https://doi.org/10.56946/jce.v4i1.599

Journal of Chemistry and Environment

<u>Research Article</u> Simulation Study of Groundwater Polluted by Landfill Leachate in Subsidence Areas Bao Hui¹, Xu Guangguan^{2*}, Liu Xiang², Selhaba Ayesha²

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Abstract

The subsidence caused by mining was used as domestic waste landfill, due to the lack of anti-seepage treatment at its bottom, the internal leachate not only contaminated the surface water, but also the goaf groundwater continuously. Therefore, taken the Datong landfill in Huainan, Anhui Province as an example, the similar simulation test was chosen to simulate the temporal variation of the concentration of pollutants in the goaf. The formed mechanism of leachate pollution of groundwater was also discussed. The results show that the leachate still caused serious pollution to groundwater after the closure of the landfill. Pollutants in groundwater mainly move from west to east under the action of convection-dispersion and adsorption. In the process of pollutant migration and diffusion from west to east, it is controlled by the boundary of the aquifer in the goaf, resulting in a large accumulation of pollutants at the end of the goaf and causing the high concentration of pollutants. In addition, due to the influence of geological environment in goaf, the pollution rate and concentration are constantly changing. The results of the test provide some reference for the groundwater remediation and treatment in the coal mine subsidence area.

Keywords: Domestic waste landfill, leachate, similar simulation test, groundwater remediation.

1. Introduction

In the past, coal mining subsidence areas were often used as landfills for domestic waste, and its leachate not only polluted the surface water [1-3], but also causes serious pollution to the groundwater in the goaf. Therefore, how to repair and control mine landfill has become a focused issue. At present, the research on groundwater pollution in landfill mainly focuses on the investigation of hydrogeological conditions and pollution status of traditional landfills, the simulation of groundwater pollution migration and the remediation of groundwater pollution. The predecessors have done a lot of research work on the investigation of the surrounding environment of the landfill and the polluted groundwater. Liang et al. [4] judged the scope of groundwater pollution through the monitoring of the Chifeng garbage dump and the surrounding groundwater environmental system, revealed the cause mechanism of groundwater pollution, providing a reference for the final groundwater remediation and treatment. Zhang et al. [5] investigated the general situation and hydrogeological conditions of a landfill site in Tangshan City. Through hole layout, sampling and monitoring, it was found that the groundwater in this area had

been polluted by landfill leachate. The main factors of pollution were COD, NO₃-N, ammonia nitrogen, total hardness and total dissolved solids. Yang et al. [6] conducted geological exploration and pollution load evaluation on an old domestic waste landfill in Xuzhou City and found that its bottom protective barrier had been polluted, and the main pollutants were Cr and Pb.

In the simulation of groundwater pollution, experimental simulation and numerical simulation are mainly used to reveal the characteristics and migration pattern of pollutants. Yin et al. [7] used Visual MODFLOW software to simulate the Huainan Datong garbage dump based on soil permeability test and conventional ion adsorption test and obtained that chloride ions continue to diffuse in shallow groundwater in the form of pollution halo mainly along the direction of water flow. Liao et al. [8] established a groundwater flow model and a solute transport model based on Visual MODFLOW software, simulated the migration of permanganate index and ammonia nitrogen in groundwater under two leakage conditions, and evaluated the environmental risk of groundwater. Fandiño et al. [9] used Modflow and MT3DMS modules to simulate the migration and diffusion of chromium, lead, copper, and zinc metals in the leachate of a landfill in Brazil, and predicted the feather epidemic behavior, finding that the landfill will cause serious pollution to the surrounding soil and groundwater after 50 years.

Many scholars have proposed a variety of remediation methods and measures to prevent landfills from polluting groundwater. Wang et al. [10] compared the pollution migration law of ammonia nitrogen and nitrate in groundwater before and after grouting in a landfill site in Shandong Province by numerical simulation method. The results showed that the pollution channel was cut off after grouting, which had a certain control effect on groundwater pollution. Xu et al. [11] used reactors with different media to simulate reactive infiltration walls to treat landfill leachate, and demonstrated that it is feasible for PRB to treat landfill leachate to pollute groundwater. Xie et al. [12] designed three schemes to control groundwater pollution in landfills, tested the effectiveness of the three methods through a

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coupled numerical model of groundwater flow and pollutant transport and found that the combination of impermeable wall and pumping well was the best solution to control pollutants.

