Study on pollution characteristics of urban pavement runoff

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ABSTRACT

Urban pavement runoff has become an important pollution source endangering the quality of urban water. This paper analyzed the characteristics of particle size distribution of road-deposited sediment (RDS). The variation of pollutants with RDS content is presented. Based on continuous sampling of runoff, the variation between pollutant concentration and rainfall characteristics is revealed. The results show that each particle group shares similar content except for the group smaller than 0.075 mm. However, the smaller particles have a stronger ability to adsorb heavy metals (Zn, Pb, Cu), and a weaker ability to adsorb chemical oxygen demand (COD). The concentrations of different pollutants have different relationships with rainfall and runoff time. The concentration of suspended solids (SS) decreases steadily with runoff time, while the concentration of heavy metals increases first and then decreases. The first 30 minutes of runoff is the best time to treat heavy metals and SS. The five-day biochemical oxygen demand (BOD\textsubscript{5}) and total petroleum hydrocarbons (TPHs) concentration are mainly affected by rainfall intensity. The result presented in this paper may provide a useful reference for the treatment of pavement runoff pollution.

Key words: characteristics of rainfall, first flush, pavement runoff, pollution, road-deposited sediment (RDS)

HIGHLIGHTS

- The variation characteristics of heavy metals, organic and petroleum hydrocarbons pollutants in road-deposited sediments with particle size are presented.
- The first flush degree of various pollutants in pavement runoff is pointed out.
- The variation of pollutant concentration with rainfall characteristics were revealed to determine the optimal time for different pollutants treatment.

INTRODUCTION

The quality of urban drinking water is closely related to the health of every urban resident. However, with the increasing number of vehicles, factories and population, the total amount of pollutants including heavy metals, organic matter and petroleum hydrocarbons pollutants is gradually increasing (Markiewicz \textit{et al.} 2017; Azimi \textit{et al.} 2018; Hou \textit{et al.} 2019). These pollutants generally adhere to the surface of roads and fine-grained sediments (such as dust). During rainfall, they enter the receiving water body or surrounding soil with pavement runoff, resulting in ecological degradation and eutrophication of urban water bodies (Wang \textit{et al.} 2020a). Ma \textit{et al.} (2018) found that 69.24\% of particulate pollutants in urban water environments come from pavement runoff. Urban pavement runoff has become an important source of urban water pollution, which has widely concerned scholars.

The research on the characteristics of pavement runoff pollution can be summarized into two aspects: RDS pollution and first flush on runoff. Some scholars have studied the accumulation law of heavy metals in RDS. \textit{Wang \textit{et al.} (2020b, 2020c)} found that 76.3\%–87\% of heavy metals are attached to particles with particle size <0.15 mm. Zinc (Zn) had a higher concentration in both coarse and fine particles, while Copper (Cu) and Lead (Pb) have a higher concentration in fine particles. \textit{Lanzetorfer} (2018) believed that the variation of heavy metal concentration with particle size has a good correlation with the power function. \textit{Hong \textit{et al.} (2020)} further pointed out that there are three main sources of heavy metals, which...
are the exhaust emissions from vehicles, the friction between the tire and the ground, and the brake lining. It can be seen that the reported studies mainly focus on the pollution characteristics of heavy metals. However, in addition to heavy metals, the RDS contains other pollutants that degrade water quality, typically including organic and petroleum hydrocarbons (Hong et al. 2019; Liu et al. 2019). At present, there are few studies on organic pollutants such as chemical oxygen demand (COD) and five-day biochemical oxygen demand (BOD5).

