in California, mainly from the California Acid Deposition Monitoring Program (CADMP). Results of this program are summarized in Fig. 3 and indicate that participation of nitrogen in wet deposition in California forests is much higher than that of sulfur (most of the sites had the equivalent $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio >1 and in addition to high NH_4^+ concentrations in precipitation). An example from the western Sierra Nevada shows that chemical composition

of cloud water was dominated by NO_3^- and NH_4^+ ions with a much lesser importance of SO_4^{2-} (average $\text{NH}_4^+:\text{NO}_3^-:\text{SO}_4^{2-}$ equivalent ratio of 4.9:3.5:1.0). Average concentrations of NH_4^+ and NO_3^- in cloud water were more than ten times of those observed in rain, and cloud water SO_4^{2-} concentrations were more than three times of those observed in rain (Collett *et al.*, 1989, 1990a,b). Precursor concentrations of aerosol NO_3^- , SO_4^{2-} , and NH_4^+ , and gas phase HNO_3 and NH_3 were sufficient to account for the observed cloud water loadings of NO_3^- , SO_4^{2-} , and NH_4^+ on a transect in Sequoia National Park (Collett *et al.*, 1990b).

On the crest of the central Sierra Nevada, precipitation acidity significantly increased between 1979 and 1986. The observed change in acidity was closely associated with changes in NO_3^- concentrations but not

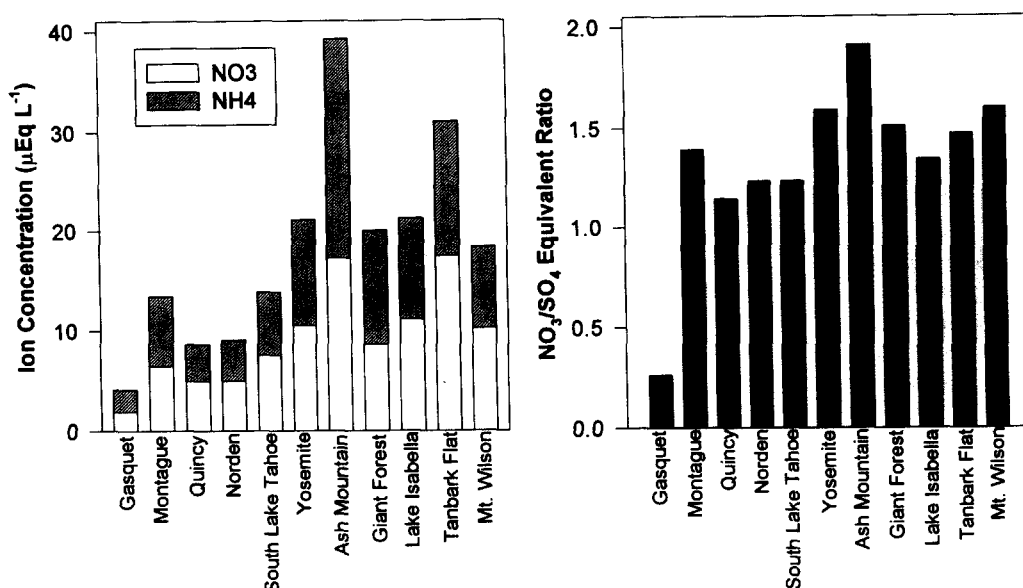


Fig. 3. Concentrations of nitrate and ammonium and nitrate/sulfate ratios in precipitation in forest locations in California. Data are for 1985–1990, from the California Acid Deposition Monitoring Program (Blanchard & Tonnessen, 1993).

of SO_4^{2-} (Byron *et al.*, 1991). The greater importance of wet deposition of N over S in California seems to apply over broad geographic areas within the State, including both urban and remote sites. The average mass deposition ratios of $\text{NO}_3^-/\text{SO}_4^{2-}$ (equivalent basis) based on several years of monitoring were: 0.7 for Northwest California; 0.95 for the Bay Area; 1.4 for central California; 1.2 for the central California coast; 1.6 for the Sierra Nevada; and 1.2 for southern California (Blanchard & Tonnessen, 1993). The range of average $\text{NO}_3^-/\text{SO}_4^{2-}$ ratios for 1990–1993 from wet deposition monitoring in 11 alpine locations of the Sierra Nevada was 1.14–1.48 (Melack *et al.*, 1995). The $\text{NO}_3^-/\text{SO}_4^{2-}$ mass deposition ratio in rain was highly pronounced (7.15) during the summer smog season at Barton Flats, a site in the San Bernardino Mountains east of Los Angeles with estimated N deposition of 5–9 $\text{kg ha}^{-1} \text{ year}^{-1}$ (Bytnerowicz *et al.*, 1995).

