SUPPORTING INFORMATION

Table S1. Area of components of vegetated cover used for scaling fertilizer use and biological N fixation. Residential fertilizer rates were scaled to the watershed by multiplying by rows (A+B+C). Non-residential fertilizer use was estimated for rows D, E, F, I, J, L, and N with individual rates determined as described in the text, scaled by the total area, and summed for each watershed. Biological N fixation rates for residential areas were scaled by multiplying by rows (A+B+C). BNF rates for non-residential areas were scaled by multiplying by row F. Watersheds are ordered from most to least residential (left to right) based on housing density.

†Areas include vacant land, industrial land, welfare facilities, "downtown" commercial properties (mostly parking ramps,

department stores) and "general" commercial properties (mostly commercial warehouses, vacant commercial property, railroad property, multi-story office buildings) not included above.

*Areas include "neighborhood" commercial properties (mostly churches, 1-2 story office buildings, shopping centers, convenience stores, supermarkets, banks, small retail properties); tax-exempt properties (mostly private schools); and office residential properties.

 5 Rates are from three golf courses, and represent the average (unweighted by total area of each golf course) of each golf course's area-weighted average for tees, greens, fairways, and rough. Average fertilization rates for each type golf course area were 15,382 (greens), 13,888 (greens), 14,128 (fairways), and 0 (rough) kg N km⁻² y⁻¹; and 1,302 (tees), 4,235 (greens), 1,318 (fairways), and 0 (rough) kg P km $^{-2}$ y $^{-1}$.

[¶]Rates are the average from two cemeteries (unweighted by total area of each cemetery).

#Rate is from the University of Minnesota.

Table S2. Inputs (% of total), outputs (% of total), outputs/inputs, and retention for N in seven study watersheds. See Table 1 for watershed name abbreviations. Watersheds are ordered from most to least residential (left to right) based on housing density.

*Dry deposition = 760 kg N km⁻² y⁻¹; wet deposition = 689 kg N km⁻² y⁻¹.

⁺Net inputs = total inputs - non-hydrologic outputs.

Table S3. Inputs (% of total), outputs (% of total), outputs/inputs, and retention for P in seven study watersheds. See Table 1 for watershed name abbreviations. Watersheds are ordered from most to least residential (left to right) based on housing density.

*Dry deposition = 0.31 kg P km⁻² y⁻¹; wet deposition = 0.18 kg P km⁻² y⁻¹.

⁺Net inputs = total inputs – non-hydrologic outputs

Table S4. Correlations of various watershed characteristics with watershed inputs and with nutrient export in stormwater runoff and baseflow (kg element km⁻² y⁻¹). Correlation coefficients (r) are shown for significant correlations (P≤0.10).

†P≤0.10, *P≤0.05, **P≤0.01; housing density and fractional tree cover over the street were highly positively correlated (r=0.88,

P=0.008); street density was highly positively correlated with impervious fraction (r=0.79, P=0.03).

Table S5. Storm drain exports of N and P. Watersheds are ordered from most to least residential (left to right) based on housing density.

Table S6. N:P ratios (mass basis) of net inputs and total storm drain exports, stormwater runoff and baseflow, leaching, and ecosystem accumulation in plants and soils.

SI Methods

Detailed methods for determining some nutrient inputs.

Atmospheric Deposition. The GEOS-Chem chemical transport model is driven with assimilated meteorological fields from the NASA Goddard Earth Observing System (GEOS-5), and we employed here a $0.5^\circ \times 0.667^\circ$ (latitude \times longitude) nested simulation over North America with 47 vertical levels and a 10-minute transport timestep. The model includes detailed HO_x-NO_x- VOC-ozone-BrO_x tropospheric chemistry coupled to aerosols (3) . Emissions were as described by Hu et al. (4, 5), and included the US EPA's National Emission Inventory (NEI08) for anthropogenic sources (6), the Model of Emissions of Gases and Aerosols from Nature (MEGANv2.1) for biogenic hydrocarbons (7), and the Global Fire Emissions Database (GFED3) for fires (8). Soil nitrogen oxide (NO_x) emissions were estimated following Hudman et al. (9). Boundary layer mixing in the model employed the non-local scheme of Lin and McElroy (10). Wet and dry deposition in GEOS-Chem were computed as described elsewhere (11-16); N deposition rates included wet $+$ dry deposition for all gas- and particle-phase N species in the model.

Atmospheric P deposition was inferred from estimates for the major river basins of Minnesota (17). For wet deposition, the values for the four river basins that include Minneapolis-St. Paul were averaged (Lower Mississippi, Upper Mississippi, St. Croix, and Minnesota Rivers). For dry deposition, the "urban" value from (17) was used.