In addition, the pollutants attached to the surface of RDS and road can be transferred to the runoff by rainwater (Zafra et al. 2017). The pollutant concentration in the early stage of runoff is usually higher than that in the later stage, which is called ‘First Flush’ (FF) (Saget et al. 1996; Deletic 1998; Larsen et al. 1998). However, FF doesn’t occur immediately with the beginning of rain. The determination of the start time of FF is beneficial to improve the treatment efficiency of pavement runoff pollutants. Geiger (1984, 1987) proposed a classic discrimination method for FF by analyzing the curve of dimensionless cumulative relative mass M of pollutants against dimensionless cumulative relative runoff volume, V. Geiger (1984, 1987) believed that when the M(V) curve has an initial slope greater than 45° or the maximum gap between M(V) curve and diagonal is greater than 0.2, FF occurs. Bertrand-Krajewski et al. (1998) proposed that FF occurs when the initial 30% of the runoff carries at least 80% of the pollution load, which is called the 30/80 standard. Similarly, there is the 20/40 standard (Deletic 1998), 25/50 standard (Vorreiter & Hickey 1994), and so on. And some scholars determined the FF by the Mass First Flush Ratio (MFFR) (Barco et al. 2008) or ultraviolet spectroscopy (Aryal et al. 2017). However, different pollutants in the RDS have different attachment states, and thus the FF of the pollutants is different as well. The FF of different pollutants needs to be determined respectively, which will benefit to improve the efficiency and pertinence of road runoff pollutant treatment.

It should be noted that the pollution characteristics of pavement runoff are a complex issue. On the one hand, the types and sources of road surface pollutants are diverse and have obvious regional characteristics. The water quality of pavement runoff may vary greatly in different regions. Even in the same region, the pollution of pavement runoff produced by different rainfall events also varies greatly (Xue et al. 2020). On the other hand, the pollutants in pavement runoff are closely related to rainfall volume, rainfall intensity, traffic condition and antecedent dry period and other factors, showing obvious randomness (Dos Santos et al. 2019; Du et al. 2019). Therefore, taking the representative cities as research objects, the pollution characteristics of pavement runoff are studied. In the research process, factors such as rainfall characteristics and road traffic are considered, which is beneficial to improve the reference significance and value of the research results.

Changsha is a node city of the middle reaches of the Yangtze River and the Yangtze River Economic Belt in China. It is also a national hub of comprehensive transportation and logistics. Changsha has a population of about 8 million, with a GDP of 1214.252 billion yuan in 2020. The pavement runoff pollution in Changsha can represent not only the main cities in China but also some cities of the world. However, there are few studies on pavement runoff pollution in Changsha. This paper takes Changsha City as the research object, and analyzes the particle size distribution of RDS, the variation of pollutant content in RDS, the FF characteristics of runoff, and the variation of pollutant concentration in runoff with rainfall intensity and rainfall volume. This paper may provide beneficial references for the treatment of runoff pollutants.

MATERIALS AND METHODS

The sampling site is located in Changsha, Hunan Province, South-central China, which has a typical subtropical monsoon climate. In 2020, the annual average daily traffic equivalent of Changsha provincial highway is about 10,879 (vehicles/day). Two typical road sections in Changsha are selected as random sampling sites. The geographic locations of sampling sites are shown in Figure 1. The first sampling site, P1, is the second section of Wanjiali South Road, Tianxin District, Changsha City, which belongs to a suburb with a small traffic volume surrounded by schools and residential areas. The second sampling site, P2, is a section on the Juyuan Overpass in Changsha City, which belongs to the urban area and has a large traffic volume. The surrounding area is mostly hotels, shopping malls and high-rise office buildings. In order to analyze the characteristics of urban runoff pollution comprehensively, this study is divided into two parts: (i) collection of solid sediments on the road surface in dry weather, and analysis of the types and relative contents of pollutants; (ii) continuous collection of runoff samples during rainfall, and analysis of the characteristics of pollutants in the runoff.

Collection method of RDS

Considering the influence of wind and road surface water on the accumulation of RDS, the time node of collecting RDS was selected when the weather remained dry for more than three days and there was no wind. Deletic & Orr (2005) pointed out that 60%–90% of solid particles are distributed within a range of 30 cm–50 cm from the curbstone. Therefore, in order to
ensure the representativeness and integrity of the sample, the sampling width was 50 cm from the curbstone, and the sampling length was 10 m in this research. As shown in Figure 2, all solid deposits were collected by brush and dustpan, and then the samples were stored in self-sealing bags (Dos Santos et al. 2019; Wang et al. 2020b).

Collection method of pavement runoff

The pavement runoff collection area of sampling sites P1 and P2 is shown in Figure 3. Polyethylene bottles were used to collect pavement runoff at the drainage outlet and drainage pipe of the sampling sites. The sampling time points of runoff were 5 min, 10 min, 20 min, 30, 45 and 60 min after runoff formation, and the sample volume was 600 ml each time. At the time of collecting runoff samples, the amount of rainfall was measured by rain gauge. According to the rainfall volume, the rainfall intensity in different periods was calculated. In this study, three rainfall process samples were taken from P1 and P2 from February 2020 to March 2020, and a total of 30 pavement runoff samples were obtained. The specific sampling time and rainfall conditions are shown in Table 1.

