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The paper was published in the proceedings of the 3rd International Symposium on Coupled Phenomena in Environmental Geotechnics and was edited by Takeshi Katsumi, Giancarlo Flores and Atsushi Takai. The conference was originally scheduled to be held in Kyoto University in October 2020, but due to the COVID-19 pandemic, it was held online from October 20th to October 21st 2021.

Organic pollutants removal by MSWI bottom ash as permeable subgrade material: experimental and numerical studies

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ABSTRACT

Considering the requirement of environment and ecosystem, permeable pavement gradually became a hot issue recently. However, the conventional permeable pavement only allowed the surface layer for drainage, while the permeable subgrade was less mentioned. In addition, the acceleration of urbanization process was accompanied by a large number of municipal solid waste (MSW) production. Among the treatment method towards MSW, incineration was widely used around the world, and resulted in a huge amount of municipal solid waste incineration (MSWI) bottom ash. Thus, the MSWI bottom ash was proposed to act as permeable subgrade material in this paper, since its low cost and eco-environmental nature. According to the test and calculated results, MSWI bottom ash presented excellent adsorption capacity towards methylene blue, around 5.44 mg/g, which was regarded as one typical organic pollutant. Through observing its microstructure and calculation based on isothermal adsorption equilibrium data, the adsorption process is mainly occurred in the form of physical adsorption. Considering above adsorption parameters, the contaminant migration and transport inside the subgrade towards surrounding environment was evaluated according to solute transport model, including influence factors like hydraulic behavior, adsorption, diffusion etc. The calculation results indicated that the adsorption capacity of MSWI bottom ash effectively retarded the diffusion of contaminant.

Keywords: MSWI bottom ash, permeable subgrade, organic pollutants adsorption, solute transport model, numerical simulation

1 INTRODUCTION

With the process of urbanization, the city's surface began to be gradually covered by water-blocking materials such as cement, making it difficult for water to penetrate, and the greenhouse effect was becoming obvious (Mohajerani et al., 2017; Palme et al., 2017; Deilami et al., 2018; Zeng et al., 2020). While the permeable pavements, especially fully permeable pavements, were regarded important to relive such a problem, which have become a hot issue recently. Different from traditional pavements, they have large porosity, which can form permeable channels and allow stormwater to pass through the surface layer (Kamali et al., 2017). They also have good texture depth, which can reduce noise and increase the friction between the wheel and the pavement (Wu et al., 2019; Zhang et al., 2019a). Because all structural layers are permeable, rainwater can seep into the ground (Saadeh et al., 2019). As rainwater enters the ground instead of being replenished as surface runoff, the flood peak water levels are lowered. Furthermore, the replenishment of groundwater can mitigate the decline of groundwater levels, purify water quality, curb seawater intrusion and improve the surround ecological environment. The widely used

permeable subgrade material is sand and gravel, which are more expensive than conventional subgrade material (Tang et al., 2015). Many local governments have banned quarrying from mountains to protect the environment, which makes them have to be transported from other areas and increases the cost of the engineers. All of these have restricted the development of permeable subgrades. Finding a low-cost permeable subgrade material has an important role in promoting the development of fully permeable pavement (Tang et al., 2014; Zhu et al., 2019).

In addition, with the economic development and population growth, the amount of municipal solid waste (MSW) has increased rapidly (Mian et al., 2017; Tang et al., 2016a; Gundupalli et al., 2017). MSW is usually treated by landfill, compost, incineration and other methods. Among them, municipal solid waste incineration (MSWI) is well recognized because it can effectively reduce the volume of MSW and generate electricity (Tang et al., 2017a). China has built 331 MSWI plants, processing 100 million tons of MSW annually. As a by-product, a large amount of MSWI bottom ash is generated. MSWI bottom ash can be used for concrete brick, cement concrete aggregate, asphalt

concrete additives, etc. (Taurino et al., 2017; Yan et al., 2020; Vaitkus et al., 2019). However, due to its continuous increasing production and limited landfill spaces in cities, MSWI bottom ash disposal and recycle became a new challenge in practice (Verbinnen et al., 2017).

