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Roofing as a source of nonpoint water pollution

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Abstract

Sixteen wooden structures with two roofs each were installed to study runoff quality for four commonly used roofing materials (wood shingle, composition shingle, painted aluminum, and galvanized iron) at Nacogdoches, Texas. Each roof, either facing NW or SE, was 1.22 m wide × 3.66 m long with a 25.8% roof slope. Thus, there were 32 alternatively arranged roofs, consisting of four roof types × two aspects × four replicates, in the study. Runoff from the roofs was collected through galvanized gutters, downspouts, and splitters. The roof runoff was compared to rainwater collected by a wet/dry acid rain collector for the concentrations of eight water quality variables, i.e. Cu^{2+}, Mn^{2+}, Pb^{2+}, Zn^{2+}, Mg^{2+}, Al^{3+}, EC and pH.

Based on 31 storms collected between October 1997 and December 1998, the results showed: (1) concentrations of pH, Cu, and Zn in rainwater already exceed the EPA freshwater quality standards even without pollutant inputs from roofs, (2) Zn and Cu, the two most serious pollutants in roof runoff, exceeded the EPA national freshwater water quality standards in virtually 100% and more than 60% of the samples, respectively, (3) pH, EC, and Zn were the only three variables significantly affected by roofing materials, (4) differences in Zn concentrations were significant among all roof types and between all roof runoff and rainwater samples, (5) although there were no differences in Cu concentrations among all roof types and between roof runoff and rainwater, all means and medians of runoff and rainwater exceeded the national water quality standards, (6) water quality from wood shingles was the worst among the roof types studied, and (7) although SE is the most frequent and NW the least frequent direction for incoming storms, only EC, Mg, Mn, and Zn in wood shingle runoff from the SE were significantly higher than those from the NW; the two aspects affected no other elements in runoff from the other three roof types. Also, Zn concentrations from new wood-shingle roofs were significantly higher than those from aged roofs of a previous study. The study demonstrated that roofs could be a serious source of nonpoint water pollution. Since Zn is the most serious water pollutant and wood shingle is the worst of the four roof types, using less compounds and materials associated with Zn along with good care and maintenance of roofs are critical in reducing Zn pollution in roof runoff.

Keywords: Precipitation chemistry; Roof runoff; Water quality; Nonpoint pollution; Roof management; East Texas

1. Introduction

Although there have been many years of efforts by governments and private sectors, nonpoint sources of pollution are still the major water quality problems in the US (Gannon et al., 1996; Griffith et al., 1999). Of the 1.35 (10^6) km rivers and streams that were surveyed in 1998, EPA (2000) reported that 40% of them were impaired by siltation and 60% of pollutants came from agriculture, the leading pollution source in the US. Traditionally, the control of nonpoint pollution has been focused on agriculture, forestry, mining, construction, livestock feedlots, urban runoff, and roads. Water pollution induced by storm runoff from different roofing materials is considered a nonpoint source and few studies have been conducted in this area.

Roof runoff is considered a potential source of nonpoint pollution for two primary reasons. First, compounds contained in roofing materials may be leached into runoff, and airborne pollutants and organic substances, such as leaves, dead insects, and bird's wastes, are added to roofs by interception and deposition. During storms, rainwater not only adds a variety of chemicals and contaminants to

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the roof watershed, the acidic nature of rainwater will react with compounds retained in or by the roof and cause many elements in the roof-runoff to leach out (King and Bedient, 1982). Second, roof temperatures are much higher than temperatures of other surfaces, due to lower albedo, greater surface inclination to direct solar radiation, and less shading effects from surrounding trees (Chang and Crowley, 1993). The higher roof temperatures may accelerate chemical reactions and organic decomposition of the materials and compounds that have accumulated on rooftops. Combining these constituents from rooftops with elements from precipitation deposition, chemical decomposition, and acid leaching make the quality of roof runoff a great concern for the household cistern system (Sharpe and Young, 1982; Ariyananda and Mawatha, 1999; Spinks et al., 2003) and on receiving streams (Che et al., 2001).