Detection methods of pollutants

For the sediment samples obtained from the pavement, the bulk impurities such as cigarette butts, leaves, and dead branches were removed first. They were then dried in the shade and weighed. The collected sediment samples were sieved with pore sizes of 0.075 mm, 0.25 mm, 0.5, 1 and 2 mm respectively. The mass of each particle group was weighed. The concentrations of heavy metals (Zn, Pb, Cu), COD and BOD5 in each solution were measured by the methods shown in Table 2. The sediment samples of different particle sizes were digested with aqua regia, and the concentrations of heavy metals were determined by atomic absorption spectrophotometry (Ministry of Ecology and Environment of the People’s Republic of China 1987). The detailed laboratory testing method refers to Zafra et al. (2017) and Dos Santos et al. (2019). The concentrations of COD and BOD5 were analyzed according to the China standard methods. The detailed laboratory testing method
refers to HJ 828-2017 (Ministry of Ecology and Environment of the People’s Republic of China 2017) and HJ 505-2009 (Ministry of Ecology and Environment of the People’s Republic of China 2009). The COD refers to the amount of oxygen required for the oxidation of organic matter in water by chemical oxidants. The BOD5 refers to the amount of oxygen required after the organic matter in water was decomposed by microorganisms for five days.

As shown in Table 2, the pollutants of suspended solids (SS) and petroleum hydrocarbons were also analyzed according to GB 11901-89 (Ministry of Ecology and Environment of the People’s Republic of China 1989) and HJ 637-2018 (Ministry of Ecology and Environment of the People’s Republic of China 2018).
Ecology and Environment of the People’s Republic of China (2018) in this paper. The above detection process complies with Chinese national standards. Meanwhile, a control test was also conducted to ensure the accuracy and reliability of the analysis results.

**Analysis method of first flush effect**

Generally speaking, the concentration of pollutants carried in the pavement runoff is relatively higher at the initial stage of rainfall. However, it is difficult to flush the pollutants when the rainfall intensity is small. Therefore, the determination of the starting point of flush and first flush effect (FFE) is a key factor in the design of pollution treatment facilities. The M(V) curves discrimination is a widely adopted method to judge the FFE (Morgan et al. 2017; Perera et al. 2019).

**RESULTS AND ANALYSIS**

**Particle size distribution of RDS**

The particle size of RDS affects the migration of particles and the ability to adsorb pollutants. In order to analyze the particle size distribution of RDS, a series of sieving tests were carried out. The mass percentage of each particle size of RDS at two sampling sites is shown in Figure 4.

It can be seen from Figure 4 that the particle size distributions of P1 and P2 are similar. The RDS were dominant with particles of medium size (0.075 < d < 2 mm), followed by coarse particles (d > 2 mm), while it contained relatively low levels of fine particles (d < 0.075 mm). This is similar to the particle size distribution of RDS in Yixing, China (Wang et al. 2020b). But the particle size of Changsha is larger than that of Yixing on the whole. The difference between P1 and P2 mainly focuses on the size groups of 0.25–0.5 mm and 1.0–2.0 mm. And the difference in mass percentage is only 4.51% and 4.09% respectively. The proportion of particles with a size of 0.25 mm–0.5 mm is the largest, with a value range of 20%–30%. The proportion of particles with a size smaller than 0.075 mm is the smallest, accounting for 2%–3% of the total particles. The particles with a size range of 0.075 mm < d < 0.25 mm, 0.5 mm < d < 1 mm, and d ≥ 2 mm account for about 20%. Particles with a size smaller than 0.075 mm have the least content of RDS, which may be because small particles are more likely to be floated into the air by vehicles.

**Analysis of pollutant types and contents in RDS**

The types and concentration percentages of pollutants at P1 and P2 are shown in Figure 5(a) and 5(b) respectively.
It can be seen from Figure 5 that the main pollutants in the RDS of Changsha are heavy metals (including Zn, Pb, Cu), COD and BOD$_5$. Among the five kinds of pollutants, the Pb content is highest, at up to nearly 60% of the total pollutant content. The other pollutants are Cu, Zn, COD and BOD$_5$ in order, of which BOD$_5$ content is the least with an average of 0.17% of the total pollutant content.

