

CHAPTER FOUR

A California Nitrogen Mass Balance for 2005

Appendix 4.2 Mass balance methods and data sources

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This is an appendix to Chapter 4 of *The California Nitrogen Assessment: Challenges and Solutions for People, Agriculture, and the Environment*. Additional information about the California Nitrogen Assessment (CNA) and appendices for other chapters are available at the Agricultural Sustainability Institute website: asi.ucdavis.edu/nitrogen

Suggested citation:

D Liptzin, RA Dahlgren, and T Harter. "Appendix 4.2: Mass balance methods and data sources for Chapter 4: A California Nitrogen Mass Balance for 2005." Online appendices for California Nitrogen Assessment: Challenges and Solutions for People, Agriculture, and the Environment. TP Tomich, SB Brodt, RA Dahlgren, and KM Scow, eds. Agricultural Sustainability Institute at UC Davis. (2016). asi.ucdavis.edu/nitrogen.

4.2 Mass Balance Calculations and Data Sources

The *imports* of new reactive nitrogen (N) for the statewide mass balance were fossil fuel combustion, biological N fixation, synthetic N fixation, agricultural feed, and fiber. The *exports* were gas/particle exports in the atmosphere, food exports, discharge of rivers to the ocean, and discharge of sewage to the ocean. *Storage terms* include soils and vegetation, reservoirs, landfills, and groundwater. We assumed no storage in the atmosphere. In addition to the calculations at the statewide level, mass balances were calculated for various *subsystems* within California: natural land, cropland, urban land, livestock, households, surface waters, groundwater, and the atmosphere. In most cases, the flows in the subsystems could be estimated with one or more independent approaches, but some flows could only be estimated by differences (e.g., groundwater in cropland).

For the calculations of flows in the three land-based subsystems, California was classified into four main land cover classes: natural land, cropland, urban land, and water. An updated version of the California Augmented Multisource Landcover (CAML) map was produced by the Information Center for the Environment at the University of California – Davis (ICE 2006). The base map layer of CAML was the 2002 Multi-Source Land Cover dataset produced by the California Department of Forestry and Fire Protection (FRAP). This layer was the source for the type of ecosystem vegetation in all of the natural land and also delineated surface waters. For biome level estimates, the FRAP vegetation types were lumped into biomes based on the California WHR13 classes: barren, desert (desert shrub and desert woodland), forest (hardwood and conifer), herbaceous, shrub, woodland (hardwood and conifer), water, and wetland. The agricultural land was further subdivided into individual crops based on the class and subclass of the polygons in the most current digitized county maps produced by the California Department of Water Resources (DWR). For counties without digitized DWR maps, agricultural land was identified based on the categories in the FRAP base layer, supplemented with crop information from pesticide use reports produced by the California Department of Pesticide Regulation. Urban areas were identified by combining the urban boundaries indicated in the California Department of Conservation Farmland Mapping Program and urban land-use types in the 2001 USDA National Land Cover Dataset. The water pixels in CAML were divided into lakes, reservoirs, and rivers in two ways: (1) areas identified as riverine and estuarine wildlife habitats were categorized as rivers, while lacustrine wildlife habitats were categorized as lakes, and (2) in pixels identified only as water, the spatial location of the pixel was compared to the USGS National Hydrography Dataset (USDI 2013); if the pixel matched a lake or reservoir, the pixel

was designated a lake or reservoir, otherwise the water pixel was considered a river. The final map was produced at a 50 m resolution.

4.2.1 Fossil Fuel Combustion

Fossil fuel combustion produces NO_x , NH_3 , and N_2O as incidental by-products which are tracked and regulated for different reasons. Nitrogen oxides are considered a criteria pollutant and all of the anthropogenic sources of NO_x included in the statewide inventory conducted by the California Air Resources Board (ARB) and the US EPA were considered emissions. The emissions from the 2002 EPA inventory (EPA 2007) were used for the calculations because that dataset was the basis for the N deposition model described below. Ammonia is an unregulated pollutant, but it has become part of the criteria pollutant monitoring program because of its role in forming secondary fine particulate matter ($\text{PM}_{2.5}$) in the atmosphere as either ammonium nitrate or ammonium sulfate. As with NO_x , the 2002 EPA dataset was used to estimate NH_3 emissions; however, only categories related to fossil fuel combustion (fuel combustion, highway vehicles, and off-highway vehicles) were included. Finally, N_2O emissions are not yet regulated, but are estimated as part of greenhouse gas inventories by both ARB and the EPA. All “included” fossil fuel combustion sources from the ARB inventory, regardless of sector, were used to calculate fossil fuel related N_2O emissions and an average for 2002-2007 was calculated.

While not necessarily exclusively from fossil fuel combustion, there is import of reactive N to the atmosphere above California from outside the boundaries of the study area. Some of this N will be transmitted completely through the state and this fraction will be ignored. However, we estimated the import of this reactive N by assuming that the offshore N deposition rate would occur across the entire state of California in the absence of any emissions from California. Based on the atmospheric deposition rates generated by the Community Multiscale Air Quality (CMAQ) model in areas off the coast of California as modeled by Tonnesen et al. (2007), the current offshore deposition rate is $1 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, split evenly between oxidized and reduced N.

4.2.2 Atmospheric Deposition

Atmospheric deposition was based on the results of Fenn et al. (2010). Their Geographic Information System (GIS) map layer uses output from the CMAQ model based on 2002 emissions data. The CMAQ model results for most of the state were available from Tonnesen et al. (2007) at a resolution of 4 km, but for northern and southeastern California only the 36 km CMAQ output from the EPA was used to create a statewide map. In certain biomes, based on the

availability of field measurements, the model output was replaced by measured deposition data. Total N deposition was partitioned statewide on the various land-use types (natural land, cropland, urban land) based on the land-cover map. However, as the composite statewide map in Fenn et al. (2010) only provided total N, the ratios of oxidized to reduced and wet to dry N deposition were calculated based on the area modeled by Tonnesen et al. (2007).

We assumed that storage was not possible in the atmosphere. Therefore, the export of NO_x and NH_3 was calculated as the difference between all inputs and N deposition. By the time the export from California occurs, secondary reactions will have occurred in the atmosphere such that NO_y (NO_x plus its oxidization products like HNO_3 or organic nitrates) and NH_x (NH_3 plus the NH_4^+) better describe the forms of N. We assumed that all of the emitted N_2 and N_2O were exported from the study area.