The concentrations of constituents in roof runoff were reported to be much higher than those in rainfall in Germany (Förster, 1998), and some elements, especially metals, far exceeded the WHO (World Health Organization) drinking water quality standards in Australia (Thomas and Greene, 1993). During and right after the application periods, concentrations of pesticides could be nine times greater than the Swiss drinking water standards (Bucheli et al., 1998). In some roof types, the average concentration of Zn in roof runoff was nine times higher than the maximum allowed value for wastewater in Germany (Quek and Förster, 1993). Malmqvist (1983) showed that roof runoff was a major source of Zn and Cu in storm runoff. In New Zealand, water quality conditions were analyzed for three metals, four bacterial indicators and five pathogen species for 125 domestic roof-collected rainwater supplies in four rural Auckland districts. The results showed that those roof-collected rainwater systems provided potable supplies of relatively poor water quality. Twenty-two (17.6%) and 70 (56%) of the systems had one or more values exceeding the New Zealand drinking water standards on chemical pollutants and fecal coliforms, respectively (Simmons et al., 2001). Zobrist et al. (2000) showed that runoff from a tile roof and a polyester roof was initially very high in concentrations, but declined to lower constant levels as storms proceeded on.

Some other studies, however, showed that roof-runoff quality was quite different than those reported above. Roof runoff from 12 cistern water supply systems located on St Thomas in the US Virgin Islands were monitored for 23 chemical elements, i.e. Ca, Mg, Na, K, Cl, SO_4^{2-}, total P, NO_2^-, NO_3^-, NH_4^+, organic N, F, Zn, Cd, Pb, Cr, Ni, Fe, Mn, Hg, specific conductance, alkalinity, and pH. The concentrations of all these elements, except for Hg, were found to be below the US EPA water quality standards for public water supplies (Lee and Jones, 1982). It was concluded that roof materials or painting of the rooftop collection system did not have a significant impact on water quality. Pazwash and Boswell (1997) also stated that roof runoff is “fairly pure and practically free of suspended matter and impurities” found in other surfaces. The variation of roof runoff quality seems to reflect differences in roofing materials, age and management, the surrounding environment, season, storm duration and intensity, and air quality conditions of the region.

Chang and Crowley (1993) conducted a preliminary study to monitor runoff quality from four residential roofs and incident rainwater over a 6-month period in Nacogdoches, Texas. They showed (1) wood shingles yielded the most precipitated elements while terra cotta clay roof yielded the least, (2) both mean and median concentrations of Zn from the four roof types exceeded the EPA fresh water quality standards, and (3) one-half of the 24 chemical variables analyzed exceeded the Standards at least once in all samples collected. Their result on Zn in roof runoff as a potential contaminant in receiving streams agreed with those studies conducted in Germany and Australia. However, the preliminary Texas study had some weaknesses, including no replicates and inconsistency in the size, inclination, aspect, age, and management among the studied roofs. The objective of the study reported here was to repeat Chang and Crowley’s (1993) preliminary roof-runoff quality study by addressing the deficiencies in their experimental design and to focus the assessment of water quality conditions on metal elements.

2. Materials and methods

2.1. Study area

The study was conducted on a 1 ha open site located at the city landfill on northwest Loop 224 of Nacogdoches, Texas. The site, surrounded by loblolly pines, is largely rural. Some industries, such as paper mills, fertilizer and animal feed production, and steel fabrication, are located to the southeast of the city, within 20 km from the study site. Large plywood mills, paper mills, saw mills, and chemical companies are located in Lufkin, Diboll, and Huntington within 60 km from the south and southeast side of the city limit. The extensive petroleum refining of Houston is about 240 km south of the city. Nacogdoches is characterized by a subtropical humid climate with hot summers and mild winters. Normal annual (1971–2000) precipitation and temperature are 1213 mm and 18.8 °C, respectively. The prevailing winds are from southerly directions in summer and virtually from all directions in late fall and early winter (Chang et al., 1996).

2.2. Experimental design

Sixteen wooden structures containing four roof types most commonly used in the area (i.e. wood shingle, composition shingle, aluminum, and metal) were installed at the study site. Each structure had two roofs of 1.22 m wide × 3.66 m long each, one facing NW and one SE.

Fig. 1. Roof structures and their set up located at the City Landfill of Nacogdoches, east Texas.

Thus, there were 32 roofs in this study, consisting of four roof types × two aspects × four replicates. Distance between adjacent structures was 3.66 m.

The roof peak on each structure was 3.05 m above the ground, slanted to 2.14 m at both ends in a 25.8% slope. Each roof was installed in accordance with commercial standards and the four replicates were arranged in alternation (Fig. 1).

