Estimating urban air pollution contribution to South Platte River nitrogen loads with National Atmospheric Deposition Program data and SPARROW model

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ABSTRACT

Air pollution is commonly disregarded as a source of nutrient loading to impaired surface waters managed under the Clean Water Act per states' 303(d) list programs. The contribution of air pollution to 2017–2018 South Platte River nitrogen (N) loads was estimated from the headwaters to the gage at Weldona, Colorado, USA (100 km downstream of Denver), using data from the National Atmospheric Deposition Program (NADP) and the SPAtially Referenced Regressions On Watershed attributes (SPARROW) model. The NADP offers wet-deposition raster created by spatial interpolation of data collected from regionally representative monitoring sites, excluding the influences from urban site data. For this study, NADP wet-deposition data obtained from sites within the Denver-Boulder, Colorado, urban corridor were included and excluded in new spatial interpolations of wet-deposition raster, which were used as input for SPARROW to model the influence of urban air pollution sources on South Platte River loads. Because urban air pollution is already incorporated into the NADP Total Deposition modeling methodology, dry N deposition was held constant for each SPARROW modeling scenario when dry deposition was included. By including the urban wet-deposition data in the model, estimated N loading to the South Platte River at Denver increased by 9–11 percent. Factoring in dry deposition at a 1:1.8 dry:wet ratio obtained from the results, urban air pollution was estimated to contribute as much as 20 percent of the nitrate Total Maximum Daily Load for Segment 14 of the South Platte River.

1. Introduction

According to the 2014 Congressional Research Report on Clean Water Act and Pollutant Total Maximum Daily Loads (TMDLs): "The most recent state 303(d) lists, most of them submitted in 2010 and 2012, identified over 41,000 waterbodies (USA) as not meeting water-quality standards and in need of a TMDL, affecting more than 300,000 miles of rivers and shorelines and 5 million acres of lakes. Nation-wide, more than 50 percent of all impairments are caused by nutrients and sediment, metals including mercury, and pathogens" (Copeland, 2014). Many impaired streams flow through urban corridors with a variety of air pollution sources, for example vehicles, power plants, factories, and homes. Watershed management agencies are challenged with controlling pollution loading into their states' 303(d)-listed waters, and air pollution is typically an overlooked source of impairment.

Atmospheric wet (precipitation) and dry deposition deliver air pollutants directly to surface water and via overland and subsurface flow (Peng et al., 2019; Driscoll et al., 1998). Many case studies have shown that nutrients such as N in atmospheric deposition (AD) are linked to water-quality degradation. For example, AD was shown to account for approximately 25 percent of Chesapeake Bay nitrate (N) loads, which comes from a variety of emission sources (Fisher and Oppenheimer, 1991; Russell et al., 1998; Loughner et al., 2016). Inputs from AD can be extremely harmful, such as in Narragansett Bay, New England's largest

estuary, where nutrients, including N in precipitation, were shown to drive summertime hypoxia events despite significant reductions in wastewater N loading to the bay (Joyce et al., 2020; Oviatt et al., 2017). Changes in stream N yields have also been linked to changes in air pollution and AD of N (Eshleman et al., 2013; Mast et al., 2014).

Despite the conclusive evidence of the afore-mentioned studies, AD contribution to surface water chemical loading, has been underrepresented by monitoring data, especially in urban areas. The NADP has traditionally produced data products using regionally representative data collected in rural and isolated areas away from urban sources and has distributed wet-deposition chemistry data for its urban sites only by request and to site sponsors. Urban wet-deposition data currently are not incorporated into the NADP's Total Deposition (TDep) data products. This is one of many reasons that the NADP is striving to monitor in urban areas and incorporate the data into interpretive products. Additionally, data assessment techniques and models are needed to accompany urban monitoring data to help watershed managers to fill important AD monitoring gaps in cities.

The case study presented here describes wet plus dry AD loading of nitrate (NO_3^-) and ammonium (NH_4^+), collectively referred to as inorganic reactive nitrogen (Nr) for the purposes of this paper, to the upper South Platte River basin; with particular focus on loading of atmospheric Nr to the river from wet-only deposition of N from urban air pollution sources. The study area is the South Platte River basin from its headwaters to the gage at Weldona, Colorado, USA (100 km downstream from Denver). This study used NADP monitoring and TDep measurement-model fusion data available from the NADP for the years 2017 and 2018 as input for the SPAtially Referenced Regressions On Watershed attributes (SPARROW) model to estimate the urban AD contribution of N loading to the South Platte River (Reference). The SPARROW model was run for two input conditions: 1) wet-only and 2) wet-plus-dry (total) atmospheric deposition, and for two annual scenarios: 1) including and 2) excluding wet deposition data from urban NADP sites, to estimate the urban AD contribution to surface-water N loads in four sub-basins upstream from the Weldona, Colorado, stream gage. Additionally, the SPARROW model results were independently checked with stream discharge and water-quality data using a second method of analysis described in the Supplemental Information section (Wetherbee, 2021).

1.1. Study area

The South Platte River's headwaters are located along the Continental Divide in the Rocky Mountains of central Colorado, USA. Most of the river's water originates as snowmelt runoff from high elevations, but inter-basin transfers add water to the South Platte River basin from the Colorado, Arkansas, and North Platte River basins. The South Platte River is highly managed along its entire course, and water is moved and stored in a complex network of ditches and reservoirs for irrigation, municipal water supply, and other uses (Sprague, 2005; Dennehy et al., 1995).

For this study, the South Platte River basin upstream from Weldona, Colorado, was divided into four sub-basins identified by sub-basin numbers 1–4 (Fig. 1). Sub-basin 1 is comprised mostly of national forest lands with some small towns and ranches. Sub-basin 2 is comprised of a variety of land uses, including wilderness, national forest, and approximately the southern half of the city of Denver and suburbs. Sub-basin 3 is urbanized and contains the northern half of the city of Denver and suburbs. Approximately 3.2 million residents live in the Denver-Boulder metropolitan area (U.S. Census Bureau, 2019 https://www.census.gov/quickfacts/, accessed May 28, 2021). The Denver County population alone was 600,000 residents in 2010 and grew to 727,211 by 2019. Sub-basin 4 contains a wide variety of land uses. The west side of sub-basin 4 includes urbanized Boulder County with 326,196 residents and Rocky Mountain National Park. Urban and suburban areas within sub-basin 4 have smaller population densities than Denver. The city of

Fort Collins, with a population of 170,243 residents, is located in the northern portion of sub-basin 4. Smaller urban areas such as Longmont, Loveland, and Greely (not shown) are located in this predominantly rural sub-basin along with intense agricultural and oil-and-gas production combined with growing residential development.