The leachate can still continue to leak for many years after the closure of the traditional landfill site, which will cause continuous pollution of the landfill site [13]. At present, most of the studies on groundwater pollution by landfill leachate are focused on existing or still operating landfills [14-16]. There are few studies on the problem of groundwater pollution caused by leachate from mine solid waste landfills that are sealed in the later stage but have not been treated with anti-seepage treatment at the bottom. Therefore, based on hydrogeological background analysis, the spatiotemporal evolution of leachate pollution components under hydrodynamic action was studied by experimental simulation method in the landfill in Datong subsidence area of Huainan City, and the mechanism of landfill leachate pollution of groundwater under this condition was discussed, which provide a reference for the further restoration and treatment of domestic waste landfill in the mine subsidence area.

2. Overview of study area

2.1. Overview of landfill area

The Datong landfill is located in the Jiulonggang coal mining subsidence area of Datong District, Huainan City. A set of Cambrian and Ordovician carbonate exposed strata is developed in the south. The coal mining subsidence area is the coal seam distribution position, and the coal seam dip angle is nearly vertical. In the outcrop area and shallow carbonate-covered area, karst is very well developed and is a karst aquifer [17], and the atmospheric precipitation flows from the piedmont to the subsidence area from west to east to recharge the coal mining subsidence area through the surface and underground runoff in the piedmont [18]. Figure 1 shows geohydrologic condition in the landfill area. Figure 1 (a) is the hydrogeological plan of the study area, and Figure 1 (b) is the geological section A-A '.

The landfill was operated in 1984 and closed in 2009. The longterm stacking of waste not only polluted the surrounding soil and air, but also produced landfill leachate inside. A series of disposal measures, such as external stacking, drainage and leachate extraction, were used to effectively control the internal pollution until 2016, but the anti-seepage system was not designed at the bottom of the garbage dump in the early stage, which still caused different degrees of pollution to the groundwater in the -goaf.



Figure 1. Hydrogeological map of the study area. (a) Plane hydrogeological map of the study area, (b) Geological profile of the A-A'.

2.2. Current status of water pollution in the study area

In order to find out the pollution of landfill leachate to the surrounding water environment, the detection indicators of water samples were determined according to the \langle Technical Specifications for Groundwater Environmental Monitoring \rangle (HJ 164-2020) and the \langle Groundwater Quality Standards \rangle

The conductivity value was determined by a conductivity meter; The TDS value is determined by the TDS instrument; DO value was determined by dissolved oxygen meter; BOD5 was determined by direct culture method or dilution inoculation method; CODcr was determined by potassium dichromate method; ammonia nitrogen was determined by Nessler's reagent spectrophotometry; The total nitrogen determination method was alkaline potassium persulfate-ultraviolet spectrophotometry; total phosphorus was determined by ammonium molybdate spectrophotometry. The results of the water samples can be seen in Table 1.



The results showed that the concentrations of ammonia nitrogen and TN in the leachate pollution index of Datong landfill were relatively high, and the pollution degree among the three was: leachate> groundwater > surface water. The conductivity of landfill leachate is much higher than that of other types of water types, indicating that the landfill leachate pollution components have polluted the groundwater. According to the worst category of single index evaluation results in the comprehensive evaluation of groundwater quality in the \langle Groundwater Quality Standard \rangle (GB / T 14848-2017) [20], the comprehensive category of groundwater quality around the landfill is V, and the index is ammonia nitrogen.

Table 1. The results of sampling point index test.

