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Buildup characteristics of roof pollutants in the Shanghai urban area, China*

Biao WANG, Tian LI†

(*State Key Lab of Pollution Control and Resource Reuse, Tongji University, Shanghai 200092, China*)

†E-mail: tianli@mail.tongji.edu.cn

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Abstract: The buildup of roof pollutants in an urban area of Shanghai, China was investigated by conducting 16 experiments between November 2007 and October 2008. Concentrations of Cu, Zn and Cd in runoff from three types of roof (concrete, aluminum and glass) exceeded USEPA National Recommended Water Quality Criteria. The solid/liquid partition of the selected metal elements was consistent for the three roof types: Al, Fe, Zn and Pb were present mainly in the particle-bound form, while the total loading of Cd was nearly 100% in the dissolved form. Atmospheric dry precipitation accounted for most of all pollutant loadings for all roof types, while roof material made only a minor contribution to the loadings. All pollutant accumulation rates except for COD showed a seasonal trend with peaks in spring (March~May) and winter (December~February) and troughs in summer (June~August) and autumn (September~November). Our results showed that a linear equation is the most reliable of commonly used buildup models to simulate the total phosphorus (TP) and total suspended solids (TSS) buildup processes on aluminum roofs and glass roofs. This study provided novel information about roof runoff in Shanghai, China, in terms of pollution status, pollution source and pollutant buildup processes, thereby aiding in rainwater utilization and non-point pollution control.

Key words: Roof runoff, Pollutant buildup, Buildup model, Seasonal trend
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INTRODUCTION

The roof is considered as a source of non-point water pollution (Bucheli *et al*., 1998a; 1998b; Chang *et al*., 2004). Water quality of roof runoff is influenced by many variables, such as roof type, locality, season and rainfall characteristics (Förster, 1996; 1998; 1999; Zobrist *et al*., 2000; van Metre and Mahler, 2003), creating uncertainties and problems for the management of non-point pollution.

Mathematical models are commonly applied to predict runoff characteristics and minimize these uncertainties. Numerous studies of models have focused on surface runoff but few have considered roof runoff (Tomanovic and Maksimovic, 1996; Cristina and Sansalone, 2000; Yan and Kahawita, 2000; Kim *et al*., 2005; Kang *et al*., 2006; Clark *et al*., 2007; Kayhanian *et al*., 2007). Currently, these models consist mainly of black-box and grey-box models as the complexity of surface runoff has not been thoroughly understood. For the black-box models, the relationship between runoff and its related factors was examined, based on the analysis of a large number of runoff events, to obtain an empirical equation (Brezonik and Stadelmann, 2002; Kayhanian *et al*., 2007). One of the main shortcomings of this kind of model is that its result is reliable only if there is a large quantity of data, which is costly to produce in terms of money and labor. By contrast, grey-box models need less information for model construction as they partially clarify how the factors affect the runoff water quality. However, they have not been widely used except for a few successful applications on simple watersheds

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(Akan *et al*., 2000; Kang *et al*., 2006).

In both kinds of models, some model parameters, such as the buildup coefficient or even model equations may change from site to site, depending on the local environment, surface properties and contaminant species. Obviously, the roof differs from the ground in terms of the generation and loss of pollutants and in surface characteristics. It is generally assumed that pollution sources of roof runoff are simpler than those of surface runoff as atmospheric precipitation and roof material are the major pollution sources of roof runoff. Furthermore, a roof is normally smoother and has a steeper slope than the ground. Therefore, there is an urgent need for more studies on roof runoff. In addition, in cities there are many different kinds of roof material which may lead to different patterns of pollutant accumulation. For example, it was estimated that a metallic roof is a source of some species of heavy metals such as Cd and Zn (van Metre and Mahler, 2003). Finally, few studies have been conducted on roof pollutant buildup in Shanghai, China.

In this work, three roofs made from different materials (concrete, aluminum and glass) were laid on the roof of a building to investigate the buildup processes of local roof pollutants. There were three specific aims: (1) to examine the effect of roof materials and seasons on pollutant loading during antecedent dry days; (2) to investigate correlations among the selected contaminants and the partitioning of accumulated metal elements between the dissolved and solid phases; (3) to establish buildup equations and related parameters suitable for different pollutants and roof materials.

MATERIALS AND METHODS

Study area

The study was conducted on the roof of a five-storey building near the intersection of an overhead road and a normal street. The building was 140 m away from the overhead road and 60 m from the street. The study area was situated in an urban area of northern Shanghai, China, which has a typical subtropical monsoon climate with the following main characteristics: (1) There are four distinct seasons: spring (March~May), summer (June~August), autumn (Sepember~November) and winter (December~February); (2) The prevailing wind is from the southeast in summer and autumn, and from the northwest in spring and winter; (3) About 60% of the annual rainfall occurs from June to September. Baosteel, an iron and steel manufacturing and coal-fired power generation complex, is located to the northwest of the study area within a distance of about 20 km.

