

Research Article

Distribution and Health Risk Assessment of Some Trace Elements in Runoff from Different Types of Athletic Fields

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Environmental risk of heavy metals and metalloids in athletic fields has raised people's attention in the recent years. Seven trace elements, including metals and metalloids, were detected in the runoff of five typical athletic fields in the university campus under three rainfall events. Except for Cr, the total concentrations of Zn, Pb, Cu, Mn, Cd, and As in artificial turf runoff are the highest among five athletic fields, followed by that of plastic runway. The concentration and first flush effect of trace elements are followed in the order of $1^{st} > 2^{nd} > 3^{rd}$ rainfall events. The strongest correlations between metals and metalloids were observed in the tennis court runoff, while the artificial turf shows the least. The release of trace elements could be directly from the surface materials and particles on the athletic field and influenced by the comprehensive factors including surface materials, rainfall events, and pollutant characters. Pollution risk assessment shows that the pollution extent of the five types of athletic field is at least "moderate" and follows the order of artificial turf > basketball court > plastic runway > badminton court > tennis court. Pb shows the highest pollution level, while Cr shows the highest healthy risk. The results can provide a theoretical basis for runoff pollution control and safety use of athletic fields.

1. Introduction

With the continuous improvement of people's living standard, people are giving more and more importance to their health. Athletic fields as one of the important sports areas are more and more popular among the public to perform exercise activities such as running and playing football, basketball, and tennis. Athletic fields are much durable than natural grass fields; thus, the number of athletic fields has increased significantly globally. As of 2020, the number of athletic fields in China has reached 55,000. The number of athletic fields in the United States has exceeded 11,000, and the number in Europe is approximately 21,000. Athletic fields are usually built in schools, parks, communities, and public entertainment venues. However, health problems were frequently found to be originated from athletic fields that have attracted people's attention. The materials of athletic fields are normally synthesized using rubber polymer, reinforcing agents (e.g., carbon black), aromatic extender oil, vulcanization additives, antioxidants and pigments, etc [1]. The commonly used rubber fillers in the athletic fields are often composed of ethylene propylene diene monomer (EPDM) and thermoplastic elastomer (TPE). In order to save manufacturing costs, recycled materials such as recycled tires and recycled EPDM rubber are often used as filling materials of athletic fields [2, 3]. With the usage of athletic fields, exogenic processes such as rainfall, weathering, and mechanical abrasion facilitate the

degradation of materials. During the aging process, the chemicals could be inevitably released from the materials, and people may get health problems via inhalation, ingestion, and dermal contact.

To make safe use of athletic fields, a number of countries have issued relevant laws and regulations to regulate the hazard chemical leaching level from materials in the athletic fields. However, in the real conditions, hazard chemicals are still inevitably released. Heavy metals and metalloids are harmful to human health. They were reported to be detected in the materials of athletic fields that may cause environmental pollution and human health problems [4]. Marsili et al. showed that the rubber particles in artificial turf contain heavy metals such as Cd, Pb, Cr, Ni, Cu, and Zn [5]. Different kinds of raw materials release different kinds of metals. Krüger et al. used the column test to analyze the leaching concentration of heavy metals from artificial turf system components [6]. They found high release of Zn in the column test for all materials, and the highest releasing concentration was 4.10 mg/L. Metals and metalloids could accumulate in athletic fields during dry weather periods and be washed off during rainfall events. Magnusson and Mácsik show that the recycled materials contain pollutants that were easier to be released into the environment, compared with nonrecycled materials [7].

Urban runoff has been reported as a major source of pollution contributing to the deterioration of the quality of receiving waters such as lakes and streams as well as being harmful to ecosystems within or close to cities [8]. Some researchers have noticed the pollution in the athletic field runoff. Celeiro et al. analysed the runoff collected directly from the artificial turf, and they detected heavy metals such as Cd, Pb, and Cr in high concentrations [9]. However, previous researches mainly focused on the specific areas such as artificial turf and plastic runway. Runoff pollution in other types of athletic fields remains unknown. Especially for heavy metals and metalloid pollution in different athletic fields, the comparison of the pollution level should be systematically conducted. In addition, environmental conditions such as rainfall intensity and duration could affect the release behavior of chemicals in the materials of the athletic field. However, there are few studies that carried out field monitoring towards heavy metals on athletic fields. Thus, field monitoring towards runoff pollution is still necessary to be carried out in artificial fields.

Heavy metals and metalloid could exist in the dusts on the road and directly gets released into the runoff with the rainfall that may cause health problems if those tiny particles are inhaled by people [10]. In addition, runoff could carry heavy metals and metalloids into the water system which eventually may cause water pollution. Human exposure to crumb rubber-derived chemicals may occur through inhalation, ingestion, or dermal contact. The dominant route by which the various chemicals in crumb rubber enter the human body will depend, in part, upon each compound's physicochemical properties. Kim et al. show that Pb in the artificial turf could enter the human body through inhalation [11]. Lead-based coloring agents used in fibers of some artificial turf can be released into the environment on the usage of athletic fields. Heavy metals have been identified as potential contaminants of concern, particularly lead, which can cause severe permanent neurological health effects in children [12]. The health risk assessment of heavy metals and metalloids in the athletic field runoff is necessary to be systematically investigated.