Sampling location		Test Indicators							
		DO /mg·L ⁻¹	BOD5 /mg·L ⁻¹	TDS /mg·L ⁻¹	Conductivity /µs·cm ⁻¹	ammonia nitrogen /mg·L ⁻¹	TN /mg·L ⁻¹	CODcr /mg·L ⁻¹	
Monitoring wells	S1	1.12	12.88	535	1050	8.15	13.4	36.8	
	S2	1.48	97.15	539	1086	10.52	18.4	128.0	
	S3	1.35	66.56	2760	5540	53.09	93.43	211.2	
	S4	1.83	8.96	693	1386	3.31	4.80	28.0	
	S5	2.42	93.28	1710	3420	43.75	67.32	176.0	
Surface water	B1	6.34	8.42	647	321	1.77	3.38	24.0	
	B2	6.62	13.44	642	321	1.48	3.76	32.0	
	В3	5.61	8.06	621	309	3.14	4.47	22.4	
Groundwater	D1	5.93	7.92	363	728	2.14	3.48	24.0	
	D2	5.09	1.62	611	1226	8.25	13.12	8.0	
	D3	2.97	5.76	453	904	1.97	4.04	16.0	

Note: DO(Dissolved Oxygen), BOD₅(Biological Oxygen Demand after five days), TDS(Total Solids Dissolved), TN(Total Nitrogen), CODcr(Chemical Oxygen Demand)

ammonia nitrogen was selected as the pollution factor in this experiment. The results indicated that ammonia nitrogen had a good indicator effect on groundwater pollution, therefore

3. Test device and process

3.1. Design of test device

(1) Design basis: Through field investigation and analysis of previous mining data, the strike length of the goaf is 2862 m, the inclined width is 230 m, and the depth is 555 m. Therefore, the model is designed with a trapezoidal sand groove with a length of 130 cm, with an upper width of 10 cm, a lower width of 8 cm, and a height of 25 cm. The similarity ratio between the model device and the actual goaf size is 1:2200.

(2) Model structure: For the caving zone and fracture zone formed in the process of coal seam mining, the former has good permeability, and the latter has relatively poor permeability. The terrain of the goaf is generally high in the west and low in the east, and the groundwater in the subsidence area flows slowly from west to east. The south side of the goaf is the piedmont recharge area (recharge boundary), and the north side

is the water barrier boundary. Therefore, the model structure is designed with two layers, the upper layer is a fine sand layer with poor permeability to simulate the fracture zone, and the lower layer is a coarse sand layer with good permeability to simulate the caving zone. The left and right boundaries are the constant head boundary, the front is the lateral recharge boundary, and the back is the water barrier boundary. The top of the device is sealed by a plexiglass cover to simulate compressive stress state. Pressure measuring holes and sampling holes of different depths are arranged on the side of the sand trough to monitor the changes of pollutants in time and space. Figure 3 shows the test device diagram.

3.2 Test material

(1) Selection of filling material

Natural quartz sand with different particle sizes and permeability was selected for the test, and the ratio was carried out according to the internal double-layer structure of the simulation area, the lower layer simulated the equivalent simulated "caving zone" with large porosity and good permeability, and the upper layer simulated the equivalent simulated " fracture zone" with relatively small porosity and poor permeability. Table 2 shows the media parameters obtained by porosity instrument and Darcy experimental instrument. (2) Calibration of solution concentration

In this experiment, 50 mg/L ammonia nitrogen solution was used to simulate the pollution components of landfill leachate. Among them, the ammonia nitrogen solution required for the test was prepared and determined according to the requirements of "Water Quality - Determination of Ammonia Nitrogen -Spectrophotometry of Nessler's Reagent" (HJ 535-2009) [21].

3.3. Test Process

(1) Test preparation: The method of layered compaction every 3 cm is adopted, and 11.5 cm thick coarse sand and fine sand were filled in layers from bottom to top to achieve the actual bulk density of the field land, and an appropriate amount of water was injected into the filling layer to make it sink and consolidate under the action of gravity.



Figure 3. Diagram of the test instrument (a) Side view of test device, (b) The overall structure of the simulated test groove.

