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Sources of Pollutants in Urban Areas (Part 2) – Recent Sheetflow Monitoring

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Two research projects that examined source area sheetflows that were conducted in the 1990s are high-lighted in this chapter. These are a comprehensive project conducted in Birmingham, AL, as part of a project developing a control strategy for critical source areas, and a series of related projects conducted in Wisconsin as part of the DNR's efforts in calibrating the Source Loading and Management Model (SLAMM). A bibliography of recent source area monitoring activities by other researchers is also included in this chapter.

These recent projects conducted in Alabama and Wisconsin are more comprehensive than the earlier monitoring efforts described in the previous chapter. These large projects included a wide range of land uses, source sources, and pollutants, in coordinated monitoring efforts. This information complements the data presented previously.

24.1 Birmingham, Alabama, Sheetflow Monitoring

Pitt, et al. (1995) studied stormwater runoff samples from a variety of source areas under different rain conditions in Birmingham, AL. All of the samples were analyzed in filtered (0.45 µm filter) and non-filtered forms to enable

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partitioning of the toxicants into "particulate" (non-filterable) and "dissolved" (filterable) forms.

Samples were taken from shallow flows originating from homogeneous source areas by using several manual grab sampling procedures. For deep flows, samples were collected directly into the sample bottles. For shallow flows, a peristaltic hand operated vacuum pump created a small vacuum in the sample bottle, which then gently drew the sample directly into the container through a Teflon tube. About one liter of sample was needed, split into two containers: one 500 mL glass bottle with Teflon lined lid was used for the organic and toxicity analyses and another 500 mL polyethylene bottle was used for the metals and other analyses.

All samples were handled, preserved, and analyzed according to accepted protocols (EPA 1982 and 1983b). The organic pollutants were analyzed using two gas chromatographs, one with a mass selective detector (GC/MSD) and another with an electron capture detector (GC/ECD). The pesticides were analyzed according to EPA method 505, while the base neutral compounds were analyzed according to EPA method 625 (but only using 100 mL samples). The pesticides were analyzed on a Perkin Elmer Sigma 300 GC/ECD using a J&W DB-1 capillary column (30m by 0.32 mm ID with a 1 μm film thickness). The base neutrals were analyzed on a Hewlett Packard 5890 GC with a 5970 MSD using a Supelco DB-5 capillary column (30 m by 0.25 mm ID with a 0.2 μm film thickness). Sample extraction was critical for these organic analyses. Liquid-liquid separation funnel extractions were necessary to provide acceptably high recoveries of the organic toxicants. Burton and Pitt (2002) describe the method development for the sample handling and analyses in detail.

Metallic toxicants were analyzed using a graphite furnace equipped atomic absorption spectrophotometer (GFAA). EPA methods 202.2 (Al), 213.2 (Cd), 218.2 (Cr), 220.2 (Cu), 239.2 (Pb), 249.2 (Ni), and 289.2 (Zn) were followed in these analyses. A Perkin Elmer 3030B atomic absorption spectrophotometer was used after nitric acid digestion of the samples. Previous research (Pitt and McLean 1986; EPA 1983a) indicated that low detection limits were necessary in order to measure the filtered sample concentrations of the metals, which would not be achieved by use of a flame atomic absorption spectrophotometer, or ICP unit most commonly available in commercial laboratories. Low detection limits would enable partitioning of the metals between the solid and liquid phases to be investigated, an important factor in assessing the fates of the metals in receiving waters and in treatment processes.

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Table 24.1 summarizes the source area sample data for the most frequently detected organic toxicants and for all of the metallic toxicants analyzed. The organic toxicants analyzed, but not reported, were generally detected in five, or less, of the non-filtered samples and in none of the filtered samples. Table 24.1 shows the mean, maximum, and minimum concentrations for the detected toxicants. Note that these values are based only on the observed concentrations. They do not consider the non-detectable conditions. Mean values based on total sample numbers for each source area category would therefore result in lower concentrations. The frequency of detection is therefore an important consideration when evaluating organic toxicants. High detection frequencies for the organics may indicate greater potential problems than infrequent high concentrations.

Table 24.1 lists stormwater toxicants detected in at least 10% of the source area sheetflow samples (Pitt, et al. 1995), and is presented in the appendix at the end of this book.

Table 24.1 also summarizes the measured pH and SS concentrations. Most pH values were in the range of 7.0 to 8.5 with a low of 4.4 and a high of 11.6 for roof and concrete plant storage area runoff samples, respectively. This range of pH can have dramatic effects on the speciation of the metals analyzed. The SS concentrations were generally less than 100 mg/L, with impervious area runoff (e.g., roofs and parking areas) having much lower SS concentrations and turbidities compared to samples obtained from pervious areas (e.g., landscaped areas).