In order to know about the occurrence and risk of trace elements from different types of athletic fields, five commonly used athletic fields including the artificial turf, plastic runway, basketball court, badminton court, and tennis court in the university campus were selected as sampling fields. Six heavy metals Zn, Pb, Cu, Mn, Cr, and Cd and one metalloid As were selected as analytical compounds. Sampling was carried out under three typical rainfall events. The objectives were (i) to distinguish the distribution of heavy metals and metalloid concentrations in runoff from different types of athletic fields; (ii) to study the effect of rainfall event characters on the release of heavy metals from different types of athletic fields; (iii) to observe the correlation between metals and metalloids; and (iv) to assess the pollution level and health risk of metals and metalloids in different types of athletic field runoffs.

2. Materials and Methods

2.1. Sampling Sites. Five types of athletic fields located in one university campus of Daxing Distrct, Beijing, China, were selected as research filed, including the artificial turf, plastic runway, basketball court, badminton court, and tennis court. The main materials of the underlying surface and the specific locations of the runoff sampling points are shown in Table 1 and Figure 1. The concentrations of seven trace elements in the five athletic field runoffs were detected under three rainfall events in the summer of 2019. Rainfall characters collected from the Meteorological Bureau of Daxing District (Beijing) are shown in Table 2. To investigate the effect of rainfall duration on heavy metal and metalloid concentration distribution, sampling was carried out at different raining time intervals. The first runoff sample was taken when the runoff was generated (defined as time 0 min). Then, sampling was collected every 30 min until the rain stops. The sampling volume is 300 mL and is collected in a brown glass without leaving the headspace. Natural rain samples (rainfall without touching the surfaces) were collected as blank samples during entire rainfall events. The samples were then taken to the laboratory and stored at 4°C in a fridge for further analysis.

2.2. Laboratory Analysis. Seven trace elements investigated in this study were Zn, Pb, Cu, Mn, Cr, Cd, and As which were chosen on the basis of their toxicities and potential environmental risks. Heavy metal and metalloid standards were purchased from Guobiao (Beijing) Testing & Certification. The trace elements in the runoff measured in this study were the total amount in both water and solid phases, including the amount adsorbed by suspended solids in runoff samples. Therefore, the concentration of the trace elements we measured is the full concentration in both

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| Sampling area | Filling materials | Catchment area (km ²) |
|---|---|-----------------------------------|
| Artificial turf | Polyethylene, rubber particles | 16.3 |
| Plastic runway | Polyurethane prepolymer rubber particles | 17.2 |
| Basketball court | Rubber | 2.90 |
| Badminton court | Acrylic acid | 1.45 |
| Tennis court | Siloxane-modified polyurethanes | 1.82 |
| Basketball court Badminton court Tennis court | Rubber Acrylic acid Siloxane-modified polyurethanes | 2.90 1.45 1.82 |

TABLE 1: Main characters of the underlying surface of the five athletic fields.



FIGURE 1: Sampling sites of five athletic fields located in one university campus of Daxing Distrct, Beijing, China.

| TABLE 2: 0 | Character | of | rainfall | events | at | sampling | locations. |
|------------|-----------|----|----------|--------|----|----------|------------|
|------------|-----------|----|----------|--------|----|----------|------------|

| Rainfall event | Date | Antecedent dry days | Rainfall duration (min) | Rainfall (mm) |
|-----------------|------------------------------|---------------------|-------------------------|---------------|
| 1 st | July 5 th , 2019 | 40 days | 90 | 8.2 |
| 2 nd | July 22 nd , 2019 | 4 days | 60 | 9.6 |
| 3 rd | July 29 th , 2019 | 7 days | 90 | 8.0 |

phases and solid particles remained in runoff, not only refers to the free dissolved concentration in water phase. The methods of measuring the trace elements were referred to Environmental Protection Standards of the People's Republic of China "Water quality-determination of 65 elements-inductively coupled plasma-mass spectrometry" (HJ700-2014). Microwave digestion (Top wave, Analytic Jena, Germany) was employed for the extraction of metals and metalloid from runoff samples. Before digestion, 0.8 mL nitric acid and 0.2 mL sulfuric acid (GR, HuShi, China) were mixed with 9 mL samples in a Teflon tube standing for half an hour. Microwave digestion was then carried out at 1000 W starting at 70°C (5 min) and reaching 180°C (15 min) after intermediate steps of 5 min at 145°C. After cooling them, the final extracts were adjusted to 50 mL with deionized water and transferred through 0.45 µm polyethylene filter (JinTeng, China) into cleaned bottle. The metal contents of runoff samples were determined by inductively coupled plasma-mass spectrometry (ICP-MS) (PerkinElmer NexION 300X) analysis. Each batch of samples was run simultaneously with two blanks.

2.2.1. Identification of First Flush Events. The first flush behavior was initially analyzed by the dimensionless cumulative pollutant load M(t) versus dimensionless cumulative runoff volume V(t), curve (M(V) curve). Assuming that the flow rate and the pollutant concentration vary linearly between two successive measurements, M(t) and V(t) can be defined as given in equations (1) and (2), respectively:

$$M(t) = \sum_{i=1}^{n} \frac{C_i Q_i \Delta t_i}{M},$$
(1)

$$V(t) = \sum_{i=1}^{n} \frac{Q_i \Delta t_i}{V},$$
(2)

where Q_i and C_i are the respective flow rate (L/s) and concentration (μ g/L) at time t_i corresponding to the *i*th measurement of an event having *n* number of measurements and *M* and *V* are the total pollutant load and the total runoff volume discharged.