According to previous research, MSWI bottom ash has good strength, and the leaching of heavy metals are limited, which means it can be used as subgrade material (Tang et al., 2018a; Huang et al., 2020). Furthermore, MSWI bottom ash is composed of particles, and drainage channels can be formed between the particles, which is similar to sandy soil. Due to its large specific surface area, MSWI bottom can adsorb some organic pollutants (Germain et al., 2017; Tang et al., 2019). Considering its huge quantity and cheap price, using MSWI bottom ash as permeable subgrade material can reduce the engineering cost. The application of MSWI bottom ash to fully permeable subgrade can reduce its amount, achieve permeability of subgrade, and adsorb some organic pollutants in rainwater.

To evaluate the eco-environmental benefits of MSWI bottom ash as a subgrade material, constant head permeability tests and adsorption tests were conducted. Based on the obtained hydraulic conductivity and adsorption capacity, a numerical simulation was performed to evaluate the transport of internal pollutants under rainwater conditions.

2 MATERIAL AND METHODS

2.1 Material

MSWI bottom ash was collected from Qizishan Municipal Solid Waste Incineration Plant in Suzhou, Jiangsu Province, China in May 2019. The incineration temperature was 850–1100°C, which means the organics in it have been fully burned. MSWI bottom ash is a gray-brown substance with a pungent odor. For avoiding the influence of moisture, it was oven-dried in a drying box at 105°C for 24 hours before the experiments. Its physical characteristics, including gradation, specific gravity, liquid limit and plastic limit, were carried out in accordance with Highway Engineering Soil Test Method (JTG E40-2007). Scanning electron microscope (SEM) was used to explore its microstructure and adsorption mechanism.

2.2 Methods

Constant head permeability tests

The constant head permeability tests were carried out under conditions of hydraulic gradients of 4 and 24. During the test, the samples were filled in the cylinder, and water flowed through the samples from top to bottom. When the head difference and the exudation flow were stable, measure the amount of water. Considering the low flow velocity, it could be considered laminar. According to Darcy's Law, the velocity is proportional to the hydraulic gradient.

$$v = k \times i \quad (1)$$

Here, v is the porous flow velocity; k is the hydraulic conductivity, which indicates the water permeability, i is the hydraulic gradient.

Adsorption tests

To obtain the adsorption capacity of MSWI bottom ash, the adsorption tests were performed. The composition of organic pollutants in rainwater is very complex, including normal paraffins, phthalates, polycyclic aromatic hydrocarbons and others. The chemical oxygen demand (COD) is generally used to evaluate the content of organic pollutants. In practice, COD varies widely with different regions, usually in 130 to 600 mg/L. Methylene blue ($C_{16}H_{18}ClN_3S$) is a commonly used chemical reagent as a typical pollutant because it widely exists in waste water. It was chosen to be the pollutant with the initial concentration of 20 mg/L in the tests.

Because the adsorption capacity, especially the physical adsorption capacity, is affected by the specific surface area, and the specific surface area is related to the size of the particles, the samples of 6 size ranges, which were < 0.075 mm, 0.075–0.1 mm, 0.1–0.25 mm, 0.25–1 mm, 1–2 mm and 2–5 mm, were tested in this adsorption tests. The tests were performed in the test tube for 25, 45, 70, 100, 200 and 300 min, and the supernatant was extracted after centrifuging for 15 minutes. The supernatant was placed in a spectrometer to test the concentration. According to the results of the experiment, isothermal adsorption equilibriums were fitted to explore the adsorption capacity and mechanism.

Numerical simulation

The numerical simulation was aimed to simulate the contaminant migration and transport inside the subgrade under various factors, including annual rainfall, hydraulic conductivity, and pollutant concentration, and consider the diffusion and adsorption. The finite element analysis software was COMSOL Multiphysics 5.5, which has been widely used in multi-physical coupled fields.

The Brinkman equation interface was used to calculate the fluid velocity and pressure field, and the porous media dilute matter transfer interface was used to calculate the concentration and transfer of substances in the pavement considering diffusion, convection and adsorption.

The parameters involved in the numerical simulation included the thickness, porosity, hydraulic conductivity, inlet velocity, maximum adsorption amount of the subgrade, and the initial concentration. According to Specifications for Design of Highway Asphalt Pavement (JTG D50-2017) and Specifications for Design of Highway Subgrades (JTG D30-2015), the highway model was constructed combined with the typical soil structure of Suzhou. Table 1 shows the specific parameters of each structural layer of the model.