Out of more than 35 targeted organic compounds analyzed, 13 were detected in more than 10% of all samples, as shown in Table 24.1. The greatest detection frequencies were for 1,3-dichlorobenzene and fluoranthene, which were each detected in 23% of the samples. The organics most frequently found in these source area samples (i.e., polycyclic aromatic hydrocarbons (PAH), especially fluoranthene and pyrene) were similar to the organics most frequently detected at outfalls in prior studies (EPA 1983a).

Roof runoff, parking area and vehicle service area samples had the greatest detection frequencies for the organic toxicants. Vehicle service areas and urban creeks had several of the observed maximum organic compound concentrations. Most of the organics were associated with the non-filtered sample portions, indicating an association with the particulate sample fractions. The compound 1,3-dichlorobenzene was an exception, having a significant dissolved fraction.

In contrast to the organics, the heavy metals analyzed were detected in almost all samples, including the filtered sample portions. The non-filtered samples generally had much higher concentrations, with the exception of zinc,

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which was mostly associated with the dissolved sample portion (i.e. not associated with the SS). Roof runoff generally had the highest concentrations of zinc, probably from galvanized roof drainage components, as previously reported by Bannerman, et al. (1983). Parking and storage areas had the highest nickel concentrations, while vehicle service areas and street runoff had the highest concentrations of cadmium and lead. Urban creek samples had the highest copper concentrations, which were probably due to illicit industrial connections or other non-stormwater discharges.

24.2 Source Area Pollutant Concentrations Observed in Wisconsin Urban Runoff

Described below are the source area concentrations collected in seven monitoring projects in Wisconsin, and one in Michigan. The monitoring was conducted by the United States Geological Survey (USGS) in cooperation with the Wisconsin Department of Natural Resources (WI DNR). All of these monitoring projects were conducted between 1991 and 1997. Contaminant concentrations for the source areas were used to calibrate SLAMM, the Source Loading and Management Model (Pitt and Voorhees 1995).

Madison, WI, runoff samples were collected during three months of 1991 (Bannerman, et al. 1993) to identify the relative pollutant loads from the most common source areas in two study areas. One study area was mostly residential with some commercial land use, while the second area was all light industrial land use. Sheetflow samples were collected from 46 sites representing roofs, streets, driveways, parking lots, and lawns in residential, commercial, and light industrial land uses. The sheetflow samplers were simple in design and were positioned to isolate the runoff from each type of source area. Runoff was delivered to the sample bottles by gravity and the bottles for most of the source areas were installed below the surface of the ground. An effort was made in all the projects to use sample collection methods and equipment that prevented the sample bottles from over-filling before the end of the runoff event. To a large extent, the source area concentrations represented a composite of the runoff occurring during the sampled events. Automated flow meters and water samplers were installed at the storm sewer outfalls for each study area for outfall verification. The sheetflow samples were analyzed for total suspended solids, total solids, total phosphorus, dissolved phosphorus, dissolved and total recoverable zinc, copper, cadmium, chromium, and lead, hardness, and fecal coliform bacteria.

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Between 7 and 10 runoff samples were collected at all the sites, except for lawns and commercial parking areas where fewer samples were collected.

Milwaukee and Madison, WI, runoff samples were collected during 1993 (Roa-Espinosa and Bannerman 1994) to evaluate different methods for collecting source area runoff samples at industrial sites. As part of this evaluation, a total of 50 sampling locations at roofs, paved areas, and lawns were sampled at five industrial facilities. The sheetflow samplers were simple in design and they were located to isolate the runoff from each type of source area. Runoff was delivered to the sample bottles by gravity and the bottles for most of the source areas were installed below the surface of the ground. The samples were analyzed for chemical oxygen demand, suspended solids, total solids, total recoverable zinc, lead, nickel, and copper, and hardness. Depending on the location, samples were collected during 5 to 7 runoff events.

Marquette, MI, runoff samples were collected during 1993 and 1994 (Steuer, et al. 1997) to characterize contaminant concentrations for eight sources in one study area. The study area (297 acres) contained a mixture of land uses including residential, open space, commercial and institutional. A total of 33 sheetflow sampling sites were located at streets, parking lots, driveways, rooftops, and grass areas. Samples were analyzed for total solids, suspended solids, ammonia N, nitrate plus nitrite, total Kjeldahl nitrogen, total phosphorus, dissolved phosphorus, hardness, total recoverable and dissolved zinc, lead, cadmium, and copper, fecal coliform, BOD, COD, and PAHs. Sheetflow samples were collected for 12 runoff events at each site. Flow and water quality were measured at the storm sewer outfall for the study area.