NITROGEN DEPOSITION INPUTS TO FOREST STANDS

Forest vegetation is an important receptor for air pollutants. Because of the reactivity of its large surface area, the foliage of forest canopies serves as a very effective receptor for airborne substances (Hosker & Lindberg, 1982). Atmospheric deposition occurs in dry and wet forms. Dry deposition is the transfer of gases and particles from the atmosphere via atmospheric turbulence and diffusion without the intervention of precipitation. Dry deposition occurs when turbulent transport or sedimentation introduces gases or particles to the laminar layer surrounding deposition surfaces. The gases and particles are transported through this layer via convection, diffusion, or inertial processes until chemical or physical capture of the pollutants by the

surface occurs (Voldner *et al.*, 1986; Böhm, 1992). In wet deposition, gaseous and aerosol pollutants may be removed from the air by rain or snow during rainout (within-cloud scavenging) and washout (below-cloud scavenging) events. Forest canopies intercept wet precipitation in various forms (rain, snow, fog, cloud, or rime ice). Dissolved ions in precipitation can be taken up directly by foliage or indirectly by roots from the soil and forest floor (Böhm, 1992). However, much of the nutrients in throughfall and precipitation can be immobilized by microorganisms in the soil and forest floor (Johnson, 1992).

Dry deposition fluxes of NO_3^- and NH_4^+ to branches of several tree species at various locations in California based on branch washing technique and estimates of N deposition on a landscape level are presented in Table 7. The values presented in Table 7 can be compared with other estimates based on throughfall analysis (both branch rinsing and throughfall analysis are based on removal of deposited airborne matter by water and therefore may be directly compared). At Tanbark Flat deposition of N was estimated as 23 $\text{kg ha}^{-1} \text{ year}^{-1}$ (Riggan *et al.*, 1985). Annual throughfall N deposition ranged from 5.6 to 12.4 kg ha^{-1} at three high-elevation (2060 m) mixed conifer sites located within 3 km of each other near Kratka Ridge in the San Gabriel Mountains northeast of Los Angeles. Differences in N deposition at the three sites were due to topographical influences on pollution transport into the sites (Kiefer, 1995). Annual N deposition was 2.0 kg ha^{-1} (Schlesinger *et al.*, 1982) in the Santa Ynez Mountains near Santa Barbara, and 1.5 kg ha^{-1} at Sky Oaks Ranch in northeastern San Diego county (Ellis *et al.*, 1983). Throughfall N inputs near Giant Forest in Sequoia National Park were 6.2 and 10.8 $\text{kg ha}^{-1} \text{ year}^{-1}$ under mixed fir and sequoia stands, with throughfall NO_3^- deposition slightly more than twice as high as NH_4^+ deposition (Chorover *et*

Table 7. Deposition fluxes of NO_3^- and NH_4^+ to native trees in California determined with a foliage rinsing technique and estimated annual dry deposition of N

Location (see Fig. 1)	Species	Deposition ($\mu\text{g m}^{-2} \text{h}^{-1}$)		Estimated annual dry deposition (kg ha^{-1})	Reference
		NO_3^-	NH_4^+		
Eastern Brook Lake	<i>Pinus albicaulis</i> , <i>P. contorta</i>	4–6 (range) 4.5 (average)	1–2 (range) 1.1 (average)	0.5	Bytnerowicz <i>et al.</i> (1992)
Emerald Lake	<i>P. contorta</i> , <i>P. monticola</i>	27–30 (range) 28 (average)	6–9 (range) 7.5 (average)	^a	Bytnerowicz <i>et al.</i> (1991)
Barton Flats	<i>P. ponderosa</i>	35–195 (range) 75 (average)	4–19 (range) 10 (average)	5–9	Bytnerowicz <i>et al.</i> (1995)
	<i>Abies concolor</i>	33–182 (range) 70 (average)	4–17 (range) 9 (average)		
	<i>Quercus kelloggii</i>	18–103 (range) 39 (average)	1–6 (range) 3 (average)		
Whitaker's Forest	<i>P. ponderosa</i> (seedlings)	6–31 (range) 14 (average)	1.5–6 (range) 3.5 (average)	1.0–1.5	Bytnerowicz and Riechers (1995)
Tanbark Flat	<i>Ceanothus crassifolius</i>	88–455 (range)	17–46 (range)	20–35	Bytnerowicz <i>et al.</i> (1987)
		224 (average)	31 (average)		
Camp Paivika	<i>P. ponderosa</i>	180 (average)	21 (average)	29	Fenn and Bytnerowicz (1993)
Camp Osceola	<i>P. ponderosa</i>	42 (average)	11 (average)	6	Fenn and Bytnerowicz (1993)