Sampling and measuring

From December 2007 to October 2008, 16 experiments (4 for each season) were performed with a period of antecedent dry days ranging from 2 to 10 d. Three man-made roofs (each 1.2 m wide×1.2 m long) made from different materials (concrete, aluminum and glass) were laid in parallel on the building roof. While atmospheric dry precipitation exhibited an uneven spatial distribution and each experimental roof had only a small area, we considered that the antecedent dry days were long enough to minimize any edge effects.

To determine the pollutant loadings, a simulated rainstorm produced using a simulator and tap water was applied to wash the experimental roofs. The simulator could produce an approximately even rainfall. Rainfall intensity was measured using a rain gauge and could be modified by adjusting the degree of opening of a pipeline valve. Methods for collecting accumulated pollutants can be classified as either ‘dry’ or ‘wet’. The method used in this study could be categorized as a wet method, capable of collecting the smallest particles left behind by traditional sampling techniques using dry vacuuming (Deletic and Orr, 2005). When the experiment was in progress, each roof in turn was placed on an iron structure (slope=2%). The structure was carefully designed so that it was able to support each roof and a PVC gutter collecting roof runoff. To prevent water from overflowing the sides of each roof and to minimize the loss of pollutants as a result of rain splash, two baffles were installed temporally and the rainfall intensity was kept as low as possible. An example of the experimental equipment with a concrete roof is illustrated in Fig.1.

Once roof runoff was generated, a 1-L PVC bottle was used to collect the runoff. When the first bottle was filled, the next bottle was applied in its place. The above process was repeated until most of the sediments on the roof surface had been removed according to tactile examination using a finger to touch the roof surface. When each experiment was finished, the simulated rainfall amount and the total volume of collected runoff were normally 8~10 mm and 10~12 L, respectively, depending on the accumulated pollutant loadings on the roofs. The amount of simulated rainfall approximated to the mean annual rainfall (10.72 mm) in Shanghai from 1985 to 2004 (Ning, 2005). After sampling, each bottle of sample was analyzed for total phosphorus (TP), total nitrogen (TN), chemical oxygen demand (COD) and total suspended solids (TSS). US Environmental Protection Agency (USEPA) approved methods (APHA, 1998) were used for all quality analyses.

Fig.1 Experimental equipment with a concrete roof

In 12 experiments during the period from March 2008 to October 2008, a composite sample was prepared for each experiment and each roof type. Both total and dissolved aluminum, chromium, iron, copper, zinc, cadmium and lead were determined in acidified samples (0.1 mol/L HNO_3). The dissolved fraction was obtained by filtration (0.45 μm polytetrafluoroethylene (PTFE) membrane) before acidification. All metal elements were measured using inductively-coupled plasma optical emission spectrometry system (ICP-OES) (Perkin-Elmer Optima 2100, USA). The particle-bound fraction was the difference between the total concentration and the dissolved fraction.

Data processing and statistical analyses

The pollutant loading per roof area was approximated by the sum of the pollutant mass of each sampling bottle divided by the roof area. Since the data failed to meet the normality assumption for parametric statistical analyses, the difference between the loadings of each roof type was tested using the Wilcoxon matched-pairs signed-ranks test. Similarly, the Spearman's rank correlation coefficient was used to determine the relationship between the loadings of all pollutants, as the assumption for the parametric Pearson's correlation coefficient was not met.

RESULTS AND DISCUSSION

Pollutant buildup rate

Table 1 lists the pollutant buildup rates of TP, TN, TSS, COD, Al, Cr, Fe, Cu, Zn, Cd and Pb for the three roof types. The buildup rates of all pollutants covered a wide range. The loadings for all metal elements on the concrete roof and the aluminum roof were sometimes lower than the detection limits. This variability demonstrated the complexity of contaminant accumulation.

As almost all water quality standards can be taken only as a reference for pollutant concentrations, the pollutant buildup rates were converted into concentrations on the basis of the rainfall characteristics in Shanghai, China, to facilitate the proper evaluation of the pollutant loadings. Assuming accumulated pollutants are completely washed away from roofs, the event mean concentrations (EMC, mg/L) of the annual average roof runoff (runoff coefficient=1) can be expressed as

$$EMC = (R \times P) / I, \tag{1}$$

where R is the pollutant buildup rate (mg/(m^2·d)), P is mean annual rainfall interval (d) and I is mean annual rainfall (mm). According to Ning (2005), the values of P and I in Shanghai are 2.97 d and 10.72 mm, respectively.