4.2.3 Biological N Fixation

Biological N fixation is also discussed in Chapter 3. A variety of field measurements of biological N fixation have been used including ^{15}N isotope methods, acetylene reduction, N accretion, and N difference, which vary in their assumptions and limitations.

4.2.3.1 Natural Land N Fixation

Based on the USDA Plants database (USDA 2013), a total of 56 native and 34 non-native, non-crop species are known to be symbiotic N fixers on natural land in California. However, field measurements of rates and the relative abundances for most of these species are poorly known. Therefore, we used three approaches, based on Cleveland et al. (1999), to estimate biological N fixation in natural land. First, the biome areas calculated from the land-use map were multiplied by the biome-specific N fixation rates compiled in this global synthesis of published rates. A range in values was estimated using the biome-specific low, medium, and high percent cover abundance of the N fixing species. Second, Cleveland et al. (1999) developed an empirical linear relationship between biome-specific modeled values of actual evapotranspiration (ET) and N fixation rates. The mean modeled statewide ET (provided by Q. Mu, University of Montana) from 2001 (33.6 cm yr^{-1}) was used because it was the only year when precipitation, modeled ET, and cropland irrigation rates were available for the entire state. Third, we used a mass balance approach. That is, we estimated all of the other N flows in and out of natural land, assumed steady-state conditions (i.e. no change in N storage) and calculated N fixation by difference.

4.2.3.2 Cropland N Fixation

Cropland N fixation rates were based on published species specific rates and harvested acreages. The most comprehensive analysis of legume N fixation rates is a meta-analysis for Australia described in Unkovich et al. (2010). These authors found highly variable rates, but a strong positive relationship, between fixed N in aboveground tissues and productivity. This may help explain, in part, the high variability in the published rates. The only crop included in this analysis that is grown on a significant acreage in California was alfalfa where the empirical relationship was $\text{aboveground fixed N (kg ha}^{-1}\text{)} = 18.2 \times \text{Production (Mg ha}^{-1}\text{)} + 0.13$. The rates for the other leguminous crops grown in California (dry beans, dry and fresh lima beans, snap beans, and clover), but not included in the analysis, were based on Smil (1999). We also include the fixation rates for rice paddies reported by Smil (1999) associated with the cyanobacteria symbiotically associated with aquatic ferns in the genus *Azolla*. Crop acreages for all legumes except clover were calculated as the 2002-2007 average of the annual harvested acreages reported in the statewide database of California Agricultural Commissioners' reports (USDA NASS 2013). Clover used to be planted widely in irrigated pastures, but now is estimated to compose only 10% of the cover in these systems (M. George, personal communication). The acreage of irrigated pasture was calculated as the average of the 2002 and 2007 Agricultural Census acreage for irrigated pasture (see Table 10 in USDA 2004 and 2009)

4.2.4 Synthetic N Fixation

Synthetic N fixation is largely the result of the Haber-Bosch process, although a small amount of ammonium sulfate is still produced as a by-product from coke oven gas during steelmaking (Kramer 2004). This industrial process converts atmospheric N_2 to NH_3 at high temperature and pressure with natural gas being the source of hydrogen and energy. National estimates of fixed N are annually compiled by the United States Geological Survey including national production, imports and exports. Fixed NH_3 is the feedstock for essentially all synthetic N fertilizers as well as a variety of industrial N-containing chemicals and explosives (Kramer 2004). Less than 2% of the national explosives use occurs in California because of the limited amount of mining (USDI 2000). Ammonium nitrate/fuel oil mixtures are the dominant form of explosives, but we assumed that the N emissions from their use was N_2 gas. Therefore explosives were not considered as part of the budget.

4.2.4.1 Non-Fertilizer Synthetic Chemicals

Non-fertilizer use of some individual compounds can be tracked, but as a whole it is typically calculated as the difference between total NH₃ fixation and fertilizer use. Other common non-fertilizer uses include synthetic chemicals, such as melamine, nylon, plastics (e.g., acrylonitrile butadiene styrene), and polyurethane (Table A4.2.1). Several estimates of synthetic N consumption are available, but the Kramer (2004) estimate was used because it breaks down the non-fertilizer N consumption most completely (Table A4.2.2). The national total for non-fertilizer consumption of N was 1,722 Gg N yr⁻¹ (Kramer 2004). Excluding synthetic N for explosives, 567 Gg N yr⁻¹ of non-fertilizer N was consumed nationally in 2002 (Kramer 2004). We scaled the national estimate to California based on the mean 2002-2007 population of California (35.6 million) and the United States (295 million) from the United States Census Bureau (2013). We used the United States Census as opposed to the California Department of Finance population estimate in order to make the most consistent estimate of California's proportion of the United States population. Most of these synthetic forms of N are assumed to be long-lasting chemicals, which become part of infrastructure and household items and eventually are disposed of in landfills (Table A4.2.1). One chemical class that is poorly tracked is N-containing compounds found in many common household products, such as surfactants and detergents that end up as part of the wastewater stream.

TABLE A4.2.1. Major Non-Fertilizer Uses of Synthetic Nitrogen in the United States. Source: Domene and Ayres 2001.

Compound	N (Gg yr ⁻¹)	End use
Acrylonitrile	173	Acrylonitrile Butadiene Styrene
Caprolactam	86	Nylon
Hexamethylenediamine	203	Nylon
Isocyanates	90	Polyurethane
Melamine	54	Laminates and surface coatings
Urea	180	Resins
Adipic Acid ¹	185	Nylon Manufacturing
Methyl methacrylate ²	102	Acrylic glass manufacturing

¹NO_x, N₂O, and N₂ emissions from the reduction of nitric acid are a byproduct of adipic acid synthesis, but nitrogen is not a component of the product.

²Ammonium sulfate, typically used as fertilizer, is produced as a by-product of methyl methacrylate synthesis.

TABLE A4.2.2. Synthetic Nitrogen Consumption (Gg N yr⁻¹) in the United States. Where possible, non-fertilizer consumption was partitioned into explosives, plastics and synthetics, and other uses.