Residential fertilizer. Based on a median and mean fertilization frequency of 1-2 times/year for households in the CRW (18) and an assumed per-fertilization event rate of 4882 kg N km⁻² (the commonly recommended fertilization rate per application of 1 pound 1000 ft⁻²) and no P fertilization, we estimated an average rate of 7323 kg N and 0 kg P km⁻² yr⁻¹ household⁻¹ (1.5 events per year) for single and multi-family households and boulevards.

Non-residential fertilizer. Non-residential fertilizer inputs for golf courses; cemeteries; college, university, and seminary campuses; the University of Minnesota St. Paul Campus Agricultural Experiment Station; public parks and recreation centers; the state fairgrounds; and public schools in the watersheds were determined through interviews with superintendents and resource managers. If no response was received after three contact attempts (three of twelve properties), we assumed that property was fertilized at the average rate of other properties in that land cover category, or at a rate of 4882 kg N km⁻² y⁻¹ (1 pound 1000 ft⁻² y⁻¹) and 0 kg P km⁻² $y⁻¹$ for unique land cover categories. Notably, only golf courses were fertilized with P. Otherwise, fertilization, when it occurred, was only with N. Public parks, recreation centers and schools were not fertilized.

For other non-residential areas, we assumed that "neighborhood" commercial properties (dominated by churches, 1-2 story office buildings, convenience stores, shopping centers, supermarkets, banks, small retail properties); tax-exempt properties (dominated by private schools); and office residential properties were fertilized at the recommended rates of 4882 kg

N km⁻² y⁻¹, 0 kg P km⁻² y⁻¹. We assumed that vacant land, industrial land, welfare facilities, "downtown" commercial properties (dominated by parking ramps, department stores) and "general" commercial properties (dominated by commercial warehouses, vacant commercial property, railroad property, multi-story office buildings) were not fertilized, given that many of these properties are unmanaged.

Pet waste. Inputs of N and P from pet waste were estimated from TCHEP, which calculated landscape N and P inputs from pet waste based on survey data of the number and mass of dogs in each household, published studies of dog metabolism, a survey of the nutritional content of dog food, and published data on dog waste pickup practices (2, 18). Average landscape inputs of N and P per household from dog waste were multiplied by the total number of single- and multi-family households in each watershed (Table S2) to determine total pet waste N and P inputs per watershed that were subsequently divided by respective watershed areas (note that this might underestimate pet ownership in multi-family households). Inputs of nutrients from domestic cats that spend time outdoors were not considered as part of pet waste, because of lack of data. Such inputs are likely small given (*i*) their mass is small on average compared to that of dogs and (ii) only a fraction of their waste deposited outdoors represents net watershed inputs, given that cats are consuming prey outdoors, and some of those nutrients are being removed from the system when they use the litter box inside and that material is exported to landfill/incinerator.

Compost. All compost taken to Ramsey County Yard Waste Collection sites within or adjacent to CRW is exported outside of the watershed. However, a fraction of finished leaf and grass clippings compost collected and composted by one county collection site well outside of CRW is distributed among the four collection sites located in St. Paul and thus represents a source of N and P to the watersheds. We estimated compost N and P input to the study watersheds based on the total wet mass of compost delivered to the St. Paul sites, its moisture content, and its total N and P content using data collected by the Environmental Health Supervisor for Ramsey County, Minnesota. We assumed that the compost was distributed among the study watersheds in proportion to the fraction of the total CRW single + two-family parcel area in each watershed.

Biological N Fixation. Cover of herbaceous legumes (predominantly *T. repens*) in residential yards was obtained from a survey of 21 residential lawns in the St. Paul-Minneapolis metropolitan area (19) (Table S7). Legume cover in city parks (which are not fertilized) was determined in five randomly selected city parks in CRW (five randomly selected 0.5 x 0.5 m quadrats per park) (Table S7). To estimate herbaceous legume aboveground biomass from percent cover, we measured legume (predominantly *T. repens*) percent cover in eleven 0.5 x 0.5 m quadrats containing a range of legume cover on the University of Minnesota St. Paul campus, in an area that received no management except mowing (no irrigation or fertilization). After measuring cover, we clipped legumes at the soil surface, dried $(65 °C)$ and weighed clipped biomass, and regressed biomass against percent cover using least-squares linear regression to develop a predictive equation. Percent cover in yards was used to predict biomass in yards

