Nitrogen deposition on coastal watersheds in the Los Angeles region

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ABSTRACT - Many waterbodies throughout the Los Angeles region are eutrophied by nutrients such as nitrogen. While the loads of nitrogen from some sources (i.e., water reclamation plants) are well monitored, the load of total nitrogen from atmospheric dry deposition is almost completely uncharacterized. The goal of this study was to estimate nitrogen deposition to coastal watersheds in the Los Angeles Basin. An atmospheric transport and transformation model was used within the airshed for dry deposition flux of total nitrogen to the Ballona Creek, Dominguez Channel, Los Angeles River, Malibu Creek, San Gabriel River, Lower Santa Ana River (below Prado Dam), Upper Santa Ana River (above Prado Dam), and Santa Clara River watersheds. The estimated mean annual flux of total nitrogen from dry deposition varied from 21 to 71 g N/ha/day among watersheds. The greatest annual loads of total nitrogen were estimated for the Los Angeles River, San Gabriel River, and Upper Santa Ana River watersheds (5,100 to 8,400 tons N/yr). These are the largest watersheds in the region, contain the greatest densities of sources that contribute atmospheric pollutants, and are located in areas of deposition maxima near the airshed boundaries (i.e., along the foothills of the coastal mountain ranges).

INTRODUCTION

Atmospheric deposition of airborne pollutants has been recognized as a potentially significant source of contaminants to coastal waters and their watersheds in other regions of the country (Baker 1997). As much as 25% of the nitrogen load to the Chesapeake Bay may come from direct deposition from the atmosphere (NOAA 1987). Atmospheric nitrogen may be deposited in coastal watersheds through one of several mechanismsBaker (1997). In the Los Angeles Air Basin, the primary emissions of nitrogen oxides to the atmosphere consist mostly of nitric oxide (NO). Atmospheric photochemical reactions subsequently convert originally emitted NO into a family of nitrogen-containing pollutants, including nitrogen dioxide (NO_2), nitric acid (HNO_3), peroxyacetyl nitrate (PAN), and aerosol nitrates (PM NO_3). Eventually, these pollutants are removed by dry and wet deposition. Dry deposition appears to contribute more pollutants than wet deposition in the Los Angeles Basin, largely because there are so few storm events in the region (Lu *et al.* 2003).

In southern California, tremendous effort has been expended on monitoring air quality for human health concerns (i.e., inhalation of fine particulates and gaseous compounds), but virtually no effort has been expended on monitoring deposition of constituents for environmental health concerns. This lack of effort occurs despite the fact that the Los Angeles Basin has among the worst air quality in the nation for smog and smog precursors such as oxides of nitrogen (NO_x) and sulfur (SO_x). Moreover, greater than 100 stream reaches or estuaries in the Los Angeles region are on the State of California's list of impaired waerbodies (SWRCB 2000).

The goal of this study was to estimate the dry deposition of nitrogen to coastal watersheds in the Los Angeles Basin. To accomplish this goal, we first begin with an estimate of nitrogen emissions using the atmospheric emissions database developed by the South Coast Air Quality Management District (AQMD). Then a Surface Meteorology and Ozone Generation (SMOG) model was used to simulate atmospheric transport and transformation within the airshed. Finally, the SMOG model was used to estimate atmospheric deposition rates and predicted annual watershed loading for NO_2, HNO_3, NH_3, and PM NO_3. Model simulations are compared among

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summer and winter dry seasons. Ultimately, the model estimates of loading were compared to estimates from other watershed sources to determine whether atmospheric deposition is an important component of nitrogen loading in these watersheds.

METHODS

Emissions Inventory

The most recent inventory of gaseous and particulate emissions in Los Angeles Basin was compiled by AQMD for the time period 1998 (AQMD 2001). The inventory consisted of emission rates for NO_x, SO_x, carbon monoxide (CO), volatile organic compounds (VOCs), and particulate matter (PM) for on-road mobile sources, off-road mobile and area sources, and major point sources. The emission inventory for on-road mobile sources (including cars, trucks, buses, and motorcycles) was a product of the CARB EMFAC7G model, with emission factors determined for 1998. The distributed area and off-road sources were quite diverse and represent best estimates by the AQMD. Area sources included dry-cleaning establishments, gasoline stations, auto body shops, and metal plating operations; off-road sources included construction equipment, ships, and aircraft. Among the stationary point sources were a variety of industrial facilities and power plants. The emissions were spatially resolved on a 5 km x 5 km grid covering the South Coast Air Basin.