Source	Year	Fertilizer	Non-fertilizer	Explosives	Plastics and synthetics	Other
Kramer 2004	2002	11,636	1,565	998	491	76
UN FAO 2010	2002	10,945	4,277			
Domene and Ayres 2001	1996	11,297	3,020	557	786	1677

4.2.4.2 Synthetic Fertilizer

Fertilizer sales, not necessarily fertilizer use, have been reported annually since the 1950s in the tonnage reports of the California Department of Food and Agriculture. These data are identical to the California data compiled by The Fertilizer Institute as part of their national survey. To prevent duplication, reporting of sales is supposed to occur when a licensed fertilizer dealer sells fertilizer to an unlicensed purchaser. The data are collected as tonnage of fertilizing materials and are converted to tons of nutrients based on the reported fertilizer grade. Fertilizer use was assumed to be on average equivalent to fertilizer sales at the state level. Because of uncertainty in these data starting in 2002, we used the average synthetic fertilizer sales for 1997-2001.

Synthetic fertilizer use was first partitioned between agricultural and urban (i.e. turfgrass) use based on data provided by the Scotts Miracle-Gro Company. Annually, an estimated 2.7 million tons of fertilizer is applied nationally to turfgrass. It is divided equally between homeowner use, commercial application to home lawns, and golf courses/athletic fields. This fertilizer tonnage was converted to N tonnage based on the typical N grade of lawn fertilizer (29%) based on the popular Scotts Turf Builder product. The national estimate was scaled down to California using remote-sensing based estimates of turfgrass acreage. California contains 11,159 km² of turfgrass, or 6.8% of the total national turfgrass acreage (Milesi et al. 2005). The Scotts Company was willing to share their sales figures for the state and reported sales of 4 Mg N sold in 2005 for the do-it-yourself homeowners market. Their research suggests that they supply approximately half of the do-it-yourself homeowners market.

Synthetic fertilizer use for cropland was calculated separately for ornamental horticulture and other crops. The amount used for environmental horticulture was based on the acreage of open grown commodities in the USDA Census of Agriculture, an annual irrigation rate of 2 m water yr⁻¹, and a N concentration of 100 ppm N assuming no recycling of N in irrigation water

(R. Evans, personal communication). Sod farms were assumed to use 400 kg N ha⁻¹ (R. Green, personal communication).

Synthetic fertilizer use on other crops was calculated by subtracting urban and environmental horticulture use from the total sales. Fertilizer use can also be validated based on crop-specific recommendations. Current (since 1999) fertilization rates by crop were extracted from UC Davis cost studies and the USDA Chemical Use Surveys and the two data sources were averaged (see Chapter 3 for further details on data). The fertilization rates were combined with the crop-specific acreages reported in the statewide Agricultural Commissioners dataset to calculate a total fertilizer recommendation that could be met with synthetic fertilizer or manure. Any difference between the calculated fertilizer use and the synthetic fertilizer sales for these crops indicates fertilization needs met by manure.

4.2.5 Agricultural Production and Consumption: Food, Feed, and Fiber

The production and consumption of food, feed, and fiber involve the majority of N flows in California. The N tonnage of all agricultural products, with the exception of wood products and ornamental horticulture, was calculated from production data compiled by the county Agricultural Commissioners (USDA NASS 2013). The 253 crop commodities in the database were consolidated into 121 classes based on similar characteristics. The 2002-2007 average N tonnage was calculated by matching each crop class to the most similar crop in the USDA Crop Nutrient Tool (USDA 2013). This database, which is the most comprehensive source of its kind, is a compilation of the nutritional content of crops from a variety of published sources, but most of the sources are at least several decades old. The only commodity not present in the database was olives whose nutritional information was based on the 2009 USDA National Nutrient Database for Standard Reference (USDA ARS, n.d.). Commodity boards in the state were contacted to determine if they had more recent and California-specific data, but only the Almond Board of California provided information. The following crop classes were considered feed crops: alfalfa hay, almond hulls, grain and silage corn, cottonseed, non-alfalfa haylage, small grain hay, grain and silage sorghum, tame hay, and wild hay.

Consumption of agricultural products was based on the population of humans, household pets, and livestock in the state. The average population of California during the period 2002-2007 was 35.6 million. The consumption of food was calculated in two ways. First, on average from 2002-2007, the national per capita food availability was 6.5 kg N yr⁻¹ (USDA ERS 2013). Second, per capita N consumption varies globally, but in the United States, 5.0 kg N yr⁻¹ is typical (Boyer et al. 2002). The waste of food by retailers, food service, and consumers has been

estimated at 27%. Combining food waste with food consumption leads to a per capita demand of 6.4 kg N yr⁻¹, almost identical to the USDA Economic Research Service (USDA ERS 2013) estimate of food availability. Thus, a per capita value of 6.4 kg N yr⁻¹ was used to calculate human food supply. Household pet populations were determined from the American Veterinary Medical Association (AVMA) survey of pet ownership (AVMA 2007)). Total household pet food consumption was based on an average body mass of dogs and cats (Baker et al. 2001) and daily N intake requirements (NRC 2006).

Nursery and floriculture N harvest was based on annual biomass production of 750 kg N ha⁻¹ (R. Evans, personal communication) and the average of the reported acreage from the 2002 and 2007 USDA Census of Agriculture for all open grown horticultural commodities. We assumed that there was no net export of horticultural commodities. Based on the value of sales reported in the 2009 Census of Horticultural Specialties (USDA 2010), California produced 20% of the total national horticultural specialty crops. However, of the nursery and annual bedding/garden plants (which likely contribute the most to harvested N), California only produced 14%, similar to the state's proportion of the national population (12%).

The N tonnage of lint cotton, the only fiber commodity harvested on cropland, was calculated identically to the food crops. Annual cotton consumption for the population of California from 2002-2007 was on average 1 Mg cotton (USDA ERS 2013). The wood harvest in California in 2004 was 56 million m³ (Morgan et al. 2004). This was converted to N production based on the specific gravity (0.5 g cm⁻³) of Douglas fir (*Pseudotsuga menziesii*), and a typical wood N content (excluding bark) of 0.15% (Cowling and Merrill 1966, USDA Forest Service 1999). The consumption of wood for California was based on the national per capita estimate of 67 ft³ per year of wood products scaled to the 2002-2007 average population of 35.6 million. This volume was converted to N tonnage with the same factors as the volume of wood harvested.