Pb mainly comes from vehicle exhaust. Zn mainly comes from tire wear. Cu mainly comes from brake lining wear. COD and BOD$_5$ mainly come from vehicle exhaust, lubricants and organic matter in tires (Markiewicz et al. 2017; Hong et al. 2020).

The percentage of Pb in P2 is higher than that in P1, and the percentage of Zn and Cu in P1 is higher than that in P2. The reason is that the P2 is located in the urban area with large traffic volume and large vehicle exhaust emissions. P1 is located at the school road section. Vehicles usually need to brake when passing this section. There is more Zn and Cu content at the sampling points P1 because of the tire wear. The pollutant contents at sampling points P1 and P2 both follow the rules of Pb > Cu > Zn > COD > BOD$_5$.

**Variation of pollutant concentration with the particle size of RDS**

Pollutants often adhere to the surface of sediments, and there are differences in the adhesion of pollutants to sediments with different particle sizes. The variation of pollutant concentration with the different particle sizes of RDS was analyzed. The curve of average value of each pollutant with particle size is shown in Figure 6.

Figure 6 shows that the content of Pb in RDS varies the most with particle size. The content of Pb in particles with a size smaller than 0.075 mm reaches 230.00 mg/kg, which is 8.52 times Changsha’s local soil background value (27 mg/kg) (China National Environmental Monitoring Centre 1990). The content of Pb in particles with a size larger than 2 mm is only 17.00 mg/kg. The content of Cu ranged from 11.40 mg/kg to 94.00 mg/kg. When the particle size was less than 2 mm, the content of Cu in the particles was higher than the background value (27 mg/kg) of Changsha soil (China National Environmental Monitoring Centre 1990). The content of Zn in each particle size is lower than the background value (95 mg/kg) of Changsha soil (China National Environmental Monitoring Centre 1990). The content of heavy metals (Pb, Cu and Zn) in RDS has the same trend with the change of particle size. It also can be seen from Figure 6 that the heavy metals content...
decreases with the increase of particle size, which is consistent with previous study (Wang et al. 2020c). Analyzing the reason, the smaller particles have a larger specific surface area. Therefore, the smaller particles have the greater adsorption capacity for heavy metals.

COD is commonly used to measure the content of organics. With the increase of COD, the pollution of organic matter is more serious. When the particle size is larger than 0.075 mm, COD content tends to rise with the increase of particle size. The overall content of BOD₅ is very low at P1 and P2, which means there is little organic matter that can be oxidized by microorganisms. However, the content of BOD₅ also shows a trend of decreasing with the increase of particle size.

Pollutants are mainly attached to the surface of particles, which means the surface area of particles affects the pollution potential of particles to a certain extent. The pollution potential of heavy metals increases with the decrease of particle size. The pollution potential of COD increases with the increase of particle size. It can be seen from the above results that the control of small size particles may effectively reduce heavy metal pollution, and the control of large size particles may effectively reduce organic pollution.

The discharge law of pavement runoff

Analysis of first flush effect (FFE) of pavement runoff

In the M(V) curves method, the cumulative relative runoff volume $f_{\text{flow}(i)}$ and the cumulative relative mass of pollutants $f_{\text{mass}(i)}$ in the $i$th sampling are respectively expressed by Equations (1) and (2):

$$f_{\text{flow}(i)} = \frac{\sum_{j=1}^{i} v_j}{\sum_{j=1}^{n} v_j}$$

$$f_{\text{mass}(i)} = \frac{\sum_{j=1}^{i} c_j v_j}{\sum_{j=1}^{n} c_j v_j}$$

where, $n$ is the total sampling times; $j$ and $i$ are the sampling times; $v_j$ is the runoff volume (L) at the $j$th sampling; $c_j$ is the runoff pollutant concentration (mg/L) at the $j$th sampling.

Because the M(V) curves are simple and the physical significance is clear, this paper uses the M(V) curves to analyze the FFE of three rainfall periods in P1 and P2. As shown in Figure 7, if the M(V) curves of pollutants are completely above the diagonal, the FFE occurs. Consider the fact that no effective data was collected due to the heavy traffic conditions at the T3 time of P2 sampling site. The data got from the T3 time of P2 are not presented here.