The calculation results are listed in Table 2. Atmospheric wet precipitation was not considered even though it is known to be one of the most important sources of roof runoff pollution. Therefore, the actual pollutant concentrations are likely to be a little or much higher than the calculated results depending on the pollutants. For example, atmospheric wet precipitation from 1998 to 2003 had an average

Table 1 Pollutant buildup rates in relation to roof material

Roof type	Statistics	Pollutant buildup rate (mg/(m^2·d))										
		TP	TN	COD	TSS	Al	Cr	Fe	Cu	Zn	Cd	Pb
Concrete	Mean	0.047^{a}	2.446^{a}	29.67^{a}	51.2^{a}	3.22^{a}	0.05^{a}	4.40^{a}	0.13^{a}	2.48^{a}	0.02^{a}	0.16^{a}
	Median	0.040	1.518	30.26	41.2	0.64	0.04	2.16	0.05	2.12	0.02	0.06
	Maximum	0.111	16.02	85.67	124.4	14.69	0.12	21.32	0.36	7.50	0.07	1.32
	Minimum	0.020	0.438	22.44	16.7	0	0	0	0	0	0	0
Aluminum	Mean	0.108^{b}	2.062^{a}	34.09^{a}	76.3^{b}	3.18^{a}	0.04^{a}	3.78^{a}	0.12^{a}	4.07^{b}	0.02^{a}	0.13^{a}
	Median	0.094	1.714	31.21	66.5	2.51	0.04	2.89	0.05	3.62	0.02	0.11
	Maximum	0.180	4.299	71.17	209.8	8.48	0.12	10.38	0.44	13.75	0.07	0.39
	Minimum	0.061	0.517	15.63	31.8	0	0	0	0	0	0	0
Glass	Mean	0.107^{b}	1.963^{a}	31.26^{a}	65.6^{b}	4.22^{a}	0.05^{a}	4.25^{a}	0.10^{a}	3.73^{ab}	0.02^{a}	0.17^{a}
	Median	0.100	2.050	26.81	66.0	2.19	0.04	3.01	0.04	2.52	0.02	0.05
	Maximum	0.169	3.626	68.14	98.6	17.13	0.17	15.21	0.52	17.38	0.07	0.79
	Minimum	0.047	0.821	9.38	32.4	0.37	0	0.72	0	0.45	0	0
	Detection limits	0.002	0.002	0.02	0.2	0.012	0.006	0.009	0.002	0.047	0.003	0.008

Note: for the convenience of statistical analysis, the value ‘0’ indicates that the pollutants were below the detection limits, which were calculated based on the concentration detection limits of the analysis method, the collected runoff volume (10 L), the roof area (1.44 m^2) and the average antecedent dry days (4.47 d). Water quality parameters with the same letters a and b are not significantly different at the 90% probability level

Table 2 Calculated pollutant concentrations based on the rainfall characteristics in Shanghai, China

Roof type	Statistics	Calculated water quality parameters (mg/L)										
		TP	TN	COD	TSS	Al	Cr	Fe	Cu	Zn	Cd	Pb
Concrete	Mean	0.013	0.678	8.23	14.2	0.893	0.014	1.220	0.036	0.688	0.006	0.044
	Median	0.011	0.421	8.39	11.4	0.178	0.011	0.599	0.014	0.588	0.006	0.017
Aluminum	Mean	0.030	0.572	9.46	21.2	0.882	0.011	1.048	0.033	1.129	0.006	0.036
	Median	0.026	0.475	8.66	18.4	0.696	0.011	0.802	0.014	1.004	0.006	0.031
Glass	Mean	0.030	0.544	8.67	18.2	1.170	0.014	1.179	0.028	1.035	0.006	0.047
	Median	0.028	0.569	7.44	18.3	0.607	0.011	0.835	0.011	0.699	0.006	0.014

TN concentration of 4.74 mg/L, so the TN concentrations of roof runoff are likely to be much higher than the values in Table 1.

Almost all means and medians of Cu, Zn and Cd (Table 2) exceeded USEPA National Recommended Water Quality Criteria (0.013, 0.120 and 0.002 mg/L, respectively) (USEPA, 2006), similar to the results of previous studies. Förster (1999) found that concentrations of Cu and Zn in roof runoff far exceeded various toxicity threshold values in Bayreuth, Germany. Chang *et al.*(2004) also reported that Zn concentrations exceeded the USEPA freshwater standards in virtually all runoff samples, while Cu concentrations exceeded the standards in more than 60% of samples. Cd concentrations in this study were one order of magnitude above those in a previous study. In Zürich, Switzerland, the mean concentrations of Cd in 0~2 mm roof runoff from an inclined tile roof and an inclined polyester roof were found to be 0.40 and 0.30 μg/L, respectively, while the mean Cd concentration of the whole roof runoff from a flat gravel roof was 0.11 μg/L (Zobrist *et al*., 2000).