^aDry deposition not estimated due to a lack of vegetation inventory data.

al., 1994). Mean annual throughfall N deposition was 1.2 kg ha^{-1} in a blue oak (*Quercus douglasii* H. and A.) stand in the Hastings Natural History Reservation in the Santa Lucia Mountains in central California over a two year period (Callaway & Nadkarni, 1991).

Because dry deposition to forests is difficult to measure directly, the use of models has been advocated (Hicks *et al.*, 1987). Most dry deposition models are based on detailed information of concentrations of gaseous pollutants of interest, microclimatological factors, and physiological activity of plants (Meyers & Baldocchi, 1988). Available models have been developed mostly for relatively flat, horizontally homologous terrain of the eastern United States (Baldocchi *et al.*, 1987) and their use in California forests is questionable. This is mainly due to much higher complexity of the mountain terrain and patchiness of the vegetation cover. An additional reason for this criticism is that the existing models are not designed for calculating deposition of particulate matter, which under the conditions prevalent in California, accounts for substantial nutrient input to forests.

Based on the results of the California Acid Deposition Monitoring Program, average wet deposition of N from $\text{NO}_3^- + \text{NH}_4^+$ was: Northern California 1.4 ; San Francisco Bay Area 1.9 ; central California 0.8 ; central California coast 0.8 ; Sierra Nevada 1.7 ; and southern California $1.5 \text{ kg ha}^{-1} \text{ year}^{-1}$ (Blanchard & Tonnessen, 1993). Annual wet deposition of N in 11 alpine Sierra Nevada locations during 1990–1993 ranged from 0.66 to 1.58 kg ha^{-1} (Melack *et al.*, 1995). At Emerald Lake in Sequoia National Park, the average annual inorganic wet N deposition was about 2.2 kg ha^{-1} (Williams *et al.*, 1995). Deposition of N to open rain collectors was 11.9 kg ha^{-1} during 5.5 months of

winter and early spring (13 December 1993 to 1 June 1994) at Camp Paivika, near Crestline on the western highly-polluted end of the San Bernardino Mountains (Fenn *et al.*, 1995). Total annual N deposition at Camp Paivika is estimated to be $35\text{--}45 \text{ kg ha}^{-1} \text{ year}^{-1}$ (Fenn & Bytnerowicz, 1993; Fenn *et al.*, in press). By comparison, total N deposition in many European forests may be higher than in most polluted California forest sites. For example, in Sweden N deposition reportedly ranges from 5 to $40 \text{ kg ha}^{-1} \text{ year}^{-1}$ (Grennfeld & Hultberg, 1986), while in the Netherlands values approaching $115 \text{ kg ha}^{-1} \text{ year}^{-1}$ have been reported (Draijers *et al.*, 1989).

EFFECTS OF N DEPOSITION ON CALIFORNIA FOREST ECOSYSTEMS

Nitrogen saturation

Northern temperate forests are generally considered to be N deficient and highly conservative of N (Aber *et al.*, 1989; Vitousek & Howarth, 1991). However, recent studies from the eastern US and Europe confirm that chronic N deposition can lead to the contrasting condition of N saturation (Aber *et al.*, 1989; Ågren & Bosatta, 1988; Dise & Wright, 1995; Driscoll & Van Dreason, 1993; Emmett *et al.*, 1993; Hultberg *et al.*, 1994; Kahl *et al.*, 1993; Stoddard, 1994) with potentially undesirable effects on forest ecosystems. An ecosystem is defined as N saturated when available N is in excess of total plant and microbial nutritional demand (Aber *et al.*, 1989), resulting in above-normal sustained N losses from the ecosystem. A forest may also be considered N saturated when vegetation no longer exhibits a positive growth response to further additions