2.3. Pollution and Health Risk Assessment

2.3.1. Pollution Index. To assess the pollution condition in athletic fields runoff, the pollution index (P_i) method was applied (Yan et al. 2015) [13]:

$$P_i = \frac{C_i}{S_i},\tag{3}$$

where C_i is the concentration of the pollutant (μ g/L) and S_i is the Chinese class III surface water quality standard for the pollutant (GB 3838–2002), where Zn was 1000 μ g/L, Pb was 50 μ g/L, Cu was 1000 μ g/L, Cr was 50 μ g/L, Cd was 5 μ g/L, and As was 50 μ g/L. It should be noted that the value of Mn is not available in the standard to calculate. We assume the urban runoff would reach nearby lakes and watershed without any treatment, and therefore, we use the threshold value for surface water for swimming and fishery area (class III). The runoff pollution were divided into five levels: unpolluted ($P_i \le 0.4$), slightly polluted ($P_i = 0.4$ –1.0), medium polluted ($P_i = 1.0$ –2.0), heavily polluted ($P_i = 2.0$ –5.0), and seriously polluted ($P_i > 5.0$) [13].

Finally, comprehensive pollution load index was used to evaluate the overall pollution situation (equation (4)) [14]:

$$IPI = \frac{1}{n} \sum_{i=1}^{n} P_i.$$
 (4)

The IPI can be divided into six different levels for pollution: unpolluted (0-0.2), subclean (0.2-0.4), slightly polluted (0.4-0.7), moderately polluted (0.7-1.0), heavily polluted (1.0-2.0), and seriously polluted (>2.0).

2.3.2. Health Risk Assessment. Due to the different chemical properties of heavy metals, human health risk assessment was classified into two categories: carcinogenic and non-carcinogenic. It was considered that the dermal absorption of contaminants by adhering to the exposed skin is the most common route for contaminants in water [15, 16]. The slope factors for carcinogenic contaminants and reference for noncarcinogenic chemicals were extracted from the Integrated Risk Information System (IRIS, USEPA). Most of the studies on the health risk assessment of toxic substances in water through skin contact use the calculation model proposed by USEPA. Health hazards caused by chemical carcinogens through skin contact risk R^p is

$$R_i^P = \frac{\left[1 - \exp\left(-\text{CDI}_i \times \text{SF}_i\right)\right]}{L},\tag{5}$$

where R_i^p is the average annual health risk caused by carcinogen *i* through dermal ingestion (a⁻¹); CDI_{*i*} is the chronic daily intake through dermal absorption in mg/kg/day; SF is carcinogenic intensity coefficient of carcinogen *i* through dermal ingestion (mg/(kg·d))⁻¹; and *L* is average life expectancy (a); for adults, 70 years of lifetime was assumed in this study.

Health hazards caused by noncarcinogenic substance through the skin contact risk R^f is

$$R_i^f = \frac{\left(\text{CDI}_i \times 10^{-6}/\text{RfD}_i\right)}{L},\tag{6}$$

$$CDI_{i} = \frac{I_{i} \times A_{sd} \times FE \times EF \times ED}{W \times AT \times f},$$
(7)

$$I_i = 2 \times 10^{-3} \times k \times C_i \times \sqrt{\frac{6 \times \tau \times TE}{\pi}},$$
(8)

where R_i^j is the average annual health risk caused by noncarcinogen *i* through dermal ingestion (a⁻¹); RfD is the daily ingestion reference dose of noncarcinogen *i* for dermal (mg/(kg·d)⁻¹); I_i is the adsorption amount of pollutant *i* per unit are of each bath (mg·m²·time); A_{sd} is the body surface area (cm²); K_p is the skin adherence factor in cm/h; *FE* is the bathing frequency; *EF* is the exposure frequency (day/year); *ED* is the exposure duration (years); *W* is the average body weight (kg); *AT* is the averaging time for noncarcinogens exposed (d); *f* is the intestinal adsorption ratio; *k* is the skin adsorption ratio (cm/h); C_i is the concentration of heavy metal *i* in runoff sample (mg/L); τ is the delay time (h); and *TE* is the duration of one shower (h).

3. Results and Discussion

3.1. Distribution of Trace Elements in Athletic Field Runoff

3.1.1. Distribution of Trace Elements in Different Athletic Field Runoffs. Total concentrations of seven metals and metalloid in runoff from different athletic fields are presented in Table 3. Among the five athletic fields, the mean concentration of seven metals and metalloids including Zn, Pb, Cu, Mn, Cd, and As is the highest in artificial turf. According to the surface materials of athletic fields (Table 1), artificial turf consisted of artificial grass and rubber particles that could release more particles than other athletic fields with flat surface. Studies have shown that the smaller the size of the particles, the more pollutants released [17]. The scouring effect of runoff enhances the frictional effect of particulate matter, making it easier for the pollution components inside the particles to dissolve out [18]. Besides the structure effect of the athletic fields, the chemical composition could also be another factor that affects the distribution of metals in runoff. For Cr, the total concentration follows in the order of badminton court > basketball court > artificial turf > tennis court > plastic runway. Cr is the main material in the tanning agent. Its main function is to maintain the stability of the rubber. In most cases, the total runoff concentration of trace elements (Zn, Cu, M,n and Cd) in plastic runway ranks the second among the five athletic fields. The reason might also be the rough surface of plastic runway that may release more particles that contains more pollutants. The surface of the tennis court, basketball court, and badminton court is smoother. The trace elements with small particles are more difficult to release from the smooth surface into the runoff [6].