Madison, WI, runoff samples were collected during 1994 and 1995 (Waschbusch, et al. 1999) to estimate the sources of phosphorus in two residential areas for further detailed calibration of SLAMM. All the source areas were in two drainage areas. One was 232 acres, with mostly residential and some commercial land uses, while the other was 41 residential acres. Sheetflow samples were collected from roofs, streets, driveways, parking lots, and lawns in residential and commercial land uses. Twenty five storms were sampled in both basins. The sheetflow samples were analyzed for total suspended solids, total solids, dissolved phosphorus, and total phosphorus. Flow and water quality were measured at the storm sewer outfalls for both study areas.

Madison, WI, runoff samples were collected during 1994 and 1995 (Waschbusch, et al. in press) to evaluate the effects of various environmental factors on the yields of pollutants washed off city streets. The environmental factors included average daily traffic count, antecedent dry time, rainfall

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intensity, rainfall depth, season, and tree canopy. Street pollutant concentrations were also used to calibrate SLAMM. Sheetflow samples were collected from five streets with different daily traffic counts. The street samplers were grouted into the street approximately 5 ft (1.5m) from the curb. The sample bottles were covered with a 6 inch (150mm) concave polycarbonate cap, set flush with the street surface. A drain hole in the cap could be constricted to control the flow into the bottle. At total of 11 or 12 runoff samples were collected at each site. Samples were analyzed for suspended solids, PAHs, hardness, and total and dissolved cadmium, lead, copper, zinc, and phosphorus.

Superior, WI, runoff samples were collected during 1995 and 1996 (Holstrom, et al. 1995 and 1996) to measure flow rates and water quality for runoff from an undeveloped site. The drainage area of the wooded lot is 76.2 acres. Flow was measured with a Parshall flume and runoff samples were collected with a volume activated water quality sampler. Sixteen storm-composite samples were analyzed for suspended solids, total solids, and total phosphorus. Samples were less frequently analyzed for COD, BOD, sulfate, chloride, nitrogen compounds, and total copper, lead, and zinc.

Madison, WI, runoff samples were collected during 1996 and 1997 (Waschbusch, et al. 1999) to verify the pollutant removal efficiency of a stormwater treatment device (Stormceptor). The device was located to treat the runoff from a 4.3 acre (1.7ha) city maintenance yard. Inlet and outlet runoff samples were collected for 45 runoff events. Samples were analyzed for total solids, suspended solids, total and dissolved phosphorus, nitrate plus nitrite, ammonia N, chloride, hardness, alkalinity, organic carbon, particle sizes, PAHs, and total and dissolved copper, cadmium, lead, and zinc. Automated sampling equipment was used to measure flow and collect flow–weighted composite samples. The inlet pollutant concentrations were used to calibrate SLAMM for industrial parking lots.

Milwaukee, WI, runoff samples were collected during 1996 (Corsi, et al. 1999) to measure the pollutant removal efficiency of a stormwater treatment device (the Multi-Chamber Treatment Train). The device was located to treat the runoff from 0.10 acres of parking lot at a city maintenance facility. Inlet and outlet samples were collected for 15 runoff events. Flow meters and automatic water samplers were used to measure flow rates and collect flow-weighted composite water samples in the inlet and outlet pipes. Samples were analyzed for total solids, suspended solids, alkalinity, BOD, COD, volatile suspended solids, ammonia as N, nitrate plus nitrite as N, chloride, sulfate, hardness, PAHs, TOC, total and dissolved phosphorus, total and dissolved

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zinc, cadmium, lead, chromium, and copper. The inlet pollutant concentrations were used to calibrate SLAMM for industrial parking lots.

Results from the eight Wisconsin studies were combined to create an average concentration for each source area (Table 24. 2). Almost all of the average concentration values represent the results from more than one study. Because the constituent list was different for each study, the sample count varies considerably between the types of source areas. Sample counts are high for suspended solids and phosphorus, since they were analyzed during all the studies. Only one project (Marquette, MI) analyzed COD and PAHs for all the source areas, so these constituents have a low sample count. Censored values (samples having less than the detection limit) are included as one-half the detection limit for some of the constituents having low sample counts.

Although loads from a source area are greatly influenced by the volume of runoff, the large differences in some of the source area concentrations can decrease the importance of volume when comparing the loads from different source areas. For example, the volume of runoff from lawns is expected to be relatively low, but concentrations of phosphorus in lawn runoff are 2 to 10 times higher than for other source areas. Because of these relatively high concentrations, lawns can contribute as much as 50% of the annual total phosphorus load in a residential area (Washbusch, et al. 1999). With PAH levels from commercial parking lots 10 to 100 times higher than from any other source area, commercial parking lots representing only 3% of an urban drainage area can contribute 60% of the annual PAH load (Steuer, et al. 1997.