It has long been known that certain pollutants in stormwater such as polynuclear aromatic hydrocarbons (PAHs) and heavy metals tend to partition onto the solid phase. In particular, smaller particles have a higher solid-phase concentration (Lau and Stenstrom, 2005). Partitioning not only has implications for washoff of pollutants and solids, but also can indicate what physicochemical mechanisms will be most effective for immobilization of dissolved and particulate-bound mass (Sansalone and Buchberger, 1997). Thus, the distribution of the studied metal elements in the solid and liquid phases was also investigated in this study. The results indicated that for most elements, the mean percentages of dissolved metal were similar for all three roof types (Fig.2). Al, Fe, Zn and Pb were present mainly in the particle-bound form, with mean percentages of 85.9%, 88.0%, 60.7% and 69.5% for the concrete roof, 86.2%, 90.2%, 74.5% and 78.2% for the aluminum roof and 93.5%, 89.0%, 70.1% and 66.4% for the glass roof, respectively. In contrast, the dissolved fraction amounted almost to the total loading of Cd for the three roof types.

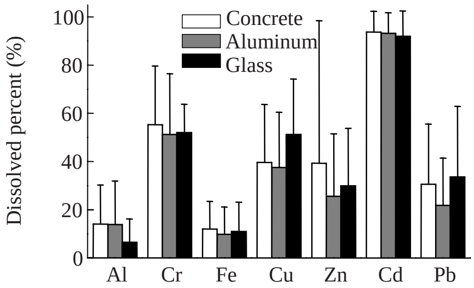

Fig.2 Partitioning of metal elements into dissolved and particle-bound fractions

Roof type

Statistical analysis (Table 1) revealed that: (1) Differences in the loadings of most pollutants among the three roof types were insignificant, suggesting that atmospheric dry precipitation is the major source of roof pollutants; (2) The aluminum roof and the glass roof yielded significantly more TP and TSS than the concrete roof at the 90% probability level; (3) The aluminum roof produced remarkably more Zn than the concrete roof.

Obviously, in this study the pollutant loadings of roof runoff came from atmospheric dry precipitation and roof material. As the aluminum and glass did not contribute to the loadings of TP and TSS, it can be concluded that they selectively absorbed more TP and TSS from atmospheric dry precipitation than the concrete. The aluminum roof contributed a large part of the Zn loading but not the Al loading of roof runoff. However, the Zn concentrations were much lower than those in roof runoff from zinc sheet roofs, which have median concentrations as high as about 17.68 mg/L (Förster, 1999). As the Zn loading of the glass roof was also greater than that of the concrete roof (although the difference was not statistically significant), the aluminum roof may have had a larger Zn loading because it absorbed more atmospheric dry deposition containing Zn, or because the aluminum roof itself may have yielded a certain loading of Zn (about 0.2% mass fraction).

To further identify the sources of pollutant loadings, an evaluation of the relationships among the pollutant loadings was conducted, which revealed some strong correlations, such as those between TP and TSS (R=0.674, p<0.05; R=0.733, p<0.01) for the aluminum roof and the glass roof. This suggested that TP and TSS for these roofs had a common source. However, the correlations were not the same for the three roof types: the relationship between TP and TSS for the concrete roof was not as close as that for the aluminum roof and the glass roof. This may be because of corrosion of the concrete roof and its rough surface which can trap some fine particles of roof runoff, considering that the TSS and TP loadings of the concrete roof were lower than those of the other roof materials.

For all roof types, the correlations among Al, Cr, Cd, Pb and between Fe and Zn were significant at the 90% level. Al came from the same source as Cr, Cd and Pb, which suggested again that the aluminum roof does not contribute much to the Al loading. It is interesting to note that the correlations between Fe and Zn (R=0.776, p<0.01; R=0.732, p<0.01) for the glass roof and the concrete roof were more significant than that (R=0.552, p<0.1) for the aluminum roof. It is known that Fe and Zn can be used as marker elements for steel-making furnaces (Huang *et al*., 1994; Zhang *et al*., 1998) and the significant correlation between Fe and Zn suggested that they were derived mostly from a common source, nearby steel-making furnaces. The lower values of the correlation coefficients for the aluminum roof suggested that its Zn loading to a certain degree may come from other sources such as the roof material.