Temporal variations in the emission rates were characterized using detailed hourly, daily (over a week), and monthly profiles. In each source category, emission inventories for individual VOCs and aerosol components were obtained by applying the latest CARB VOC and PM speciation profiles to the total VOC and PM emissions for that category. The on-road mobile PM emissions for 1998 were assigned as follows: 40.6% from diesel exhaust, 9.1% from gasoline exhaust, and 50.3% from tire and brake wear, based on studies for Los Angeles County (*Xinqiu Zhang*, personal communication). The total PM mass emission was partitioned into four cumulative size fractions; i.e., for diameters <1 µm, <2.5 µm, <10 µm, and the total emission. Each chemical species was assigned a fraction of the cumulative mass at each size cut. Hence, the emissions for each species in the individual size bins (<1 µm, 1-2.5 µm, 2.5-10 µm, and >10 ?m diameter) were calculated by difference in the cumulative masses.

The AQMD emission database estimates roughly 1,000 mt of NO_x are released as a yearly mean, 24-h average input to the Los Angeles Basin (Table 1). Nearly half (54%) of the emissions arise from mobile sources. One third (34%) of the No_x emissions are attributable to area and off-road sources. Point sources have the smallest contribution (12%).

The SMOG Model

The SMOG model was developed by Lu *et al.* (1997a). It is a three-dimensional, integrated air pollution modeling system developed for studying air pollution on urban and regional scales. The system includes detailed treatments of meteorology, tracer transport and dispersion, chemical and aerosol microphysical transformations, and solar and terrestrial radiation transfer. The performance of the air pollution modeling system has been evaluated by comparing with the intensive measurement data collected during the Southern California Air Quality Study (SCAQS) (Lu *et al*., 1997b). The model has been used to study pollutant transport patterns in the Los Angeles Basin (Lu and Turco, 1995), analyze three-dimensional distributions of ozone and other pollutants and the formation of elevated pollution layers in the basin (Lu and Turco, 1996), and trace metal deposition in Santa Monica bay and watersheds (Lu *et al.*, 2003).

In the present simulations, the model domain covers the entire southern California area, with a configurable inner domain over the Los Angeles Basin used for the air quality calculations (Figure 1). The nominal uniform grid spacing is 0.050° longi-

Table 1. Estimated cumulative South Coast Air Basin emission inventory for three different source categories in 1998 (emission rates in metric tons/day).

Source	On-Road Mobile	Off-Road and Area	Point	TOTAL
CO	3,427	1,440	142	5,008
NO_x	588	365	134	1,088
So_x	14	43	23	80
VOC	448	1,020	218	1,687
PM	26	1,392	23	1,441
TOTAL	4,503	4,261	540	9,304

tude x 0.045 ◦ latitude (roughly 4.6 x 5 km), and applies to both the outer and nested grids. In the vertical direction, variable grid spacing is used with higher resolution in the boundary layer. Meteorological parameters, including wind vectors, temperature, pressure, and relative humidity are predicted over the larger domain (with 85 x 55 x 20 grid cells).

Seasonally averaged meteorological conditions to simulate nitrogen dispersion and deposition across the Los Angeles Basin were utilized. Composite upper air soundings for these conditions were obtained from three meteorological stations around the South Coast Air Basin; DRA (Desert Rock, NV), KNX (Miramar, San Diego, CA), and VBG (Vandenburg Air Force Base, Lompoc, CA). Each of these soundings was averaged over two periods—from July 1 to August 31, 1998, and from November 1, 1998 to January 31, 1999—to represent mean summer and winter conditions, respectively. These soundings were used to initialize summer and winter simulations characteristic of the region.