Generally, the concentration of Zn in the five athletic fields runoff was the highest among the seven trace elements. The maximum concentration of Zn of the artificial turf,

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| | 0.1 | , , , , , , , , , , , , , , , , , , , | | | |
|----------------------|------------------|---------------------------------------|---------|---------|---------------|
| Metals and metalloid | Sites | Range | Mean | SD | CNEQS-SW-V |
| | Artificial turf | 361.83-4887.83 | 1696.91 | 1237.53 | |
| | Plastic runway | 248.94-4229.94 | 1285.44 | 1123.25 | |
| Zn | Basketball court | 268.39-2662.94 | 980.13 | 678.05 | 2000 |
| | Badminton court | 322.39-2080.06 | 927.81 | 598.52 | |
| | Tennis court | 306.8-3469.06 | 1197.88 | 883.56 | |
| | Artificial turf | 109.17-2181.72 | 485.92 | 456.45 | |
| | Plastic runway | 25.22-1225.67 | 234.40 | 281.91 | |
| Pb | Basketball court | 63.00-1769.78 | 433.78 | 420.61 | 100 |
| | Badminton court | 55.61-3605.94 | 701.72 | 871.35 | |
| | Tennis court | 49.78-379.17 | 162.72 | 86.92 | |
| | Artificial turf | 29.61-368.39 | 102.99 | 76.32 | |
| | Plastic runway | 18.00-188.61 | 78.85 | 55.17 | |
| Cu | Basketball court | 16.33-174.06 | 70.21 | 55.38 | 1000 |
| | Badminton court | 20.83-284.17 | 76.14 | 64.66 | |
| | Tennis court | 19.22-262.44 | 99.92 | 78.37 | |
| | Artificial turf | 19.28-4236.94 | 495.23 | 884.46 | |
| | Plastic runway | 12.56-1247.11 | 229.95 | 336.71 | |
| Mn | Basketball court | 12.50-246.89 | 87.20 | 73.95 | Not available |
| | Badminton court | 9.78-443.39 | 130.28 | 130.85 | |
| | Tennis court | 18.11-855.33 | 154.87 | 221.06 | |
| | Artificial turf | 27.56-470.50 | 110.88 | 108.83 | |
| | Plastic runway | 15.78-158.06 | 64.78 | 49.34 | |
| Cr | Basketball court | 35.00-465.17 | 140.60 | 109.56 | 100 |
| | Badminton court | 18.83-810.67 | 150.13 | 186.31 | |
| | Tennis court | 27.77-170.78 | 71.53 | 42.60 | |
| | Artificial turf | 2.17-51.44 | 9.60 | 12.06 | |
| | Plastic runway | 0.89-8.56 | 4.50 | 2.42 | |
| Cd | Basketball court | 0.94-11.61 | 4.09 | 2.75 | 10 |
| | Badminton court | 1.44-20.28 | 4.29 | 4.03 | |
| | Tennis court | 1.44-8.56 | 4.12 | 1.99 | |
| | Artificial turf | 1.72-44.48 | 13.22 | 11.01 | |
| | Plastic runway | 0.17-21.94 | 6.84 | 6.73 | |
| As | Basketball court | 0.56-26.83 | 6.50 | 7.08 | 100 |
| | Badminton court | 0.56-26.83 | 7.29 | 11.95 | |
| | Tennis court | 0.78-22.83 | 7.34 | 7.16 | |

TABLE 3: Metalloid and metal (total) concentrations (μ g/L) in runoff from different athletic fields.

*CNEQS-SW V: China National Environmental Quality Standards for Surface Water (GB3838-2002).

plastic runway, tennis court, basketball court and badminton court is 4887.83 µg/L, 4229.94 µg/L, 3469.06 µg/L, 2662.94 µg/L, and 2080.06 µg/L, respectively. The maximum Zn concentrations in the five athletic field all exceed the Chinese Class V surface water quality standard (CNEQS-SW-V, GB 3838-2002, 2000 µg/L). Previous studies also reported high Zn concentration leaching from athletic field. Krüger et al. reported that the leaching concentration of Zn from artificial turf could reach $4100 \,\mu\text{g/L}$ by column leaching test [6]. The high concentration of Zn detected from athletic fields could be due to two reasons. First, Zn could be originated from zinc oxide as a common addictive in the tire [18]. Waste tire as common materials could be reused as rubber in the surface materials of athletic fields. Another reason could be due to the easier leachable character of Zn from the materials [19]. Wachtendorf et al. reported that the release of Zn from the aged raw materials of artificial turf and plastic runway is significant [19]. In the raining event, the Zn prefers to be released more from rubber materials; as a consequence, the high concentration of Zn was detected in the runoff.

Even though Zn possesses the highest concentration among all the metals and metalloids studied in runoff, the pollution extent is less than Pb. In Table 3, it is shown that the maximum concentration of Pb of the badminton court, artificial turf, basketball court, plastic runway, and tennis court is 3605.94 µg/L, 2181.72 µg/L, 1769.78 µg/L, 1225.67 µg/L, and 379.17 μ g/L, respectively. The maximum concentration is 3-37 times of the CNEQS-SW-V. Pb could originate from lead oxide as a colorant and important component of pigments commonly used in rubber. A large amount of raw materials containing lead oxide is used in the laying process of the athletic fields. In outdoor environments, athletic fields are exposed to oxidizing agents, light, high temperature, and rainfall that may reduce the mechanical resistance of athletic field materials and enhance the release of toxic agents [19]. Pb was reported to be the typical pollutant in the runoff of traffic area, and its concentration is 173.0 µg/L [20]. The Pb concentration in the athletic field runoff is much more than Pb in the traffic road runoff. The Pb pollution in athletic field runoffs should attract people's wide attention.

