
Contribution of trace metals from atmospheric deposition to stormwater runoff in a small impervious urban catchment

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ABSTRACT

The contribution of atmospheric deposition to emissions of trace metals in stormwater runoff was investigated by quantifying wet and dry deposition fluxes and stormwater discharges within a small, highly impervious urban catchment in Los Angeles. At the beginning of the dry season in spring 2003, dry deposition measurements of chromium, copper, lead, nickel, and zinc were made monthly for one year. Stormwater runoff and wet deposition samples also were collected, and loading estimates of total annual deposition (wet + dry) were compared with annual stormwater loads. Wet deposition contributed 1% to 10% of the total deposition inside the catchment, indicating the dominance of dry deposition in semi-arid regions such as Los Angeles. Based on the ratio of total deposition to stormwater, atmospheric deposition potentially accounted for as much as 57% to 100% of the total trace metal loads in stormwater within the study area. Despite potential bias attributable to processes that were not quantified in this study (e.g., resuspension out of the catchment or sequestration within the catchment), these results demonstrate atmospheric deposition represents an important source of trace metals in stormwater to waterbodies near urban centers.

INTRODUCTION

Urban stormwater runoff can be highly contaminated with heavy metals and other toxic compounds, representing a significant non-point source of pollution to waterbodies within and adjacent to urban centers (Sansalone and Buchberger 1997, Smullen *et al.* 1999, Buffleben *et al.* 2002). In Southern California, mass emissions from urban stormwater runoff can be higher than from point sources, e.g., wastewater treatment plants and industrial discharges (Schiff *et al.* 2000). Furthermore, urban stormwater runoff can be toxic

to aquatic organisms, and trace metals may be one of the constituents responsible for this toxicity (Marsalek *et al.* 1999, Schiff *et al.* 2002, Greenstein *et al.* 2004).

While future water quality improvements in urban areas may depend on contaminant reduction from stormwater, many of the trace metal sources to urban stormwater have not been well characterized. In semi-arid regions such as Southern California, pollutants may build-up on impervious surfaces during the extended dry season, and subsequently wash-off into nearby waterbodies once the wet season begins. Atmospheric deposition may be especially important as a source of pollutants to stormwater in these regions because significant quantities of trace metals and other pollutants are emitted into the atmosphere daily (SCAQMD 2003), and the ultimate fate of the trace metals in particular is unknown.

Yet despite this potential, there are relatively few studies specifically targeting the pollutant contribution of atmospheric deposition to urban stormwater runoff in Los Angeles. The majority of atmospheric deposition research has focused on areas such as the Great Lakes and Chesapeake Bay regions (Lin *et al.* 1993, Baker *et al.* 1997, Paode *et al.* 1998). These areas have different atmospheric emissions and climatic parameters, and greater precipitation than Southern California, which may increase the importance of wet vs. dry deposition. Studies specific to urban atmospheric deposition have been limited even though urban areas have been shown to have higher deposition rates for a number of pollutants, including trace metals (Galloway *et al.* 1982, Yi *et al.* 2001). The present research was designed to quantify the contribution of atmospheric deposition of trace metals to stormwater loadings in a small urban catchment in Los Angeles.

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METHODS

Los Angeles has a semi-arid climate, with an average annual rainfall of 33 cm. Typically, the bulk of this precipitation occurs from December to March. Starting with the beginning of the dry season in May 2003 and continuing for one year, dry deposition and atmospheric concentrations of chromium, copper, nickel, lead and zinc were measured for 48-hours once a month, on days without rain within a defined catchment in Los Angeles. Concentrations of trace metals in rain and stormwater within the catchment were measured from December 2003 to March 2004. The data were used to estimate the contribution of atmospheric deposition to stormwater loadings within the catchment. The site was selected to minimize sources of trace metals to stormwater within the catchment other than urban background atmospheric deposition.