Livestock feed was determined based on animal populations and dietary needs. For non-cattle livestock that are raised for meat (broilers, turkeys, pigs), the population was the average of the 2002 and 2007 USDA Agricultural Census quantity of animals sold. The feed requirements for these types of livestock were estimated on a grow-out basis (Van Horn 1998). For dairy cattle, steers, and layers the population estimates were the 2002-2007 average of the USDA National Agricultural Statistics Service annual year-end inventory. All beef cows, beef replacement heifers, and all calves were assumed to be grazed on rangelands. We assumed that all dairy cattle were on feed, as more than 95% of the dairy cows were located in the counties of the Central Valley or in the Chino Basin (USDA NASS 2012) where confinement is the typical practice. The feed requirements for dairy cows were from Chang et al. (2005) with the assumption that for one-

sixth of the year the cows were dry. The feed requirement for dairy replacement heifers was based on a 440 kg Holstein heifer (ASAE 2005). Although horses are included in the USDA Agricultural Census, this survey underestimates their population because it excludes animals that are not working animals. Instead, the horse population was based on the AVMA (2007) survey of pet populations and N intake requirements were from NRC (2007). Unlike dogs and cats, the horse population was estimated regionally: the California horse population was estimated assuming that the number of horses per household was the same across the entire Pacific region (Washington, Oregon, and California). In addition, there is anecdotal evidence that horse owners in California feed alfalfa to horses in the state because it is perceived to be higher quality feed (C. Stull, personal communication). A diet of 100% alfalfa feed with the suggested dry matter intake would provide 50% more N to horses than is needed.

Livestock-based food production (milk, eggs, meat) was based on 2002-2007 average production estimates from USDA annual surveys with the exception of broilers which were the average production from the 2002 and 2007 USDA Agricultural Census (USDA 2004, USDA 2009). The N content of various products was from NRC (2003), except that turkey N content was assumed to be the same as broilers (Table A4.2.3).

Product	N content (%)
Hogs, beef	2
Milk	0.5
Eggs	1.8
Broilers, turkeys	2.3

TABLE A4.2.3. Assumed Nitrogen Content of Animal Products.
Source: NRC 2003.

4.2.6 Manure Production and Disposal

Manure production was calculated based on the populations used for feed requirements and animal-specific excretion rates. For dairy cows, excretion was 169 kg N head⁻¹ yr⁻¹ for lactating cows and 81 kg N head⁻¹ yr⁻¹ for dry cows (Chang et al. 2005). It was assumed that all cows were dry for one-sixth of the year and lactating for five-sixths of the year resulting in an average manure production of 208 kg N head⁻¹ yr⁻¹. Dairy replacement heifers excreted 43 kg N head⁻¹ yr⁻¹ (ASAE 2005). Excretion rates for beef steers, pigs and poultry were based on Van Horn (1998). Horse excretion was assumed to be equivalent to feed intake (i.e. what was consumed was excreted). As with the calculations for feed intake, we assumed that all beef cows, replacement heifers, and calves were permanently on range with insignificant N inputs and outputs.

Manure N from confined animals was either leached to groundwater from the animal facilities, emitted to the atmosphere, or applied on cropland. The leaching of manure N was based on the amount of dairy manure N produced and the fraction leached from facilities reported by van der Schans et al. (2009). One source of data for livestock NH_3 emissions was the 2005 EPA NH_3 emission inventory for California (EPA 2008). A second method to estimate NH_3 emissions was multiplying the manure production estimates described above by animal-specific NH_3 emission factors from EPA (2004). Nitrous oxide produced prior to land application of manure was based on the average for the 2002-2007 manure management subsector of the ARB greenhouse gas inventory (CARB 2013). There are few quantitative estimates of N_2 emissions from the housing and production portion of dairies, but they are suggested to be small (Rotz 2004).

The predominant source of manure produced in California is confined dairies. The N content of solid and liquid excreta from dairy cattle is well established. However, the manure that is applied to cropland in solid and liquid form represents a mixture of N from urine and feces diminished in magnitude by volatilization and leaching. There are no data currently that would allow for partitioning the manure applied on and off dairies into solid and liquid form. However, if the nutrient management plans required by the Central Valley Regional Water Quality Control Board become publically available, they will be an invaluable resource for understanding N flows in the dairy-forage system. The manure from pigs, poultry, feedlot beef cattle, horses, and sold dairy manure was also assumed to be applied to cropland.

4.2.7 Household Waste Production and Disposal

Per capita N availability nationally for 2002-2006 was reported as 110 g protein day^{-1} or 6.4 kg N yr^{-1} (USDA ERS 2013). Statewide per capita N consumption (4.9 kg N yr^{-1}) was estimated based on actual protein consumption reported for various demographic groups and the populations of these groups in the United States Census for 2003-2007 following Baker et al. (2001). The consumed N was assumed to end up as sewage N. The difference between available food (228 Gg N) and food consumption (174 Gg N) was assumed to be waste. This 54 Gg N, or 23%, in waste is close to the 27% food waste reported at the retail and consumer level (Kantor et al. 1997). Food waste has several potential fates: down the sink to wastewater, composted and applied to urban land or cropland, and disposed in landfills. While the number of communities collecting household green waste is growing, we assumed that food waste went to landfills.

The tonnage of N discharged as wastewater without advanced treatment in areas with centralized sewage was calculated directly from measurements of wastewater N effluent. A list of

facilities classified as wastewater dischargers was obtained from the State Water Resources Control Board's (SWRCB) publically available database, the California Integrated Water Quality System (CA SWRCB 2013). This list was supplemented based on manually examining the list of dischargers without a category or those in the 'other' category. In addition, effluent discharge, and in many cases effluent N concentrations, was obtained. An empirical relationship was developed between design flow, which is included as part of the SWRCB facility database, and the discharge of NH_3 for all of the facilities in the state that serve more than 100,000 people. Like the SWRCB, we refer to the sum of NH_3 and NH_4^+ in effluent as NH_3 . In addition, NH_3 concentration and flow data were available electronically for facilities within the San Francisco Bay Regional Water Quality Board. Because the flow and N tonnage varied by more than 5 orders of magnitude, a log-log relationship was used with a polynomial fit (Figure A4.2.1). While NH_3 is the only N constituent commonly measured in effluent, in a few cases, organic N and/or NO_3^- were also monitored in facilities with no N treatment and they were <10% of the total N load. A minor amount of the N loading to wastewater treatment is from sink disposals and household chemicals (e.g., Baker et al. 2007), but these are typically insignificant sources of N.