Similar to Pb, the maximum concentration of Cr in runoff of the five athletic fields exceeds the CNEQS-SW-V (100 g/L). The highest concentration is 2–5 times that of the CNEQS-SW-V. The main source of Cr should be a stabilizer for athletic fields. Human activities will accelerate the aging of the athletic field materials. The aging process could induce the degradation process in the polymers, which leads to the formation of new surfaces allowing water to access additional reservoirs of leachable compounds. As a consequence, it could cause pollutant emissions [19]. Especially, the excessively high Cr concentration in the badminton court runoff may be caused by human activities. Efforts should be paid to reduce the Cr pollution in athletic field runoffs especially in badminton court. The mean concentrations of Cd in the five athletic fields is lower than CNEQS-SW-V (10 g/L), while the maximum concentration exceeded this standard. Shajib et al. reported that the Cd in the runoff of traffic area is $2.10 \,\mu\text{g/L}$ [20]. They conclude automobile could be the source in the traffic area. The mean concentration of Cd in the athletic field runoff is 2-4 times higher than traffic area [20]. The source of Cd could be the plastic stabilizers, paints, and pigments added in the materials. The concentration of As, Cu, and Mn in the runoff of the athletic fields does not exceed that of the CNEQS-SW-V. Compared with the traffic area runoff (Shajib et al.), the Cu concentration in the athletic field runoff is lower. The main sources of Cu in traffic area runoff are surface particulate matter and automobile brake pads [20]. Therefore, atmospheric dry deposition and surface particulate matter may be the main sources of Cu in the athletic field runoff. The concentration of Mn in the athletic field runoff is similar to traffic area (Shajib et al.). The main sources of Mn and Cu in traffic area runoff could be similar [20].

3.1.2. Temporal Variation of Metal Concentrations under Different Rainfall Events. Figure 2 shows the seven trace elements concentration in different athletic fields under the three rainfall events. Different trace elements show different patterns on the temporal variation of concentrations under different rainfall events. For the 1st rainfall event on July 5th, the trace element concentration in the five athletic runoff seems to be random with the rainfall duration and intensity. For the 2nd rainfall event on July 22nd, most of the trace element concentration (especially for Zn, Pb, Cu, Cr, and As) in the five athletic field runoff decreased over rainfall time, and their concentration decreased with the increase of rainfall intensity. For the 3rd rainfall event on July 29th, the trace element concentration in the five athletic field runoffs was much lower than the above two rainfall events, and it is irrelevant with the rainfall duration and intensity. The main sources of trace elements in the runoff of athletic fields may be the composed materials of the athletic fields and surface dust particles. The rain event on July 5th is the 1st heavy rain event in the year of 2019. Experiencing long antecedent dry days (Table 2), the surface materials experienced complex weathering process, and the surface dust accumulation could be significant. The pollutants accumulated in the surface dust and field materials for a long period could be released in

the runoff. Therefore, due to the complex factors, the released amount in runoff is random with the rainfall duration and intensity. For the 2^{nd} rainfall event on July 22^{nd} , the release of pollutants from surface dust is not important as the surface materials since most of the dust has been rushed out by the 1^{st} rainfall. The source of pollutants is mainly derived from the surface materials of the athletic field. With the increase in the rainfall duration, the trace elements released from surface materials could be less over time, since the pollutants were diluted with continuous rushing. The higher the rainfall intensity, the pollutants were more diluted that attributes to the lower concentration. For the 3^{rd} rainfall event on July 29^{th} , the concentration of pollutants is much lower and the regulation of concentration with duration and intensity could be minimized.

It could be clearly seen that the metal and metalloid concentration in all fields is the highest under the 1st rainfall event, while the concentration is the lowest under 3rd rainfall event. The rainfall characters such as the date of rainfall, antecedent dry days, rainfall duration, and rainfall intensity could all affect the distribution of pollutants in the runoff. Several studies have reported that antecedent dry days show a significant influence on the pollutant's concentration in the urban road runoff. Shajib et al. reported the Pb, Mn, Cu, and Zn concentrations in residential road runoff linearly increased with the antecedent dry days [20]. However, in our study, the trace element concentration did not linearly increase with the increase of antecedent dry days. Our observation shows that the surface dust may not be the main source of runoff pollution from the athletic field. Different from the urban road normally constructed by concrete, athletic fields were composed of polymer, reinforcing agents (e.g., carbon black), aromatic extender oil, vulcanization additives, antioxidants, etc [21]. The surface materials are not resistant to the oxidizing agents, light, temperature, and rainfall [19]. The release of toxic agents from athletic field surface materials is much likely significant than that from the dust on the athletic field. Therefore, the antecedent dry days seem not to be the main influencing factor on the distribution of pollutants in the athletic field runoff. With the more rainfall rushing events occurs, the release amount should be diluted. As a consequence, the pollutant concentration in runoff was lower than that under the later rain event. The rainfall date should be the main influencing factor on the metals and metalloids in the athletic field. The rain event on July 5th is the 1st heavy rain event in the year of 2019. The surface materials have experienced a long period of weathering that facilitates the release of pollutants. Thus, a high concentration of trace elements was detected. On the 3rd rain event on July 29th, under several rainfall events of scouring, pollutants released from the surface materials are less than before, thus attributing to the low concentration of pollutants in the runoff.

3.1.3. First Flush Effect Identification. Figure 3 shows the effect of the first flushing of pollutants in the five athletic fields. The relationship between cumulative pollutant quality and cumulative runoff was plotted, and the variation curve







FIGURE 2: Concentration of heavy metals and metalloid under different rainfall events and durations in five athletic fields.