Site description

The catchment was located in the San Fernando Valley of Los Angeles, California, within the grounds of a water reclamation plant. This site was suitable for this study as (1) the land surface was relatively flat; (2) the plant was surrounded by an earthen berm, preventing surface runoff from surrounding areas from entering the catchment; (3) sources of metals inside the plant were limited because of restricted access and lack of major industrial activities within the plant. Virtually all of the surface flow from the catchment was routed through a single catch basin, which was the site of runoff collection. The estimated drainage area to the catch basin was 5 ha based upon facility storm drain plans, discussions with the Plant Engineer, visual inspection, and on site measurements. The drainage area consisted primarily of impervious surfaces including asphalt roads, concrete sidewalks, and concrete structures with monolithic poured foam roofs. Unpaved dirt and vegetated areas covered <20% of the drainage area. A runoff coefficient of 1.0 was assumed because pervious areas were not subject to substantial infiltration. Evaluation of this assumption led to minimal bias and any overestimation of the runoff volume would result in conservative estimates of stormwater discharges. Traffic inside the plant was limited to ~50 vehicles per day, and streets were cleaned weekly.

Instrumentation

Dry deposition measurements were made using a

modification of surrogate surfaces used by Paode *et al.* (1998) and Lin *et al.* (1993). Surrogate surfaces for this study were comprised of a circular PVC deposition plate, 33 cm in diameter, with a sharp edge (<10 degree angle), covered with a Mylar® sheet coated with Apezion L grease. The grease was liquefied by heating and then painted onto the Mylar® film to obtain a thin, uniform 10 mm layer. During sampling, the plate was mounted onto a tripod at a height of 2 m. Atmospheric concentrations of trace metals on total suspended particulate (TSP) were collected using a filter based sampling system attached to a vacuum pump. The open-faced inlet was loaded with a 37 mm, 2.0 µm pore Teflon® filter, and sampling was done at a flow rate of 10 l/min. The open-faced inlet was expected to reduce large particle losses to the walls and inlets typical of conventional impactor samplers. Wind speed and direction, temperature, and relative humidity were measured using a portable meteorological station (PortLog, Rain Wise, Inc., Bar Harbor, Maine).

Event-based wet deposition samples were collected using an automated rainwater collector developed by the National Atmospheric Deposition Program (NADP; 1997). The cover opened during periods of precipitation and closed when precipitation ended, eliminating evaporation from the sampler and preventing contamination of the sample. A pre-cleaned container was used for each event.

Flow-weighted composite stormwater samples were collected during each storm in 500 ml plastic bottles using an ISCO 6700 automated stormwater sampler, which also logged flow to determine runoff quantity.

Sample preparation and analysis

For the deposition plates, Mylar® sheets were cut into 30 cm diameter circles, wiped with methanol and soaked in 10% nitric acid followed by methanol for 5 min each, then rinsed with distilled water, and allowed to air dry. Each sheet was coated with a thin layer of grease, mounted on a deposition plate, and stored in clean, airtight containers for transport to the field. After sampling, the Mylar® sheets were removed, folded (greased side inward), and placed inside a clean glass jar. In the laboratory, Mylar® sheets were cut into ten smaller pieces and rinsed three successive times with 15 ml of n-hexane. The rinses were combined into a 50 ml centrifuge tube. The Mylar pieces were then rinsed with 5% Optima Grade nitric acid and the acid and hexane rinses were combined. The hexane was evaporated in a

50°C water bath and the sample was acid-digested at 65°C under sonication for a minimum of 24 hours.

Prior to sampling, a clean Teflon® filter was loaded into the TSP sample holder, and the sample holder was stored in a clean plastic bag for transport to the field. After sampling, the filter was stored in a clean petri dish prior to analysis. In the laboratory, Teflon® filters were placed into clean 15 ml plastic centrifuge tubes and 10 ml of 5% Optima Grade nitric acid was added and the tubes capped tightly. The samples were acid-digested at 65°C under sonication for a minimum of 24 hours.

For wet deposition and stormwater analyses, collection vessels were cleaned with soap and water, soaked in 10% nitric acid and rinsed with distilled water. All stormwater samples from a given storm were acidified to pH 2 with ultra-pure nitric acid and stored at 4°C. A representative composite from each storm was digested by acidification to pH <2 using HNO₃ for a minimum of 16 hours.