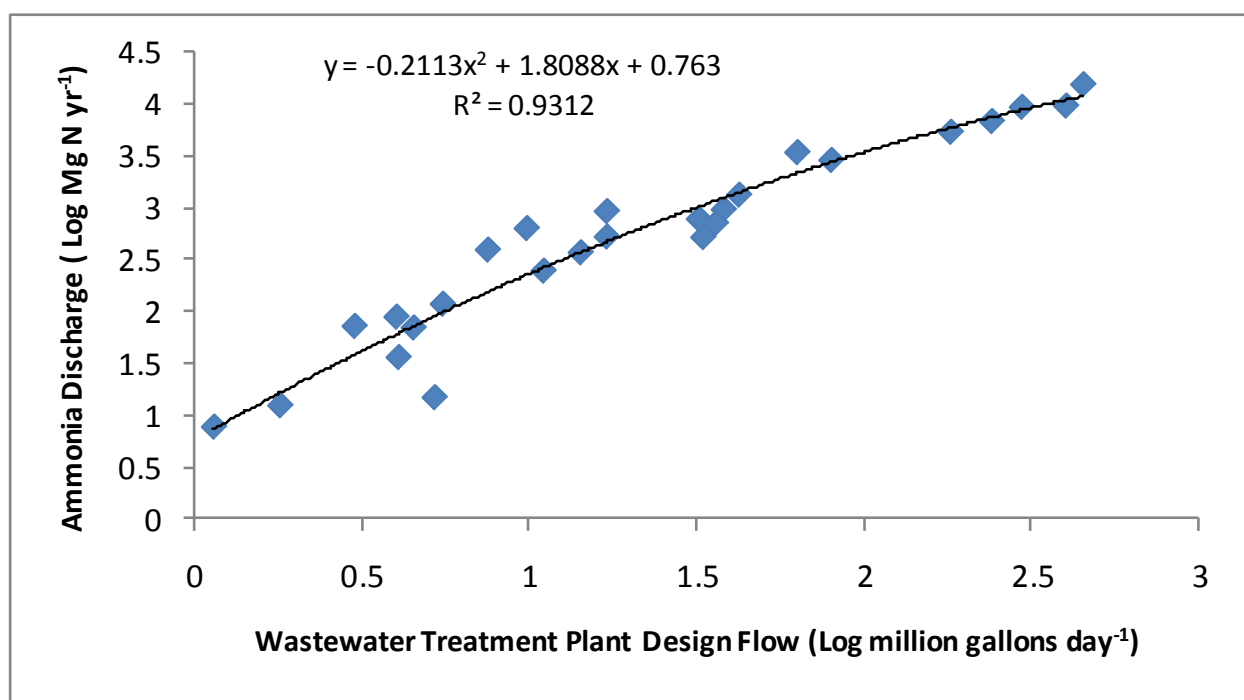


FIGURE A4.2.1. Relationship Between Wastewater Treatment Plant Design Flow and Nitrogen Discharge in California. Design flow was chosen as the predictor because it is reported by essentially all facilities to the State Water Resources Control Board. Population served is also a strong predictor of nitrogen discharge, but is not

necessarily reported as part of the Waste Discharge Requirements. The data points represent the mean value for each facility from data available for 2002-2007. The facilities chosen for this analysis included all of the large treatment plants in the state (population served >100,000) as well as all of the treatment plants in Region 2 because it is the only region with an electronic database of monitoring data.

The level of treatment in the known facilities was determined based on three data sources. First, the orders issued by the Regional Water Quality Control Boards were examined for the facilities with large (>10 mgd) flows. Second, data on treatment level were compiled as part of a brine survey by the United States Bureau of Reclamation for coastal areas of southern California (USDI 2009). Third, the SWRCB wastewater user survey contains information on the treatment level of sewage agencies (CA SWRCB 2008). This database was matched based on the agency name in the California Integrated Water Quality System database. In some cases these databases disagreed, often because some facilities have a small water reclamation capability with advanced treatment, but the majority of the flow receives no advanced N removal treatment. In cases where the databases disagreed, the orders were assumed to be correct, followed by the United States Bureau of Reclamation report, followed by the SWRCB wastewater use survey. Facilities with no information were assumed to have no advanced treatment. The average N load removed from these facilities with advanced treatment was ~50% based on dividing the median inorganic N ($\text{NH}_3 + \text{NO}_3^-$) concentration of the facilities with treatment by the median NH_3 concentration of facilities without treatment. Dissolved organic N is rarely measured by itself and was assumed to be a minor portion of the flow and unaffected by treatment. The decrease in inorganic N associated with advanced N removal was assumed to be converted to N_2 gas through denitrification.

The fate of discharged wastewater N was based on the permit type and facility location. Facilities with a National Pollutant Discharge Elimination System permit were assumed to discharge to surface water and are regulated by the United States EPA and subject to the federal Clean Water Act. Facilities with a NON15 Waste Discharge Requirement Program, regulated by the SWRCB, were assumed to discharge to land. If a facility had both permit types, the discharge was assumed to go to surface water. For facilities with National Pollutant Discharge Elimination System permits, the surface water body receiving the effluent is listed as part of the permit. In many cases, the receiving water body was the Pacific Ocean. In addition to facilities discharging directly to the ocean, facilities that discharged to San Francisco Bay, San Pablo Bay, Carquinez Strait, or Suisun Bay (as well as Sacramento and Stockton which discharge downstream of the river gauging stations on the Sacramento and San Joaquin Rivers) were also included in calculations of wastewater discharge to the ocean. In some cases, land applied effluent is applied

to fields growing crops, while in others it is applied to the surface of recharge basins. However, we assumed that all wastewater N discharged to land would flow completely to groundwater with no gaseous outputs or plant uptake after application. To calculate the N load in rivers associated with sewage discharge, a point vector layer of the georeferenced facility addresses was created and joined with the polygon layer of major (>1000 km²) watersheds in the state based on the United State Geological Survey Hydrologic Units in ArcGIS.

In addition to dissolved forms of N in effluent, wastewater treatment also results in the production of waste biosolids and gaseous forms of N. The two most common uses for the treated solids, or biosolids, are application as an organic amendment to soils, often in degraded areas, or use as an alternative daily cover in landfills. We assumed that all of the biosolids were used on urban land equally split between land application and landfills. The tonnage and fate of biosolids in the state were estimated by the California Association of Sanitation Agencies. The biosolids N content was assumed to be 3% (Tchobanoglous et al. 2002).