Table 2.	List	of	simulated	medium	parameters
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simulated layer	filling medium	permeability coefficient /cm·s ⁻¹	porosity
first layer (fracture zone)	fine sand	0.00136	0.25
Second layer (caving zone)	coarse sand	0.00312	0.36

Finally, a 2 cm thick test soil sample was filled to simulate the aquiclude. After filling, seal with a plexiglass lid to achieve a

pressure-bearing state. The trapezoidal test tank indirectly controls the inlet water level through the upstream water supply

tank, and indirectly controls the outlet water level through the downstream outlet water tank. The inlet water level of the test tank is 28.0 cm, and the outlet water level is 27.4 cm, so that the water flow is in a continuous and stable flow state. The test water is all ultrapure water obtained from the ultrapure water mechanism.

(2) Test process: When the flow rate reaches a stable state, 50 mg/L ammonia nitrogen solution is uniformly injected into the test tank through a peristaltic pump, and the ammonia nitrogen concentration value is measured and recorded at intervals. When the mass concentration of the tail of the test tank is similar to the mass concentration of pollutant injection, the test is stopped.

4. Results

4.1. The variation of the concentration of pollution components with time

The breakthrough curve of ammonia nitrogen solution in the sand tank is shown in figure 4 (A-C). The junction of different sand layers represents the interaction position of the fracture zone and the falling zone. In the figure, A-E represent the sampling holes in columns 1-5, and the numbers 1-3 represent the fracture zone, junction zone, and caving zone. The experimental results show that: The mass concentration of

ammonia nitrogen at each measurement point basically shows the same change trend, that is, the concentration first rises in a straight line and then tends to be stable. Therefore, the process of the change of pollutant component concentration with time is divided into two stages: rising period and stable period. The first stage (rising stage): the concentration of ammonia nitrogen in the exudate at each measurement point increased rapidly from the initial value of 0 mg/L, indicating that the rapid convective diffusion of ammonia nitrogen occurred under the action of groundwater flow. The second stage (stable period): when the concentration of ammonia nitrogen reaches the peak, it gradually tends to be stable, but the time taken by this change law is different at different locations, and the maximum concentration does not reach 50 mg/L. This phenomenon is mainly caused by adsorption action and dilution action. The pollution components migrate faster in the fracture zone and the boundary layer, and reach the relative concentration peak earlier, while the pollution components migrate slower in the caving zone, and reach the relative concentration peak later. This indicates that the filling material level has a certain influence on the penetration rate of pollutants, and the interception ability of different permeable media to pollutants is different.



Figure 4. The variation of ammonia nitrogen concentration with time at different positions (a) fracture zone, (b) junction zone, (c) caving zone.



Figure 5. Distribution of ammonia nitrogen concentration on the plane at different times (a) prophase (47 h) and (b) anaphase (779 h).

4.2. The variation of the concentration of pollutant

components with space

4.2.1. The variation law of pollution components in different depth plane of goaf

The concentration change of the selected pollutant ammonia nitrogen on the plane is shown in figure 5 (A & B). The two moments of the early period (47 h) and the late stage (779 h) were selected as the main time nodes to describe the spatial variation of ammonia nitrogen concentration, and the results showed that:

Along the direction of groundwater flow: In the early stage of the test, the pollutant concentration on the plane of different depths in the goaf showed a trend of increasing first and then decreasing. Among them, the concentration difference in the upper part of the goaf (fracture zone) is larger, and the concentration difference in the lower part of the goaf (caving zone) is smaller. The results show that the pollutants in the early stage of the test have serious pollution to the upper part of the goaf, the pollution speed is faster, and the pollution to the lower part of the goaf is lighter. In the later stage of the test, the concentration of each layer increased first and then decreased. The maximum mass concentration of ammonia nitrogen in the fracture zone and the boundary layer was between columns A and B, and was basically equal to the concentration of pollutant injection. The minimum concentration of pollution components in the caving zone is in column A, indicating that the pollution level is low

Vertical to the direction of groundwater flow: Most of the concentration contours are curved arcs rather than straight lines perpendicular to the direction of water flow. The concentration distribution shows a phenomenon of high in the south and low on both sides of the north, which is caused by the lateral recharge and water barrier boundary in front of the mountain.