Seasonal trends

Although the differences between pollutant loadings among roof materials were mostly not significant, accumulation rates showed an obvious seasonal variation for almost all pollutants. The accumulation rates of all pollutants except COD for the three roof types reached minimum values in summer or autumn (Fig.3). Indeed, some heavy metals such as Cr, Cd and Pb could not be detected in summer or autumn. This phenomenon must result from factors associated closely with the seasons. Shu *et al*.(2001) reported that when the wind comes from the north, the

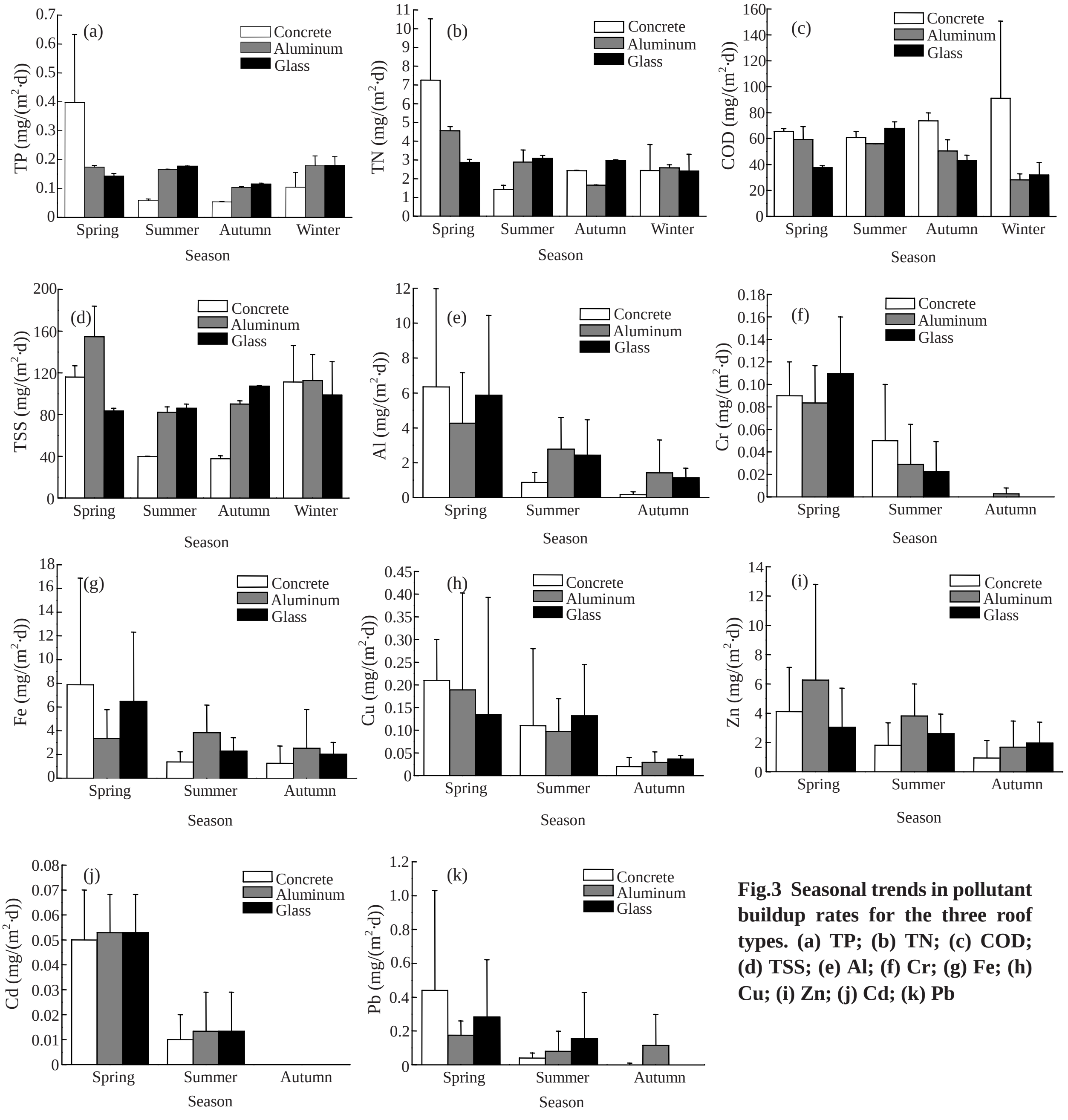

Fig.3 Seasonal trends in pollutant buildup rates for the three roof types. (a) TP; (b) TN; (c) COD; (d) TSS; (e) Al; (f) Cr; (g) Fe; (h) Cu; (i) Zn; (j) Cd; (k) Pb

main contributions to total suspended particles (TSPs) in Shanghai are products of coal combustion, with lesser contributions from construction sites, vehicle emissions, windblown soil and steel-making furnaces. Furthermore, they found that the mean TSP concentrations at a location downwind of Baosteel were higher than those at other non-downwind areas and that TSP concentrations for all sites were higher with northwest winds and lower with southerly wind directions. Our experiment area was situated at a location downwind of Baosteel and the results agreed well with Shu *et al.*(2001). The seasonal variation can be well explained by the dominant wind direction of the seasons. In spring and winter, northwest winds prevail, which contain a large number of particles derived from the main iron and steel manufacturing factory and coal-fired power generation plant. Furthermore, the nearby heavy-traffic highway may also contribute to a certain degree to the metals loading. In summer and autumn, the prevailing winds coming from the sea are southeasters and atmospheric dry precipitation contains low concentrations of metal elements.