In order to calculate the nitrogen deposition fluxes onto watersheds in the Los Angeles Basin, aerosol sizes were resolved into 12 diameter bins covering the range from 0.01 µm to 40 µm (Table 2). For the present work, the aerosol was characterized by one internally mixed particle type having 34 components, with each component defined by its volume fraction in each size class. The main components included nitrate, sulfate, sea salt, organic and elemental carbon, as well as trace metals (Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn), and water. The fixed size bins corresponded to the equivalent "dry" aerosol, consisting of all of the volatile and nonvolatile components except water. In practice, the model predicts the concentration and composition of the particles in each size bin, at every spatial cell in the three-dimensional regional domain. The water content of the particles in a given size/space bin therefore changed with ambient relative humidity. The corresponding "wet" aerosol sizes were used to calculate coagulation, sedimentation, and deposition rates.

Using the SMOG model, dry deposition fluxes of gases and particles from the atmosphere to the surface were assumed to be proportional to the pollutant concentration and deposition velocity at a reference height of approximately 20 m above the surface. This height is the middle of the lowest model layer in this study. The physical model used to calculate dry deposition velocities was based on an "electrical resistance" analog, where the "voltage" is the vapor concentration and the "current" is the molecular flux across the region of interest. The deposition velocity for gases at the reference height then was defined as the inverse of the total resistance to pollutant deposition, which includes: (1) turbulent transport through the overlying atmosphere to the laminar viscous surface layer; (2) the resistance related to molecular scale diffusive transport through the viscous surface sub-layer; and (3) the resistance associated with the uptake of the pollutant gas molecules at the surface. The surface resistance was a function of surface type and the reactivity of the depositing vapor with the surface material, and may be expressed as an "accommodation" coefficient. For example, the surface resistance for HNO_3 was essentially zero whereas surface resistance for NO is fairly large, so that the direct deposition of NO is limited. The turbulent resistance was controlled by atmospheric stability and calculated based on the similarity theory. The resistance of the viscous sub-layer depends on the dimensionless Schmidt number, *Sc* (Wesely *et al.*, 1977), which is a function of kinematic viscosity of air and the molecular diffusivity in air.

In the case of particle dry deposition, the effects of Brownian diffusion, inertial impaction, and particle gravitational sedimentation were accounted for in addition to turbulent transport

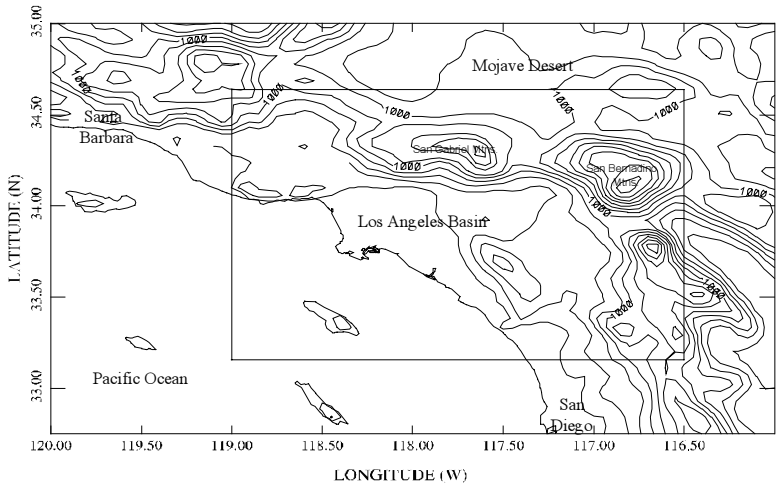

Figure 1. Model simulation domains. The outer domain is for meteorological predictions and the inner domain is used for the calculation of air chemistry and deposition.

Table 2. Discrete aerosol size bins used in the simulations.

Bin No.	Mean Diameter (µm)	Lower Boundary (µm)	Upper Boundary (µm)
1	0.017	0.010	0.020
2	0.033	0.020	0.040
3	0.066	0.040	0.080
4	0.131	0.080	0.158
5	0.261	0.158	0.316
6	0.520	0.316	0.630
7	1.038	0.630	1.257
8	2.069	1.257	2.506
9	4.126	2.506	4.997
10	8.228	4.997	9.964
11	16.41	9.964	19.871
12	32.72	19.871	39.625

and surface accommodation. If particles adhere to the surface upon contact without resuspension, the dry deposition velocity is related to particle gravitational settling velocity, which varies by particle size. Over ocean surfaces, particle dry deposition velocities were calculated using the model of Williams (1982). This model takes into account the effects of wave breaking and spray formation in high winds, and particle growth in the humid sub-layer near the air/water interface.