All acid-digested samples were transferred to a centrifuge tube and analyzed for metals per EPA Method 200.8 using inductively coupled plasma-mass spectroscopy. Method detection limits (MDL) ranged from 0.5 - 1.0 ng corresponding to <0.01 µg/m²/day for flux and <0.03 ng/m³ for concentration based on the sampling times and air volumes collected. The MDL for metals in precipitation and stormwater was <0.1 µ/L for all metals. A five-point external calibration curve, laboratory blank, matrix spike, and matrix spike duplicate were measured with each batch of fifteen or less samples to ensure quality. Matrix spike recoveries were within 99-107% for all metals. Matrix spike duplicates were within 10% of the original spike for all metals (relative percent difference or RPD). All laboratory blanks were nondetectable. Field blanks (greased Mylar® sheets mounted on a deposition plate, Teflon® filters loaded into a TSP sampling cartridge, stormwater sample bottles filled with distilled water) were prepared, taken to the field, and analyzed along with the samples. All field blanks contained detectable levels of trace metals, and all samples were corrected for their respective field blank. Field blank corrections were typically <20% of the sample mass for copper, lead and zinc, but up to 100% of sample mass for chromium and nickel. Field duplicates indicated the precision of the deposition plates for each of the five metals, on average, was 31% (chromium), 25% (copper), 24% (lead), 87% (nickel) and 47% (zinc) RPD. This was an acceptable level

of precision for field duplicates because differences of less than a factor of two between fluxes measured during different sampling events were not considered significant.

Mass loading calculations

Annual dry deposition mass loadings were calculated for each metal by multiplying the mean daily flux from each sampling event by the number of dry days between that sampling event and the next. These loadings were then summed to obtain the total annual load inside the catchment. It was assumed no dry deposition occurred during periods of rain. Any errors introduced by this assumption would be small because of the limited number of days with precipitation that occurred during the year.

The annual event mean concentration (EMC) was calculated for both rainwater and stormwater using Equation 1:

$$C_m = \frac{\sum_{i=1}^n (C_i V_i)}{\sum_{i=1}^n V_i} \quad (1)$$

Where: C_m = Annual EMC for population j ; C_i = Concentration during storm event i ; V_i = Weighting factor - total volume sampled for event i ; and n = Number of storm events sampled.

For wet deposition, the total mass loading for each metal was then calculated by multiplying the rainwater annual EMC by the area of the catchment and the total volume of rainfall during the year, which was obtained from published precipitation data from the Sepulveda Dam Rain Gauge (NOAA 2003, 2004), located less than 1.5 km from the catchment.

The individual wet deposition flux for each storm was calculated by multiplying the rainwater EMC by the catchment area and the volume of rainfall from a single storm. The mean storm flux provided a better comparison to the mean daily dry deposition flux because it more closely approximated a daily wet deposition value than the annual flux.

The mass loadings of trace metals in stormwater were calculated by multiplying the stormwater annual EMC (Equation 1) by the total volume of runoff during the storm season. To obtain stormwater volumes, standard hydrologic equations were used based on water level, slope, and roughness of the storm drain pipe from which samples were collected. Water level was measured using a bubbler. Pipe slope and roughness were provided by the facility manager. Flow estimates were calibrated using the

relationship between rainfall, measured runoff volumes, and catchment area (Figure 1) to account for uncertainties in the inputs and assumptions used in the flow calculations (e.g., estimated slope, assumption of uniform flow, etc.). The relationship between rainfall and runoff was significant ($R^2 > 0.99$) and this regression was used to estimate runoff volume for storms that were not sampled, providing a good approximation of the total runoff volume inside the catchment for the entire year. From the y-intercept of the regression equation, when rainfall was < 0.15 cm, runoff volume was zero. This was supported by observations at the site.