2.3. Sample collection

Roof runoff was caught by a commercial galvanized gutter of 1.22 m wide installed at the end of each roof. The collected runoff was drained into a sample collection apparatus through a galvanized down spout. There were two splitters in the collection apparatus (Fig. 2), each collecting 25% of the runoff. The two splitters were installed at two different heights above the runoff sample containers so that the runoff finally draining into the sample container was only 6.25% of the total. The residual runoff was drained into a second container to be used as a spare, or overflowed to the ground. About 1000 ml of roof runoff per storm were collected for chemical analyses. Thus, the samples collected under the system reflected the total metal concentrations of the entire storm. However, the concentrations might have been affected because excessive runoff samples were allowed to overflow from the sampling containers during intense rainfall events. Samples of rainwater were also collected using an automatic sensing wet/dry acid rain collector (Aerochem Metrics Model #301, Scientific Sales, Inc., Lawrenceville, NJ) installed about 7 m north of the roof structures. Rainwater obtained from the wet/dry acid rain collector was used as the control in this study.

The 16 wooden structures with two roof facets on each structure were installed in summer 1997; data collection started in October 1997 and ended in December 1998. Roof runoff in each sample container was agitated before collection. When necessary, sample volumes were supplemented with water from the spare container to make up a volume of 1000 ml required in the laboratory analyses. Water samples were collected in high-density polyethylene

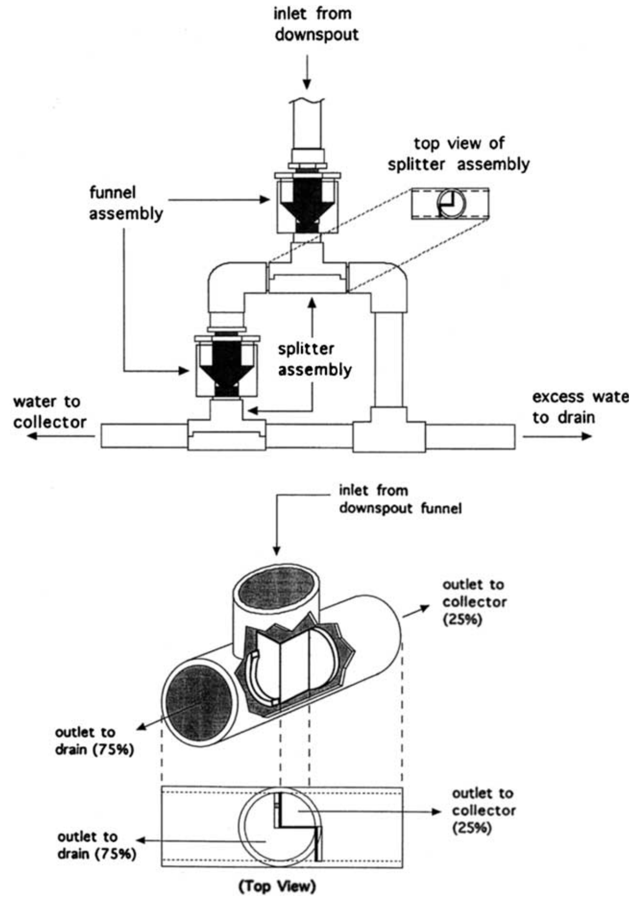

Fig. 2. The roof runoff collection apparatus (top) and the splitter design (bottom).

(HDPE) containers that had been washed in phosphate-free laboratory detergent, followed by a soak/rinse in a 0.1 N HCl bath and by 3× rinse in distilled water. The collected roof runoff and rainfall samples were transported (6 km) in an ice chest to the laboratory for immediate determination of electrical conductivity and pH and then stored in a refrigerator for other chemical analyses within 48 h. A total of 31 storm-runoff samples were collected, analyzed, and reported in this paper.

2.4. Chemical analysis

The collected runoff and rainfall samples were stored in ice chests and brought back to the laboratory for analyses of pH, electrical conductivity (EC), and the concentrations of aluminum (Al^{3+}), magnesium (Mg^{2+}), manganese (Mn^{2+}), lead (Pb^{2+}), zinc (Zn^{2+}), and copper (Cu^{2+}) within 48 h. These ions were the most significant in Chang and Crowley's (1993) study. All pH measurements were made at ambient laboratory temperature. Runoff water was acidified and digested by HNO_3 as specified by APHA 4500 prior to analysis on the ICP by APHA method 3120. Methods of chemical analyses and their detection limits are given in Table 1. The analyses were conducted in the Soil