Concentrations for some of the pollutants can be compared between roofs and streets for all three land uses. Streets in industrial areas are likely important sources of suspended solids, total phosphorus, and zinc whenever they are compared to commercial and residential streets. But concentrations of these three pollutants in industrial roof runoff is similar to, or lower than, the other two land uses.

Enough data are compiled in Table 24.2 to determine pollutant loads from many of the typical urban source areas. However, more monitoring is needed to expand the list of constituents, especially in areas having low sample counts. Additional source areas also need to be monitored. Pesticides are an example of a pollutant not included in Table 24.2. Both lawns and undeveloped areas do not have any concentrations for PAHs. The sample count is very low for source areas having PAH, COD, and BOD data. Sample counts are less than five for 40 out of the 334 concentrations on the table. Gas stations are an example of a missing specific source area. Also missing are runoff concentrations from a range of parking lot types. Parking lots with high turnover are expected to experience higher pollutant concentrations than those

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used for long-term employee parking, for example, and need to be better represented. *Table 24.2 (Wisconsin Source Area Monitoring Data) has been placed in the appendix at the end of this book.*

24.3 Other Source Area Stormwater Investigations

24.7.1 General Sources

Nowakowska-Blaszezyk, et al. (1996) studied the sources of wet-weather pollutants in Poland. It was found that storm runoff from parking areas and streets had the greatest concentrations of suspended solids (SS), chemical oxygen demand (COD), five-day biochemical oxygen demand (BOD_5), and Pb, while phosphorus was mostly contributed from landscaped-area storm runoff. Storm runoff from roofs covered with roofing paper was also a significant contributor of many pollutants.

Gromaire-Mertz, et al. (1999) collected stormwater runoff from 4 roofs, 3 courtyards and 6 streets on an experimental catchment in central Paris, France, and analyzed the samples for SS, VSS, COD, BOD_5, hydrocarbons, and heavy metals both in dissolved and particulate fractions. The street runoff showed large SS, COD and hydrocarbon loads, but the roof runoff had high concentrations of heavy metals.

Davis, et al. (2001) presented loading estimates of lead, copper, cadmium, and zinc in stormwater from different sources. They reviewed available data from the literature, and conducted controlled experiments and other sampling. Specific sources that they examined included building siding and roofs; automobile brakes, tires, and oil leakage; and wet and dry atmospheric deposition. The most important sources they identified were building siding for all four metals, vehicle brake emissions for copper, and tire wear for zinc. Atmospheric deposition was an important source for cadmium, copper, and lead.

24.3.2 Atmospheric Deposition Studies

Jordan, et al. (1997) estimated that 40% of the nitrogen (N) loading to the Chesapeake Bay watershed comes from atmospheric deposition, 33% from livestock waste, and 27% from fertilizer. Ahn and James (2001) reported that atmospheric deposition is a substantial source of phosphorus to the Florida Everglades. Phosphorus has been measured on a weekly basis since 1974, but

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the results were highly variable: the average mean and standard deviation of the calculated P deposition rates for 13 sites were 41± 33 mg P m^{-2} yr^{-1}. They found that the atmospheric P deposition load showed high spatial and temporal variability, with no consistent long-term trend. Because of the random nature of P deposition, the estimated P deposition loads have a significant amount of uncertainty, no matter what type of collection instrument is used, and replicate sampling is highly recommended.

Atasi, et al. (1999 and 2001) conducted source monitoring using specialized sampling equipment and ultra-clean analytical methodology to quantify the concentrations and fluxes of mercury, cadmium, and polychlorinated biphenyl in ambient air, precipitation, runoff, sanitary sewer, and wastewater treatment plant influent. The relationships between the atmospheric deposition and runoff on controlled surfaces were also examined. Atmospheric deposition was found to be the primary source of these pollutants in runoff. They concluded that wet weather flows, not atmospheric deposition, contributed the main portion of these pollutants to the Detroit Wastewater Treatment Plant. Garnaud, et al. (1999) studied heavy metal concentrations in dry and wet atmospheric deposits in Paris, France, for comparison with urban runoff. Samples were continuously collected for 2 to 13 months at each of four test sites. Comparisons of median values of metal concentrations showed that rainwater contamination with heavy metals was only slightly higher in the center of Paris than at Fontainebleau (48 km SE of the city) which illustrates the medium range transport of atmospheric contamination. Glass and Sorensen (1999) examined a six-year trend (1990-1995) of wet mercury deposition in the Upper Midwest of the United States. The annual wet mercury deposition averaged 7.4 μg Hg/m^2-y and showed significant variations between sites and illustrated significant increasing trends over the monitoring period. Warm (rain) season wet mercury deposition was found to average 77% of total annual wet deposition.