Therefore, in the early stage of the experiment, the simulated concentration of ammonia nitrogen in the goaf increases first and then decreases with the change of geological environment, and the same trend was also shown in the later stage of the experiment. The migration speed of pollutants in the goaf caving zone is faster than that in the fracture zone.

4.2.2. The variation law of pollution components in different sections of goaf at different positions

The concentration of pollutants in different position profiles was monitored. and the distribution. The distribution and overall change trend of ammonia nitrogen concentration in different position profiles are shown in figure 6, and the test results show that:

Along the depth direction (from high to low): In the early stage of the test, with the deepening of the depth, the concentration of pollution components in the fracture zones of columns A and B was greater than that in the caving zone, indicating that the pollutants migrate from top to bottom in the vertical direction. The concentration of pollutant ions in the junction layer of columns C, D, and E was the largest, which is due to the accumulation of contaminants due to the difference in the permeability of the filling medium. At the later stage of the test, the concentrations of pollutants in columns A and B decreased with the increase of depth, and the concentrations of pollutants in most of the parts close to the junction were the largest, but the concentrations in columns C, D and E were opposite. With the deepening of the depth, the concentration contour distribution of the caving zone at the end of the goaf was loose, the concentration contour distribution of the fracture zone was dense, and the goaf had been completely polluted.

Perpendicular to the depth direction: the peak concentration was basically at the left of the center line of the boundary layer of the goaf, and the concentration isoline on the left side of the center line was dense. The mass concentration of ammonia nitrogen near the aquifuge boundary in the caving zone was higher than that around the lateral recharge area.

Therefore, the ammonia nitrogen content in column D, column E (downstream) and column A in the later stage of the experiment was similar, and the migration rate of pollutants in the caving zone of the goaf had slowed down.



Figure 6. Distribution of ammonia nitrogen concentration on the profile at different times (a) 47 h and (b) (b) 779 h



Figure 7. The change curve of ammonia nitrogen concentration at different times, (a) fracture zone and (b) caving zone

The pollution concentration was close to the peak concentration, indicating that the ammonia nitrogen had gradually moved from the highest concentration column (upstream) to the farthest column (downstream) from the pollution source, and the goaf had been completely polluted.

5. Discussions

Groundwater recharge, runoff, and discharge are important hydrogeological control factors of groundwater in landfill leachate pollution subsidence areas [22,23]. Therefore, identifying the influencing factors of recharge, runoff and discharge processes in the groundwater system is —a prerequisite for revealing the mechanism of pollutant transport.

5.1. Recharge

Atmospheric precipitation changes the groundwater level through the lateral recharge of the exposed piedmont carbonate rocks in the south of the dumpsite, which affects the migration of pollutant components.

Due to the effect of lateral recharge and aquifuge boundary in goaf, the contour of pollutant concentration was curved along the direction of water flow, which affects the direction of groundwater flow. The pollutant not only moves with the water along the water flow direction, but also moves to the water resisting boundary and gathers, so that the peak concentration was basically on the left side of the center line of the trough junction, showing that the concentration contour on the left side of the center line was dense and the concentration contour on the right side was loose.

5.2. Runoff

The change of pollutant concentration in the process of groundwater pollution in goaf was mainly affected by convection-diffusion and adsorption [24-26].

5.2.1. Convection-diffusion action

(1) Convection action

According to the test data, the variation curves of ammonia nitrogen concentration in different monitoring wells with time were plotted, as shown in figure 7. Under the action of convection, the pollutants migrate horizontally in the fracture zone and the caving zone respectively. The increase of pollutant concentration in A-B column reaches the high peak value of pollutant concentration. This is followed by a low peak of pollutant concentration in columns B-C. This is due to the injection of pollution sources near column A. Pollutants migrate along the flow direction with water molecules under convection, and the concentration of pollutants decreases with the increase of migration distance. The D-E column was located at the end of the goaf. Due to the existence of bound water and the occlusion of the goaf environment, the pollutant

discharge was not smooth, and the concentration of pollutants at the end of the goaf was rising.