A small fraction of the wastewater N is emitted as N₂O during treatment, which is tracked as part of the statewide greenhouse gas inventory by both the California ARB and the United States EPA. In addition, N₂ can be produced most commonly in facilities that promote nitrification followed by denitrification during advanced wastewater treatment. Emission as N₂ would be expected during advanced secondary or tertiary treatment (see above for calculations), but we assumed that no N₂ was emitted in the absence of advanced N removal treatment.

According to the 1990 United States Census (United States Census Bureau 1992), approximately 10.4% of households in California were not on centralized sewage systems and the percentage with on-site waste treatment (i.e. septic systems) was essentially unchanged in 1999 (TCW Economics 2008). Based on Lauver and Baker (2000) we assumed that the N removal efficiency was 9%, which is already accounted for in the flow of biosolids from wastewater treatment plants. We assumed that the other 91% of the N from septic systems leached to groundwater.

Households produce other forms of N-containing waste besides sewage. Food waste was described earlier in this section, but a fraction of household and yard waste is disposed of in landfills. Surveys of the materials transported to landfills are conducted periodically by the California Department of Resources Recycling and Recovery. Landfill N disposal was calculated based on the tonnage of organic materials and their N content (Table 4.10 in CNA).

Household pet waste and feed intake requirements were calculated based on the average body mass of dogs (20 kg) and cats (3.6 kg) (Baker et al. 2001, NRC 2006)). Feed intake calculations assumed that all feed intake was excreted. Populations of dogs and cats for 2006 were

taken from AVMA (2007). We follow the approach of Baker et al. (2001) by assuming that 100% of dog waste and 50% of cat waste is added to urban soils. Ammonia emissions from dog (24%) and cat (12%) waste were from Sutton et al. (2000).

4.2.8 Gaseous Emissions

Gas emissions were tracked by individual gas (NO_x , N_2O , N_2 , NH_3) for all sources. Fossil fuel combustion (section 4.2.1), upwind sources (section 4.2.2), manure (section 4.2.6), wastewater (section 4.2.7), and surface waters (section 4.2.9) all emit one or all of these gases, but are described elsewhere. This section provides the methods for gaseous emissions from soils and forest wildfires.

Total N volatilization during natural land fires was estimated as the product of average annual acreage burned (H. Safford, personal communication) and an average areal N emission rate of 100 kg N ha^{-1} during fires (Johnson et al. 1998). The emission of NO_x and NH_3 from fires was based on the 2005 EPA National Emission Inventory (EPA 2008) while N_2O emissions were determined to be an insignificant flow based on the ARB greenhouse gas inventory (CARB 2010). The balance of the volatilized N was assumed to be N_2 .

Ammonia emissions for natural land soils were estimated from the biome-specific rates modeled by Potter et al. (2003) for California and extrapolated to the entire state based on the land cover map. Statewide emissions of NO and N_2O from soils on natural land were scaled up with the land cover map using the average of published sources reporting typical biome-specific rates (Table 4.5 in CNA).

For cropland, unlike the natural land biomes, we also compiled published estimates of gaseous emissions in California. The only source of field NO emissions in California was the average daily flux of all crops reported in Matson et al. (1997). For N_2O , the median rate was calculated across all crops and management practices for N_2O emissions for California published in the last decade (Appendix 4.1). A second unique approach for estimating N_2O emissions from cropland combined the estimate based on an emission factor for fertilizer with background emissions unrelated to fertilizer use. We assumed a direct emissions factor of 1% for both synthetic fertilizer and manure applied to cultivated cropland based on the ARB methodology in the greenhouse gas inventory (CARB 2010). However, we also include a background soil emission rate of $1 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ (Stehfest and Bouwman 2006) in order to estimate total N_2O emissions and not just anthropogenic emissions. This background rate is higher than most natural ecosystems, but there are no current estimates of N_2O emissions in California cropland soils that don't receive fertilizer. For both cropland and natural land, N_2 emissions were based on

the mean $N_2:N_2O$ ratios reported for natural land (1.03) and cropland (1.66) (Schlesinger 2009). Cropland NH_3 emissions for synthetic fertilizer were based on the direct emissions factor reported in Krauter et al. (2006). On average, across the range of fertilizer types and crops with varying agronomic practices that were studied, 3.2% of applied synthetic fertilizer was volatilized as NH_3 , but emissions ranged from 0.1 to 6.5% of applied fertilizer. Based on the crop mix in California, Krauter et al. (2006) suggested that the actual emission factor was only 2.4%. While the emission factor for urea can be significantly higher, most other fertilizers are reported to have an emission factor of less than 5% (Battye et al. 2003). Using the values in Battye et al. (2003) and the reported sales of fertilizer in California during the study period, the emissions factor ranges from 4% to 5%. Ammonia emissions associated with manure application on cropland were based on the reported values for each class of livestock in EPA (2004), ranging from 3% for beef cattle to 15% for poultry.

For urban land, gaseous emissions were assumed to occur only from turfgrass soils related to fertilization. Gaseous emissions were based on data compiled in Petrovic (1990) on the direct emissions of fertilizer N. The median fraction of fertilizer that volatilized as NH_3 or was denitrified in turfgrass areas was calculated for all the reported data. Total emissions were calculated based on the total synthetic N fertilizer use in urban areas.

4.2.9 Surface Water Loadings and Withdrawals

Only 55% of California's land area drains to the ocean. This area does not include the Tulare Basin, which is now essentially a closed basin because of water management. The only point source of N to surface waters was the discharge of wastewater effluent as described in Section 4.2.7. We did not include any discharge of food processors to surface water. These facilities are regulated by Regional Water Quality Control Boards in either the stormwater program or in the wastewater program. To get a sense of the potential for discharge to surface water from food processors, we calculated total N discharge for the 162 facilities in the Central Valley included by HydroGeoPhysics Inc. as part of the Hilmar Supplemental Environmental Project (HydroGeoPhysics 2007). While many facilities do not have monitoring data, the sum of the loading from those that do was $\sim 2 \text{ Gg N yr}^{-1}$. Because of the lack of complete data for these discharges, we do not include them in the calculations. We estimate atmospheric N deposition on surface water bodies by summing the modeled CMAQ deposition (described in Section 4.2.2) for all of the surface water pixels in the land-use map.