Tsai, et al. (2001) described their pilot study, conducted from August 1999 through August 2000, that estimated the loading of heavy metals from the atmosphere to San Francisco Bay. Dry deposition flux of copper, nickel, cadmium, and chromium was approximately 1100 +/- 73, 600 +/- 35, 22 +/- 15, and 1300 +/- 90 $\mu g/m^2/y$, respectively. The volume-weighted average concentrations of these trace metals in the rain water were 1.2, 0.4, 0.1, and 0.2 μg/L, respectively. Direct atmospheric deposition onto Bay waters, from both dry deposition and rainfall, contributed approximately 1900, 930, 93 and 1600 kg/y of copper, nickel, cadmium and chromium, respectively. Stormwater runoff contributed approximately twice as much as the loading

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from direct atmospheric deposition. Direct atmospheric deposition was therefore found to be a minor contributor to the total load of these pollutants to the Bay. A mass balance of all known sources and sinks for heavy metals (Ag, Cd, Cu, and Pb) in New Haven Harbor, CT, was conducted by Rozan and Benoit (2001). Sources included direct atmospheric deposition, rivers, treated sewage effluent, combined sewer overflows, and permitted industrial discharges. All of the fluxes were directly measured, and the uncertainties were quantified. River inputs accounted for most of the total yearly metal discharges, while the salt marshes removed about 20 to 30% of the metals from the rivers before reaching the harbor. Atmospheric deposition is of minor importance, and is comparable to sewage effluent discharges. Schiff and Stolzenbach (2003) investigated the heavy metal contribution of atmospheric deposition to Santa Monica Bay and compared the atmospheric deposition loading to the loading from other sources. The annual atmospheric deposition of chromium, copper, lead, nickel and zinc exceeded the estimated annual effluent loads from industrial and power generating stations to Santa Monica Bay.

24.3.3 Roof Runoff and Other Building Materials

Sakakibara (1996) investigated roof runoff quality in Ibaraki prefecture, Japan, in order to determine the feasibility of using roof runoff in urban areas for various beneficial uses. Eighty three samples were collected during one year and analyzed for pH (averaged 6.1), BOD_5 (averaged 1.6 mg/L), COD (averaged 3.2 mg/L), and SS (averaged 12 mg/L). It was concluded that roof runoff could be used for toilet flushing and landscaping watering with minimal treatment or problems.

Heavy metals and major ions in roof-runoff were investigated by Förster (1996) in Bayreuth, Germany. It was found that the major ions were from the rain, while very high Cu and Zn concentrations were from metal flashings used on the roofs. It was concluded that the best option would be to abandon the use of exposed metal surfaces on roofs and walls of buildings. Pesticides present in rainwater do not pose a greater groundwater contamination problem during artificial roof runoff infiltration (a practice in Switzerland to reduce runoff) than does the direct application in agriculture (Bucheli, et al. 1998a); however, the herbicide (R,S)-mecoprop, a root protection agent in Preventol B 2 commonly applied to roofs, is of the same order of magnitude as loads from agricultural applications (Bucheli, et al. 1998b). Förster (1999) and Förster, et al. (1999) summarized their studies investigating roof runoff as a stormwater

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pollutant source. Runoff samples were taken from an experimental roof system containing five different roofing materials and from house roofs at five different locations in Bayreuth, Germany. It was found that local sources (e.g. PAH from heating systems), dissolution of the roof systems' metal components, and background air pollution were the main sources of the roof runoff pollution. They found that the first flush from the roofs often was heavily polluted and should be specially treated. They concluded that roofs having metal surfaces should not be connected to infiltration facilities as concentrations of copper and zinc far exceed various toxicity threshold values. They also examined a green (vegetated) roof for comparison. These roofs were found to act as a source of heavy metals which were found to be in complexes with dissolved organic material. Leaching from unprotected zinc sheet surfaces on the green roofs resulted in extremely high zinc concentrations in the runoff. In contrast, the green roofs were a trap for PAHs.

Davis and Burns (1999) examined lead concentrations in runoff from painted surfaces. In many tests, high lead concentrations were found (using 100 mL of wash water over 1600 cm^2 of surface). Lead concentrations from 169 different structures followed the following order (median concentrations in the wash water): wood (49 μg/L) > brick (16 μg/L) > block (8.0 μg/L). Lead concentration depended strongly on paint age and condition, with the lead levels from washes of older paints being much higher than from freshly painted surfaces. Lead from surface washes were found to be 70%, or greater, in particulate lead form, suggesting the release of lead pigments from the weathered paints.