Model Application

The SMOG model was applied to determine the flux of nitrogen in g N/ha/day as NO, NO_2, HNO_3, NH_3, and PM NO_3 in each of eight coastal watersheds in the Los Angeles Basin. The total nitrogen flux was calculated as the sum of the five nitrogen species. The model simulates both a summer and winter condition; seasonal mean flux represented the average of deposition for all surface layer grid cells within the watershed boundary. Annual mean flux was calculated as the average of the summer and winter condition. Loading of total nitrogen to each of the watersheds was calculated similarly, except a time constant was applied to achieve mass. The eight watersheds included Ballona Creek, Dominguez Channel, Los Angeles River, Malibu Creek, San Gabriel River, lower Santa Ana River, upper Santa Ana River, and the Santa Clara River (Table 3, Figure 2). These watersheds were chosen based on three equally weighted criteria: (1) environmental concerns regarding nutrient or nutrient-related impacts within the watershed; (2) density of potential pollutant sources within the watershed; and (3) size and location of the watershed relative to potential sources and prevailing surface flow patterns. In all three cases, a range of conditions was preferable across the watersheds of interest.

The results from the model application was examined in four fashions. First, the mean annual flux of total nitrogen among the eight watersheds of interest were compared. Second, a similar comparison of total nitrogen flux was conducted, but this time using the summer and winter conditions to determine whether seasonality was a factor in loading. Third, the mean annual flux was compared among the major nitrogen species responsible for deposition. Finally, the annual loading of total nitrogen from dry deposition was compared among watersheds.

Results

The Los Angeles River had the greatest estimated mean annual dry deposition flux of nitrogen (71 g N/ha/day) compared to the other seven watersheds examined in the coastal Los Angeles Basin (Table 4). The San Gabriel River, Ballona Creek, and Santa Ana River watersheds had similar, but slightly lower mean annual nitrogen dry deposition flux rates compared to the Los Angeles River watershed. The estimated dry deposition flux rates ranged from 60 to 69 g N/ha/day among these watersheds. The Dominguez Channel watershed had moderate dry deposition flux rates compared to the other watersheds examined (ca. 44 g N/ha/day). The Malibu Creek and Santa Clara River watersheds had the lowest dry deposition flux rates of all the watersheds

Table 3. Watershed sizes used for modeling nitrogen dry deposition in the Los Angeles Basin.

Watersheds	Area (ha)
Ballona Creek	33,762
Dominguez Channel	30,906
Los Angeles River	215,602
Malibu Creek	28,407
San Gabriel River	186,201
Santa Ana River (above Prado Dam)	384,132
Santa Ana River (below Prado Dam)	49,215
Santa Clara River	266,067

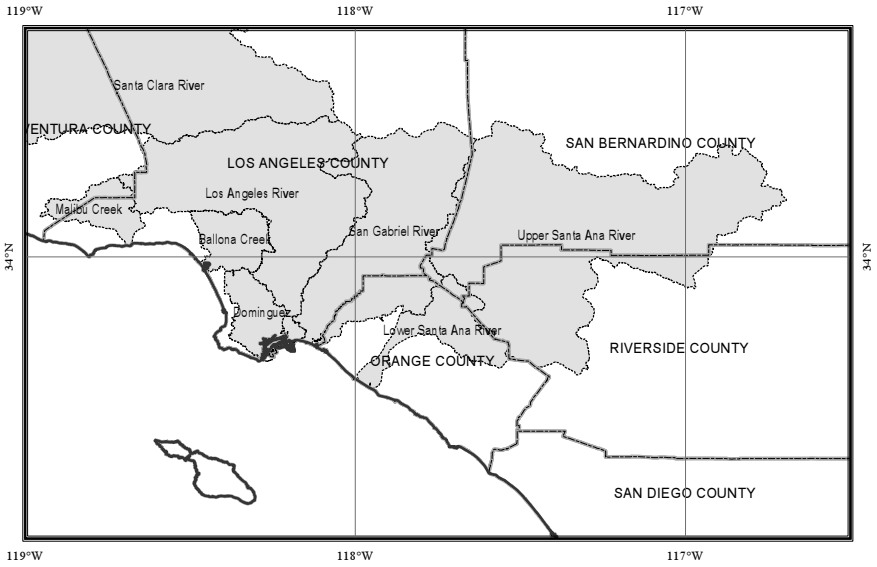

Figure 2. Locations of major watersheds examined in this study.