Table 1
Methods of chemical analyses and detection limits

Parameter	Analysis method	Detection level[a]
pH	Electrometric pH meter	0–14 pH units
Conductivity	Conductivity meter	0.010 μS/cm
Aluminum	APHA 4500 digestion, 3120 ICP[a]	0.008 mg/l
Magnesium	APHA 4500 digestion, 3120 ICP[a]	0.001 mg/l
Manganese	APHA 4500 digestion, 3120 ICP[a]	0.001 mg/l
Lead	APHA 4500 digestion, 3120 ICP[a]	0.025 mg/l
Zinc	APHA 4500 digestion, 3120 ICP[a]	0.030 mg/l
Copper	APHA 4500 digestion, 3120 ICP[a]	0.001 mg/l

[a] Total metals acid digestion (HNO_3) with analysis on inductively coupled plasma (ICP) atomic emission spectrometer (APHA, 1998).

Testing Lab, Agricultural Department, Stephen F. Austin State University at Nacogdoches, Texas.

2.5. Data analysis

USEPA's (1999) water quality standards were used as a reference to evaluate roof runoff quality conditions as described by the water quality parameters. Since the data failed to meet the assumption of normality for parametric statistical analyses, the nonparametric Kruskall–Wallis test as described by Hollander and Wolfe (1999) and SAS Institute, Inc. (1999) was employed to determine differences in concentrations among the four roof types and between roof types and rainwater. The Wilcoxon's rank sum procedure was used to evaluate multiple comparisons where the Kruskall–Wallis test found differences to be significant at $\alpha = 0.05$. Correlation analysis was used to determine the degree of association between water quality parameters and storm characteristics such as total rainfall, storm duration, maximum intensity, and storm interval. Again, the Spearman's rank correlation coefficient was used since data failed to meet the assumptions for the parametric Pearson's correlation coefficient.

3. Results and discussion

3.1. Precipitation chemistry

Average annual pH of precipitation in North America ranges from 4.12 at the Hubbard Brook Experimental Forest, New Hampshire (Buso et al., 2000) and the central Appalachians to 6.50 in eastern Alaska and central Canada (Cowling, 1983). Precipitation pH at Nacogdoches during the study period ranged from 4.20 to 7.03 with an arithmetic average of 5.55 (Table 2). The average pH was within the pH range in East Texas, but was higher than the 4.77 monitored at the Nacogdoches International Paper Company by the Texas Air Control Board in 1983 (Driscoll and Porter, 1984).

It is difficult to assess pH values for two locations that were monitored in two different periods. However, air pollutants such as organic ash emitted from the International Paper Co. might have more impacts on precipitation acidity at its own site than at the City Landfill site about 20 km away. On the other hand, bulldozer activities and organic matter decomposition at the city landfill may add alkaline pollutants such as calcium and magnesium carbonates, ammonia, and other soil-derived material to the air. Whether the combined impacts of the site conditions had made precipitation pH higher at the city landfill and lower at the International Paper Co. requires additional observations and studies.

Average concentrations of the six metal ions (Cu^{2+}, Mn^{2+}, Pb^{2+}, Zn^{2+}, Mg^{2+}, and Al^{3+}) in precipitation at Nacogdoches were comparable to the average values of the US (Lazrus et al., 1970), but were one order of magnitude higher than the two stations in the Northeast, i.e. Greenville in ME and Essex in NY (Table 3). The higher concentrations of these ions at Nacogdoches also came along with an electrical conductivity 27.6 μS/cm, higher than the 22.4 and 12.3 μS/cm recorded at Greenville and Essex. Nacogdoches is located about 240 km north of Houston, the fourth largest city and the largest international seaport in the US. There, oil and gas exploration, petroleum refining, petrochemical production, and many other manufacturing processes may have great impacts on precipitation chemistry. In addition, paper mills, plywood mills, timber treatment plants, sawmills, and chemical companies are located south and southeast of town within 60 km. Since prevailing storm winds in Nacogdoches areas come from southerly directions (Chang et al., 1996), the anthropogenic sources of pollutants may be the Houston area, East Texas, or even Mexico.