Table 1. Parameters of road structures.

Structural layers	Parameters	Value
Pavement surface	Thickness	0.18 m
	Porosity	0.3
	Hydraulic conductivity	4×10^{-3} m/s
Pavement base	Thickness	0.36 m
	Porosity	0.25
	Hydraulic conductivity	4×10^{-3} m/s
Subgrade	Thickness	3 m
	Porosity	0.25
	Hydraulic conductivity	Obtained from experiments
	Q_m	Obtained from experiments
Land fills	k_L	Obtained from experiments
	Thickness	5 m
	Porosity	0.475
	Hydraulic conductivity	2×10^{-6} m/s
Silty clay	Thickness	30 m
	Porosity	0.47
	Hydraulic conductivity	3.5×10^{-6} m/s

Considering the different rainfall in different climates, the annual rainfall was divided into five groups of 100, 500, 1000, 1500 and 2000 mm. Among them, the rainfall in areas with annual rainfall less than 100 mm can be ignored, and the annual rainfall of 2000 mm corresponded to the rainfall in tropical rainforests.

COD is a chemical method to measure the amount of reducing substances that need to be oxidized in water samples. 5 groups used in this simulation were 50, 100, 200, 300 and 500 mg/L.

It was assumed that the rainwater only entered from the surface, the subgrade slopes and floors were impermeable, and the sides and bottom of the soil layer were rainwater and pollutant outlets. Because the adsorption was mainly occurred above the soil layer in this simulation, the rainwater seepage from the ground had little effect on the highway structure, thus it could be ignored.

It should be noted that the unit of the simulation results was mol/m³, not mg/kg. They can be converted by the molecular weight of methylene blue and the density of MSWI bottom ash.

3 RESULTS AND DISCUSSION

3.1 Physical properties

The physical properties of MSWI bottom ash mainly include gradation, specific gravity, liquid limit and plastic limit, specific surface area, as shown in Table 2. Among them, gradation have great influence on water permeability and adsorption performance. According to the calculation, the nonuniformity coefficient C_u was 12.5, the curvature coefficient C_c was 1.125. This means

that the soil particles were unevenly distributed and the porous were filled with small particles (Hani, 2012). This was good for strength, however, bad for permeability. If necessary, the gradation can be redesigned to improve permeability under the premise of strength.

Table 2. Physical properties of MSWI bottom ash.

Grain size range (mm)	Proportion	Specific gravity	Liquid / Plastic limit (%)	Specific surface area (m ² /g)
< 0.075	3.6	1.90	34.1/15.3	12.61
0.075-0.1	2.3	1.97		14.00
0.1-0.25	7.6	2.06		9.71
0.25-1	26.2	2.37		6.71
1-2	20.3	2.57		4.30
2-5	22.4	2.69		3.23
> 5	17.6	2.81		6.39

3.2 Constant head permeability tests

The constant head permeability tests were performed to obtain its hydraulic conductivity, and the results were shown in Fig. 1. The hydraulic conductivity decreased at first, and obtained a stable value after 30 s, which was consistent with the situation of other types of soil. Under different hydraulic gradients, the hydraulic conductivity became larger with the decrease of hydraulic gradient. The larger hydraulic gradient makes the flow velocity faster, and the resistance in the soil became greater with the increase of flow velocity. Furthermore, as the flow rate increases, the particles become denser, which will also lead to a decrease in the permeable captivity.

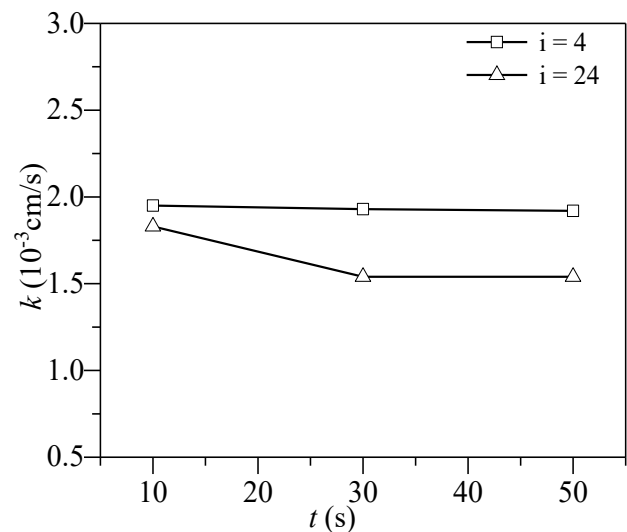


Fig. 1. Hydraulic conductivity on different hydraulic gradient.