examined. The estimated dry deposition of nitrogen ranged from 21 to 22 g N/ha/day, respectively. The flux of total nitrogen is generally greater in the summer than in the winter (Table 4). Six of eight watersheds had greater estimated dry deposition of nitrogen in summer than in winter. On average, summer deposition flux rates of total nitrogen were 48% higher than winter flux rates. The watershed with the greatest differential between summer and winter deposition flux rates of total nitrogen was the Santa Clara River watershed. The only two watersheds with decreasing flux rates of total nitrogen from winter to summer was the Ballona Creek and Dominguez Channel watersheds.

The flux of nitrogen deposition in coastal watersheds was dominated by three nitrogen species (Table 5). Altogether, these three species comprised greater than 99% of the estimated total nitrogen deposition flux. The dry deposition of NO_2 averaged 34.7% of the total nitrogen deposited among all watersheds, comprising the major species in the Los Angeles River, San Gabriel River, and lower Santa Ana River watersheds. The dry deposition of NH_3 averaged 33.5% of the total nitrogen among all watersheds, comprising the major species in the Ballona Creek, Dominguez Channel, Malibu, and Santa Clara watersheds. The dry deposition of HNO_3 averaged 31.3% of the total nitrogen deposition among all watersheds, comprising the major species in upper Santa Ana River watershed. The flux of NO_3^- and NH_4^+ represented less than 0.1% of the total nitrogen deposited to the coastal watersheds in the Los Angeles Basin.

Approximately 23,400 mt N/yr are deposited to the eight coastal watersheds examined in this study (Table 5). The range of loading varied from approximately 500 to 8,400 mt N/year among watersheds. The greatest loading was in the upper Santa Ana River watershed (36.1% of total nitrogen load). The Los Angeles River and San Gabriel River watersheds comprised 23.8% and 20.2% of the total nitrogen load, respectively. Cumulatively, the Ballona Creek, Dominguez Channel, Malibu Creek, lower Santa Ana River, and Santa Clara watersheds comprised less than 20% of the total nitrogen load. The dry deposition in the Los Angeles Basin was a reflection of the flux rates observed for the various species; deposition was dominated by HNO_3, NO_2, and NH_3, while PM NO_3 and NH_4^+ was inconsequentially small.

Table 4. Seasonal and annual mean dry deposition flux of total nitrogen to selected coastal watersheds in the Los Angeles Basin.

	Mean Flux (g N/ha/day)		
Watersheds	Summer	Winter	Annual
Ballona Creek	61.6	75.6	68.6
Dominguez Channel	37.5	50.4	44.0
Los Angeles River	87.2	54.1	70.6
Malibu Creek	22.8	19.6	21.2
San Gabriel River	85.9	52.8	69.3
Santa Ana River (above Prado Dam	76.8	43.6	60.2
Santa Ana River (below Prado Dam	65.8	39.0	52.4
Santa Clara River	31.6	12.9	22.3

Table 5. Estimated annual dry deposition of total nitrogen and loading among nitrogen species in eight watersheds of the Los Angeles Basin.

Watersheds	Mean Flux (percent of total load)					Annual Loading (metric tons)
	HNO_3	NO_2	NO_3^-	NH_4^+	NH_3	
Ballona Creek	12.7	43.1	0.3	0.3	43.6	845.2
Dominguez Channel	12.4	37.2	0.3	0.4	49.6	495.8
Los Angeles River	28.8	40.4	0.2	0.2	30.4	5559.2
Malibu Creek	19.6	26.5	0.2	0.1	53.6	219.9
San Gabriel River	30.7	38.6	0.2	0.2	30.2	4711.3
Santa Ana River (above Prado	35.4	33.2	0.3	0.2	30.9	8446.4
Santa Ana River (below Prado	35.2	35.8	0.3	0.3	28.4	941.4
Santa Clara River	34.7	13.9	0.1	0	51.3	2160.0