Colorado's 303(d) accounting system tracks water-quality impairment by segmentation of streams into discrete reaches based on water-quality characteristics and pollution loading. The segment of the South Platte River extending from the Chatfield Reservoir outlet to the Burlington Ditch Headgate at the Denver – Adams County line is known as Segment 14 (Fig. 1). The Colorado Water Quality Control Commission has determined that Segment 14 should be suitable for use for recreation, aquatic life, drinking water, and agriculture, and they have set standards, which include numerical criteria for NH_3 and nitrite (NO_2^-), to protect aquatic life and for NO_3^- in drinking water. Nitrate is a dominant constituent of concern in the South Platte River (Water Quality Control Division (WQCD), 2004). In 1998, Segment 14 was placed on the State of Colorado's 303(d) list of impaired waters for exceedances of the NO_3^- standard, and a TMDL for NO_3^- was approved by the EPA in 2004, which establishes the standard concentration of 10 mg/L at the Burlington Ditch Headgate (USEPA, 2004). The TMDL identified three primary wastewater sources that need to be reduced to address exceedances of the NO_3^- standards in low-flow conditions (Water Quality Control Division (WQCD), 2004). The TMDL prompted each wastewater treatment plant to implement load-reducing actions, such as secondary NO_3^- treatment, which resulted in notable decreases in NO_3^- concentrations in Segment 14 as measured at station N25E (Fig. 2) after one of the plants was decommissioned in 2006. Other, minor point and non-point sources of NO_3^- were identified but were determined by the TMDL process to have minimal effects on water quality.

2. Material and methods

2.1. Atmospheric deposition

Atmospheric deposition of Nr to the South Platte River basin was estimated from data collected by the NADP. The NADP data used for this study are available at: https://doi.org/10.5066/P9OOIQ0E (Wetherbee et al., 2021). Each NADP site consisted of a continuously recording rain gage and an automated precipitation sample collector, which was designed to open during precipitation events to collect wet-only atmospheric deposition samples. Weekly samples were collected routinely on Tuesdays and shipped to the NADP Central Analytical Laboratory in deionized-water rinsed Nalgene bottles, and samples were analyzed per NADP chemical analysis and quality-control protocols (http://nadp.slh.wisc.edu/lib/qaplans/QAPNADPLab2020.pdf, accessed September 22, 2020). The Nr concentrations were calculated by summing the concentrations of NH_4^+ as N and NO_3^- as N for each sample. Precipitation depth data were collected in 15-min intervals, and these data were summed for the collection period for each, approximately weekly sample.

Annual precipitation-weighted mean Nr (wetfall) concentrations were calculated for 39 NADP sites within Colorado and the closest NADP sites in adjacent states for calendar years 2017 and 2018 (Table 1). Spatially interpolated raster data sets (4 km² resolution) were generated by interpolating the annual precipitation-weighted mean concentrations using cubic inverse distance weighting with a 100 km interpolation radius in ArcGIS[1] 10.7.1 (Esri, 2019). A raster of annual wet Nr deposition (wetfall), in units of kilograms per hectare (kg•ha^{-1}), was generated for 2017 and 2018 by multiplying the raster of precipitation-weighted mean concentrations by a raster of precipitation amount. The precipitation amount raster, which is at the same 4 km² resolution, was developed by the Parameter-elevation Relationships on Independent Slopes (PRISM) model (2019), and data are available at: https://prism.oregonstate.edu/ (accessed September 22, 2020).

Percent Land Use

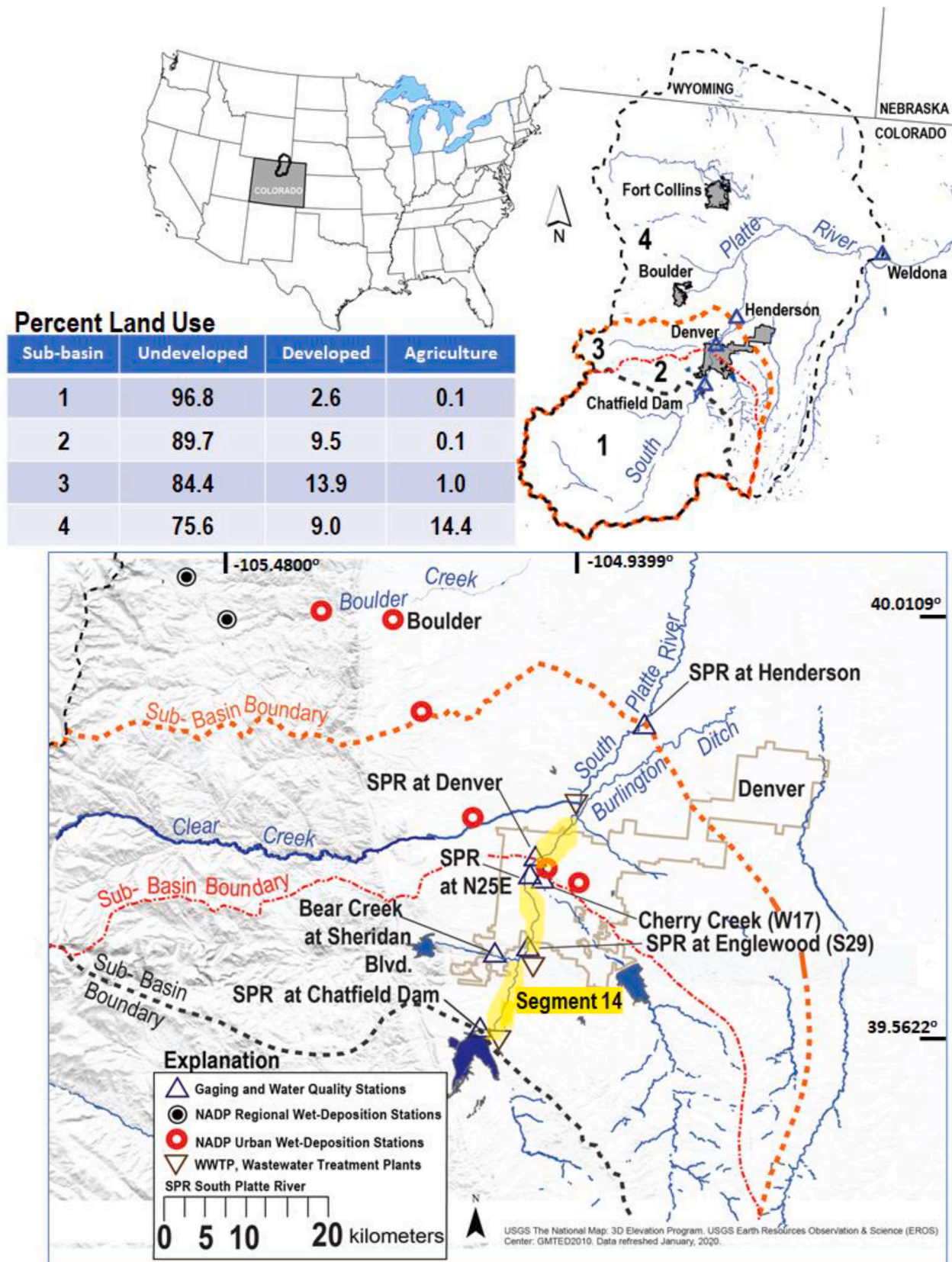| Sub-basin	Undeveloped	Developed	Agriculture
1	96.8	2.6	0.1
2	89.7	9.5	0.1
3	84.4	13.9	1.0
4	75.6	9.0	14.4