According to previous research, the hydraulic conductivity of fine sand is 1.2×10^{-3} – 6.0×10^{-3} cm/s, and hydraulic conductivity of gravel is 6.02×10^{-2} – 1.8×10^{-1} cm/s. The permeability of MSWI bottom ash is close to that of fine sand. Considering the rainwater velocity

and the storage capacity of subgrades and pavements, it can already meet the requirements of permeable subgrades.

3.3 Adsorption tests

Adsorption isotherm refers to the curve of adsorption capacity changing with equilibrium concentration, which indicates the corresponding relationship between equilibrium adsorption capacity Q_e and equilibrium concentration C_e of adsorbate at a certain temperature (Tang et al., 2010). In this paper, adsorption isotherm models include Langmuir isotherm and Freundlich isotherm were used.

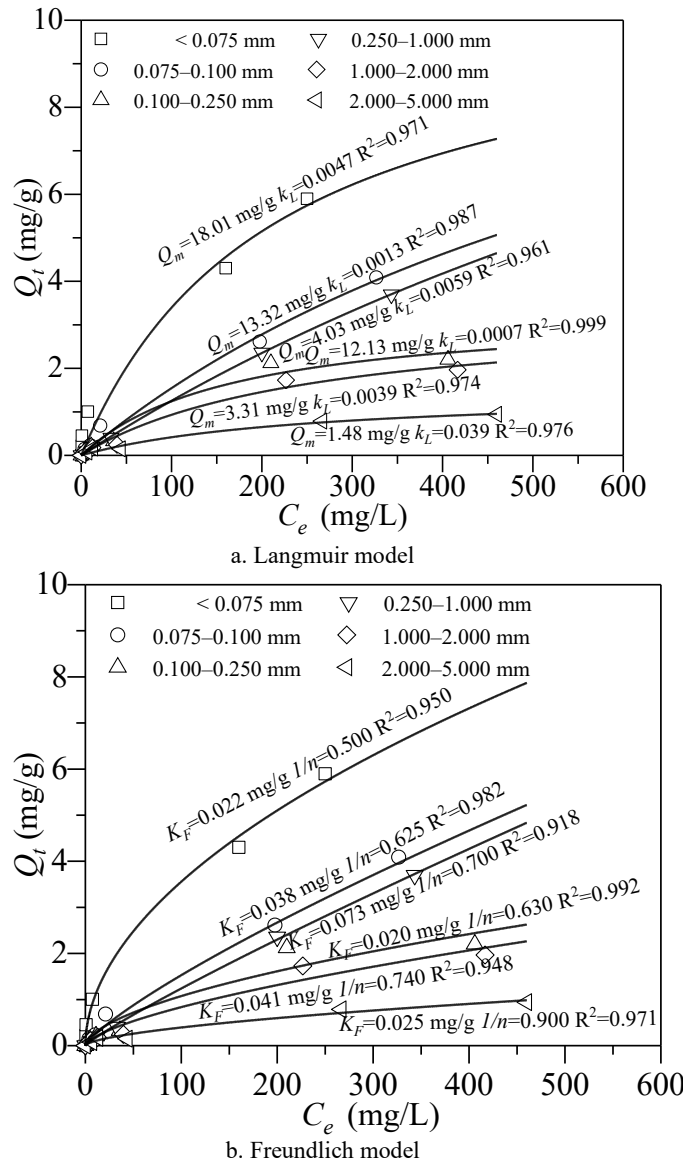


Fig. 2. Isothermal adsorption lines of different size.