FIGURE 3: Cumulative mass and volume curves of metals and metalloids in the five athletic field runoffs.

of pollution load with rainfall was shown. If the pollutant scour rate in the basin is proportional to the flow rate, the curve is the same as the bisector line. If the rate of pollution load is higher than that of storm runoff, the curves will be above the bisector line, which indicates that the pollutant has first flush effect. The difference between the curves and the bisector can be used to indicate the initial of first flush [22, 23]. From Figure 3, generally, trace elements under the 1^{st} and 2^{nd} rainfall events show stronger first flush effect than the 3^{rd} rainfall event. From the above session, it is seen that the trace element concentration in runoff under the 1^{st} and 2^{nd} rainfall events is higher than that in the 3^{rd} rainfall event. The reason could be due to the easier migration behavior of the higher pollutant concentration in runoff. Rainfall intensity is also one of the factors that affect the first flush [24]. However, the analysis result did not show any significant relationship between the first flush and rainfall intensity. Li et al. show that, if the strong rainfall intensity appears earlier during a rainfall event, it would cause more distinctive first flush [23]. On the other hand, if the strong rainfall intensity appears later during a rainfall event, the first flush effect is weak. In our study, the maximum rainfall intensities of both of 1st and 2nd rainfall appeared later (Figure 2) that should show a weak first flush effect. However, the flush effect of trace elements under 1st and 2nd is significant. Therefore, we conclude that both of the pollutant concentration and rainfall intensity could affect the first flush effect, and the former plays a major role.

From Figure 3, the first flush effect of trace elements is different in different athletic fields. Among the five athletic fields, trace elements in artificial turf (1st and 2nd rainfall), plastic runway (2nd rainfall), and basketball court (1st and 2nd rainfall) show stronger first flush effect than other athletic fields. The first flush effect in the badminton court and tennis court was weak under the three rainfall events. Besides the rainfall characters, the roughness and impervious extent of the field materials could also affect the first flush effect. Lee and Bang [25] concluded that first flush occurs strongly as the proportion of impervious area is higher. The impervious area of the five athletic fields decreased in the order of artificial turf > plastic runway > basketball court > tennis court > badminton court. Therefore, parts of the first flush effects are stronger in the artificial turf, plastic runway, and basketball court, while the effects are weak in the tennis court and badminton court.

Different types of trace elements have different first flush effects in different fields and rainfall events. For example, in the artificial turf, the first flush effect is decreased in the order of As > Pb > Mn > Cd > Cr > Cu > Zn under the 1^{st} rainfall, while the first flush effect is decreased in the order of Cu > Pb > Cr > Cd > Cr > Zn > As > Mn under 2^{nd} rainfall. The first flush effect is not distinguished under 3^{rd} rainfall that might be because the effect under the 2^{nd} rainfall is decreased in the order of Mn > Pb > As > Cr > Zn > Cu > Cd. From the above analysis, we could see that the first flush effect of trace elements in the athletic field runoff could be the complexity process.

3.1.4. Correlations between Trace Elements. The results of Pearson's correlation analysis between the total trace element concentrations in the athletic field runoffs are presented in Table 4. The correlation between trace elements is different from the type of athletic fields. Among all types of athletic fields, the tennis court shows the strongest correlations between trace elements. For the tennis court, all of the correlations between trace elements are significant as shown in r > 0.60. Among them, there are 11 correlations showing "very strong" correlations (r > 0.80), which are As towards Cu, Zn, Cr, Mn; Cu towards Zn, Cr, Mn; Zn towards Cr, Mn; and Cr towards Mn; Pb towards Cd. The rest 11 couples show "strong" correlations. The strong correlation between pollutants indicates that the source of the pollutants is the same. This may be due to the

uniform distribution of materials on the surface of the tennis court. As discussed in the above session, because the surface of the tennis court is smoother than other fields, the metals and metalloids distributed are evenly in the surface materials. Therefore, the strong correlation shows that pollutants may be mainly originated from the surface materials of the tennis court. On the other hand, among all types of athletic fields, the artificial turf shows the least strong correlations between metals and metalloids. For the artificial turf, there are 14 correlations between trace elements which is significant as shown in r > 0.60. Among them, there are only 4 correlations showing "very strong" correlations (r > 0.80), which are As towards Mn; Cr towards Zn, Pb, and Mn. The rest 7 couples show "weak" or "moderate" correlations. The strong correlation between the 14 couples of trace elements indicates that the source of the pollutants in artificial turf is similar and may mainly come from the surface particles released from the rough surface. While the other source of the trace elements may be atmospheric fry deposition. Among all the metals and metalloids, the correlation between As and Zn, As and Pb, As and Mn, Cu and Pb, Cu and Mn, Pb and Mn is always "strong" in the five types of athletic fields. Besides from the same source, the strong correlation could be due to the consistent behavior of those pollutants.

3.2. Pollution and Health Risk Assessment

3.2.1. Integrated Pollution Index (IPI) and Pollution Index (P_i) . The integrated pollution index values of the total trace elements in the five athletic fiee ld runoffs are summarized in Figure 4. The IPI value ranges are 0.05-3.36, 0.02-4.46, 0.02-4.02, 0.02-4.11, and 0.03-2.6 for artificial turf, basketball court, badminton court, plastic runway, and tennis court, respectively. Most of the IPI values have exceeded the line of "subclean" which indicated heavy metals and metalloids in the five athletic fields causing runoff pollution to different extents. The mean IPI value is in the order of artificial turf > basketball court > plastic runway > badminton court > tennis court (the corresponding IPI values are 1.29, 1.19, 1.01, 0.95, and 0.86). From the perspective of mean IPI value, trace elements in runoff from three athletic fields including the artificial turf, basketball court, and badminton court are classified as "heavily polluted." The pollution degree of plastic runway and tennis court was classified as "moderately polluted." Attention should be drawn especially to the heavily polluted athletic field runoffs to avoid the subsequent environmental pollution that might be caused. Since some IPI values also appear in the "heavily polluted" range, the moderate polluted athletic fields also should not be ignored.