RESULTS

Dry atmospheric deposition fluxes and atmospheric concentrations

The TSP detection frequency was 100% for all trace metals except chromium, which was 92%. Atmospheric concentrations of trace metals on TSP were relatively stable over time (Figure 2a). The ranges of chromium, copper, lead, and zinc concentrations were all within factors of two during the year-long survey, while nickel concentrations were the most variable, but still within a factor of four. Deposition plate detection frequencies were 100% for all metals except nickel, which was only detected in $\sim 50\%$ of the samples. With the exception of a single event, discussed below, dry deposition fluxes were normally distributed and not highly variable over time during the course of this study (Figure 2b). Dry deposition fluxes of all metals ranged within factors of 2 to 5 from their mean values. For all five metals, deposition fluxes were not significantly correlated with meteorological parameters including mean daily wind speed, temperature and relative

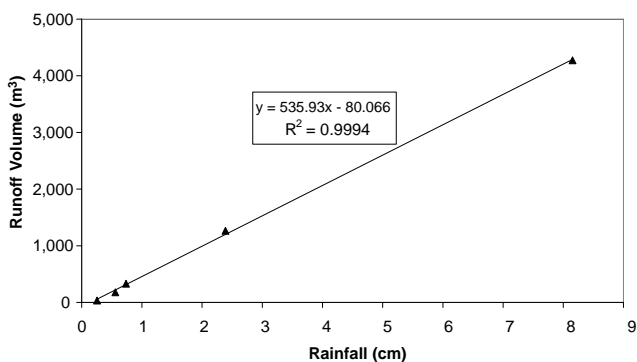


Figure 1. Linear Regression of rainfall vs. runoff measured in an urban Los Angeles catchment.

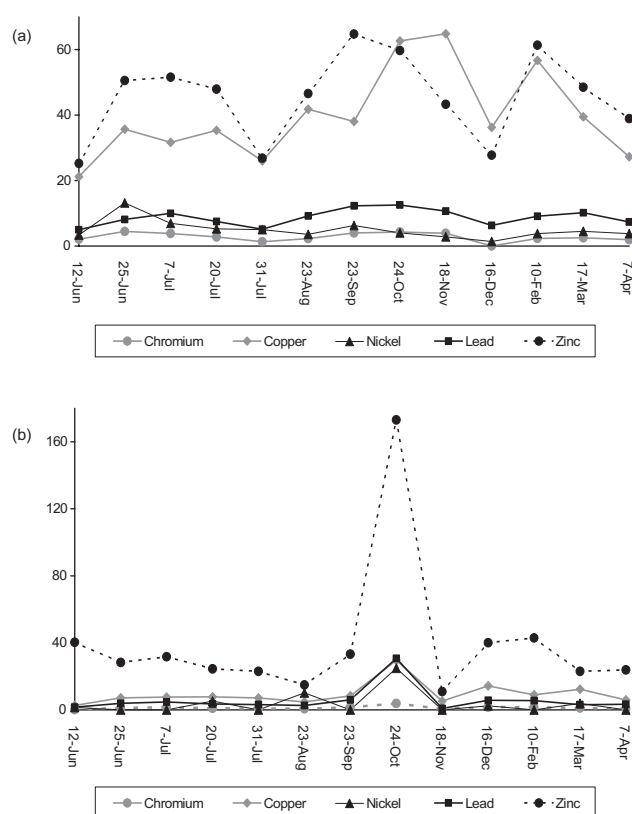


Figure 2. Time series of atmospheric concentrations (a) and dry deposition fluxes (b).

humidity, maximum 10-minute wind speed, and antecedent rainfall days ($p > 0.05$).

Forest fires in nearby mountains and offshore (i.e., Santa Ana) wind conditions occurred during a single sampling event in October. The highest fluxes for all metals were measured during this unique event. While the sample size limited the application of statistical tests of significance, it is interesting to note the fluxes measured during these unusual conditions of forest fires were factors of four (chromium and copper), six (zinc), eight (lead), and thirteen (nickel) times greater than the mean fluxes for all other sampling events.

Storm events

There were twenty-one rainfall events inside the catchment during the period from October 2003 through April 2004 (Figure 3). The total amount of rainfall from these events was 20 cm, with $\sim 75\%$ of the total rainfall for the season produced by only three storms. Samples of rainwater were collected from seven events, comprising $\sim 70\%$ of the total rainfall for the season. Ten rainfall events had sufficient volume to generate runoff within the catch-