of N (Nilsson, 1986), even though other factors are not yet limiting (Ågren & Bosatta, 1988; Brown *et al.*, 1988). A number of forests with moderate or high N deposition inputs are N saturated (Dise & Wright, 1995; Feger, 1992; Foster *et al.*, 1989; Johnson & Fernandez, 1992; Johnson *et al.*, 1991b; Van Breemen *et al.*, 1987).

Most studies on the effects of N deposition to terrestrial ecosystems are from Europe and eastern North America, where concern over forest damage due to acid rain began in the early 1980s (Blank *et al.*, 1988). However, the effects of N deposition on ecosystems of California and much of the western US may not always parallel the effects of N deposition on ecosystems in Europe and eastern North America because of major differences in climate, vegetation, soil characteristics, and in the forms of pollutants which predominate in each region.

An array of N status indicators (ideally including long-term baseline data) may be needed to evaluate the degree of N saturation of an ecosystem. Examples of possible indicators of N enrichment of an ecosystem include: high nutrient cycling rates in soil and in the forest floor (Fenn, 1991; Van Miegroet *et al.*, 1992), elevated emissions of trace N gases from the soil (Matson *et al.*, 1992; Melillo *et al.*, 1989), altered plant nutrient ratios (Ericsson *et al.*, 1993; Katzensteiner *et al.*, 1992; Mohren *et al.*, 1986), high soil acidity and low base saturation (Falkengren-Grerup, 1989; Schulze, 1989; Tamm & Hallbäck, 1988), foliar accumulation of NO_3^- (Fenn *et al.*, in press; Stams & Schipholt, 1990) or amino acids (Ericsson *et al.*, 1993), and high NO_3^- concentrations in the soil solution (Katzensteiner *et al.*, 1992) and in water emanating from polluted watersheds (Riggan *et al.*, 1985, 1994; Stoddard, 1994).

Naturally occurring N saturation

Nitrogen saturation in Western forest ecosystems can occur from biological or geological sources of excess N (Dahlgren, 1994; Johnson *et al.*, 1991a; Van Miegroet & Cole, 1984). Douglas-fir forests in the Pacific Northwest with a significant red alder (*Alnus rubra*) component experience major N losses annually (Johnson, 1992). Nitrogen fixation was estimated to be $70 \text{ kg ha}^{-1} \text{ year}^{-1}$ in a red alder stand, an amount equivalent to NO_3^- leachate loss from the ecosystem (Johnson, 1992; Van Miegroet & Cole, 1984).

In the Klamath mountains in northern California numerous barren areas (except for scattered growth of the prostrate herb *Calyptridium umbellatum*) up to 20 ha in size are found interspersed among adjacent stands of healthy coniferous forest. It was reported that soil acidification and N saturation occur in these areas as a result of the weathering of ammonium in mica schist bedrock. Nitrogen saturation developed in sites presumably cleared of vegetation by a disturbance such as lightning-induced fire, followed by soil erosion. Nitrogen saturation conditions were not found in adjacent sites with coniferous vegetation (Dahlgren, 1994). The C:N ratio of soil from barren and healthy forest sites

was < 10 due to the weathering of geological N. It was proposed that ammonium released from the parent soil material is rapidly nitrified, leading to acidification and high NO_3^- and aluminum concentrations in the soil solution. The N saturated sites presumably remain barren due to extreme cation depletion, high concentrations of labile Al in the soil solution, and soil acidification (Dahlgren, 1994).

Nitrogen deposition in southern California forests—a test case

Chaparral, sagebrush, mixed conifer forests, and transition communities are the major terrestrial ecosystems within the region of greatest chronic air pollution exposure (Miller, 1983, 1992). Only recently have the ecological effects of chronic N deposition in these ecosystems been addressed (Fenn, 1991; Fenn & Bytnerowicz, 1993; Fenn *et al.*, in press; Riggan *et al.*, 1985, 1994). The N status of forests within the Los Angeles Air Basin spanning a range of N deposition inputs during the last 40–50 years serves as a test case of the capacity of summer-dry Western forests to process and retain fixed atmospheric N. The more extreme cases of air pollution exposure increase the likelihood that signals of air pollution effects will be more clearly discernible above the background variation inherent in natural ecosystems. As discussed by Lang *et al.* (1982), extreme cases of atmospheric deposition may exaggerate unnoticed effects of air pollutants that are occurring elsewhere. Effects may be magnified or hastened, thus providing a forewarning of ecological effects that may occur in less-polluted areas. Excess N deposition in the West may become a more widespread problem in coming years as urban centers continue to grow (Böhm, 1992).