Zobrist, et al. (2000) examined the potential effects of roof runoff on urban stormwater drainage from three different types of roofs: an inclined tile roof, an inclined polyester roof and a flat gravel roof. Runoff from the two inclined roofs showed initially high ("first flush") concentrations of the pollutants with a rapid decline to lower levels. The flat gravel roof showed lower concentrations of most of the pollutants because of the ponding of the water on the roof surface acting like a detention pond. Pollutant loadings were similar to atmospheric deposition, with the exception of copper from drain corrosion (rate about 5 $g/m^2/y$). Tobiason and Logan (2000) used the whole effluent toxicity (WET) test to characterize stormwater runoff samples from four outfalls at Sea-Tac International Airport. Three of the four outfalls met standards; the source of the toxicity at the fourth outfall was found to be zinc-galvanized metal rooftops. Typically, more than 50% of the total zinc in the runoff was in dissolved form and likely bioavailable. Polkowska, et al. (2002) presented the results of testing roof runoff waters from buildings in Gdafisk, Poland. More than half of the samples (25) were found to be toxic, with

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inhibition exceeding 20%. The toxicity was weakly correlated to the levels of organonitrogen and organophosphorus pesticides in runoff waters. It was established that at least in some cases the roofing material affected the levels of the pollutants found in the samples. Heijerick, et al. (2002) investigated the bioavailability of zinc in runoff from roofing materials in Stockholm, Sweden. Chemical speciation modeling revealed that most zinc (94.3-99.9%) was present as the free Zn ion, the most bioavailable speciation form. These findings were confirmed by the results of the biosensor test (Biomet™), which indicated that all zinc was indeed bioavailable. Analysis of the ecotoxicity data also suggested that the observed toxic effects were due to the presence of Zn^{2+} ions. Gromaire, et al. (2002) investigated the impact of zinc roofing on urban pollutant loads in Paris. On an annual basis, runoff from Parisian zinc roofs would produce around 34 to 64 metric tons of zinc and 15 to 25 kg of cadmium, which is approximately half the load generated by runoff from all of Paris.

Karlen, et al. (2002) investigated runoff rates, chemical speciation and bioavailability of copper released from naturally patinated copper roofs in Stockholm, Sweden. The results show annual runoff rates between 1.0 and 1.5 g/m^2 year for naturally patinated copper of varying age with rates increasing slightly with patina age. The total copper concentration in investigated runoff samplings ranged from 0.9 to 9.7 mg/L. The majority (60 – 100%) of the released copper was present as the free hydrated cupric ion, $Cu(H_2O)_6{}^{2+}$, the most bioavailable copper species. The copper-containing runoff water, sampled directly after release from the roof, caused significant reduction in growth rate of green algae. Wallinder, et al. (2002) studied the atmospheric corrosion of naturally and pre-patinated copper roofs in Singapore and Stockholm. Measured annual runoff rates from fresh and brown prepatinated were 1.1-1.6 g/m^2 and 5.5-5.7 g/m^2, in Stockholm and Singapore, respectively. Naturally aged copper sheet (130 years old) and green pre-patinated copper sheet showed slightly higher (1.6-2.3 g/m^2), but comparable runoff rates in Stockholm. In Singapore, runoff rates from green pre-patinated copper sheet were 8.4-8.8 g/m^2. Comparable runoff rates between fresh and brown-patinated copper sheet and between green naturally patinated and green pre-patinated copper sheet at each site were related to similarities in patina morphology and composition. Boller and Steiner (2002) investigated the emission and control of copper from roofs and roads in urban surface runoff. A large copper façade was used to investigate the concentrations of copper emitted. The concentrations ranged from 1 – 10 mg/L. Michels, et al. (2003) investigated the environmental impact of stormwater runoff from a

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copper roof. It was shown that the runoff became less toxic as it passed through the drainage system.

Clark, et al. (2003) studied the potential pollutant contributions from commonly-used building materials (roofing, siding, wood) using a modified Toxicity Characteristic Leaching Procedure (TCLP) test. Results of particular interest included evidence of elevated levels of phosphate, nitrate and ammonia in the leachant following exposure of common roofing and siding materials to simulated acid rain.

Lebow, et al. (2003) investigated the release of preservatives, primarily arsenic, from CCA-treated wood under simulated rainfall and the ability of wood finishes to prevent/reduce their release. Water repellent significantly decreased the amounts of these elements in the runoff, while UV exposure increased the leaching of preservatives from the wood.

24.3.4 Highway and other Roadway Runoff

Wada and Miura (1996) examined storm runoff from a heavily traveled highway in Osaka, Japan. A significant “first-flush” for COD was found and the amount of small rubber pieces from tire wear in the highway storm runoff was more than 20 times greater than for an “ordinary” road. The primary factors affecting storm runoff concentrations were the amount of traffic (and related exhaust emissions and tire wear) and the fraction of the total traffic that was comprised of trucks and buses. Montrejaud-Vignoles, et al. (1996) collected storm runoff from a heavily used six-lane motorway in the Mediterranean area of France. The very irregular rainfall in this area and associated very-long dry periods can result in storm runoff that is much more polluted than elsewhere in France. As an example, during the one-year study, a single rain of only 10 mm but having an antecedent-dry period of 35 days produced more than 12% of the annual COD discharges. Ball, et al. (1996) and Ball 2000 examined roadway pollutant accumulations in a suburb of Sydney, Australia. It was concluded that the local heavy winds have a significant effect on pollutant accumulations that commonly available stormwater models do not consider, and that historical United States’ data on roadway-pollutant accumulations are much greater than found in their area.