However, it should be noted that the contribution of Baosteel to the loadings of metal elements and other pollutants may be different from those of other sources. Metals came largely from Baosteel, while most TP, TN, COD and TSS were derived from other sources such as traffic, construction sites or large-dimension dry precipitation, as their loadings were still high in summer and autumn when the study area was no longer downwind of Baosteel.

The seasonal trends in pollutant loading have implications for the treatment and utilization of roof runoff. Pollutant buildup rates in summer and autumn would be lower than those in winter and spring. However, most rainfall in Shanghai occurs in summer and autumn. Thus, it can be inferred that the water quality of roof runoff in summer and autumn would be cleaner than that in winter and spring. Therefore, it is beneficial to harvest and utilize rainwater in summer and autumn and non-point pollution control should be focused more on the treatment of roof runoff in spring and winter.

Buildup model

Normally, four functions are used to model the pollutant buildup process: linear, power, exponential, and Michaelis-Menton. Among these various models, the most widely employed is the exponential function (Chen and Adams, 2006), while only a few researchers have applied linear equations (Soonthornnonda *et al.*, 2008). All these functions assume that pollutant loadings increase with time, as shown by a large number of studies (e.g., Kang *et al.*, 2006; Kim *et al.*, 2006). However, a very different result was obtained by Deletic and Orr (2005) suggesting that the number of preceding dry days could have a weak negative influence on the loading. These contradictory results indicate that the pollutant buildup process may be site-specific.

The pollutant loadings of all metal elements in this study showed no obvious trend when the buildup duration increased. However, yields of TP, TN, COD, TSS and turbidity increased as the number of dry hours increased. An example of TP loading in relation to the number of dry hours for the aluminum roof is shown in Fig.4.

The four buildup equations were applied to fit the TP buildup process and the following linear equation (Eq.(2)) was found to be the most suitable:

$$y = Kx, \tag{2}$$

where y is the pollutant buildup load (mg/m^2), x is the number of dry hours (h) and K is the slope of the linear equation (mg/(m^2·h)).

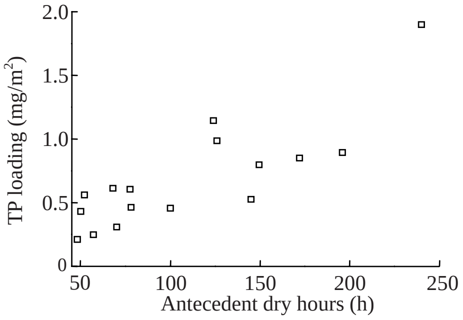

Fig.4 Relationship between TP loadings of the aluminum roof and the antecedent dry hours

The detailed procedure for establishing the linear model was as follows: Firstly, 12 random experiment points were chosen for a resampling method, namely bootstrapping; Secondly, these selected points were resampled 1000 times to form 1000 samples; Thirdly, 1000 K values were calculated using linear regression for the 1000 samples and the distribution, mean value and standard deviation of the K values were obtained; Finally, the linear equation with the mean K value was used to test against the remaining 4 experiment points. For example, the distribution of K values for TP is shown in Fig.5. The mean K value was 6.5×10^{-3} with a standard deviation of 7.9×10^{-4}.

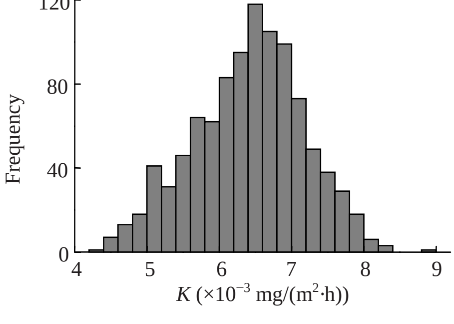

Fig.5 Distribution of *K* values for TP

A comparison between the predicted and the measured TP loadings for four independent experiments is shown in Fig.6. The measured and simulated results were in reasonably good agreement with relative errors ranging from 7.7% to 29.3%.