Effects of N deposition in mixed conifer forests in the San Bernardino Mountains

Photochemical oxidant damage to ponderosa pine in the San Bernardino Mountains (SBM) was observed in the 1950s, although the cause of the damage was not confirmed until 1963 (Miller, 1992; Miller *et al.*, 1963). An ozone gradient, with decreasing concentrations primarily from west to east corresponded with a gradient of decreasing ozone injury (Miller, 1992). Dry deposition of N also decreases along the west to east ozone gradient in the SBM (Fenn & Bytnerowicz, 1993). Nitrogen deposition at Camp Paivika (CP, a western site in the SBNF) is estimated to be $35\text{--}45 \text{ kg ha}^{-1} \text{ year}^{-1}$, compared to $5\text{--}9 \text{ kg ha}^{-1} \text{ year}^{-1}$ at Camp Osceola (CAO, an eastern site in the San Bernardino Mountains (Fenn & Bytnerowicz, 1993; Fenn *et al.*, in press).

A number of nutritional and edaphic parameters are listed in Table 8 indicating that the mixed conifer forest at CP is N saturated (Fenn *et al.*, in press). The most diagnostic indicators of N saturation at CP are the maintenance of high concentrations of NO_3^- in foliage, the soil solution and in soil extracts throughout the growing season, and elevated NO emissions from soil. High soil NO_3^- concentrations and NO emissions

Table 8. Nutritional and edaphic parameters indicating N saturation in high-deposition sites in the San Bernardino Mountains^a

Parameter	Status
Soil NO ₃ ⁻	Concentrations in soil solution and soil extracts 14–44 times higher at CP ^b than at CAO ^b
Foliar NO ₃ ⁻	Significantly greater accumulation at CP compared to CAO
NO fluxes from soil	Elevated at CP compared to CAO and other forests in North America
Soil C:N	Low at CP compared to low-deposition sites
Foliar N:P	Greater at CP
Soil pH	Low at CP. Decreasing soil pH in high-deposition sites in the San Bernardino and San Gabriel Mountains since the 1970s (Wood <i>et al.</i> , 1992)
Percent base saturation	Significantly lower at high-deposition sites
L-litter decomposition	Significantly greater in high deposition sites (Fenn & Dunn, 1989); not N-limited at CP (Fenn, 1991)
N-mineralization rates in soil and litter	Greater in high deposition sites than low-deposition sites
Foliar growth response to N fertilization	Tree growth not N limited at CP, but was limited at low-deposition sites

^aUnless indicated otherwise, results are from Fenn *et al.* (in press); ^bestimated annual N deposition at CP (Camp Paivika) is 35–45 kg ha⁻¹, and 5–9 kg ha⁻¹ at CAO (Camp Osceola).

suggest available soil N in excess of biological demand. Katzensteiner *et al.* (1992) also concluded that the Norway spruce stands in their study were N saturated based on high NO₃⁻ concentrations in soil solution during the growing season. Excess available soil N at CP is further evidenced by lack of foliar growth in ponderosa pine in response to N fertilization in two experiments (Fenn, unpublished data). N:nutrient ratios in soil and foliage and enhanced litter decomposition and N mineralization rates also indicate N enrichment at CP (Table 8). Percent base cation saturation and soil pH were also lowest in the high-pollution sites. Soil acidification has also occurred during the last 20 years in soils of high-deposition sites in the San Bernardino and San Gabriel mountains (Fenn *et al.*, in press; Wood *et al.*, 1992). Field data on soil solution NO₃⁻, soil acidification, and base cation depletion corresponded closely with earlier simulation model results using the Nutrient Cycling Model NuCM (Fenn *et al.*, in press).