Fig. 1. Study area within the upper South Platte River basin, including Denver, Boulder, and Fort Collins metropolitan areas, sub-basins delineated by gaging stations, and stream and atmospheric deposition sampling locations. Sub-basin land use data obtained from National Land Cover Database, 2016) accessed June 2, 2021 at https://www.mrlc.gov/data, Homer et al. (2012).
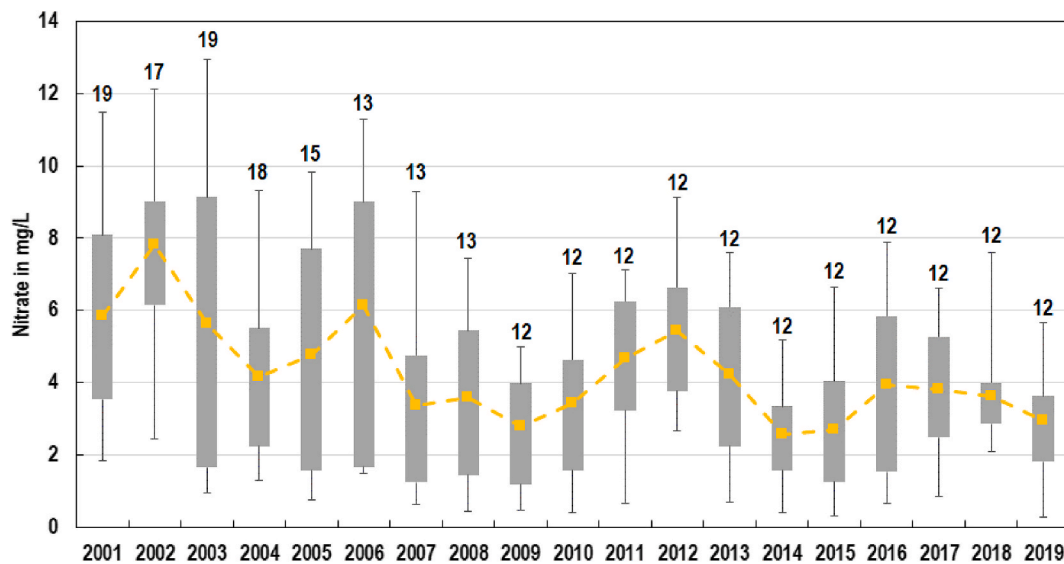
Fig. 2. Box and whisker plots showing changes in nitrate concentrations in the South Platte River immediately upstream of the confluence with Cherry Creek at station N25E, 2001–2019. The yellow dashed line connects mean annual nitrate concentrations. The number above each box plot is the number of samples collected that year. A wastewater inflow was removed in 2006. Data obtained from Denver Department of Public Health & Environment, Environmental Quality Division. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
NADP sites used for inorganic reactive N deposition raster data; urban sites shaded.

State	NADP Site Identifier	Latitude (decimal degrees)	Longitude (decimal degrees)	Included in 2017 raster	Included in 2018 raster
Colorado	CO00	37.4414	−105.8653	x	x
	CO01	38.1178	−103.3161	x	x
	CO02	40.0553	−105.5883	x	
	CO06	39.7512	−104.9876	x	x
	CO08	39.4031	−107.3411	x	x
	CO09	39.4031	−107.3411	x	x
	CO10	38.9581	−106.9850	x	x
	CO11	39.8003	−105.1000	x	x
	CO15	40.5075	−107.7019	x	x
	CO19	40.3642	−105.5819	x	x
	CO21	39.1011	−105.0919	x	x
	CO22	40.8064	−104.7547	x	x
	CO84	40.0138	−105.3463		x
	CO85	40.0109	−105.2422	x	x
	CO86	39.9128	−105.1886	x	x
	CO87	39.7386	−104.9399	x	x
	CO90	40.0367	−105.5440	x	x
	CO91	37.4686	−106.7903	x	x
	CO92	39.4272	−107.3797	x	x
	CO93	40.5347	−106.7800	x	x
	CO94	39.9939	−105.4800	x	x
	CO96	37.7514	−107.6853	x	x
	CO97	40.5378	−106.6764	x	x
	CO98	40.2878	−105.6628	x	x
	CO99	37.1981	−108.4903	x	
Kansas	KS31	39.1022	−96.6092	x	x
	KS32	38.6717	−100.9164	x	x
	KS97	39.7603	−95.6358	x	
Nebraska	NE15	41.1528	−96.4912		x
	NE99	41.0592	−100.7464	x	
New Mexico	NM07	35.7817	−106.2675	x	x
Oklahoma	OK29	36.5908	−101.6175	x	
Utah	UT09	38.4550	−109.8217	x	x
	UT98	38.9983	−110.1653	x	
	UT99	37.6186	−112.1728		x
Wyoming	WY00	41.3761	−106.2594	x	x
	WY02	42.7339	−108.8500	x	x
	WY95	41.3647	−106.2408	x	x
	WY98	43.2228	−109.9911	x	x

Annual dry Nr deposition (dryfall) was obtained from the TDep raster data products available at: http://nadp.slh.wisc.edu/committees/tdep/tdepmaps/(accessed September 22, 2020). The TDep raster data were described by Schwede and Lear (2014) as a measurement-model fusion product created using the Community Multiscale Air Quality (CMAQ) model (a.k.a., dryfall) combined with the NADP wet-deposition raster data (a.k.a., wetfall). Raster data sets of spatially interpolated annual total N deposition (wetfall plus dryfall) were calculated by adding each annual NADP TDep dry deposition raster to the corresponding annual Nr wet-deposition raster for two scenarios: 1) including and 2) excluding the urban NADP site data. Total and wet-only deposition raster data were averaged for each of sub-basins 1–4 shown in Fig. 1. This resulted in four raster input data sets for SPARROW modeling conditions and scenarios for each year: 1) wet-only deposition with no urban data; 2) wet-only deposition with urban data; 3) wet plus dry deposition with no urban data, and 4) wet plus dry with urban data. The raster calculation and SPARROW modeling methodology is schematically illustrated in Fig. 3.