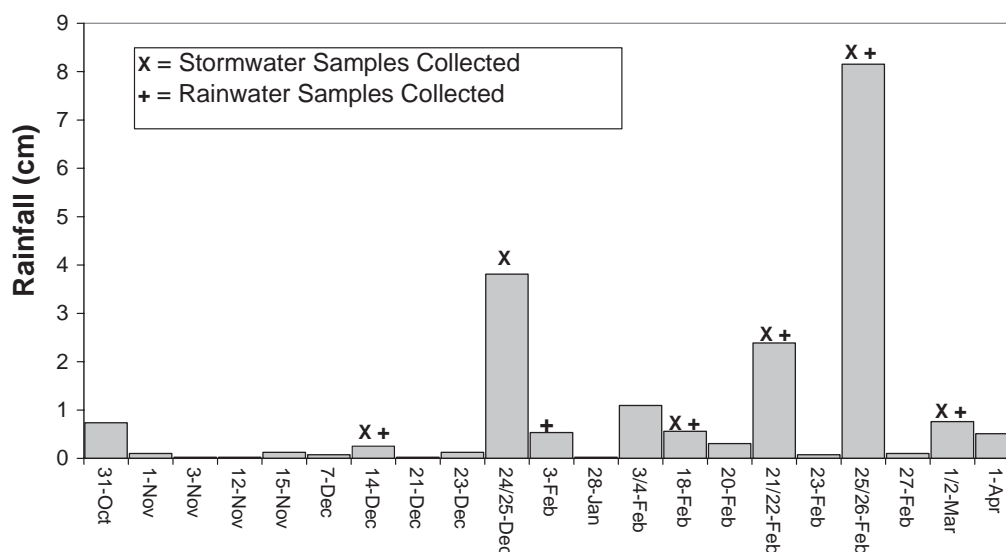


Figure 3. Rainfall in the catchment during the 2003-2004 storm season.

ment. Stormwater runoff samples were collected from six of these events, comprising ~50% of the total stormwater runoff inside the catchment during the season.

Detection frequencies in rainwater were low for most metals (Table 1). The highest concentrations for all metals were from the December 14, 2003 storm. The rainwater annual EMCs for each of the five metals were an order of magnitude lower than the rainwater concentrations from the December 14th storm. The relative proportions of metals in rainwater and on atmospheric TSP at the site were similar, indicating particle scavenging from the atmosphere was the likely source of these metals in precipitation (Figure 4).

Chromium, copper, nickel, lead, and zinc were detected in 100% of the stormwater runoff samples (Table 1). The highest concentrations of chromium and zinc were observed during the first sampled storm of the season (December 14, 2003), while the highest concentrations of copper, lead, and nickel were observed during the largest storm of the season (February 25, 2004). No relationship was evident between stormwater concentrations and parameters such as storm intensity, mean or peak flow rates, or antecedent rainfall days using regression models ($p > 0.05$). Thus, the annual EMCs were used to estimate the loads of trace metals in stormwater runoff within the catchment.

Table 1. Concentrations of metals in precipitation and stormwater inside the catchment.

	Detection Frequency	Range (µg/l)	Annual Event Mean Concentration ± standard error (µg/l)
Precipitation			
Chromium	14%	b.d.-2.2	0.09 ± 0.06
Copper	86%	b.d.-14	1.0 ± 0.6
Lead	29%	b.d.-5.0	0.15 ± .09
Nickel	71%	b.d.-3.2	0.19 ± 0.12
Zinc	43%	b.d.-210	7.8 ± 4.9
Stormwater			
Chromium	100%	2.1-20	3.1 ± 1.6
Copper	100%	5.9-37	27 ± 24
Lead	100%	1.2-16	12 ± 10
Nickel	100%	2.1-8.5	6.6 ± 5.2
Zinc	100%	32-320	160 ± 130

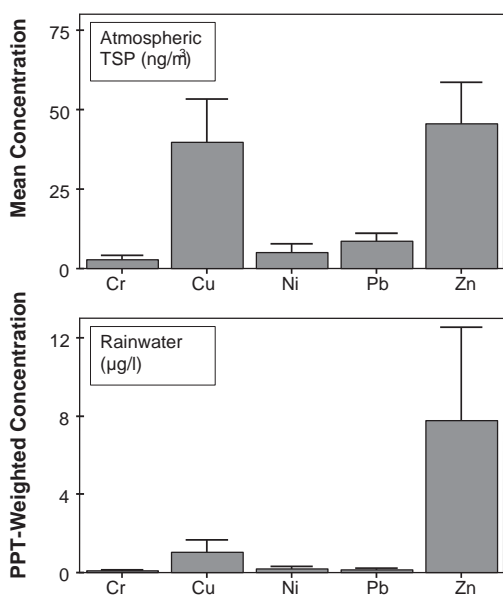


Figure 4. Mean trace metal concentrations on atmospheric TSP (a) and in precipitation measured in the catchment (b). Error bars represent the standard error of the mean.