In reality, differences in rainfall ion concentrations obtained from different studies are not practically comparable due to sample size, sample frequency, method of analysis, and detection limit. Based on a one-year field study comparing the rainfall ion concentrations of weekly measured samples and weekly values derived from daily samples in Georgia, Kansas, and Vermont, Topol et al. (1987) reported that the weekly measured ion concentrations were generally larger with differences up to 34% but less than 10% in most cases. Also, ion concentrations in rainwater generally do not follow a normal distribution. Their means can be greatly affected by one or two large values in the sampled data on one hand and by the detection limit of the analysis method on the other hand. For example, the detection limit for zinc concentrations is 0.1 mg/l by the Hach's (1995) DR-2000 spectrophotometer digestion procedure, but is 0.003 mg/l by the ion chromatography method. A water sample with 0.003 mg/l of Zn will be noted as <0.1 mg/l if analyzed by the Hach procedure

Table 2
Descriptive statistics for eight water quality variables for runoff from four roofing materials in 31 storms and rainwater observed between October 1998 and December 1999 at the City Landfill, Nacogdoches, TX

Variable	Statistics	Roof runoff				Rainwater	Standard[a]
		Wood shingle	Composition shingle	Aluminum	Galvanized iron		
pH	Mean	*5.07*a	6.69b	*6.20*c	6.59d	5.55e	6.5–9.0
	Median	*5.03*	6.73	*6.22*	6.63	*5.69*	
	Maximum	6.89	8.25	7.26	7.41	7.03	
	Minimum	3.33	4.08	4.78	3.62	4.20	
EC (μS/cm)	Mean	38.78a	30.19b	14.53d	20.34c	27.60bc	
	Median	28.00	22.00	10.00	17.00	19.00	
	Maximum	232.00	179.00	57.00	172.00	79.00	
	Minimum	7.00	6.00	2.20	4.00	7.00	
Al (mg/l)	Mean	0.382a	0.495a	0.381a	0.435a	0.354a	<0.750
	Median	0.224	0.181	0.169	0.194	0.251	
	Maximum	*2.343*	*6.736*	*4.077*	*6.884*	*2.047*	
	Minimum	0.008	0.008	0.008	0.008	0.008	
Mg (mg/l)	Mean	0.982a	0.713b	0.372a	0.362c	0.823b	
	Median	0.646	0.368	0.292	0.246	0.487	
	Maximum	6.680	5.063	1.478	3.659	4.739	
	Minimum	0.082	0.023	0.004	0.001	0.053	
Mn (mg/l)	Mean	0.044a	0.028a	0.015b	0.017b	0.030b	<0.050
	Median	0.022	0.011	0.010	0.010	0.017	
	Maximum	*0.404*	*0.369*	*0.117*	*0.252*	*0.339*	
	Minimum	0.001	0.001	0.001	0.001	0.001	
Cu (mg/l)	Mean	0.029a	0.025a	0.026a	0.028a	0.043a	<0.013
	Median	0.022	0.018	0.020	0.020	0.021	
	Maximum	5.410	0.126	0.248	0.224	0.174	
	Minimum	0.001	0.001	0.001	0.001	0.001	
Pb (mg/l)	Mean	0.045a	0.038a	0.037a	0.049b	0.034a	<0.065
	Median	0.025	0.025	0.025	0.025	0.025	
	Maximum	*0.700*	*0.203*	*0.134*	*0.255*	*0.116*	
	Minimum	0.025	0.025	0.025	0.025	0.025	
Zn (mg/l)	Mean	*16.317*a	*1.372*d	*3.230*c	*11.788*b	*0.139*e	<0.120
	Median	*9.717*	*0.859*	*2.248*	*8.219*	0.085	
	Maximum	*109.7*	*13.590*	*16.600*	*212.330*	*0.978*	
	Minimum	0.039	0.043	*0.514*	*0.124*	0.003	

Italized values exceed the USEPA water quality standards and means with the same letter are not significantly different at the 95% probability level.
[a] See Table 3.

Table 3
Mean concentrations for selected metal ions (mg/l), electrical conductivity (μS/cm), and pH ($\log_{10}^{-1}$) in bulk precipitation at Nacogdoches, TX and a few locations in the North America

Location	Cu^{2+}	Mn^{2+}	Pb^{2+}	Zn^{2+}	Mg^{2+}	Al^{3+}	EC	pH	Reference
Nacogdoches, TX	0.044	0.030	0.030	0.139	0.823	0.354	27.6	5.55	
Greenville, ME	0.005		0.004	0.014	0.022		22.4	4.60	Smath and Potter (1987)
Essex, NY		0.002	0.003	0.052	<0.1		12.3		Peters and Bonelli (1982)
Berkeley, CA	0.005	0.003		0.016	0.070		13.2	5.0	McColl and Bush (1978)
Glen Ellyn, IL[a]	0.080		0.04	0.057		0.684			Gatz et al. (1984)
USA	0.021	0.012	0.034	0.107					Lazrus et al. (1970)
Hamilton, Ont.[b]	0.01–0.04	0.148–1.497		0.042–0.214	0.540–5.30	0.227–2.41			Landsberger et al. (1987)

[a] Median.
[b] Range.

and would be entered as 0.05 mg/l for statistical calculations (Newman et al., 1989). Concentrations could be one order of magnitude different between these two methods.