Currently, less than 10 percent of NADP National Trends Network (NTN) monitoring sites are classified as "urban," which are defined as sites within 15 km of an area with a population density exceeding 400 people km^{-2}. The NADP's interpolated annual wet-deposition raster products exclude data from urban sites. This is also true for the Total Deposition (TDep) raster products. Therefore, separate urban- and non-urban influenced wet Nr deposition raster data were generated specifically for this study by including and excluding NADP data from urban deposition sites located in the Denver – Boulder, Colorado metropolitan area (Table 1). For each year, the deposition raster that excluded the urban sites was subtracted from the deposition raster that included the urban sites to generate maps of urban wet Nr deposition (Fig. 4). Data for NTN sites NE99 and OK29 were excluded from the interpolations for 2018 due to incomplete data, which likely caused less constrained spatial influence of the urban site data in the northern and eastern regions of the wet deposition maps for 2018 as compared to 2017. However, the region outside of the drainage basin does not affect the surface-water loading analysis, which is constrained to the drainage basin.

2.2. SPARROW modeling

Hydrologic, chemical, and biological attenuation of the
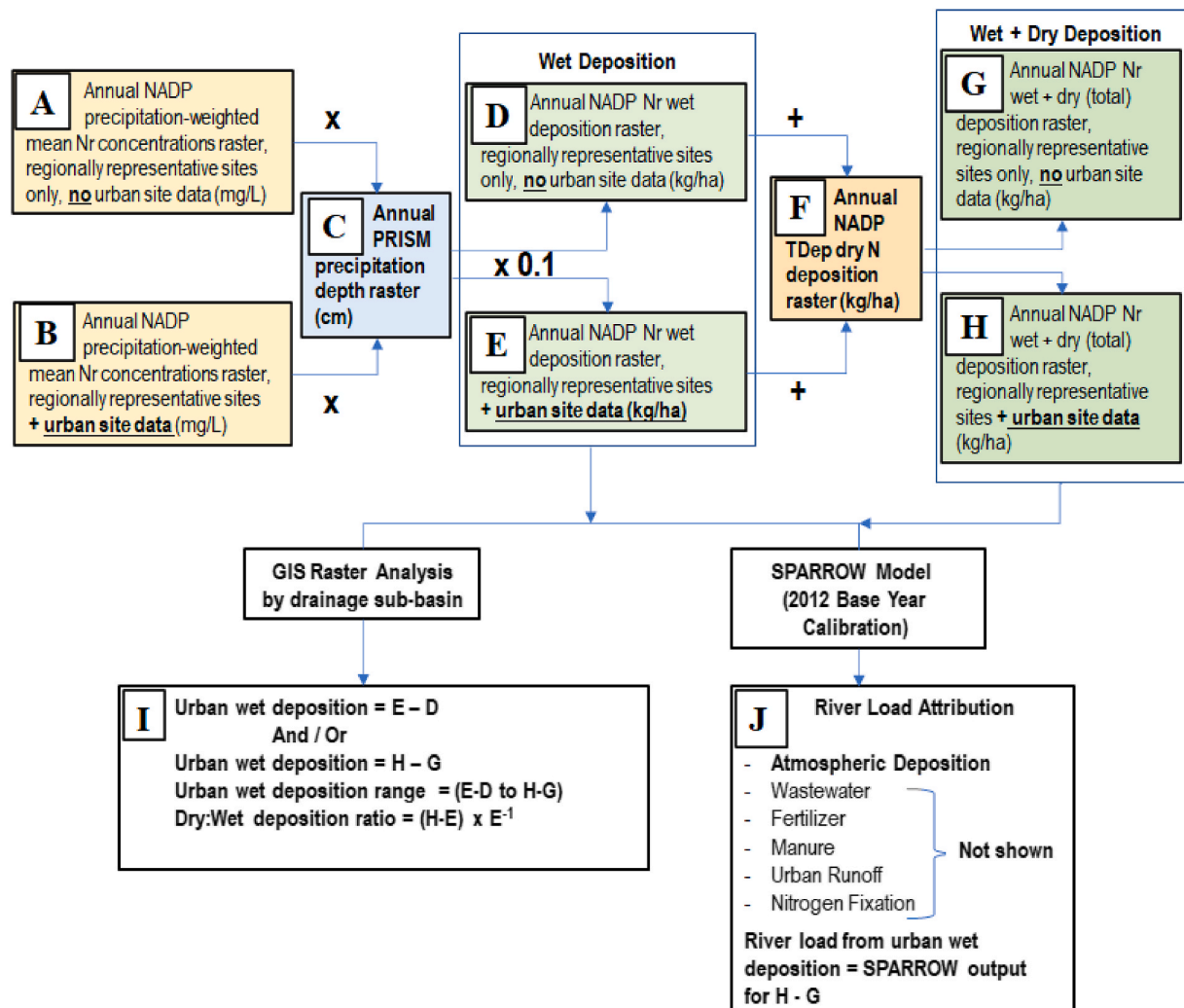
Fig. 3. Methodology for raster calculations and SPAtially Referenced Regressions On Watershed attributes (SPARROW) modeling scenarios used to estimate atmospheric deposition contributions to upper South Platte River loads for 2017–2018.
[NADP, National Atmospheric Deposition Program; TDep, NADP Total Deposition data products accessed at https://gaftp.epa.gov/castnet/tdep/grids/; mg/L, milligrams per liter; kg/ha, kilograms per hectare; PRISM, Parameter-elevation Relationships on Independent Slopes model; GIS, geographic information system; raster mathematical operators: x, multiply; +, add].

atmospherically deposited Nr is complex in urban environments, and precise accounting for these processes over large-scale watersheds and associated airsheds is infeasible with monitoring data alone. Therefore, rather than assuming Nr is transported in a conservative manner, such as with a simple delivery coefficient, the SPARROW model was used to estimate the contribution of AD to the South Platte River using process-based algorithms. SPARROW uses a hybrid mass-balance/statistical approach to estimate and track the non-conservative transport (i.e., transport with losses) of a constituent throughout a study area in relation to landscape properties describing the sources of a given constituent, such as fertilizer input or the amount of urban land, and factors affecting its transport, such as climate, soils, and instream/reservoir properties (Smith et al., 1997; Schwarz et al., 2006).