Fig. 2 is the isotherms of different particle sizes. It can be observed from Fig. 2 that the unit adsorption amount increased with the increase of the equilibrium concentration C_e , and the growth rate decreased rapidly and then gradually reached equilibrium. When the unit adsorption amount was the largest, except for the particle

size group of 0.1–0.25 mm, the unit adsorption amount increased as the particle size decreases. The correlation coefficients of the fitting results were very closed to 1. The Langmuir model had better fitting effect (Tang et al., 2017b).

According to the fitting result of Langmuir model, when the particles were smaller than 0.075mm, their adsorption capacity was the strongest, which was 18.01 mg/g. According to the particle size gradation of this test, the adsorption capacity of MSWI bottom ash was about 5.44 mg/g. This was more than double the silt adsorption capacity (Tang et al., 2018b). Therefore, MSWI bottom ash has good adsorption capacity.

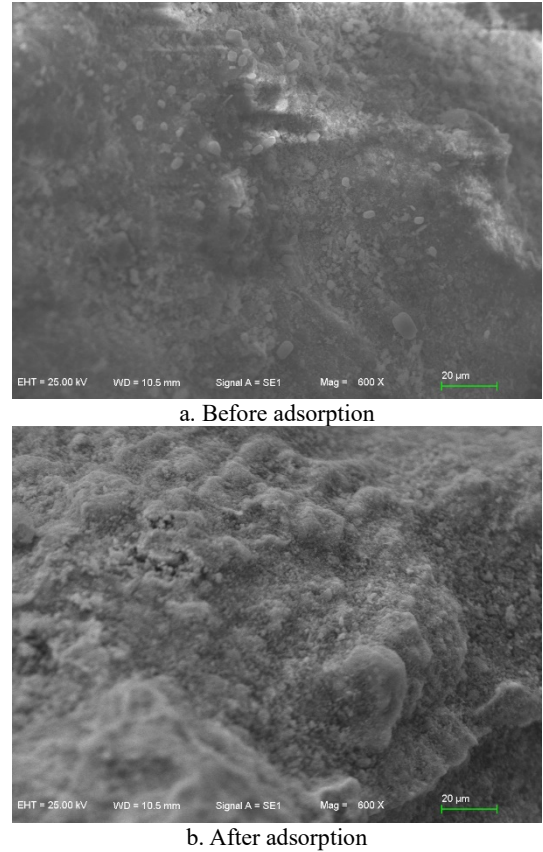


Fig. 3. SEM images.

As the adsorption progressed, a part of the soluble salt dissolved and a part of methylene blue was adsorbed on the particle surface. It can be found by SEM in Fig. 3 that after the adsorption was completed, the crystalline material on the surface was significantly increased, which may be caused by a part of methylene blue adsorbed on the surface. Thus, it can be seen that there were agglomerates on the surface of the particles (Tang et al., 2016b; Tang et al., 2018c).

4 NUMERICAL SIMULATION

The flow net was calculated by Brinkman equation shown in Fig. 4. According to the simulation results, under the condition of the same annual rainfall, the

velocity of the road surface, basement and subgrade was relatively close to inlet flow velocity. Due to the excellent permeable capability of the pavement, rainwater could quickly enter into the structure layer, rather than ran off the surface (Tziampou et al., 2020). Even if the water in other structural layers couldn't penetrate the soil layer in time, it would be stored in the pavement and roadbed, thus it was reasonable to assume that all the rainwater could penetrate the soil. Because the flow rate of each section was approximately the same at the same time interval, the flow velocity was only related to the area flowing through. Therefore, the velocity of the entire structural layer was substantially on the same order of magnitude.