3.2. Roof runoff quality

Descriptive statistics including mean, median, maximum, and minimum of the eight water quality variables for roof runoff collected during the study period are presented in Table 2. Water quality standards are also listed in the table for reference.

Of the eight variables studied, no published standards are available for magnesium (Mg) and electrical conductivity (EC). The typical concentration of Mg in streams is about 4 mg/l, but may be as high as 379 mg/l (McCutcheon et al., 1992). The Mg means and medians in runoff and rainwater from the four roof-types were all less than 1.0 mg/l, and the maximum recorded concentration in this study was 6.68 mg/l. Thus, Mg in runoff from the four roofing materials should not be a concern for its potential adverse effects on stream water quality. As for EC, the mean annual values for 43 USGS benchmark stream stations in the US ranged from 15.12 to 1150.81 μS/cm with a mean of 163.28 μS/cm (Binkley and Brown, 1993). Values of EC in runoff from the four roof types ranged from 2 to 232 μS/cm with means less than 40 μS/cm. These concentrations were in the lower range of US streams.

Runoff samples from the four roofing materials revealed concentrations of the other six variables (i.e. Al^{3+}, Mn^{2+}, Cu^{2+}, Pb^{2+} Zn^{2+} and pH) exceeding established standards (USEPA, 1999) at least 5% of the time (Table 4). The violations in water quality standards were the most severe for Zn^{2+} and Cu^{2+}. Zn^{2+} exceeded the national freshwater standards in virtually all runoff samples, while Cu^{2+} exceeded the standards in more than 60% of the samples, including all means and medians. At Bayreuth, Germany, Förster (1999) reported that concentrations of copper and zinc in roof runoff could far exceed various toxicity threshold values. No means and medians of Al^{3+}, Mn^{2+}, and Pb^{2+} in runoff from the four roof types exceeded the standards.

Results showed that the Zn concentration of the rainwater increased as it contacted the four types of roof material. Concentrations of Zn^{2+} ranged from 0.039 to 212.330 mg/l for runoff from the four roof types as compared to from 0.003 to 0.978 mg/l for rainwater in the open. The median ranged from 0.859 mg/l for composition shingle to 9.717 mg/l for wood shingle as compared to 0.085 mg/l for rainwater. Differences in Zn concentrations between runoff from the four types of roof and rainwater were statistically significant. The sources of Zn on the roof are believed to come from galvanized gutters and downspouts, nails, solder, dry-deposition of aerosols, fungi resistant materials, coating, and decomposition of organic matter. At Bayreuth, Germany, Zn concentrations in runoff from the zinc sheet roof, with median as high as 17.78 mg/l, were two to three orders of magnitudes above those in runoff from a roof without any metal components such as fibrous cement roof (Förster, 1999).

In addition to roof runoff, all means and medians of Cu concentrations in rainwater also exceeded the US national freshwater quality standards. Differences in Cu concentrations between roof runoff and rainwater and between runoff from any roof types were statistically insignificant. This means that the four roofing materials did not contribute appreciable quantities of Cu to roof runoff. Also, no significant differences in mean Al^{3+} runoff concentrations were measured for the four roof types. However, mean Mn^{2+} concentrations in runoff from wood and composition shingle roofs were found to be significantly greater than in runoff from painted aluminum or galvanized roofs. Furthermore, mean runoff concentrations of Pb^{2+} from galvanized iron roofs were significantly greater than the runoff concentrations from the other three roofing materials.

All mean pH values of runoff from the four roof types were statistically different than that of rainwater, but the effects seemed to be mixed. Wood shingle caused mean and median pH to be lower than those of rainwater, both exceeding the national freshwater quality standards.