The SPARROW model was calibrated by relating water-quality measurements made at a network of monitoring stations to watershed attributes (Schwarz et al., 2006). Spatial variability in the environmental setting, described with land-to-water delivery variables in the model, is used to describe differences in how input from each nutrient source (in this case AD) is mediated by watershed characteristics during transport to the stream/river network. The SPARROW N model used in this study was calibrated for the Midwest part of the U.S. using data for the 15-year period 2001–2014 (median year 2012) (Robertson and

Saad, 2019). All model parameters and inputs from the non-atmospheric sources (shown in Table 2) were held constant for the model runs for this study.

Atmospheric Nr deposition rasters were substituted into the calibrated SPARROW model for each modeling scenario to estimate AD loading attribution. Wet-only deposition raster grids that excluded the TDep dry-deposition raster data were input to SPARROW to test the model's consistency and to compare the output for loads calculated using wet-only and total deposition independently. Urban wet deposition contributions to the river were estimated by subtracting the SPARROW-estimated loads without urban wet AD data from the SPARROW-estimated loads that included urban wet AD (Fig. 3). The SPARROW-estimated South Platte River N loads for each of the four sub-basins were compared for each scenario.

Dry AD is an important component of urban air pollution deposition (Decina et al., 2020; Bettez and Groffman, 2013; Lovett et al., 2000). However, no measurements of urban dry N AD or modeling approaches to compartmentalize urban dry AD were attempted for this study. Instead, the ratio of wet AD to dry AD was estimated from the raster data for each sub-basin, and the ratio was multiplied by the urban wet AD to estimate the urban dry AD. The calculated urban dry AD values were used to estimate contributions to South Platte River Segment 14 N loads.
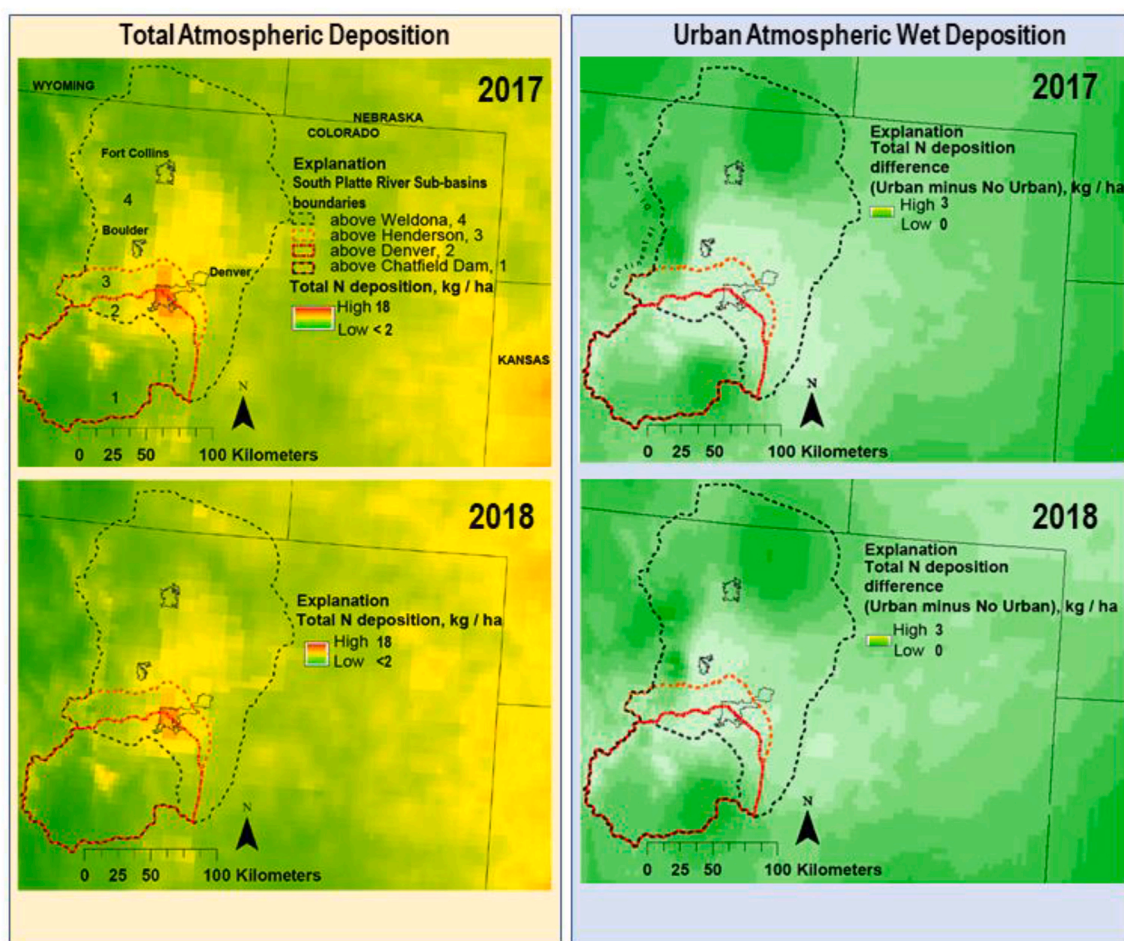
Fig. 4. Spatially interpolated National Atmospheric Deposition Program data for total (wetfall plus dryfall) atmospheric inorganic reactive nitrogen deposition (left) and urban wet-only deposition (right) calculated by subtracting the raster with no urban data from the raster with urban data, for 2017–2018.

Table 2
SPARROW input total nitrogen loads for original 2012 base-year calibration and 2017–2018 atmospheric deposition scenarios [Sub-basin numbers shown in Fig. 1; N, nitrogen; t, tonnes; WWTP, wastewater treatment plant; ha, hectares; data from Robertson and Saad, 2019].

Sub-basin/gaging station	2012 Base Year						
	Input N to sub-basins (t)						
	Drainage area (km²)	WWTP	Fertilizer	Manure	Atmo-spheric deposition (total)	Percent Atmospheric deposition	N-fixing crop area (ha)
4/Weldona	34,201	1971	16,263	38,227	11,505	17	523
3/Henderson	12,349	3272	201	161	1553	30	2
2/Denver	10,013	3764	87	269	1319	24	1
1/Chatfield Dam	7830	184	55	761	2953	75	0

	2017–2018 Atmospheric Deposition Scenarios					
	Input Atmospheric wet + dry N deposition to sub-basins (t)					
Sub-basin/gaging station	Original base-year		2017 including urban NADP data	2017 excluding urban NADP data	2018 including urban NADP data	2018 excluding urban NADP data
4/Weldona	11,505	958		818	935 808	
3/Henderson	1553	462		363	445 359	
2/Denver	1319	337		263	325 263	
1/Chatfield Dam	2953	287		226	278 227	