Total loading to surface water from non-point sources was calculated based on the export coefficients for cropland ($EC_C = 11.9 \text{ kg N ha}^{-1} \text{ yr}^{-1}$), natural land ($EC_N = 2.4 \text{ kg N ha}^{-1} \text{ yr}^{-1}$), and

urban land ($EC_U = 9.3 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) (Wickham et al. 2008). To check if these values were reasonable for California, we calculated export coefficients for 25 of the subwatersheds of the San Joaquin and Sacramento Rivers in the Central Valley measured by Kratzer et al. (2011) and the area of cropland, urban land, and natural land from our land-use map; we excluded two drainages as outliers (Colusa Basin Drain and Sacramento Slough). Using the Solver function in Excel, we calculated the best fit EC_C , EC_U , and EC_N for the Central Valley. We then solved for the export coefficients by minimizing the sum of the squared difference between the measured and predicted yields with the predicted yield calculated as $EC_C * \% \text{ Cropland} + EC_U * \% \text{ Urban Land} + EC_N * \% \text{ Natural Land}$. Similar to Wickham et al. (2008), we estimated $EC_C = 14.2 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, $EC_N = 1.6 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, and $EC_U = 7.0 \text{ kg N ha}^{-1} \text{ yr}^{-1}$.

The N loading to the ocean was estimated in two distinct ways. First, for the major watersheds ($>1000 \text{ km}^2$) where measured N discharge has been reported, we used the measured values from Sobota et al. (2009), Schaefer et al. (2009), and Kratzer et al. (2011). In watersheds where measurements had not been made, we used adjusted estimates from the export coefficients. The export coefficients provide a means to predict N loading to surface water, but not necessarily the N discharge to the ocean because of gaseous emissions and sedimentation in reservoirs. We calculated the log-log relationship between the measured values and predicted values for the eight watersheds with measured data. We used the regression of this relationship $\log [(\text{Measured N}) = 0.5685 * \log (\text{Predicted N}) + 1.2991 (R^2=0.71)]$ for these ungauged watersheds to adjust the predicted N discharge from the export coefficients to predict the actual discharge of N. We report the values predicted by the export coefficients, the adjusted values predicted by the export coefficients and the measured values for the watersheds in the state (Table 4.15 in CNA). Nitrogen loads for the urbanized areas in the San Francisco Bay watershed and along the southern coast from Santa Barbara to the Mexican border were estimated in Davis et al. (2004) and Ackerman and Schiff (2003), respectively. However, in both cases the estimates are for stormwater inputs of inorganic N only, so they likely underestimate the total N load.

Water withdrawals for irrigation were considered an output from the surface water subsystem. The volume of water for irrigation was based on Hutson et al. (2004), which reported $26 * 10^{12} \text{ L yr}^{-1}$ withdrawn for California in 2000. An average of $7.8 * 10^{12} \text{ L yr}^{-1}$ of this water was pumped from the Delta from 2000-2004. The water pumped from the Delta was not included in the surface water mass balance as it was actually considered a N import to the state because of the location of USGS river gauges. That is, for the purposes of our N budget, the Delta pumps are located outside of the study area, so that the dissolved N in this water is considered a N import to the state. The water quality at the Harvey O. Banks Pumping Plant (Station number KA000331),

where water is pumped from the Delta, was historically monitored each month (CA DWR 2013). The total N concentration for 2002-2007 was on average $\sim 1 \text{ mg N L}^{-1}$, and was split almost evenly between nitrate and dissolved organic N. The N concentration was assumed to be the same for the $18.2 \times 10^{12} \text{ L yr}^{-1}$ withdrawn from other surface water bodies in California. A smaller volume of surface water was withdrawn for domestic use ($4.6 \times 10^{12} \text{ L yr}^{-1}$): we ignored this flow as the majority of this water is used for indoor residential and industrial use which would likely be accounted for in wastewater effluent to surface water or the ocean (Gleick et al. 2003).

Gaseous outputs from surface water were only significant in the form of N_2 and N_2O , predominantly from denitrification. For rivers, gas emissions were estimated based on the areal rates of $2.8 \text{ kg N}_2\text{O-N ha}^{-1}$ (Beaulieu et al. 2011) and $51 \text{ kg N}_2\text{-N ha}^{-1} \text{ yr}^{-1}$ (Mulholland et al. 2009). The gaseous emissions from lakes and reservoirs were also based on these sources given the similarity in denitrification rates in rivers and lakes reported in Seitzinger et al. (2006). The acreage of rivers, lakes and reservoirs was based on a comparison between the USGS National Hydrography Dataset and the CAML land-use map. Waterbody pixels in the land-use map not identified as lakes or reservoirs in the USGS dataset were categorized as rivers.

The burial of N in lake and reservoir sediments was considered surface water storage and was estimated by difference for the purposes of the mass balance. However, there are two potential independent approaches to calculating the amount of N retained. The first provides an estimate for just reservoirs, and the second, for both lakes and reservoirs. First, the total volume of sediment in all California reservoirs was estimated by Minear and Kondolf (2009). Based on the reservoir age, an annual sedimentation rate was calculated. The annual rate of N sedimentation was calculated by assuming a bulk density of 1 g cm^{-3} (Verstraeten and Poesen 2001), a carbon content of these sediments of 1.9% (Stallard 1998) and a C:N ratio of 10 (Vanni et al. 2011). Second, Harrison et al. (2008) estimated that a global average of $306 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ was retained in reservoirs. These authors also estimated that lakes retain $\sim 30 \text{ kg N ha}^{-1} \text{ yr}^{-1}$. The total annual N retention was calculated from the area of reservoirs (180,000 ha) and lakes (350,000 ha) in the state by partitioning the National Hydrography dataset. The difference between retention and denitrification as calculated above provides an estimate of burial in sediments.