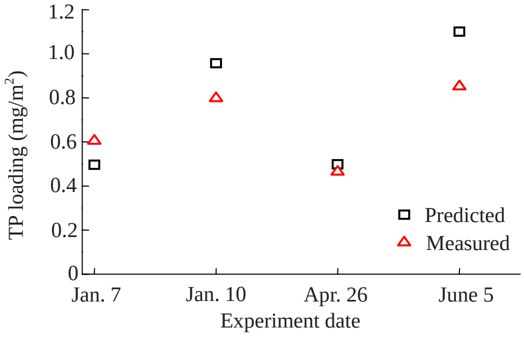

Fig.6 Linear equations applied to four independent experiments in 2008

The linear model could represent only the TP and TSS buildup processes on the aluminum roof and the glass roof with good prediction performance (relative errors=14.3%~32.5%). The corresponding linear equations are shown in Table 3.

Table 3 Linear equations of the TP and TSS buildup processes for the aluminum roof and the glass roof

Roof type	Linear equation	
	TP	TSS
Aluminum	y=0.0065x (0.00077)	y=4.3x (0.29)
Glass	y=0.0070x (0.00078)	y=3.7x (0.39)

Note: values in parentheses are the standard deviations of *K* values

CONCLUSION

Three roofs, each made from different materials (concrete, aluminum and glass), were placed on the roof of a building in Shanghai, China and a total of 16 simulated storms were conducted throughout the year to examine the characteristics of roof pollutant buildup. The conclusions were as follows:

(1) Concentrations of Cu, Zn and Cd in runoff from all three types of roof exceeded USEPA National Recommended Water Quality Criteria; Cu, Zn, Cd, Al, Fe, Zn and Pb exist mainly in the particle-bound form; by contrast, almost all Cd loading was found in the dissolved fraction.

(2) There were no significant differences between the loadings of most pollutants for the three roof types, implying that the main contribution to roof pollutant loading was atmospheric dry precipitation rather than the roof material. However, the aluminum roof and the glass roof contributed more loadings of TP and TSS than the concrete roof and the aluminum roof also produced significantly more Zn loading than the concrete roof.

(3) All pollutant accumulation rates (except for COD) showed an obvious seasonal variation and the accumulation rates for the three roof types reached minimum values in summer or autumn. This can be explained by the dominant wind direction in each season and the location of the main pollution sources, a steel manufacturing plant and a coal-fired power generation complex, to the northwest of the study site.

(4) Among four commonly used equations, the linear equation was the most reliable model to simulate the TP and TSS buildup processes on the aluminum roof and the glass roof.

References

Akan, A.O., Schafran, G.C., Pommerenk, P., Harrell, L.J., 2000. Modeling storm-water runoff quantity and quality from marine drydocks. *Journal of Environmental Engineering*, **126**(1):5-11. [doi:10.1061/(ASCE)0733-9372(2000)126:1(5)]

APHA (American Public Health Association), 1998. Standard Methods for the Examination of Water and Wastewater (20th Ed.). Washington, D.C.

Brezonik, P.L., Stadelmann, T.H., 2002. Analysis and predictive models of stormwater runoff volumes, loads, and pollutant concentrations from watersheds in the Twin Cities metropolitan area, Minnesota, USA. *Water Research*, **36**(7):1743-1757. [doi:10.1016/S0043-1354(01)00375-X]

Bucheli, T.D., Müller, S.R., Voegelin, A., Schwarzenbach, R.P., 1998a. Bituminous roof sealing membranes as major sources of the herbicide (R, S)-mecoprop in roof runoff waters: potential contamination of groundwater and surface waters. *Environmental Science & Technology*, **32**(22):3465-3471. [doi:10.1021/es980318f]

Bucheli, T.D., Müller, S.R., Heberle, S., Schwarzenbach, R.P., 1998b. Occurrence and behavior of pesticides in rainwater, roof runoff, and artificial stormwater infiltration. *Environmental Science & Technology*, **32**(22):3457-3464. [doi:10.1021/es980317n]

Chang, M., McBroom, M.W., Beasley, R.S., 2004. Roofing as a source of nonpoint water pollution. *Journal of Environmental Management*, **73**(4):307-315. [doi:10.1016/j.jenvman.2004.06.014]

Chen, J., Adams, B.J., 2006. Analytical urban storm water quality models based on pollutant buildup and washoff processes. *Journal of Environmental Engineering*, **132**(10):1314-1330. [doi:10.1061/(ASCE)0733-9372(2006)132:10(1314)]

Clark, S.E., Burian, S., Pitt, R., Field, R., 2007. Urban wet-weather flows. *Water Environment Research*, **79**(10):1166-1227. [doi:10.2175/106143007X218386]

Cristina, C.M., Sansalone, J.J., 2000. Kinematic wave model of urban pavement rainfall-runoff subject to traffic loadings. *Journal of Environmental Engineering*, **126**(7):629-636. [doi:10.1061/(ASCE)0733-9372(2003)129:7(629)]