2.3. Estimated South Platte River total N loads for 2017–2018

Annual total N loads were estimated from streamflow and water-quality data for the South Platte River at Denver gage (sub-basin 2, Fig. 1). Streamflow data for sub-basin 2 were obtained from the National Water Information System (https://waterdata.usgs.gov/nwis; U.S. Geological Survey, 2021) and the Colorado Division of Water Resources

web site at: (https://dwr.state.co.us/Tools/Stations, (accessed February 5, 2021). Instantaneous and daily mean discharge were obtained for the South Platte River gaging stations at Chatfield Dam, Denver, Cherry Creek at Denver (W17), Bear Creek at Sheridan Boulevard, and the Englewood and Centennial wastewater reclamation plants (Fig. 1). Stream water-chemistry data were obtained from NWIS and the Denver Department of Public Health and Environment (Denver Department of

Public Health and Environment (DDPHE), 2020) for the N25E and W17 sites and the South Platte River at Denver and Cherry Creek at Denver gages. All streamflow and water-quality data used for this study are contained in an online data release at: https://doi.org/10.5066/P9UP346K (Wetherbee, 2021). The data release also contains details pertaining to sample collection and analysis, nitrogen loading estimation, and the calculation and use of runoff coefficients to estimate AD Nr delivery to the South Platte River at the Denver gage (Supplemental Information; Wetherbee, 2021).

Annual total N loads were estimated for the N25E and W17 sites using the U.S. Geological Survey LOADEST program adapted for R (rloadest) using instantaneous total N concentrations and measured flows (R Core Team, 2013 Runkel et al., 2004; https://github.com/USGS-R/rloadest). The 'automated model selection' function in the package was used to select the best regression model for predicting concentration based on the minimum AIC value of 10 predefined models that included discharge and time (seasonality and time) terms. Statistical diagnostic information for the rloadest models are available in the afore-mentioned data release (Wetherbee, 2021). LOADEST is based on correlations between N load and discharge. The selected models indicated acceptable correlations of N load and discharge with r-square values of 0.35 and 0.86 for 2017 and 2018, respectively. A file of daily discharge was used to estimate daily loads from the regression model using the predLoad function in the package. The daily loads were summed for each year to obtain annual loads for sites N25E and W17, and then the loads from these two sites were summed to estimate annual N loads of 781 and 521 tonnes (t) for 2017 and 2018, respectively at the Denver gage downstream from the Cherry Creek confluence.

2.4. Alternative approach to South Platte River N load attribution

A second approach to estimating the AD contribution to South Platte River N loads was implemented as a check on the SPARROW results. For this approach, it was necessary to estimate the portion of the annual total Nr AD that was delivered to surface water. For simplicity, it was assumed that the Nr AD in the part of the precipitation that contributes to streamflow was conservatively transported to surface water. The South Platte River hydrology is highly managed and complicated by several large reservoirs, imported water from other river basins, and numerous diversions for beneficial use. Therefore, estimation of annual runoff from precipitation was only attempted in the reach of the river between Chatfield Dam and the Denver gage (sub-basin 2) where there are few inflows and diversions. The results from this approach, which were similar to the SPARROW model output, are provided in the Supplemental Information.

3. Results

3.1. Atmospheric deposition

Results for total (wet plus dry) Nr AD and urban wet-only AD (the portion of the total attributed to urban air pollution) estimated within each of the four South Platte River sub-basins 1–4 are shown in Fig. 5. In the relatively undeveloped sub-basin 1 above Chatfield Dam (Fig. 1), urban Nr accounted for 25–28 percent of wet-only AD. Similarly, in sub-basin 4, urban Nr accounted for 22–27 percent of wet-only AD. As expected, urban Nr accounted for higher proportions of wet-only AD in the more intensely urbanized sub-basins 2 (Chatfield Dam to Denver) and 3 (Denver to Henderson) ranging 38–48 percent. Urban wet-deposited Nr accounted for 11–18 percent of total (wet + dry) Nr AD in the less urbanized sub-basins (1 and 4) and 20–25 percent in the urbanized sub-basins (2 and 3). Fig. 4 illustrates that urban air pollution effects extend well beyond the urban area, which was shown by Wetherbee et al. (2019).
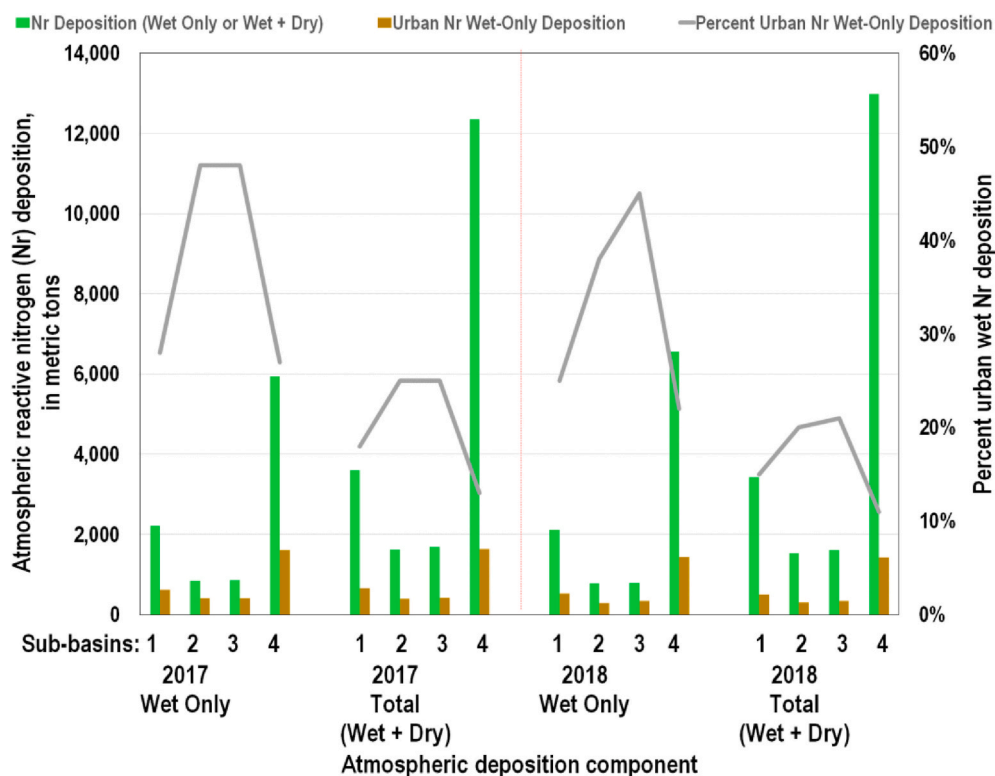

Fig. 5. Atmospheric wet-only and total reactive nitrogen (Nr) deposition to upper South Platte River sub-basins estimated from spatially interpolated National Atmospheric Deposition Program wet-only deposition data and Total Deposition (TDep) data products. Sub-basins 2 and 3 have higher urbanization intensity relative to sub-basins 1 and 4.