Discussion

Dry deposition of nitrogen in the coastal watersheds of the Los Angeles Basin is partly a function of meteorology. The local climate in southern California is controlled primarily by large-scale pressure systems, especially the semi-permanent high over the North Pacific Ocean (DeMarrais *et al.* 1965). Crucial to the dispersion of air pollution in this region are thermally forced winds, including sea-land breezes and mountain-valley flows, as well as winds channeled by terrain (Lu and Turco 1994, 1995; Wakimoto and McElroy 1986). The predominant daytime wind flow observed in the summer is the southwesterly sea breeze that carries pollutants from major sources in western Los Angeles toward the San Fernando Valley, into the eastern basin, and through mountain passes. Onshore flow acts to improve air quality in coastal and near-offshore regions. By contrast, nocturnal summer winds become light and unorganized across the basin and at the coast. In the winter, the daytime sea breeze has a flow pattern similar to that in the summer, but with a much weaker intensity. The nighttime winter winds have a stronger easterly and northeasterly offshore component due to drainage from the local mountains. The down-slope flow is driven by radiative cooling of land surfaces during the long winter night, bringing potentially higher air concentrations to the smaller coastal watersheds.

Pollutant transport and transformation provides the second key to understanding dry deposition of nitrogen in the air basin. Most primary pollutants, such as NO, are emitted in the western Los Angeles Basin. As pollutants are transported inland on strong sea breezes, NO is converted to NO_2 and then oxidized to HNO_3 by photochemical reactions. Because of its relative short lifetime during the day, NO_2 tends to deposit closer to its pollutant source region and is confined largely in the air basin with sharp gradient near the boundaries (Figure 3). Deposition is bounded therefore by surrounding mountain barriers with large deposition on the flanks and valleys facing the polluted urban area. As a result, deposition maxima are found near the southern slope of the San Gabriel Mountain range. The gradients appear less steep during the winter months when the sea breezes are lighter.

Significant amounts of HNO_3 can be found in photochemically aged air because it has much longer lifetime compared to reactive nitrogen oxides. The primary sink of HNO_3 is the dry and wet deposition at the surface. Since HNO_3 has very low surface resistance to enhance deposition, it has much larger impact on regions where severe photochemical air pollution occurs. The deposition of HNO_3, therefore, extends from the basin further inland to the high desert region as pollutants are transported through mountain passes (Figure 4). Smaller HNO_3 deposition rates are found in the winter when the solar radiation, and therefore photochemistry, is reduced.

Ammonium nitrate aerosols are formed in photochemically aged air. However, PM ammonium and nitrate tend to stay in the accumulation mode between 0.2 to 1 μm in diameter. The deposition velocities are small because effective deposition in this size range is limited. For example, the deposition fluxes of PM nitrates are significantly smaller than the gas phase deposition rates of NO_2 and HNO_3 (Figures 5 and 6). Near the coast, deposition rates increase slightly as the result of aerosol growth with high relative humidity.

The atmospheric transport and transformation of nitrogen species, coupled with the local meteorology,

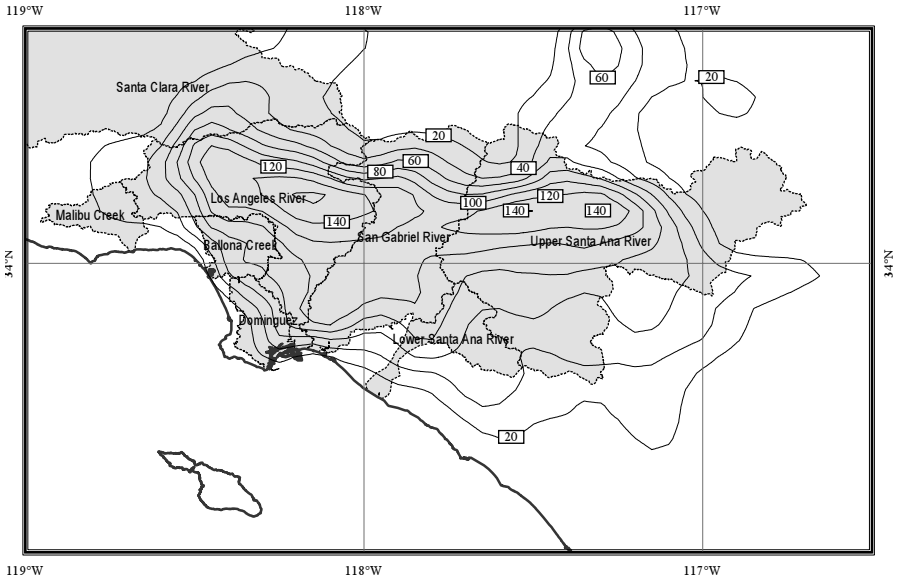

Figure 3. Deposition rates of nitrogen dioxide (NO_2) for the summer case. Contour interval is 20 grams hectare^{-1} day^{-1}.