Deletic, A., Orr, D.W., 2005. Pollution buildup on road surfaces. *Journal of Environmental Engineering*, **131**(1):49-59. [doi:10.1061/(ASCE)0733-9372(2005)131:1(49)]

Förster, J., 1996. Patterns of roof runoff contamination and their potential implication on practice and regulation of treatment and local infiltration. *Water Science & Technology*, **33**(6):39-48. [doi:10.1016/0273-1223(96)00329-0]

Förster, J., 1998. The influence of location and season on the concentrations of macroions and organic trace pollutants in roof runoff. *Water Science & Technology*, **38**(10):83-90. [doi:10.1016/S0273-1223(98)00736-7]

Förster, J., 1999. Variability of roof runoff quality. *Water Science & Technology*, **39**(5):137-144. [doi:10.1016/S0273-1223(99)00095-5]

Huang, X., Olmez, I., Aras, N.K., Gordon, G.E., 1994. Emissions of trace elements from motor vehicles: potential marker elements and source composition profile. *Atmospheric Environment*, **28**(8):1385-1931. [doi:10.1016/1352-2310(94)90201-1]

Kang, J.H., Kayhanian, M., Stenstrom, M.K., 2006. Implication of a kinematic wave model for first flush treatment design. *Water Research*, **40**(20):3820-3830. [doi:10.1016/j.watres.2006.09.007]

Kayhanian, M., Suverkropp, C., Rubyb, A., Tsay, K., 2007. Characterization and prediction of highway runoff constituent event mean concentration. *Journal of Environmental Management*, **85**(2):279-295. [doi:10.1016/j.jenvman.2006.09.024]

Kim, L.H., Kayhanian, M., Zoh, K.D., Stenstrom, M.K., 2005. Modeling of highway stormwater runoff. *Science of the Total Environment*, **348**(1-3):1-18. [doi:10.1016/j.scitotenv.2004.12.063]

Kim, L.H., Zoh, K.D., Jeong, S.M., Kayhanian, M., Stenstrom, M.K., 2006. Estimating pollutant mass accumulation on highways during dry periods. *Journal of Environmental Engineering*, **132**(9):985-993. [doi:10.1061/(ASCE)0733-9372(2006)132:9(985)]

Lau, S.L., Stenstrom, M.K., 2005. Metals and PAHs adsorbed to street particles. *Water Research*, **39**(17):4083-4092. [doi:10.1016/j.watres.2005.08.002]

Ning, J., 2005. Study on Rainstorm Intensity Formula for Short Duration and Design Hyetograph of Shanghai. MS Thesis, Department of Environmental Engineering, Tongji University, Shanghai, China (in Chinese).

Sansalone, J.J., Buchberger, S.G., 1997. Partitioning and first flush of metals in urban roadway storm water. *Journal of Environmental Engineering*, **123**(2):134-143. [doi:10.1061/(ASCE)0733-9372(1997)123:2(134)]

Shu, J., Dearing, J.A., Morse, A.P., Yu, L., Yuan, N., 2001. Determining the sources of atmospheric particles in Shanghai, China, from magnetic and geochemical properties. *Atmospheric Environment*, **35**(15):2615-2625. [doi:10.1016/S1352-2310(00)00454-4]

Soonthornnonda, P., Christensen, E.R., Liu, Y., Li, J., 2008. A washoff model for stormwater pollutants. *Science of the Total Environment*, **402**(2-3):248-256. [doi:10.1016/j.scitotenv.2008.04.036]

Tomanovic, A., Maksimovic, C., 1996. Improved modeling of suspended solids discharge from asphalt surface during storm event. *Water Science & Technology*, **33**(4-5):363-369. [doi:10.1016/0273-1223(96)00253-3]

USEPA (US Environmental Protection Agency), 2006. National Recommended Water Quality Criteria-Correction. Office of Water EPA 822-B-00-021, Washington, D.C.

van Metre, P.C.M., Mahler, B.J., 2003. The contribution of particles washed from rooftops to contamination loading to urban streams. *Chemosphere*, **52**(10):1727-1741. [doi:10.1016/S0045-6535(03)00454-5]

Yan, M., Kahawita, R., 2000. Modelling the fate of pollutant in overland flow. *Water Research*, **34**(13):3335-3344. [doi:10.1016/S0043-1354(00)00053-1]

Zhang, J., Cheng, Z., Wang, W., 1998. Source apportionment on fine particles in atmosphere in Beijing. *Acta Scientiae Circumstantiae*, **18**:62-67.

Zobrist, J., Müller, S.R., Ammann, A., Bucheli, T.D., Mottier, V., Ochs, M., Schoenenberger, R., Eugster, J., Boller, M., 2000. Quality of roof runoff for groundwater infiltration. *Water Research*, **34**(5):1455-1462. [doi:10.1016/S0043-1354(99)00290-0]