3.2. SPARROW model-estimated load attribution

The SPARROW model results attributed 25–44 percent of the estimated South Platte River total N load at the Denver gage to AD, approximately one third of which was attributed to urban wet Nr deposition (Table 3). By comparison, SPARROW indicated that AD Nr accounted for 80–84 percent of the model-estimated South Platte River total N load at Chatfield Dam, approximately one fifth of which was attributed to urban wet Nr AD. Sub-basin 1 above Chatfield Dam is predominately forested, whereas.

Sub-basin 2 below the dam contains both forest and urban land uses. In sub-basins 3 and 4, SPARROW indicated that AD Nr accounted for 14–22 percent of the annual total N loads at Henderson and Weldona. Sub-basins 3 and 4 contain mixed land uses with sub-basin 3 being much more urbanized than sub-basin 4.

The 2017 estimated total N load at the Denver gage (sub-basin 2) of 781 t compares well with the SPARROW model estimated total N load of 768 t when the NADP urban wet-deposition data are included in the model input (Table 3). The SPARROW base model was calibrated for conditions averaged over a 15-year period, and 2017 was a normal streamflow year. Conversely, the estimated 2018 total N load at the Denver gage was 521 t; comparing poorly with the 2018 SPARROW-estimated load of 756 t. Lower than normal precipitation and streamflow in 2018 resulted in lower N loads than normal. Therefore, the SPARROW base model calibration was likely more representative of 2017 conditions than 2018 conditions and suggests that contributions of Nr AD to total N loads in the South Platte River during 2018 were likely underestimated by SPARROW. There are other factors intrinsic to the model that others have shown to have caused underestimation of loads by SPARROW (Burns et al., 2021).

4. Discussion

The results of this study are not necessarily transferable to other watersheds. Comparison of the South Platte River estimates to other studies reveals the watershed-specific nature of AD loading. For example, estimated AD Nr contributions to the waters of the Blue Ridge Mountains (7 percent; Buell and Peters, 1988) and Narraganset Bay (8 percent; Morris, 1991) are much lower than contributions estimated of the South Platte River. Several studies of AD N contributions to Chesapeake Bay are cited and synthesized by Burns et al. (2021), including investigations using SPARROW. Although 60–73 percent reductions in both wet and dry AD of oxidized nitrogen and dry AD of reduced nitrogen occurred during 1995–2019, AD contribution to Chesapeake Bay N loading is still approximately 25 percent (Burns et al., 2021). Another study by Burian et al. (2001) found that approximately 88 percent and 90 percent of stormwater runoff NO_3^- and NH_4^+ loads, respectively, were from wet deposition in a Los Angeles catchment, and their results are consistent with other studies in Pennsylvania and Florida, which they cite. Watershed characteristics for the catchments in each of these studies were not directly compared, but others have shown that they affect the wide range of surface-water N loads attributed to AD (Mustard et al., 1987;, Campbell et al., 2000; Clow et al., 2018). Because the SPARROW model accounts for watershed characteristics, it must be run for unique urbanized watersheds to estimate urban contributions of Nr AD to surface-water loads. In addition to watershed-specific characteristics, differences in the results may be due to study methods. Inclusion of urban wet AD data is unique to this study's methodology. This points to the need for more AD measurements in urban areas as well as improved source attribution models and data analysis techniques.

The SPARROW results suggest that Nr AD contributes approximately 25–44 percent of the annual total N load in the South Platte River at Denver, about one-third of which is attributed to wet deposited Nr from urban air pollution. Using the LOADEST model-estimated loads and SPARROW model source attribution, approximately 344 and 224 t N from AD passed the Denver gage in 2017 and 2018, respectively, approximately 78 and 42 t of which was from wet Nr AD of urban air pollution. Using the 15-min discharge values from the Denver gage and the Segment 14 TMDL model NO_3^- concentration of 12 mg/L for this stream gage (https://cdphe.colorado.gov/tmdl-south-platte-river-basin), estimated Nr wet-only AD from urban air pollution accounted for approximately 11 percent of the TMDL for 2017 and 2018, assuming complete nitrification of the wet AD Nr simply for comparison purposes. Factoring in dry deposition at a 1:1.8 dry:wet ratio, according to the

Table 3

Results of the SPAtially Referenced Regressions On Watershed attributes (SPARROW) 2012 base model with substituted 2017 and 2018 raster data for atmospheric inorganic reactive nitrogen (Nr) deposition for selected South Platte River sub-basins. Two scenarios where modeled for each year with urban site data included and excluded. [AD, atmospheric deposition; t•yr⁻¹, tonnes per year; kg/ha•yr⁻¹, kilograms per hectare per year; NADP/TDep, National Atmospheric Deposition Program/Total Deposition measurement-model fusion; N, annual total nitrogen; %, percent].

Sub-basin/gaging station	NADP/TDep annual AD N loads (t•yr⁻¹)		SPARROW-estimated AD contribution to South Platte River N load (range, %)	SPARROW-estimated South Platte River N load [t•yr⁻¹ (kg/ha•yr⁻¹)]		SPARROW-estimated urban wet AD contribution to South Platte River N load [t•yr⁻¹ (kg/ha•yr⁻¹) %]	
	NADP urban data			NADP urban data			
	Included	Excluded		Included	Excluded		
Scenario: 2017 Total (wet + dry) atmospheric deposition							
4/Weldona	958	818	20–22	4284 (*2.0*)	4144 (*1.9*)	**141** (*0.06*)	3
3/Henderson	462	362	14–17	2715 (*12*)	2616 (*11*)	**100** (*0.43*)	4
2/Denver	337	263	38–44	768 (*3.5*)	694 (*3.2*)	**75** (*0.34*)	10
1/Chatfield	287	226	80–84	343 (*0.44*)	283 (*0.36*)	**60** (*0.08*)	18
Scenario: 2017 Wet only atmospheric deposition							
4/Weldona	557	416	11–14	3883 (*1.8*)	3742 (*1.7*)	**141** (*0.06*)	4
3/Henderson	296	196	8–12	2549 (*11*)	2449 (*10*)	**100** (*0.43*)	4
2/Denver	221	146	25–34	652 (*3.0*)	577 (*2.6*)	**75** (*0.34*)	12
1/Chatfield	192	131	70–77	249 (*0.32*)	188 (*0.24*)	**61** (*0.08*)	24
Scenario: 2018 Total (wet + dry) atmospheric deposition							
4/Weldona	935	808	20–22	4261 (*1.9*)	4134 (*1.9*)	**128** (*0.06*)	3
3/Henderson	445	359	14–16	2698 (*12*)	2612 (*11*)	**86** (*0.37*)	3
2/Denver	325	263	38–43	756 (*3.5*)	694 (*3.2*)	**62** (*0.28*)	8
1/Chatfield	278	227	80–83	334 (*0.43*)	284 (*0.36*)	**50** (*0.07*)	15
Scenario: 2018 Wet only atmospheric deposition							
4/Weldona	524	396	11–14	3850 (*1.8*)	3722 (*1.7*)	**128** (*0.06*)	3
3/Henderson	276	189	8–11	2529 (*11*)	2442 (*10*)	**87** (*0.37*)	3
2/Denver	207	145	25–32	639 (*2.9*)	576 (*2.6*)	**63** (*0.29*)	10
1/Chatfield	182	130	70–76	238 (*0.30*)	187 (*0.24*)	**51** (*0.07*)	22