Foliar N:P ratios are reportedly better indicators of excess N than foliar N concentrations alone (Ericsson *et al.*, 1993; Mohren *et al.*, 1986; Zinke, 1980). According to the N saturation hypothesis, plant P levels will decrease in N saturated sites as N limitations are removed (Aber *et al.*, 1989). The N:P ratio in foliage of ponderosa pine, California black oak, and bracken fern was 1.7–2.5 times greater at CP than at CAO (Fenn *et al.*, in press). Zinke (1980) reported greater N:P ratios in bigcone Douglas-fir (*Pseudotsuga macrocarpa* (Vasey) Mayr) in the high-pollution sites than in the less-polluted sites in several mountainous areas of southern California. More recently, it was reported that foliar N concentrations in bigcone Douglas-fir in the SBM were negatively correlated with P concentrations, with higher N:P ratios in study plots of the high-pollution region compared to the low-pollution plots (Poth *et al.*, 1991).

Recent studies in the San Bernardino and San Gabriel mountains provide preliminary evidence for N saturation in sites experiencing N deposition loads of *ca* 25 kg ha⁻¹ year⁻¹ or greater, but not in sites with < 15 kg ha⁻¹ year⁻¹ (Fenn *et al.*, in press; Kiefer, 1995; Riggan *et al.*, 1985). Three mixed-conifer stands (2060 m eleva-

tion) near Kratka Ridge in the San Gabriel Mountains northeast of Los Angeles were N limited based on a large foliar growth response following N fertilization (Kiefer, 1995). Annual N deposition in throughfall at the three sites ranged from 5.6 to 12.4 kg ha⁻¹. Chaparral watersheds in the San Gabriel Mountains with annual throughfall N inputs of *ca* 24 kg ha⁻¹ were N saturated, and N inputs to the N saturated mixed conifer forest at Camp Paivika are *ca* 35–45 kg ha⁻¹ year⁻¹. Based on data for 65 forest stands in Europe, Dise and Wright (1995) also found the deposition of 25 kg N ha⁻¹ year⁻¹ to be a threshold above which N saturation (based on N leaching losses) occurred. At N deposition levels between 10–25 kg ha⁻¹ year⁻¹ N leaching occurred at some sites, while N saturation was not induced below 10 kg ha⁻¹ year⁻¹.

Although N saturation may not result in discernible ecological effects in the short-term, chronic N deposition can potentially cause a number of modifications in ecosystem processes, including: growth stimulation (Kenk & Fischer, 1988), induced nutrient deficiencies or imbalances (Boxman *et al.*, 1994; Schulze, 1989), interactions with other pollutants (e.g. O₃, CO₂; Gifford (1994); Melillo *et al.* (1993); Tjoelker & Luxmoore (1991)), direct phytotoxic or plant growth effects (e.g. from PAN, NO, NO₂, HNO₃), shifts in root/shoot ratios (Sullivan, 1993), effects on mycorrhizal diversity, sporophore formation and root development (Ter-morshuizen & Schaffers, 1991), altered drought stress and frost tolerance (Aber *et al.*, 1989), increased soil acidification (Falkengren-Grerup, 1989), vegetation community shifts (Van Breemen & Van Dijk, 1988), altered plant-pest relationships (Skeffington & Wilson, 1988), and NO₃⁻ contamination of groundwater and streamwater (Riggan *et al.*, 1985).

Nitrogen deposition and streamwater NO₃⁻ in chaparral watersheds in the San Gabriel Mountains

In two reports Riggan *et al.* (1985, 1994) reported elevated NO₃⁻ concentrations in streamwater emanating from chaparral watersheds exposed to high N deposition in the San Gabriel Mountains. Throughfall N

input to a *Ceanothus* chaparral canopy in the same study was $23.3 \text{ kg ha}^{-1} \text{ year}^{-1}$. It was estimated that the N deposition rate was 2.3 times higher than the N accretion rate of the *Ceanothus* stands there. Annual discharge rates in two chaparral watersheds over 4 years ranged from 0.04 to $10.0 \text{ kg N ha}^{-1} \text{ year}^{-1}$, and discharge rates for two grassland watersheds ranged from 0.11 to $19.4 \text{ kg ha}^{-1} \text{ year}^{-1}$. Streamwater NO_3^- concentrations in watersheds heavily influenced by urban air pollution were one to three orders of magnitude greater than in watersheds within and outside of the Los Angeles Basin which were not exposed to high concentrations of air pollution. Relatively low NO_3^- concentrations occurred during base flow in the summer, probably due to biological NO_3^- uptake within the aquatic system. Sharp NO_3^- concentration peaks occurred during the first winter storms when the ecosystem is loaded with N accumulated during the dry summer months. The NO_3^- yield in these watersheds was apparently limited by the amount of water percolating through the soil.