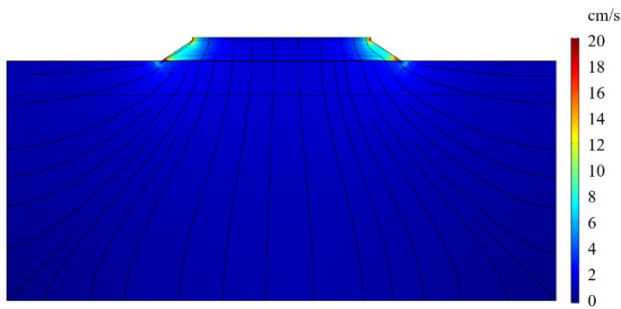
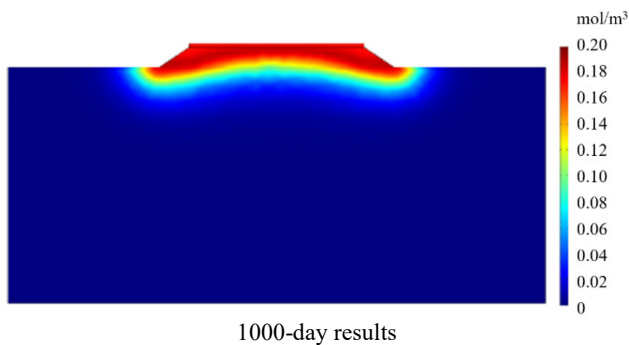
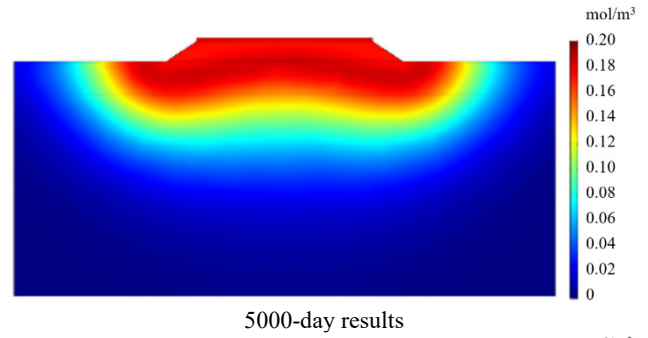


Fig. 4. Streamline by simulation.

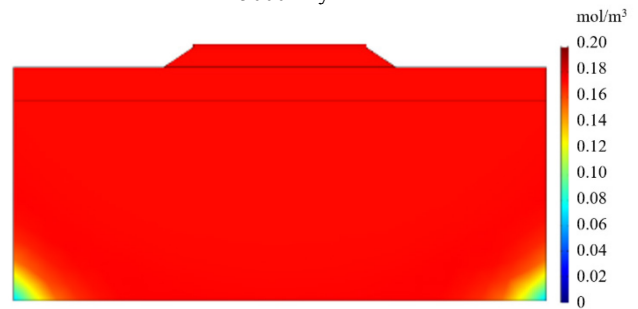
When the COD concentration was 100 mg/L and the annual rainfall was 1000 mm, the comparison with and without adsorption is shown in Fig. 5. Without considering the adsorption conditions, the pollutants had reached the soil within 500 days. In consideration of adsorption conditions, the time would be extended to more than 50,000 days (about 137 years). This means that the highway would have reached its design life at this time. By comparison, it can be found that MSWI bottom ash could effectively hinder the migration of pollutants. This phenomenon is mainly caused by three reasons. (1) The adsorption capacity of MSWI bottom ash was excellent. (2) The maximum adsorption capacity was high due to the huge amount of MSWI bottom ash in the subgrade. (3) The pollutant content was related to the concentration of pollutants and rainfall, which led to a low total amount of pollutants.



1000-day results



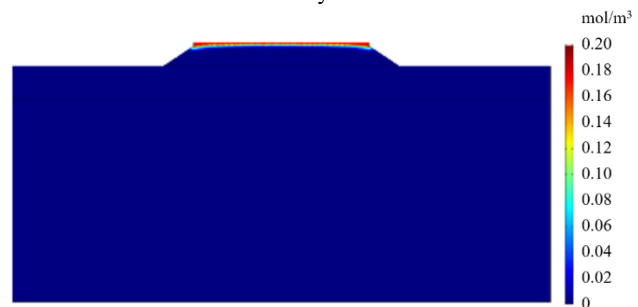
5000-day results



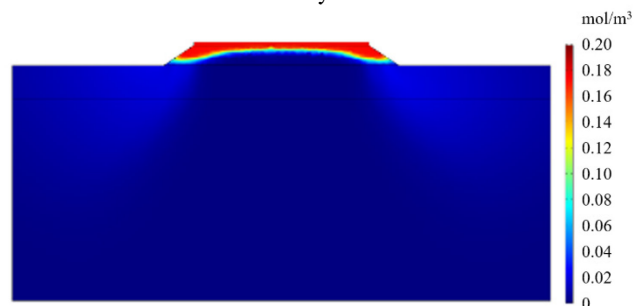
50000-day results
a. Without adsorption



1000-day results



5000-day results



50000-day results
b. With adsorption

Fig. 5. Results under different time.