Different types of elements have different pollution indexes. According to the selected value of S_i and P_i classification, Figure 5 shows the single factor pollution index for individual trace elements (Zn, Pb, Cu, Cr, Cd, and As) in the five athletic field runoffs. Among the six trace elements, Pb shows the highest pollution level as P_i is distributed in the

| Artificial | | As | Cu | Zn | Cr | Pb | Mn |
|---------------|----|--------------|-------------|---------|--------------|--------------|-------------|
| Turf | Cu | 0.111 | 1 | | | | |
| | Zn | 0.653** | 0.301 | 1 | | | |
| | Cr | 0.785** | 0.504^{*} | 0.919** | 1 | | |
| <i>n</i> = 24 | Pb | 0.686** | 0.756** | 0.695** | 0.895** | 1 | |
| | Mn | 0.925** | 0.080 | 0.799** | 0.854** | 0.686** | 1 |
| | Cd | 0.248 | 0.776** | 0.473* | 0.640** | 0.795** | 0.299 |
| Plastic | Cu | 0.888** | 1 | | | | |
| Runway | Zn | 0.730** | 0.831** | 1 | | | |
| | Cr | 0.899** | 0.870** | 0.875** | 1 | | |
| <i>m</i> = 22 | Pb | 0.711** | 0.636** | 0.406 | 0.686** | 1 | |
| n = 22 | Mn | 0.774^{**} | 0.708** | 0.523* | 0.790** | 0.960** | 1 |
| | Cd | 0.612** | 0.610** | 0.485* | 0.512* | 0.545** | 0.477^{*} |
| Badminton | Cu | 0.519* | 1 | | | | |
| Court | Zn | 0.687** | 0.586** | 1 | | | |
| | Cr | 0.521* | 0.584** | 0.701** | 1 | | |
| <i>n</i> – 22 | Pb | 0.660** | 0.704** | 0.705** | 0.952** | 1 | |
| n - 22 | Mn | 0.657** | 0.798** | 0.844** | 0.877** | 0.933** | 1 |
| | Cd | 0.878** | 0.362 | 0.472* | 0.230 | 0.402 | 0.380 |
| Basketball | Cu | 0.638** | 1 | | | | |
| Court | Zn | 0.374 | 0.837** | 1 | | | |
| | Cr | 0.614^{**} | 0.886** | 0.717** | 1 | | |
| m = 24 | Pb | 0.589** | 0.640** | 0.502* | 0.869** | 1 | |
| n = 24 | Mn | 0.687** | 0.959** | 0.792** | 0.954** | 0.784^{**} | 1 |
| | Cd | 0.383 | 0.472^{*} | 0.535** | 0.646** | 0.774^{**} | 0.620** |
| Tennis | Cu | 0.893** | 1 | | | | |
| Court | Zn | 0.875** | 0.820** | 1 | | | |
| | Cr | 0.934** | 0.885** | 0.904** | 1 | | |
| <i>n</i> = 23 | Pb | 0.746** | 0.773** | 0.764** | 0.748^{**} | 1 | |
| | Mn | 0.876** | 0.818** | 0.840** | 0.881** | 0.775** | 1 |

TABLE 4: Pearson's correlation coefficients for correlation between seven metals and metalloid concentrations in athletic field runoffs.

** Correlation is significant at the 0.01 level (2-tailed). * Correlation is significant at the 0.05 level (2-tailed). The range of absolute value of *r* is 0.00–0.19 (very weak); 0.20–0.39 (weak); 0.40–0.59 (moderate); 0.60–0.79 (strong); 0.80–1.0 (very strong) [20].



FIGURE 4: Box-plots for integrated pollution index (IPI) of metals and metalloids in five athletic field runoffs. For reference, dashed lines indicate the different pollution classes by Yan et al. (2015) [13].

"heavily polluted" to "seriously polluted" range. Therefore, Pb pollution should be concerned in athletic field runoff regardless of the type of the athletic fields. Cr ranks the second highest polluted metals as P_i is distributed in the "slightly polluted" to "heavily polluted" range. The maximum and the mean P_i values of Cr (9.30 and 2.81, respectively; corresponding concentrations are 465 and 140.5 μ g/L) are observed in the basketball court which indicated that Cr pollution is the most significant one among the five types of athletic fields.