4.2.10 Groundwater Loading and Withdrawals

Groundwater inputs included leaching from septic tanks and wastewater treatment discharge (Section 4.2.7), cropland soils, and natural land soils. For cropland, leaching to groundwater was calculated in two ways. First, the average NO_3^- concentration in water leached below the rooting

zone in crop soils was calculated from a compilation of California literature (See Chapter 7 for details on data). The N concentration (38 mg N/L) was multiplied by the total volume of recharge in agricultural regions, where the majority of groundwater recharge occurs. All of the recharge was assumed to occur in the Central Valley (9.6×10^{12} L; Faunt 2009), Salinas Valley (2.3×10^{11} L; Montgomery Watson 1997) and Imperial Valley (3.0×10^{11} L; Montgomery Watson 1995) groundwater basins. Second, the median fraction of applied fertilizer that leached was calculated from a compilation of California literature (see Chapter 7 for further details on data). This fraction (38%) was multiplied by the sum of statewide fertilizer use in cropland (synthetic fertilizer + manure). In natural land, groundwater inputs were assumed to occur only in areas lacking drainage to the ocean. Leaching inputs in the driest portions of the state which occur in closed basins have been estimated based on the N stock in the subsurface that has accumulated over millennia. The annual N flow was calculated as the product of a leaching rate of $0.6 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ (Walvoord et al. 2003) and an area of 18 million ha. Leaching from turfgrass was estimated as the median of the fraction of applied fertilizer that leached as summarized by Petrovic et al. (1990).

Groundwater outputs were only from water pumped from the ground. Nitrogen removal from groundwater was calculated as the product of groundwater volume withdrawn and average groundwater N concentration. The volume of groundwater withdrawal was reported in both Hutson et al. (2004) and CA DWR (2003). However, we used the former for the calculations because it partitioned use into municipal vs. irrigation and also provided estimates of surface water withdrawals. N concentrations were calculated as the average of all wells available in the USGS Groundwater Ambient Monitoring and Assessment and EPA STORET databases for the years 2002-2007 available on the Geotracker website (CA SWRCB, n.d.).

We calculated groundwater denitrification in three ways. (1) We estimated N inputs to groundwater since 1940 and used literature values for the half-life of N to estimate denitrification losses. Green et al. (2008) report a half-life of 31 years at one site near Merced. These authors found limited evidence for denitrification in aquifers below cropland soils in California, with 50% N removal in groundwater after 31 years. This represents a loss rate of $2.3\% \text{ yr}^{-1}$. A second estimate of the half-life can be made from the $^3\text{H}/\text{He}$ and N_2 excess reported in Landon et al. (2011). The data from this study, which covered a much larger area of the Central Valley, would result in a half-life of 80 years or a loss rate of only $0.9\% \text{ yr}^{-1}$ (C. Green, personal communication). Because of the more regional nature of this study, we chose the value calculated from Landon et al. (2011). We assumed that groundwater recharge of N has increased linearly since 1940 with only 10 Gg N of natural inputs occurring prior to 1940. We chose this starting

date based on the trend in fertilizer use (sales of synthetic fertilizer plus dairy manure since 1980). Manure production was assumed to start in 1980 because dairies had largely transitioned to confined feeding by then. Manure production was calculated based on milk production reported by USDA NASS (2012) with an assumed efficiency of 25%. Manure applied as fertilizer was calculated assuming 38% of manure production was volatilized. The x-intercept of the fertilizer-time relationship was 1940. Finally, groundwater N extraction was assumed to be zero in 1940 and increased linearly to 2005. Starting in 1940 10 Gg N was leached, 0 Gg N was extracted, 0.23 Gg N was denitrified, and 9.67 Gg N was stored. This process was assumed to continue with 0.9% of the annual input plus the groundwater storage denitrified annually. (2) We used the product of a concentration-based denitrification rate and the total volume of groundwater. Liao et al. (2012) reported denitrification in Merced County to be $0.2 \text{ mg N L}^{-1} \text{ yr}^{-1}$. Based on the data in Landon et al. (2011), a more regional value of groundwater denitrification was estimated to be $0.04 \text{ mg N L}^{-1} \text{ yr}^{-1}$. The volume of recharge water contaminated with N was assumed to be constant between 1940 and 2005 and was estimated the same way as for determining the load of N leaching from soils. (3) We used the average proportion of groundwater N inputs that were denitrified as reported for Europe (46%; Leip et al. 2011) and globally (40%; Seitzinger et al. 2006). The groundwater denitrification was the average of the three independent estimates.

We assumed that the net N exchange between groundwater and surface water was essentially zero. For the Central Valley aquifer, if anything, the flow of water ($0.2 \times 10^{12} \text{ L yr}^{-1}$) moves from surface water to groundwater (Faunt 2009). At a N concentration of 1 mg N L^{-1} as measured in the Delta representing the water in the Sacramento and San Joaquin rivers, this represents an insignificant flow of N. Nitrogen storage was calculated as the difference between inputs and withdrawals.

4.2.11 Storage

Storage in cropland and natural land subsystems was calculated by difference. That is, storage was equal to the difference of N flows in and out. This storage could occur in either soils or perennial vegetation. Storage in urban systems has three components. First, landfills are considered storage and the methods of calculating N flows to landfill are described in Section 4.2.7. Second, land (soils + vegetation) storage was calculated as the difference between inputs of fertilizer, atmospheric deposition, and dog waste and the outputs in the form of soil gaseous emissions and surface runoff. Finally, other storage was calculated as the difference between synthetic chemical and wood N inputs and landfill N storage.

The storage terms calculated for the surface water and groundwater subsystems are described in Sections 4.2.9 and 4.2.10, respectively.

Box A4.2.1 The Haber-Bosch process and cropland nitrogen

Synthetic fertilizer, which is almost exclusively produced by the Haber-Bosch process, is the largest source of N to cropland. However, Haber-Bosch derived N is not limited to the annual application of synthetic fertilizer. The N in applied manure also originates in part from feed that was grown with synthetic fertilizer and in part from biological N fixation by alfalfa. Of the 537 Gg N yr⁻¹ needed to feed livestock, 170 Gg N yr⁻¹ of the feed was in the form of alfalfa. Thus, alfalfa contributed 30% of the N supply in livestock feed, and presumably an equivalent fraction of manure N. The remaining manure (184 Gg N yr⁻¹) presumably originates as Haber-Bosch N. A large fraction of the biosolids applied to cropland also comes from Haber-Bosch N. The N applied in irrigation water could originate from any land use, but synthetic fertilizer application to cropland is likely the dominant source. Atmospheric deposition is a mixture of fossil fuel combustion with some contribution of reduced N from livestock manure NH₃ volatilization. If we assume that irrigation water was derived from synthetic N while atmospheric deposition was fossil fuel combustion, a total of 69% of N entering the cropland subsystem (Figure 4.4 in CNA) was from synthetic N fixation. At the statewide level, there is also the import of grain crops, largely corn, to California from the Midwest that is largely Haber-Bosch N as well.

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