(single and multi-family households) and boulevards (Table S7); percent cover from parks was used to predict biomass in parks, recreation centers, schools, and the State Capitol grounds, all of which were unfertilized (Table S7). Other more highly degraded unfertilized areas (vacant land, industrial land, welfare facilities, "downtown" and "general" commercial properties) were assumed to have zero legume biomass. Legume biomass also was assumed zero in nonresidential areas known to receive fertilizer (i.e., golf courses, cemeteries, university campuses, some commercial areas, etc.), confirmed using observations and interviews with golf course superintendents. To estimate biological N fixation rates per areal cover of legumes (F), we regressed estimates of *F* against clover dry biomass across two different experiments (forcing the linear fit through the origin) presented in Jørgensen et al. (20) for white clover growing in pure stands and in mixtures with ryegrass in experimental fields in Denmark (Table S7). With this equation, we used the average estimated dry mass of clover for residential yards to estimate BNF for yards and boulevards, and the estimated dry mass of clover for city parks to estimate BNF for city parks and recreation centers, public schools, and the State Capitol grounds for each watershed. Total BNF per watershed was divided by watershed area.

^{*}Estimated from percent cover using the equation $B = -0.60 + 1.11$ ^{*}C, where $B = \text{biomass (g/m}^2)$ and *C* = cover (%), R²=0.94, P<0.0001, n=11.

⁺Estimated from (20) using the equation $F = 0.39$ ^{*}*B*, where $F = N$ fixation (kg N ha⁻¹ y⁻¹) and *B* = biomass (g/m^2) , P<0.001.

Uncertainties regarding nutrient inputs. Of the different inputs, non-residential fertilizer, residential fertilizer, and atmospheric N deposition were best constrained, whereas pet waste, atmospheric P deposition, weathering, compost, and biological N fixation were less certain. For atmospheric N deposition, the largest uncertainties are associated with NO_x and NH_x emission and deposition rates. In addition, we treated all watersheds as a single grid cell, which ignores potential spatial variability in deposition associated with proximity to sources, such as major roadways (21, 22). For atmospheric P deposition stem, we lacked measurements within the actual study watersheds, and were unable to determine sources of P deposition and whether they originated from within versus outside of the watersheds. The primary uncertainties associated with estimates of residential N fertilizer stem from the use of self-reported survey data on frequencies of fertilization by homeowners and from the assumption that homeowners are fertilizing at recommended rates. Lawn care companies, which were used by 78% of yards fertilized at rates exceeding recommended rates (23), provided relatively precise information regarding fertilization rates (18). Estimates of non-residential fertilizer use in golf courses,

cemeteries, and campuses were well constrained, as superintendents and resource managers kept detailed records of fertilization practices. Estimates of fertilizer use in other nonresidential areas are highly uncertain, given lack of data on their management. As described in the main text, the primary uncertainties associated with estimates of pet waste nutrient inputs stem from the limited number of published self-reported survey data on pet waste pickup practices (24) and the exclusion of outdoor cats. The primary uncertainties associated with estimates of BNF stem from the use of published values for *F* and from the cover data from yards and parks used for scaling to the watershed. Given their importance to watershed nutrient budgets and their relatively high uncertainty, estimates of residential fertilizer use, pet waste, and atmospheric deposition (especially of P) should be targets for future refinement.

Detailed methods for determining nutrient outputs.

Residential Leaf Litter and Grass Clippings. We obtained leaf litter and grass clippings N and P production rates per area of lawn for households visited during TCHEP field surveys that were located in CRW (n=86) (18). Those values were scaled to household N and P export rates based on a larger mail survey of CRW households (n=590) that indicated that 20.6% of households exported clippings and 57.5% exported leaf litter. Mean rates of N or P export per unit yard area for clippings were 594.3 kg N and 98.9 kg P km⁻² yard area y⁻¹, respectively, and for leaf litter were 1026.2 kg N and 73.1 kg P km⁻² yard area y⁻¹, respectively. All yard waste taken to Ramsey County Collection Sites in St. Paul (i.e., in or immediately adjacent to CRW) is exported to a distant site for composting and thus represents a nutrient export.

Street Sweeping. The majority of streets in CRW are swept twice each year, typically once in April and once in October. Swept material is hauled out of the watersheds by the St. Paul Public Works Department, so street sweeping represents a watershed nutrient export. We estimated this export using multivariate regressions developed in a study in Prior Lake, MN (ca. 50 km southwest of St. Paul) that predicted the mass of N and P removed by street sweeping as a function of month of year, sweeping frequency, and tree cover over the street (25). In applying the Prior Lake model to our study watersheds, we assumed the lowest street sweeping frequency regime (once every four weeks) in the months of April and October, and average canopy cover over streets in a watershed (Table S1). As street canopy in the study watersheds exceeded the maximum in Prior Lake (20%), we assumed that nutrient export in street sweeping was linearly related to tree canopy cover at canopy covers > 20%.