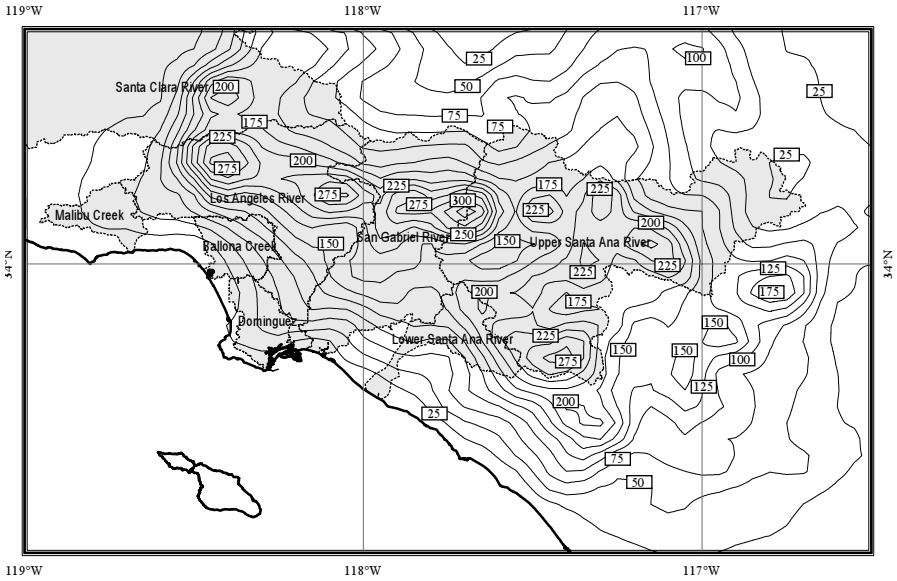

Figure 4. Deposition rates of nitric acid (HNO_3) for the summer case. Contour interval is 25 grams hectare^{-1} day^{-1}.

explain the differences in dry deposition among the watersheds examined in this study. At one end of the spectrum are the smaller watersheds, such as Malibu Creek, Ballona Creek and Dominguez Channel. Because of their reduced size, total nitrogen loadings via dry deposition are much smaller compared to larger watersheds. However, this also reflects their proximity to the coast, which is in the opposite direction of surface flows and upwind of many pollutant sources. Instead, these watersheds are exposed to ocean air masses, which are generally lower in pollutant concentrations. Larger watersheds, such as the Los Angeles River, San Gabriel River, and Santa Ana River, not only have more surface area to receive dry deposition of pollutants, but many of the sources reside within their watershed boundaries. A cumulative factor that increases the dry deposition in many of these watersheds is that their headwaters extend to the borders of the air basin, where atmospheric transport generates air basin dry deposition maxima. One good example is the Los Angeles River watershed, which contains a great density of atmospheric pollutant sources (e.g., downtown Los Angeles) and has watershed boundaries that extend into the San Fernando Valley and surrounding foothills.

The two major mechanisms of dry atmospheric deposition are direct deposition (deposited directly onto a water surface) and indirect deposition (deposited onto surrounding terrestrial surfaces in the watershed). Direct deposition is a very small proportion of the total atmospheric input in southern California because water surfaces represent <0.1% of the total watershed acreage; most rivers are only a couple meters wide and lakes are generally few. As a result, direct dry deposition appears to be a relative minor source of nitrogen compared to other direct sources including water reclamation plants (WRPs; Table 6). The much larger fraction of nitrogen loading by indirect deposition occurs as nitrogen is washed off the watershed during subsequent storm events. However, the actual load attributable to indirect deposition by wash-off is unknown because the fraction of deposited nitrogen that is consumed by terrestrial plants, transformed within the soils by bacteria, and abiotically degraded remains unquantified.