It can be seen from Figure 7 that the cumulative relative mass curves of each pollutant in pavement runoff are all above the diagonal. This indicates that there is FFE in pavement runoff. Among them, the M(V) curves of SS are on the outermost side, indicating that SS is the easiest to be flushed. The reason is that SS is suspended solid particles. And solid particles are more easily flushed away by runoff under the flushing effect of rainwater. Therefore, SS tends to show a stronger FFE. The M(V) curves of Pb, Cu and Zn are not significantly different in the same rainfall event. This indicates that the flush effect degree of the three heavy metals is similar, which may be due to the similar occurrence state of heavy metals in runoff. For organic pollutants, except for rainfall events of P1 at T2 time, the M(V) curves of COD in other rainfall events are all outside BOD₅. This indicates that COD is more likely to be flushed than BOD₅. By analyzing the reasons, it can be seen from Figure 6 that the COD content in RDS is much higher than BOD₅. The COD and BOD₅ in the runoff mainly come from RDS. The
COD content in the sediment is higher, so it is easier to be flushed by the runoff. Except for rainfall events of P1 at T2 time, the M(V) curves of TPHs in other rainfall events are closer to the diagonal than other pollutants. This indicates that the FFE of TPHs is weak. It may be because TPHs mainly come from asphalt pavements. The flush effect of rainwater is difficult to flush petroleum hydrocarbons out of asphalt pavements.

According to Figure 7, the pollutant content carried by initial 30% runoff is calculated by linear interpolation method, as shown in Table 3.

As shown in Table 3, the order of the first flush degree of pollutants in pavement runoff is SS > COD > Zn > Pb > Cu > BOD₅ > TPHs. For the whole rainfall event, 30% of the runoff at the initial stage contains 78%–90% SS, 40%–63% Pb, 41%–69% Zn, 47%–63% Cu, 38%–72% COD, 44%–57% BOD₅ and 56%–55% TPHs. These results are similar to the researches in Beijing (Zhang et al. 2021) and Xi’an (Chen et al. 2017). But the FFE in Changsha is more significant than that in other cities. The treatment of pollutants in the early runoff of pavement can greatly improve the purification efficiency.

Analysis on the variation of runoff pollutant concentration with rainfall characteristics

Pavement flush and pollutants in runoff are closely related to rainfall intensity and rainfall volume, based on continuous observation of rainfall intensity and rainfall volume in three periods of T1, T2 and T3. The shortest observation time is
65 min and the longest is 120 min. Among them, the rainfall intensity and rainfall volume obtained from T1 time of sampling site P1 have continuous changes, which are typical and representative. In this study, the data at T1 time of sampling site P1 is taken as an example to further analyze the dynamic change of pollutants in pavement runoff. The variation curve of pollutant concentration, rainfall volume and rainfall intensity in runoff are shown in Figure 8.

It can be seen from Figure 8 that the variation of different pollutant contents with runoff time is different. SS shows a monotonically decreasing trend with runoff time. However, heavy metals, TPHs, BOD$_5$ and COD all fluctuate in different periods. This is mainly because, on the one hand, flushing out different pollutants needs different rainfall volumes and rainfall intensities. On the other hand, SS is the easiest to be flushed, as analyzed above. The concentration of SS decreases by 70% within

Figure 8 | Variation curve of different pollutants with rainfall volume and rainfall intensity. (a) Concentration curve of SS. (b) Concentration curve of Pb, Zn and Cu. (c) Concentration curve of COD. (d) Concentration curve of TPHs and BOD$_5$. 

10 min of runoff. Then, the concentration of SS decreases with the continuous runoff. When the runoff lasts for 45 min, the concentration of SS reaches the lowest value, which is about 5% of the initial concentration.

The flush effect of heavy metals and COD is different from SS. The concentrations of heavy metals and COD increase firstly and then decrease with runoff time. The rainfall intensity is least at the 10th minute, while the concentrations of heavy metals and COD reach the peak point. Analyzing the reasons, heavy metals and COD mainly come from RDS; the initial runoff will flush them out from RDS. Although runoff can take away some pollutants, the flush effect of runoff on different pollutants is different. After 10 minutes, the concentrations of heavy metals and COD decrease with the increase of rainfall intensity and the continuation of runoff. When the runoff lasts for 45 min, the pollutant concentrations reach the lowest point and remain at a relatively low level. The variation curve of heavy metals with rainfall characteristics has the same trend as the study of heavy metal pollution in runoff in Nanjing, China (Xue et al. 2020).