Considering the design service life of the highway,

the time for observing the migration of pollutants was determined as 20,000 days (about 54.8 years). With the COD concentration of 100 mg/L and consideration of adsorption, different rainfall conditions were adjusted, and the results are in Fig. 6. As rainfall increased, pollution occurred in the same time, and the migration was obviously. However, even at 2000 mm, which was the rainfall in a tropical rain forest climate, the soil was still not reachable. The depth of migration of pollutants in the subgrade was basically proportional to rainfall. This means that the performance of the permeable subgrade is very good, and it can effectively inhibit the movement of pollutants in the heavy rainfall.

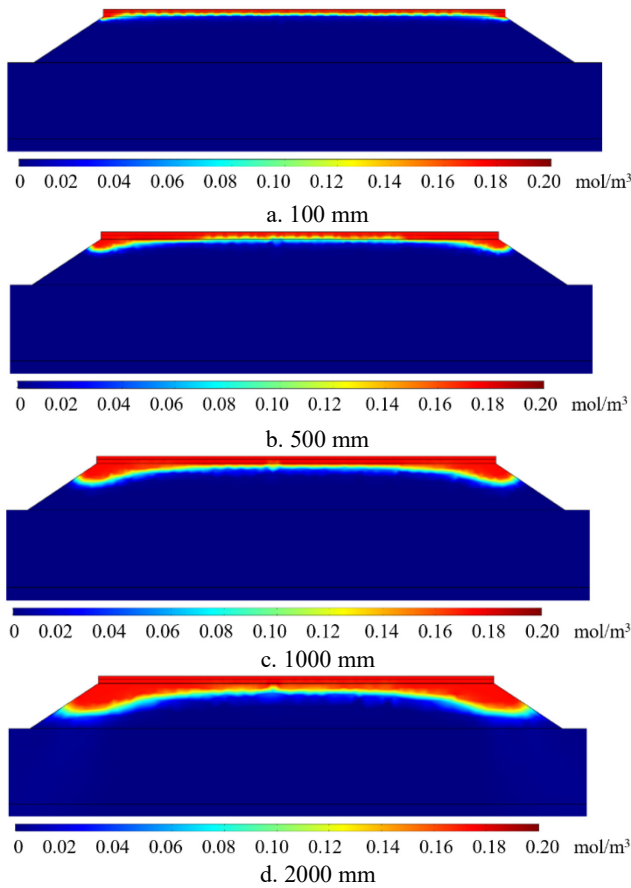


Fig. 6. 20000-day results under different rainfall conditions with adsorption.

Contaminant concentration is also an important factor affecting pollutant transport. With an annual rainfall of 1000 mm and considering the conditions of adsorption, the concentration of COD was adjusted. After simulation, the results at 20,000 days are in Fig. 7. It can be found through simulation that as the concentration of pollutants increased, the range and concentration of pollutant diffusion increased. The increase of the diffusion range was mainly due to the larger concentration, which led to the increase of the diffusion (Tang et al., 2018d).