Table 4
Percentages of time the runoff samples from four types of roof and rainwater in the open at Nacogdoches, TX exceeded the USEPA (1999) water quality standards

Variable	Roof runoff				Rainwater	Standard (mg/l)
	Wood shingle	Composition shingle	Aluminum	Galvanized Iron		
Al^{3+}	13.6	17.7	12.3	15.9	8.0	0.750[a]
Mn^{2}	27.7	14.7	4.9	6.4	8.0	<0.050[b]
Cu^{2+}	76.2	59.6	77.9	77.7	72.0	0.013[c]
Pb^{2+}	15.1	10.8	12.8	20.3	8.0	<0.065[c]
Zn^{2+}	99.5	99.5	100.0	100.0	68.0	<0.120[c]
pH	98.7	29.7	78.2	37.6	82.8	6.5–9.0[a]

[a] EPA freshwater quality for nonpriority pollutants.
[b] EPA drinking water quality for nonpriority pollutants.
[c] EPA freshwater quality for priority toxic pollutants.

The roughness and cracks of wood shingle roofs may trap water, which allows wood-rotting organisms to penetrate deeper into the wood, plants to grow, and organic matter to decay, and provide homes to an array of insects and other organisms. Additional H^+ ions that lower the pH values in wood shingle runoff may be accordingly released due to the weathering process of wooden material (cedar, red wood, or cypress in most cases) and the decomposition of wood-destroying fungi, lichens, mosses, debris, growing plants, insects, and other organic matter. On the contrary, composition shingles, painted aluminum, and galvanized iron roofs caused the pH of runoff to be significantly higher than the pH of rainwater. Apparently, ions released from these three roof types must be dominated by OH^- and consequently raise the pH values in rainwater draining through these roof surfaces.

3.3. Roof types

Of the four roofing materials, wood shingles had the lowest mean and median for pH and the highest means and medians for the other seven elements studied. However, differences in concentrations of these variables among the four roof types were insignificant for Al, Pb, and Cu, somewhat significant for EC, Mg, and Mn, and significant for pH and Zn.

Most wood shingle and shake roofs are made from western red cedar (*Thuja plicata*), red wood (*Sequoia sempervirens*), and cypress (*Taxodium distichum*). They are often impregnated with preservative chemicals such as copper naphthenate, copper octoate, and zinc naphthenate, and fungi killing chemicals most notably zinc sulfate, copper sulfate, and zinc chloride (Niemiec and Brown, 2002). Wood shingles are subject to weathering when exposed to sunlight and precipitation. The rough surfaces and cracks created by tension, compression, and shrinkage trap water, aerosols, and debris which in turn enhance fungi and moss development, attract insects and other organisms as a residence, and allow plants to grow. The organic matter retained on the roof is subject to accelerated decomposition due to heat energy from the sun, trapped moisture, and acid ions from rainwater. The results are more ions released from the roof, causing pH to be lowest and other water quality variables to be highest among the four roof types, especially Zn and EC. Thus, care and maintenance become important in element concentrations in roof runoff.

Zn concentrations from the four roof types were one to two orders of magnitudes higher than those in rainwater and most often violated EPA water quality standards. The mean and median ranged from the lowest 1.372 and 0.859 mg/l, respectively, for composition shingle to the highest 16.317 and 9.717 mg/l for wood shingle. Differences in Zn concentrations between any pair of roofs were statistically significant at the 95% level. Zn is soluble in water and the high concentrations in wood-shingle roof-runoff, in addition to galvanized gutters and downspouts mentioned above, apparently come from the treated Zn compounds. This suggests the necessity of using less Zn compounds and less galvanized materials in reducing Zn concentrations in roof runoff.

Although the mean and median of Al concentrations for painted aluminum roofs, respectively, 0.381 and 0.169 mg/l, were the lowest of the four roof types, differences were insignificant at the 95% level. The top and bottom sides of aluminum sheets are usually coated with resin paints such as polyester and silicone resins and fluoropolymers. This may help reduce the leaching of Al in aluminum roof runoff.

Compared to Chang and Crowley's (1993) study, Pb concentrations for both wood shingles and composition shingles were about the same, but the mean and median Zn concentrations were one order of magnitude higher in the present investigation. The wood-shingle roof used in the previous study was more than 20 years old. Apparently, a great portion of the impregnated chemicals in wood shingles had been leached out over the years.