Sansalone and Buchberger (1996) studied metal distributions in stormwater and snowmelt from a major highway in Cincinnati, OH. Zn and Cd were mostly in filterable (dissolved solids) forms in the storm runoff, while lead was mostly associated with particulates. A receptor-source model was used to apportion source contributions for PAH in street and creek

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sediments. The model showed that vehicles along with the coke ovens, are the major contributors to PAH in street sediments (Sharma et al., 1997). Measurements of conductivity and turbidity taken in a study of the Crum Creek which runs through the suburbs of Philadelphia, Pa. indicated two stages during the first three hours of wet weather runoff: a dissolved solids flush followed by a suspended solids (SS) flush (Downing and McGarity 1998). In San Francisco, Calif., vehicle emissions of both ultrafine (< 0.12 μm) and accumulation mode (0.12 —2 μm) particulate polycyclic aromatic hydrocarbons (PAH) are derived from diesel vehicles while gasoline vehicles emit higher molecular weight PAH primarily in the ultrafine mode. Heavy duty diesel vehicles were found to be important sources of fine black carbon particles (Miguel, et al. 1998). In a European study, 90% of the particles from a contaminated highway runoff catchment were smaller than 100 μm. The constituents of the contaminants smaller than 50 μm were further analyzed by X-ray diffraction, thermogravimetry and specific mass and contained 56% clay, 15% quartz, 12% chalk, 9% organic matter, 5% feldspars, and 2% dolomite (Roger, et al. 1998).

Waschbusch, et al. (1999) investigated sources of phosphorus in stormwater and street dirt from two urban residential basins in Madison, Wisconsin. They collected numerous sheetflow runoff samples from throughout the test watersheds and used SLAMM, an urban stormwater quality model, to quantify the significance of the different phosphorus sources. Lawns and streets were found to be the most significant sources of phosphorus in the test basins, contributing about 80% of the total annual loading. In the Kerault Region of France, the effects of pollution were studied using solid matter from a section of the A9 motorway. This study analyzed both settled sediments from collecting basin and characteristics of sediments in the water column during and after eight storm events between October 12, 1993, and February 6, 1994. Settled sediments were used to measure particle sizes, mineral content, and related characteristics, whereas water samples were used to document total suspended solids, mineral content, and heavy metals (Andral, et al. 1999). Highway runoff contains significant loads of heavy metals and hydrocarbons, and according to German regulations, it should be infiltrated over embankments to support groundwater recharge. To investigate the decontaminating effect of greened embankments, soil-monoliths from highways with high traffic densities were taken. Soils were analyzed to characterize the contamination in relation to distance and depth for lead, zinc, copper, cadmium, PAH and MOTH (Dierkes and Geiger 1999).

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An investigation by Drapper, et al. (2000) showed that the pollutant concentrations (heavy metals, hydrocarbons, pesticides, and physical characteristics) in ‘first flush’ road runoff in Brisbane in southeast Queensland, Australia was within the ranges reported internationally for highways. Traffic volumes were the best indicator of road runoff pollutant concentrations, with interevent duration also being statistically significant factor. Exit-lane sites were found to have higher concentrations of acid-extractable copper and zinc, likely due to brake pad and tire wear caused by rapid deceleration, and laser particle sizing showed that a significant proportion of the sediment in runoff was less than 100 µm. Krein and Schorer (2000) investigated heavy metals and PAHs in road runoff and found that, as expected, an inverse relationship existed between particle size and particle-bound heavy metals concentration existed. However, particulate bound PAHs were found to be bimodally distributed. Three-ring PAHs were mostly found in the fine sand fraction, while six-ring PAHs were mostly concentrated in the fine silt fraction. Sutherland, et al. (2000) investigated the potential for road-deposited sediments in Oahu, Hawaii, to bind contaminants, and thus transporting these bound contaminants to the receiving water as part of the runoff. In the sediment fractions less than 2 mm in diameter, the origins of the aluminum, cobalt, iron, manganese and nickel were determined to be geologic. Three of the metals concentrations, copper, lead and zinc, were found to be enhanced by anthropogenic activities. Sequential extraction of the sediment determined the associations of the metals with the following fractions: acid extractable, reducible, oxidizable, and residual).