In general, Zn is less polluted than Cr even though it processes the highest concentration among all the metals and metalloids (Table 3). Zn is "medium polluted" in the tennis court, plastic runway, and artificial turf (0.31-3.47, 0.25-4.23, and 0.36-4.88, respectively; corresponding concentrations are 306.8-3469.06, 248.94-4229.94, and $361.83-4887.83 \,\mu g/L$), while "slightly polluted" in the badminton court and basketball court (0.32-2.08 and 0.27-2.66; corresponding concentrations of 322.39-2080.06 and $268.39-2662.94 \,\mu g/L$). The above three heavy metals (i.e., Pb, Cr, and Zn) show heavier pollution than other metals and metalloids (i.e., Cd, As, and Cu). Cu does not cause pollution as P_i is distributed in the "unpolluted" range. The pollution condition of As is also not serious as most of P_i is distributed in the "unpolluted" range except for "slightly polluted" in the artificial turf (0.03-0.89; corresponding concentration of $1.72-44.48 \,\mu g/L$). For Cd, the pollution situation of the



FIGURE 5: Box-plot of the single factor pollution index (P_i) of seven metals and metalloids in runoff from athletic fields. For reference, the dashed lines indicate the different pollution class by Yan et al. (2015) [13].

five fields is relatively similar. The P_i is mainly distributed in the "slightly polluted," range and a small amount in "moderately polluted" Range. Since different heavy metals and metalloids could cause different pollution levels, efforts should be taken especially for the heavily polluted metals such as Pb, Cr, and Zn in the athletic field runoffs.

3.2.2. Health Risk Assessment. Metals and metalloids in runoff from the athletic fields that cause health problems is one of the important sources [26]. The health risk assessment model by USEPA was also applied to analyze the noncarcinogenic risk of trace elements via dermal contact. The hazard quotient (HQ) has the same meaning as R^{f} in this study. For hazard index (HI), it was described as the sum of all the HQ of different intake pathways, i.e., ingestion, inhalation, and dermal. However, in this study, dermal contact is considered to be the only pathway considered for intake. Our practice is similar to that of Ravivndra et al.'s [27]. They conducted the health risk assessment of arsenic and selected heavy metals in Groundwater of Chandigarh, India, who also consider ingestion is the only pathway consider for intake. Figure 6 shows the health risk assessment results of heavy metals and metalloids in runoff of the five athletic fields. The International Collaborative Research Program (ICRP) suggests that maximum acceptable risk level is 5×10^{-5} . It could be seen that Cr has the highest health risk index among the seven trace elements, and the R^p of Cr of the five athletic field runoffs exceeded 5×10^{-5} . The mean health risk index is in the order of basketball court > badminton court > artificial turf > tennis court > plastic runway (the corresponding health risk values are 2.59×10^{-4} , 2.29×10^{-4} , 1.82×10^{-4} , 1.74×10^{-4} , and 1.57×10^{-4} , respectively). This indicates that Cr posed serious health concerns to the athletic field user via dermal contact. From the pollution risk assessment, Cr ranks the second highest polluted metals among the seven trace elements. Both high health and pollution risk should raise the concern of people about Cr in the athletic field runoff. Next to Cr, the R^p of As is also high, and parts of As in the five



FIGURE 6: Health risk assessment result of heavy metals and metalloid in five athletic field runoff.

athletic fields runoff exceeded 5×10^{-5} . The mean health risk index of As is in the order of artificial turf > tennis court > badminton court > plastic runway > basketball court (the corresponding health risk values are 1.23×10^{-4} , 7.11×10^{-5} , 7.03×10^{-5} , 6.62×10^{-5} , and 6.29×10^{-5} , respectively). Even though the pollution risk of As is "slight," the health risk of it could not be ignored. US EPA indicated that the target health risk index exceeding 1×10^{-4} could increase the probability of cancer. Cr in the five athletic fields and As in the artificial turf, plastic runway, basketball court, and tennis court may increase the probability of cancer through dermal contact. Cd is a slightly polluted metal, and the health risk index is between 1.0×10^{-6} and 4.6×10^{-5} in the five athletic fields, but there are still a few samples whose R^p is higher than 5×10^{-5} . This shows that Cd through dermal contact may cause slight health concerns. R^f values of Zn, Pb, Cu, and Mn are around 1.0×10^{-8} which are much lower than 5×10^{-5} . The pollution of Pb and Zn is much serious; however, these metals had little health threat via dermal contact.

4. Conclusion

In this study, seven trace elements including heavy metals and metalloid were detected in five typical athletic field runoffs under three rainfall events. The occurrence and the risk of pollution and health were analyzed. The main conclusion could be drawn as follows:

- (i) Except for Cr, the total concentration of the rest trace elements in artificial turf runoff is the highest among five athletic fields, followed by plastic runway. Most of the trace element concentration is higher than that of traffic road runoff and exceeds that of CNEQS-SW-V. The high concentration of trace elements in runoff is due to their release directly from surface materials and the particles on athletic field.
- (ii) Patterns between trace elements concentration and temporal variation as well as the first flush under different rainfall events are both affected by the comprehensive effect including rainfall intensity and surface materials. Concentration and first flush effect of trace elements in athletic field runoff is followed in the order of $1^{st} > 2^{nd} > 3^{rd}$ rainfall events. Besides the effect of antecedent dry days, the rainfall date should be the main influencing factor on the trace element distributions.
- (iii) Tennis court shows the strongest correlations between trace elements, while the artificial turf shows the least. This may be due to the uniform distribution of materials on the surface of the tennis court and indicates the similar source of trace elements. The correlation between As and Zn, As and Pb, As and Mn, Cu and Pb, Cu and Mn, Pb and Mn is always "strong" in the five types of athletic fields, indicating the consistent behavior of those pollutants.
- (iv) Pollution risk assessment shows that the pollution extent of the five types of athletic field is at least "moderate," and follows the order of artificial turf > basketball court > plastic runway > badminton court > tennis court. Three trace elements (i.e., Pb, Cr, and Zn) show heavier pollution than of other metals and metalloid (i.e., Cd, As, and Cu), and Pb is the "heaviest polluted" metal. Health risk assessment show that Cr, As, and Cd could cause more risk than Pb, Zn, Cu, and Mn.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The data used to support this study are available within the article.

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