(2) Diffusion action

Due to the difference in pore size and permeability between the upper and lower layers of the goaf, that is, the porosity and permeability coefficient of the caving zone are greater than the porosity and permeability coefficient of the fracture zone, so that the concentration contour density of the caving zone is greater than that of the fracture zone, and the migration and diffusion speed of pollutants in the caving zone is faster than that in the fracture zone. In addition, under the action of molecular diffusion, pollutants not only migrate along the direction of water flow, but also diffuse vertically.

Therefore, after the pollutants enter the groundwater, the water migrates from the beginning to the end of the goaf under the action of convection, and the pollutants mainly migrate with the movement of water. In addition, under the action of hydrodynamic dispersion (molecular diffusion and mechanical dispersion), pollutants migrate from the high-concentration area in the double-layer medium of the goaf to the lowconcentration area, and finally reach the saturation state of pollutants.

5.2.2 Adsorption action

In addition to the convection-dispersion action, there was also adsorption action when the pollutants enter the groundwater, and the pollutants will adhere to the solid surface with the groundwater flow to slow down the groundwater flow rate. Therefore, due to the adsorption of solid particles on the surface, the concentration of pollutants in the goaf was less than 50 mg/L.

5.3 Discharge

The pollutants in the groundwater in the goaf are transported to the tail end of the goaf for discharge through convectiondispersion and adsorption. As can be seen from figure 7, the ammonia nitrogen concentration in the second half of the goaf shows an upward trend. The main reason for this phenomenon is that the difference in the properties of the aquifer in the outer area of the goaf and the inner area of the goaf leads to the poor discharge of water at the tail end of the goaf and the decrease of the water flow velocity, which indirectly affects the degree of convection-diffusion action, resulting in the accumulation of pollutants at the end of the goaf and the increase of the concentration of pollutants at the end of the goaf.

6. Conclusions

In this paper, the migration and diffusion process of pollutants in groundwater is designed and simulated for the pollution of goaf groundwater by landfill leachate in coal mining subsidence area. The main conclusions are as follows.

(1) The migration of pollutant components in the goaf is mainly affected by the factors of recharge, runoff and excretion, and the migration and diffusion are carried out under the convection-diffusion and adsorption effects.

(2) In the process of migration and diffusion of pollutants from west to east in the goaf, they are controlled by the water separation boundary of the goaf, resulting in a large number of pollutants accumulating at the tail end of the goaf, resulting in a large concentration of pollutants.

(3) Due to the influence of the permeability mutation and adsorption difference of the aquifer between the water conduction fracture zone and the caving zone, the pollutant concentration tends to increase when the pollutants infiltrate vertically through the fracture zone to the caving zone.

Authors contribution

Xu Guangquan devised the project, the main conceptual ideas and proof outline. Bao Hui worked out almost all of the technical details, and designed experiments, analysis of results, wrote the manuscript. Liu Xiang helped in sampling from study area and test simulation.

Ayesha Selhaba helped in editing and formatting of

manuscript.

Conflicts of Interest

There are no conflicts of interest reported by the writers.

Acknowledgment

I wish to extend my heartfelt appreciation to my supervisor, Prof. Xu Guangquan, for his expert guidance and unwavering support during the entire process of this academic endeavor. His profound insights and meticulous feedback were instrumental in shaping the direction of my research. I am deeply indebted to him for both my academic growth and the accomplishments reflected in this work. Additionally, I extend my sincere thanks to my research colleagues for their collaborative spirit and constructive discussions, which greatly enriched the quality of this research.

Data Availability statement

The data presented in this study are available on request from the corresponding author.

Funding: Not applicable(N/A).

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How to cite this article:

Bao H., Xu G., Liu X., Selhaba A. (2025). Simulation Study of Groundwater Polluted by Landfill Leachate in Subsidence Areas. *Journal of Chemistry and Environment*. 4(1). p. 41-52.