Wildfire was found to mobilize N accumulated from chronic N deposition in chaparral watersheds (Riggan *et al.*, 1994). Nitrogen concentrations as high as 69 mg liter^{-1} were found in streamwater from severely burned watersheds exposed to high levels of N deposition. The mechanisms for N mobilization by wildfire include soil erosion, nitrification in soil and sediment, and streamflow. However, high NO_3^- concentrations in streamwater from burned chaparral ecosystems is apparently not the norm. Burned and unburned chaparral watersheds in Arizona (Longstreth & Patten, 1975) and in relatively unpolluted areas in the Los Angeles Air Basin (Riggan *et al.*, 1985, 1994) did not exhibit high NO_3^- levels in streamflow. It was concluded from these studies that N deposition is a dominant factor in the N cycle of polluted chaparral watersheds, and is contributing to 'a developing N saturation or inability to sequester additional N' (Riggan *et al.*, 1994). Depletion of available C in soils is suggested as a possible factor inhibiting the capacity of the chaparral soils to sequester N, since low available C would limit the immobilization of mineral N by heterotrophic microorganisms. Streamwater NO_3^- may contribute to existing groundwater NO_3^- pollution, but judicious application of prescribed burning could mitigate the severity of NO_3^- contamination of streamflow in these chaparral ecosystems (Riggan *et al.*, 1994).

FUTURE MONITORING AND RESEARCH NEEDS

With increasing urbanization of California, especially of its southern and central regions, N deposition will likely continue to increase in overall magnitude and geographic extent. In the future, even larger forested areas, including National Parks and large acreages of the Sierra Nevada, will be increasingly affected by elevated concentrations of various air pollutants. The effects of elevated N deposition will possibly lead to interactions

with changing climatic conditions and other stresses occurring in the state, such as drought, air pollution, and forest pests and diseases (Smith *et al.*, 1994). For example, significant N $\times$ ozone interactions may occur, especially with ozone-sensitive species such as ponderosa pine. Low soil N availability has been shown to severely limit positive growth responses of vegetation to increased CO_2 (Curtis *et al.*, 1994). Greater N availability in forests with elevated N deposition is thus likely to increase the capacity of forests to respond to increased CO_2 concentrations until other factors become limiting (Sinclair, 1992). Following are specific recommendations for further research on the effects of N deposition in forests of the western US and for California in particular:

- In order to be able to accurately calculate or measure N deposition to various landscape forms (including forest stands) more research on mechanisms of dry and wet uptake of various pollutants is urgently needed. Most of the available literature is from the eastern United States or Europe where climatic conditions and air pollution composition are quite different from the generally arid conditions in the West. Very little is known of the deposition of HNO_3 vapor, which due to its high deposition velocity, may be a major N input to some forests.
- Ecosystem characteristics that are important for N retention need to be studied in order to develop and implement management practices that increase the capacity of forests to retain and utilize N and to reduce NO_3^- runoff in streamwater. Also the fate and processing of N pollutants within the forest N cycle needs further study.
- The long-term effects of N deposition on understory and overstory vegetation community composition and succession, mycorrhizal symbioses, and soil acidification are also important research issues for ecosystem protection.
- Further research is needed to quantify trace gas emissions from soil in sites with chronic N deposition and high soil NO_3^- concentrations, and under different management practices (harvesting, prescribed fires, etc.).
- Further testing and development of diagnostic indicators of N saturation are needed to determine the geographic extent of N saturation in California forests. More data correlating N inputs with the degree of N saturation would help define the N retention capacity of forests and the levels of N deposition at which N saturation is induced in forest ecosystems of California.
- Models simulating stand-level deposition which take into account the patchy and mountainous landscape and Mediterranean climate of California forests should be developed. The available models are not appropriate for California since they were developed for agricultural crops or the relatively uniform canopies of eastern US forests.

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