3.4. Roof orientation

Most storms in the Nacogdoches area come from southerly directions in summer and fall and virtually from all directions in winter and spring (Chang et al., 1980). Thus, southeast (SE) is the most frequent and northwest (NW) the least frequent directions for incoming storms in the region. However, only runoff from wood shingles was affected by the orientation. EC, Mg, Mn, and Zn were significantly higher on the SE exposure than those on the NW. No other elements were affected by the two aspects for the other three roof types, or for all roofs combined. Note that the roof had a 25.8% or 14.5° slope. If a storm comes from the SE with 14.5° inclination, the rainfall catch by the NW roof is cos 29° or 87.5% of that by the SE. This is the maximum effect on rainfall interception for a storm with 14.5° inclination. The blocking effect on rainfall interception of the NW roof may be more significant if the SE-roof slope is greater. Orientation only affected runoff quality from wood shingles, possibly a result of its surface roughness as discussed in Section 3.3. Aerosols, particulate matter, industrial emissions, and other air pollutants brought from industrial regions in Lufkin and Houston by the southeast prevailing wind are expected to be deposited and retained more on wood shingles than on other roofing materials. As a result, only the SE aspect of wood shingles produced greater metal concentrations than the NW aspect; orientation imposed no effects on the water quality of the other three roofing materials.

3.5. Storm conditions

In addition to storm track pattern that may affect sources of airborne particulates (Hanson and Norton, 1982; Chang and Crowley, 1993), other factors may include storm conditions such as rainfall amount, intensity, duration, and occurrence interval. Spearman's correlation analyses showed that rainfall amount, intensity, and duration, in most cases, had a negative impact on the concentrations of the six metal elements, although the impact was weak with most of the correlation coefficients in the −0.20s and −0.10s. The highest correlation coefficient was −0.42 between rainfall intensity and Zn in aluminum roof runoff. However, the impacts of rainfall occurrence interval were positive for Al, Mg, Mn, and Zn, but negative for Cu and Pb. Longer storm interval implies more airborne fallout and more weathering and deposition processes, resulting in more release of ions. The negative effect of rainfall interval on Cu and Pb was unclear; perhaps the coefficients were too low to be significant.

4. Conclusions

Roofs can be a nonpoint source of water pollution. However, results of roof runoff studies have been variable. The variation reflects differences in roofing materials, industrial treatments, care and maintenance, age, climatic conditions, orientation and slope of roofs, and air quality of the region. In the East Texas area, concentrations of Cu and Zn in rainwater already exceed the EPA freshwater quality standards even without pollutant input from roofs. This is probably due to industrial emissions from petroleum refining, petrochemical production, and forest products production in the Houston and East Texas areas. Of the eight roof runoff quality variables studied, only pH, EC, and Zn were significantly affected by the types of roofing materials. However, concentrations of Al, Mn, Cu, Pb, Zn, and pH in roof runoff exceeded the national quality standards at least 5% of the time. Zn and Cu most often violated standards. Concentrations of Zn exceeded the standard in all roof runoff samples (100%), while Cu exceeded the standard more than 60% of the time. Although the mean and median concentrations of Cu exceeded the standard in all roof types, differences in concentrations between any two roofs and between roofs and rainwater were statistically insignificant.

The mean and median Zn concentrations were the highest for wood shingles, followed by galvanized iron, painted aluminum, and composition shingle. They were all significantly higher than for rainwater; all exceeded the EPA freshwater quality standards, and were significantly different among each other. Zn concentrations in runoff from new wood-shingle roofs were significantly higher than those from the aged wood-shingle roofs in a previous study in the same area. Since Zn is the major pollutant in roof runoff regardless of roof type and the major sources of Zn come from Zn compounds associated with chemical treatments and fungi protection, galvanized gutters and downspouts, and nails, it may become necessary to use less Zn compounds and less galvanized materials to reduce Zn concentrations in roof runoff.

Runoff quality from wood shingles was the worst of the four roof types studied. Leaf litter, pine needles, and other debris often accumulate between wood shingles, in the valleys of the roof structure, and in gutters. They tend to retard the drainage of water, reduce the circulation of air, trap more aerosols, allow water penetration into the wood, enhance the growth of molds and fungi, and accelerate organic matter decomposition. This makes care and maintenance of wood shingle roofs very important with respect to roof life and runoff quality.

All the four roof types caused runoff pH to be significantly different than rainwater pH, but only the means and medians for wood shingle and painted aluminum exceeded the water quality standards. Wood shingles tended to lower runoff pH, while the other three roof types tended to raise runoff pH values. Although SE is the most frequent and NW the least frequent direction for incoming storms, only EC, Mg, Mn, and Zn in wood shingle runoff from the SE were significantly higher than those from the NW. The two aspects affected no other elements in runoff from the other three roof types.

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