Uncertainties regarding nutrient outputs. Of the different watershed outputs, storm drain and crop exports were most constrained, whereas yard waste and street sweeping exports were less certain. The primary uncertainties associated with estimates of yard waste nutrient export stem from the use of self-reported survey data on yard waste management, from estimates of leaf litter production based on allometric relationships with tree size and of grass clippings based on a biogeochemical model, and from the exclusion of other types of yard waste besides leaf litter and grass clippings. Linear extrapolation to the high tree canopy cover over streets of the study watersheds was the main source of uncertainty related to street sweeping.

Methods for estimating water balances for study watersheds.

We estimated the annual amount of water leaving the watershed through groundwater pathways (not including baseflow) using a simplified water balance for each watershed. Estimated groundwater exports were calculated as precipitation inputs (Table S8a) minus evapotranspiration (ET) (Table S8b) and storm drain outputs (stormflow and baseflow) (Table S8); we did not account for changes in soil water storage (Table S9).

Precipitation data collected at the University of Minnesota weather station were used for all watersheds. ET outputs were calculated for multiple land cover types (Table S8b). For vegetated areas, we calculated ET using the BROOK90 model (1). For impervious surfaces, we used recommended coefficients from the Minnesota Stormwater Manual (26) to estimate that 14.5% of annual precipitation evaporated. For open water areas, we estimated evaporation as 70% of pan evaporation rates, using annual pan evaporation measured at the University of Minnesota (27). Unclassifiable patches in the remotely sensed data were assigned the same evaporation rate as impervious surfaces.

Land-cover percentages were estimated from remotely sensed data, as described in the main body of this paper. However, for the water balance we defined impervious surface area as only the area of impervious surfaces without overlying tree canopy cover (Table S11). We further subdivided tree canopy area into evergreen canopy, deciduous canopy over pervious surfaces (which we modeled as a turfgrass understory), and deciduous canopy over impervious surfaces, using methods from Nidzgorski and Hobbie (1) (Table S11).

Table S8. Annual precipitation (a) and evapotranspiration (b) rates.

(a)

(b)

Table S9. Storm drain output (runoff and baseflow) for each study watershed.

Table S10. Estimated annual groundwater export rates for each study watershed.

*Storm drain flow for AHUG was not measured for 2010 and 2011, so groundwater export could not be calculated for those years. Negative values for PC were due to unusually high storm drain baseflow, possibly indicating that the storm drains (which follow former creek paths) are draining a larger groundwatershed than the PC surface watershed and/or a deeper groundwater aquifer.

Table S11. Area (m²) of vegetation cover components used for scaling leaching to groundwater and constructing water balance.

Table S12. Incorporation of ecosystem N and P accumulation, leaching, and denitrification into watershed N and P budgets. Residual flux is the sum of inputs, accumulation, outputs, leaching, and denitrification. Negative residual flux indicates that accumulation and outputs exceed inputs. Positive residual flux indicates that inputs exceed accumulation and outputs. Watersheds are ordered from most to least residential (left to right), based on housing density.

⁺scaled from (2) by the total area of single- and multi-family households and boulevards

*scaled from (1) by the total area of turfgrass and tree cover in the watershed

 $\frac{1}{2}$ scaled from (28) by the total fertilized vegetated area in each watershed (single- and multi-family yards, boulevards, golf courses,

cemeteries, campuses, and other fertilized areas; rows A, B, C, D, E, I, J, L, and N in Table S1),

 n inputs + outputs + ecosystem uptake + leaching + denitrification

 $*$ inputs + outputs + ecosystem uptake + leaching

SI Discussion

An alternative explanation for the decrease in the N:P ratio of storm drain exports versus net

inputs is that household sewage (which has an N:P ratio of 5.8) (29) was leaking into the storm

drainage network. This explanation is unlikely, however, for several reasons (30). First, the N:P

ratio in baseflow (22.7:1 on average, Fig. 5) greatly exceeds the N:P ratio in human sewage.

Second, St. Paul has a relatively new sanitary waste system, and an active sanitary sewer

maintenance program that regularly inspects sanitary drains for leaks. Third, CRWD monitoring

detected very few illicit discharges in the storm drains, and when they have occurred, their

contribution to total nutrient loads have been negligible (31).

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