The concentration curves of TPHs and BOD$_5$ are always at the lowest level and kept consistent with the rainfall intensity curve. In the first 20 minutes of runoff, the concentrations of TPHs and BOD$_5$ decrease. When the rainfall intensity is the maximum, the concentrations of TPHs and BOD$_5$ also reach the peak position at the same time. The reasons may be that petroleum hydrocarbon pollutants mainly come from asphalt pavement. With the increase of rainfall intensity, the flush effect of rainwater on asphalt pavement also increases, resulting in the increase of TPHs concentration in runoff. BOD$_5$ mainly comes from RDS. With the increase of rainfall intensity, the flush effect of rainwater increases, which makes BOD$_5$ be flushed out from the sediment. Therefore, the concentration of BOD$_5$ in runoff changes with rainfall intensity.

The variation of pollutant concentration with runoff is the result of the comprehensive effect of rainfall volume and rainfall intensity. With the increase of rainfall volume, the concentrations of each pollutant show a downward trend. However, due to the different flush effects of different pollutants, the concentrations show different development rules with runoff time. SS is the easiest to flush. Once runoff is formed, SS decreases monotonously with rainfall volume. The occurrence state of heavy metals in runoff is similar. The peak concentration of heavy metals and COD in runoff is not at the initial stage of flush, and there is a certain lag phenomenon. The peak value of heavy metals and COD in this test appears at the 10th minute of runoff formation. TPHs and BOD$_5$ are mainly affected by rainfall intensity. When the rainfall intensity reaches the peak, the treatment can effectively reduce the harm caused by these pollutants.

CONCLUSION

(i) The particle size distribution of RDS is relatively uniform. The proportion of particles of 0.25 mm–0.5 mm group is largest, about 26.5%. The proportion of particles smaller than 0.075 mm is smallest, accounting for 2%–3% of the total particles. However, the smaller particles have the stronger ability to carry heavy metals and BOD$_5$ and the weaker ability to carry COD.

(ii) The concentrations of pollutants in RDS are generally higher than the reference values of the local soil. The types of pollutants include heavy metals (Zn, Pb and Cu), COD and BOD$_5$. More than 50% of the pollutants are Pb, followed by Cu, Zn, COD and BOD$_5$.

(iii) There is an obvious first flush effect in pavement runoff, and the flush difficulty of each pollutant is different. The order from easy to difficult is SS > COD > Zn > Pb > Cu > BOD$_5$ > TPHs. The pollutants in the pavement mainly focus on the initial runoff, especially in the first 30 minutes of runoff. Therefore, the first 30 minutes of initial runoff is the key time for pollutant disposal.

(iv) Rainfall volume, rainfall intensity and first flush affect the variation of pollutant concentration with runoff. The variation of different pollutants with runoff is different. The peak concentration of SS appears at the beginning of runoff. The peak concentrations of COD and heavy metals appear about 10 minutes after the formation of runoff, while the peak concentrations of TPHs and BOD$_5$ are mainly affected by rainfall intensity. It is an effective method to reduce the pollutant content of runoff to classify and treat the runoff in different periods.

It is noteworthy that only seven pollutants were considered in this study. To better study the pollution characteristics of urban pavement runoff, other more representative pollutants should be taken into consideration in future research. Furthermore, traffic volume is an important factor affecting the pollution of pavement runoff. It is necessary to analyze the relationship between the concentration of pollutants in pavement runoff and traffic volume in future research.
ACKNOWLEDGEMENTS

The research was supported by the Special Funds for the Construction of Innovative Provinces in Hunan, China (grant number: 2019SK2171); and the Science and Technology Innovation Program of Hunan Province (grant number: 2020RC4048); and the Excellent Youth Project of Hunan Providence Department of Education (grant number: 19B030); and the National Science Foundation of Hunan Province, China (grant number: 2020JJ5576).

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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First received 24 May 2021; accepted in revised form 27 August 2021. Available online 13 September 2021.