Wet vs. dry deposition flux compared to stormwater loading estimates

Based on the total annual flux, dry deposition fluxes were substantially greater than wet deposition fluxes (Table 2). Wet deposition comprised 1% to 10% of the total annual deposition (wet+dry) inside the catchment. For all five metals, the mean wet deposition fluxes per storm (which typically lasted <1 day) were the same order of magnitude as the daily dry deposition fluxes (Table 2); the differences between individual storm wet fluxes and daily dry fluxes ranged from a factor of one to four for all metals. Only zinc had a higher mean wet deposition flux per storm compared with the daily dry deposition flux.

For each metal, the estimated mass of cumulative wet and dry atmospheric deposition to the catchment was similar to the estimated mass of trace metals discharged from the catchment through stormwater runoff (Table 3). Annual wet and dry deposition mass ranged from 57% (for zinc) to approximately 100% (for nickel and lead) of the

Table 2. Comparison between wet and dry deposition fluxes; ranges indicated in parenthesis.

Metal	Wet Deposition Fluxes				Dry Deposition Fluxes			
	Annual Flux ^a (µg/m ² /yr)		Average Flux Per Storm (µg/m ² /storm)		Annual Flux ^b (µg/m ² /yr)		Average Daily Flux (µg/m ² /day)	
Chromium	18	(0-45)	0.84	(0.07-1.6)	440	(250-620)	1.3	(0.7-1.8)
Copper	200	(0-520)	9.6	(0.9-18)	3,211	(1,800-4,600)	9.4	(5.3-14)
Lead	29	(0-74)	1.4	(0.1-2.6)	2,000	(390-3,600)	5.8	(1.1-10)
Nickel	38	(0-96)	1.8	(0.2-3.4)	1,300	(0-2,700)	3.7	(0-8.0)
Zinc	1,500	(0-3,900)	73	(6-140)	13,000	(4,900-22,000)	39	(14-64)

^aOctober 2003 - April 2004 storm season

^bMay 2000 - April 2004 dry days

Table 3. Comparison of metal loadings from atmospheric deposition and stormwater runoff from may 2003 - April 2004 (g/year).

	Chromium	Copper	Nickel	Lead	Zinc
Wet Deposition	1	10	2	1	77
Dry Deposition	22	160	63	99	670
Stormwater Runoff	32	230	59	93	1300
Wet Deposition/Stormwater	0.03	0.04	0.03	0.01	0.06
Dry Deposition/Stormwater	0.69	0.70	1.07	1.06	0.52
Total Deposition(Wet+Dry)/Stormwater	0.72	0.74	1.10	1.08	0.57

Table 4. Comparison of measured air concentrations and fluxes of trace metals.

	Year	Chromium	Copper	Lead	Nickel	Zinc
Air Concentration (ng/m³)						
This Study	2003-2004	2.8	40	9	4.9	46
Los Angeles ^a	2002-2003	4.9	52	14	9.2	84
Los Angeles ^b	1998-1999	4.9	39	25	8.7	106
Total Deposition Flux: wet+dry (mg/m²/year)						
This Study	2003-2004	0.46	3.4	2.0	1.3	14.5
Lake Michigan ^c	1993-1994	0.20	1.9	1.6	0.6	6.0
Lake Superior ^c	1993-1994	0.21	3.1	1.5	0.8	8.8
Lake Erie (urban influenced) ^c	1993-1994	1.06	4.2	1.8	0.7	16.5
Chesapeake Bay Atmospheric Deposition Study (non-urban) ^d	1990-1992	0.35	0.60	1.2	0.93	3.7
Wye	1990-1992	0.25	0.67	1.1	0.71	3.5
Elms	1990-1992	0.20	0.85	1.2	1.1	7.1
Haven Beach	1990-1992	0.20	0.85	1.2	1.1	7.1

^aLim *et al.* In press

^bSCAQMD 2000

^cSweet *et al.* 1998

^dBaker *et al.* 1997

annual stormwater load. Annual dry deposition had the greatest potential for influencing stormwater mass emissions. Between 52% (for zinc) and approximately 100% (for nickel and lead) could be attributed to dry deposition alone. Moreover, rainwater concentrations were typically more than an order of magnitude lower than concentrations in stormwater runoff (Table 1).