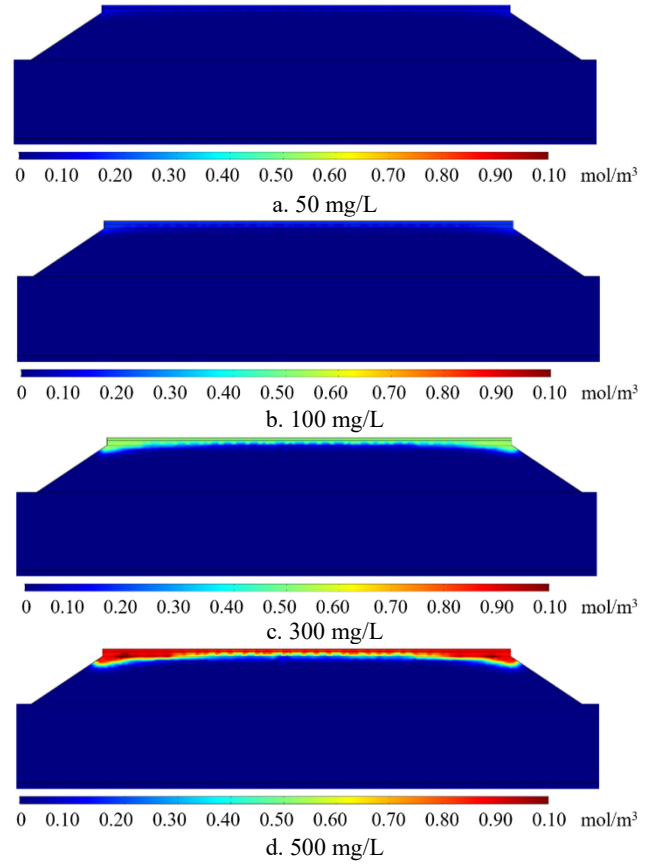
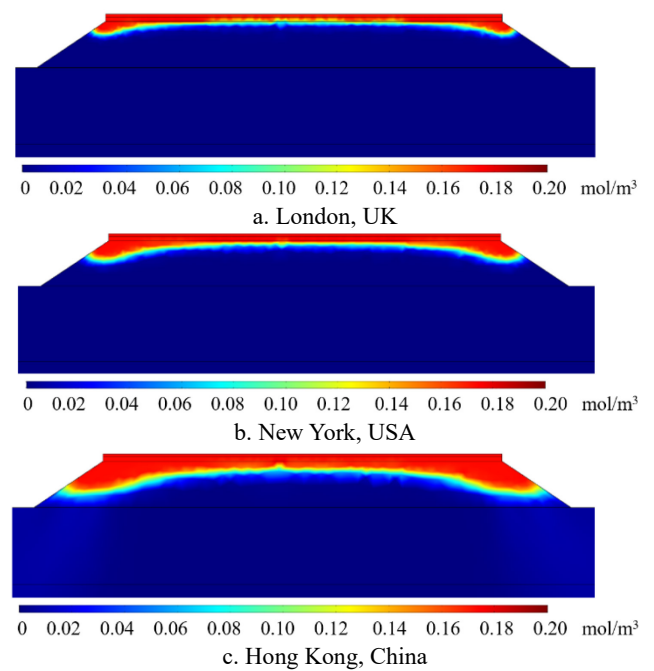


Fig. 7. 20000-day results under different COD concentration conditions.

In addition, 4 typical cities in different regions and climates were selected and simulated in Fig. 8. By comparing the results of various cities, the retardation effect on the transport of pollutants was very obviously. MSWI bottom ash permeable subgrade can meet the needs of various regions.



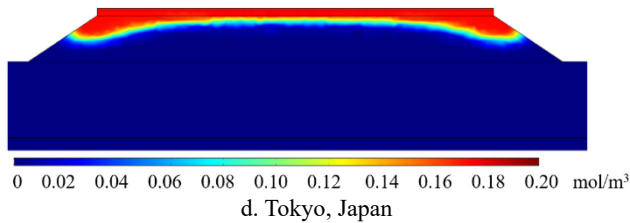


Fig. 8. 20000-day results in different areas with adsorption.

5 CONCLUSIONS

Through the above experimental results, calculations and analysis, the following conclusions can be obtained:

1. MSWI bottom ash as subgrade material, has excellent hydraulic behaviors, which about 0.00002 m/s, and can meet the requirements of permeable subgrade.
2. MSWI bottom ash has a certain adsorption capacity about 5.44 mg/g, and its adsorption form is mainly physical adsorption.
3. MSWI bottom ash permeable subgrade can effectively inhibit the movement of organic pollutants under various engineering conditions, including concentration, rainfall, etc.

ACKNOWLEDGEMENTS

This study was supported by the National Natural Science Foundation of China (51778386, 51708377, 51608059), Natural Science Foundation of Jiangsu Province (BK20170339), Natural Science Fund for Colleges and Universities in Jiangsu Province (17KJB560008), Open Fund of National Engineering Laboratory of Highway Maintenance Technology (Changsha University of Science & Technology) (kfj180105) and project from Jiangsu Provincial Department of Housing, Urban-Rural Development (2016ZD18 and 2017ZD002).

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