Stenstrom, et al. (2001) studied freeway runoff from three sites in the west Los Angeles area. Each site was sampled for 14 storms during the 1999-2000 rain season. Samples were collected very early in the storm in order to compare water quality from the first runoff to water quality from the middle of the storm. A large range of water quality parameters and metals were analyzed. The data showed large first flushes in concentration and moderate first flushes in mass emission rates. Neary, et al. (2002) studied the pollutant washoff and loadings from parking lots in Cookeville, Tennessee. The monitoring results indicated that the washoff response from small parking lot catchments was also affected by other factors that included antecedent dry conditions and rainfall intensity. Ma, et al. (2002) investigated the first-flush phenomenon for highways. Most pollutants showed median mass first flushes where 30% of the mass is released in the first 20% of the runoff. Pollutants representing organic contaminants had the highest first flush ratios. Lau, et al. (2002) studied whether a first flush of organics (COD, oil and grease, and PAHs) would be seen in highway runoff. The three highway sites exhibited a

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first flush in most cases for most parameters. The mass first flush ratio (the ratio of the normalized transported mass of pollutant to the normalized runoff volume) generally was above 1.8 for the first 25% of the runoff volume, and in some cases as high as 2.8.

Vaze and Chiew (2003) studied pollutant washoff from small impervious experimental plots and showed that the energy of the falling raindrops was important at the beginning of the event where the concentration/prevalence of easily detachable pollutants is greatest. The authors suggest that meaningful characteristic curves that relate event total suspended solids (TSS) and total phosphorus (TP) loads to storm durations for specific rainfall intensities could be developed from the experimental data. Kayhanian, et al. (2003) investigated the relationships between annual average daily traffic (AADT) numbers and highway runoff pollutant concentrations from California Department of Transportation highway sites. No direct linear correlation was found between highway runoff pollutant event mean concentrations (EMCs) and AADT, but multiple linear regression showed that AADT, as well as antecedent dry period (ADP), drainage area, maximum rain intensity and land use, influenced most highway runoff constituent concentrations. Mishra, et al. (2003) developed hysteresis and normal mass rating curves for runoff rate and mass of 12 dissolved and particulate bound metal elements from Cincinnati, OH. Zinc was found to increase with antecedent dry period (ADP). Shinya, et al. (2003) evaluated the factors influencing diffusion of highway pollutant loads in urban highway runoff. Particulates (suspended solids, iron and total phosphorus) were inclined to be washed off in heavier rainfall; event mean runoff intensity and cumulative runoff depth were correlated with cumulative runoff load of the constituents except total nitrogen (TN). ADP and traffic flow volume were not correlated with cumulative runoff load (except TN). Sutherland (2003) investigated lead concentrations in six grain-size fractions of road-deposited sediment from Oahu, HI. Significant Pb concentrations were seen in all samples and the median labile Pb concentration was 170 mg/kg (4 to 1750 mg/kg), with the silt plus clay fraction containing 38% of the total sediment in this fraction.

Landscaped Areas

Emerson (2003) discussed Plymouth, MN restrictions on the use of lawn fertilizers containing phosphorus. The result of the program has been an improvement in water quality due to reducing phosphorus in the runoff. Strynchuk, et al. (2003) studied the decomposition of grass and leaves and the subsequent input of nutrients to receiving waters in Brevard County, FL.

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Release rates for these nutrients have been calculated, which can then be used to select and determine maintenance frequencies of stormwater control practices that treat nutrients.

24.4 Conclusions

The greatest detection frequencies for organic toxicants were for 1,3-dichlorobenzene and fluoranthene, which were each detected in 23% of the Birmingham area samples. The organics most frequently found in these source area samples (i.e., fluoranthene and pyrene) were similar to the organics most frequently detected at outfalls in prior studies. In contrast to the organics, the heavy metals analyzed were detected in almost all samples, including the filtered sample portions.

The volume of runoff from Wisconsin lawns is expected to be relatively low, but concentrations of phosphorus in lawn runoff are 2 to 10 times higher than for other source areas. Because of these relatively high concentrations, lawns can contribute as much as 50 % of the annual total phosphorus load in a residential area. Similarly, with PAH levels from commercial parking lots 10 to 100 times higher than from any other source area, commercial parking lots representing only 3% of an urban drainage area can contribute 60% of the annual PAH load

Many studies throughout the world have investigated stormwater pollutant sources. Pavement is usually identified as the most important source for toxicants (heavy metals and PAHs), while landscaped areas are important sediment and nutrient sources. Stormwater managers need to understand the likely sources of critical pollutants in their areas in order to make reasonable decisions pertaining to source area and outfall controls, and when developing regulations. The information in this chapter should also be of use to stormwater modelers.

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