results in Table 3, urban air pollution could account for approximately 17 percent of the TMDL-modeled daily load for NO_3^- at the Denver gage. The South Platte River Segment 14 NO_3^- TMDL is evaluated at the Burlington Ditch headgate at a concentration of 10 mg/L (https://cdph e.colorado.gov/tmdl-south-platte-river-basin), which is 17 percent less than the TMDL-model target concentration for the Denver gage. Therefore, Nr AD of urban air pollution accounted for as much as 20 percent of the NO_3^- TMDL at the Burlington Ditch headgate.

An important consideration for attempting to control urban air pollution loading to surface water in the Denver metropolitan area is the proximity of emission sources located outside of the urbanized watersheds, which is common among many cities (Chang et al., 2019; Singh and Kulshrestha, 2014; Hu et al., 2014). Sources of N emissions, in both inorganic and organic forms were identified and correlated with detection of trace gases and meteorology, in the northern portion of the South Platte River sub-basin, by Benedict et al. (2018). They traced air pollutants measured in Rocky Mountain National Park, located on the western edge of the South Platte River basin, to emissions from urban centers, agricultural operations, and oil-and-gas development in northeastern Colorado. These emissions are transported more than 100 km west to Rocky Mountain National Park by easterly upslope flow, and it can be argued that such events also affect Nr AD in the Denver metropolitan area (Wetherbee et al., 2019).

Regional transport of N emissions in the South Platte River basin by upslope processes is well documented in numerous studies pertaining to their effects on Rocky Mountain National Park (Malm et al., 2013; Benedict et al., 2013). East to west NH_3 concentration gradients across the South Platte River basin discovered by ambient air sampling over several years (Li et al., 2017) suggest that animal feeding operations located north of the Denver metropolitan area have the potential to affect regional air quality for at least an 80 km distance. Therefore, the mixture of urban and agricultural emissions, especially during upslope events, complicates strategies for reducing surface-water N loading from AD in Segment 14 of the South Platte River.

Additional urban NADP data are needed to evaluate the relative importance of urban, agricultural, and regional background atmospheric N loading to streams. Regionally representative NADP sites are also needed within approximately the same distance as the spatial interpolation radius (e.g., 50–100 km) to the urban sites, to avoid unrealistic expansion of the radius of influence of urban Nr AD in the deposition raster data. This was especially true for this study in the forested and predominantly undeveloped and agricultural sub-basins 1 and 4, respectively. An example of preferred monitoring site spatial density is in sub-basin 4 west of Boulder where NADP sites at the Betasso Preserve (CO84) and Sugarloaf (CO94) are considered to be urban sites by NADP criteria due to their proximities to Boulder (5 km and 10 km, respectively), even though they are in forested, rural environments. The CO84 N wet AD concentrations are very similar to those measured at the NADP site CO85 in Boulder, whereas the CO94 N concentrations are much lower, demonstrating a concentration and AD gradient within just 10 km from the urban area (Wetherbee et al., 2019, 2021). Controlling the spatial interpolation of NADP data was particularly problematic for the 2018 deposition raster (Fig. 4), which suffered predominantly from missing data for sites NE99 and OK29. Data from NE99 and OK29 constrained the radius of influence of the urban site data for the 2017 deposition raster. Several new NADP/NTN sites began monitoring AD north of the Denver metropolitan area in 2021, and these data might be useful to help constrain interpolation of the urban Nr concentrations for wet-deposition mapping.

5. Conclusions

The National Atmospheric Deposition Program (NADP) wet-only and wet-plus-dry (TDep) atmospheric deposition (AD) data products were used with the SPAtially Referenced Regressions On Watershed attributes (SPARROW) model to discover that AD contributes appreciably to South Platte River annual total nitrogen (N) loads, in both urbanized and adjacent undeveloped areas. In Segment 14 of the South Platte River, reactive nitrogen (Nr, nitrogen from nitrate (NO_3^-) plus ammonium) from urban air pollution alone accounted for up to 20 percent of the TMDL for NO_3^-. Controlling sources of AD loading to surface water is complicated by numerous emission sources located both within and outside of the urbanized watersheds combined with local meteorological conditions. Interpolation of AD data using two slightly different spatial distributions of monitoring stations produced remarkably different interpolated AD raster data sets for 2017 and 2018, indicating the need for monitoring not only in the urban corridor but also just outside of it where N emissions are expectedly lower. This demonstrates that locations and density of AD monitoring stations in a watershed must be carefully optimized to evaluate AD effects on the landscape and surface water.

This study demonstrates the value of integrated urban atmospheric deposition and surface-water quality monitoring combined with watershed modeling to evaluate air-pollution effects on water resources, which are commonly overlooked. Monitoring of the air-pollution sources of Nr with the intention of affecting water quality in streams could lead to engineered controls and policy considerations that reduce N impairment loads and remove waters from the 303(d) lists.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.jenvman.2021.113861.

Credit author statement

Gregory Wetherbee: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, writing – both draft and final versions, Visualization, Supervision, Project administration, Funding acquisition. Michael Wieczorek: Methodology, Software, Formal analysis,. Dale Robertson: Methodology, Software, Data curation, Writing – review & editing. David Saad: Software Resources, Writing – review & editing. Jon Novick: Formal analysis, Data curation, writing of draft. M. Alisa Mast: Formal analysis, Validation, writing of draft, Writing – review & editing.

Availability of data and material

All data are available from the NADP at: http://nadp.slh.wisc.edu/l ib/qaplans/QAPNADPLab2020.pdf; the USGS ScienceBase Catalog at: https://doi.org/10.5066/P9OOIQ0E and https://doi.org/10.5066

/P9UP346K; and the U.S. Geological Survey National Information Water System (U.S. Geological Survey, 2021, https://doi.org/10.5066/F7P55KJN).

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