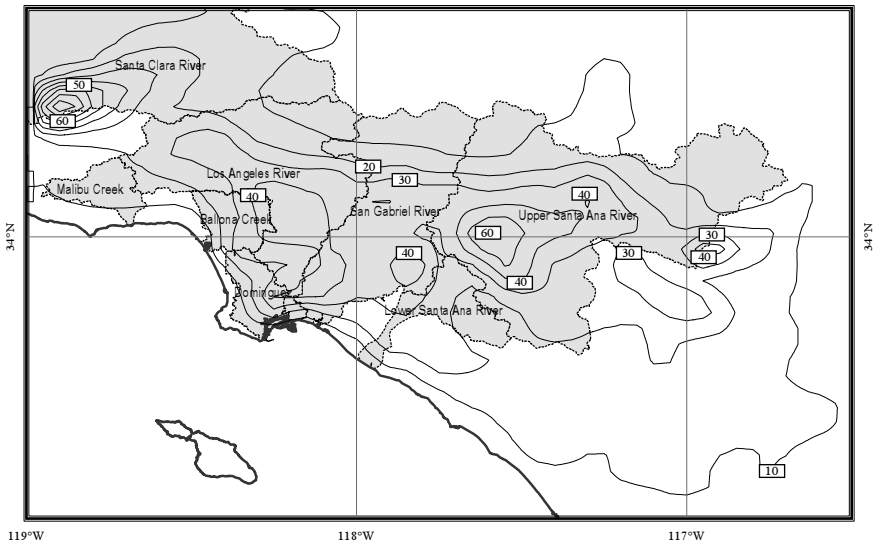

Figure 5. Deposition rates of ammonia (NH_3) for the summer case. Contour interval is 10 grams hectare^{-1} day^{-1}.

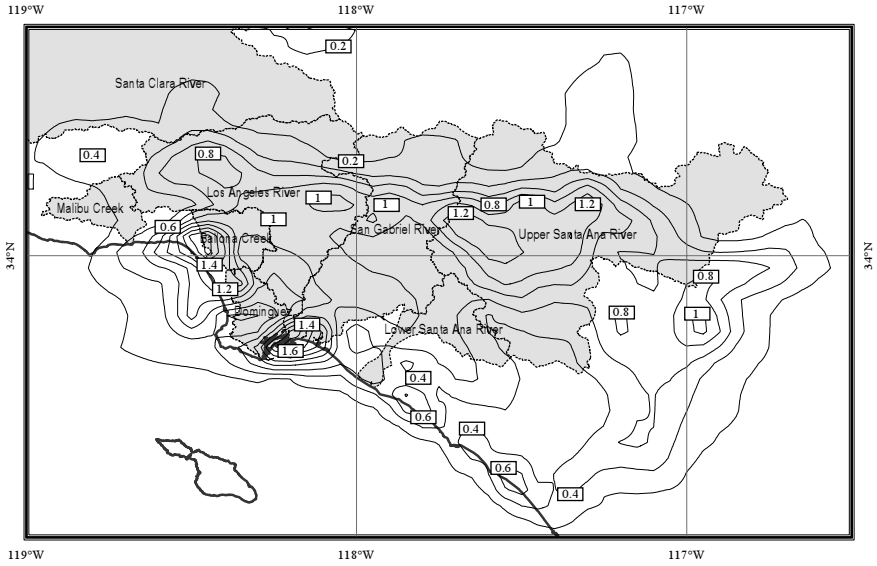

Figure 6. Deposition rates of PM nitrate (NO_3^-) for the summer case. Contour interval is 0.2 grams hectare^{-1} day^{-1}.

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Table 6. Comparison of total nitrogen loading by reclamation plants (WRPs) and direct dry deposition to water surfaces in coastal rivers and creeks of the Los Angeles Basin.

	Annual Load (mt/yr)	
Watersheds	Water Reclamation Plants	Direct Dry Atmospheric Deposition
Los Angeles River	1,042	0.2
Malibu Creek	142[b]	0.1
San Gabriel River	2,380	1.4

[b]Includes effluent and effluent irrigation loads.