DISCUSSION

Deposition fluxes

Atmospheric deposition of trace metals in semi-arid urban areas has unique characteristics not observed in previous studies (Table 4). The magnitude of the total deposition fluxes measured in urban Los Angeles in the present study was significantly higher than the fluxes measured at non-urban sites. This demonstrates the importance of anthropogenic sources in urban areas to higher deposition rates. Also, annual wet deposition fluxes were significantly lower than dry deposition fluxes, indicating the dominance of dry deposition in arid regions compared with other areas of the country. For example, wet deposition comprised only 1% to 10% of the total deposition flux in the present study, while measurements near Chesapeake Bay, where annual rainfall is typically three times that of Los Angeles, indicated wet deposition accounted for 20% to 50% of the total flux (Baker *et al.* 1997). Thus, dry deposition appears to be the dominant mechanism for transfer of atmospheric pollutants to watershed surfaces because of the low rainfall quantity in semi-arid regions like Los Angeles.

Temporal variability of dry deposition fluxes was low, in agreement with the findings of Sabin *et al.* (2004) for urban Los Angeles. The exception was the sampling event during Santa Ana winds and forest fires, which produced high fluxes for all metals, suggesting these anomalous conditions contributed to high fluxes. Other seasonal or meteorological variables were not correlated with fluxes due, in part, to the limited range of meteorological data resulting from the mild climate in Southern California. These results suggest daily, chronic conditions are primarily responsible for the majority of the dry deposition mass of trace metals in Southern California, as demonstrated by computer modeling developed by Lu *et al.* (2003).

While direct measurements of trace metal deposition fluxes have not been made extensively in Southern California, other data for Los Angeles indicate atmospheric TSP concentrations of trace metals

at the study site were approximately half the concentrations measured at other urban sites in Los Angeles (Table 4). This result was not unexpected, since the site in the present study was located in a relatively suburban area of the city, and predominantly upwind of significant point and mobile sources. Because dry deposition is directly proportional to atmospheric concentrations near the surface (Hicks *et al.* 1984), higher deposition fluxes, and subsequently higher loadings from deposition, would be expected in heavily urbanized areas which have higher atmospheric concentrations.

Contribution to stormwater loading

The data from the present study indicate atmospheric deposition is an important contributor to stormwater runoff in urban catchments. Assuming the total quantity deposited onto the catchment was available for removal in stormwater runoff, atmospheric deposition potentially accounted for as much as 57% to 100% of the total trace metal loads in annual stormwater discharges. The finding that atmospheric deposition and stormwater loadings were approximately the same order of magnitude is in agreement with previous studies in this region (Lu *et al.* 2003, Sabin *et al.* 2004), and further demonstrates atmospheric deposition should not be ignored when assessing sources of trace metal pollution to contaminated waterbodies near urban centers.

There are several limitations to these findings. First, not all of the trace metal loads estimated from the average daily deposition measurements may be effectively available for immediate washoff. Some fraction of the deposited material may be removed from surfaces by means other than stormwater runoff due to processes we have not quantified, including resuspension out of the catchment or sequestration within the catchment through uptake by vegetation, accretion, adsorption, and other means (James and Shivalingaiah 1985, Novotny *et al.* 1985). Second, material remaining on the surface may not be completely washed off during storm events (Vaze and Chiew 2002). The amount of material mobilized during surface flows depends on a number of factors, such as surface type (e.g., impervious vs. natural surfaces), street cleaning practices, and rainfall intensity and duration (Novotny *et al.* 1985, Vaze and Chiew 2002). This material may then be available for removal at a later time, and thus some portion of the runoff load may be due to materials that were deposited earlier than the period of measurement.

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