Experimental study on clogging and water quality change during artificial recharge

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Abstract. In this paper, an experimental sand column was used to investigate the clogging process and water quality changes simultaneously during artificial recharge. Clogging was described quantitatively according to the changes of hydraulic conductivity, whilst several important water quality parameters, such as SS, total bacterial count, DO, TOC, total nitrogen, total phosphorus were measured at regular time intervals during recharge. After 80 hours of continuous recharge, the hydraulic conductivity of the upper column (10-15cm) and the lower(15-55cm) decreased from 1.23×10^-4 m/s to 9.84×10^-5 m/s and from 1.23×10^-4 m/s to 1.06×10^-4 m/s, respectively. This was thought to be due to physical clogging by suspended solids and then biological clogging by biomass accumulation along the columns. The sand column had significant self-purification effect on suspended matter, bacteria, TOC, permanganate index, total nitrogen, total phosphorus, copper, etc. The filtration and adsorption effect of the upper column were much stronger than that of the lower, so the clogging of the upper was more severe. The results of this study will provide technical support for the implementation of large-scale artificial recharge in the Liaolan groundwater funnel area of Qingdao.

1. Introduction
Due to the limitation of sustainability of the conventional use of water resources approaches in some arid areas, it is very critical to advocate water reclamation and appropriate reuse. Due to the continuous drought and excessive exploitation of groundwater, the groundwater level gradually drops, forming a large-scale funnel area centered on Liaolan Town, in Qingdao. Artificial groundwater recharge is one of the most important ways to relieve the water crisis and environmental geology problems at present[1,2,5]. However, the negative effects often encounters aquifer clogging and water pollution during artificial groundwater recharge[1, 2]. Aquifer clogging is a complicated physical and chemical process, which was influenced by particle size characteristics of aquifer medium and the quality of recharge water. It has been shown that suspended solids (SS), namely physical clogging, is the first reason to cause aquifer clogging [3-5]. While, biological clogging is another important reason which cannot be ignored. nonfecal coli forms were usually found in recovered water, when injection and extraction period were not continuous, even though the recharge water was pre-chlorinated. [5-7]
As for changes in water quality, many laboratory studies have been carried out to simulate its variation during artificial recharge. The aquifer has significant self-purification effect on some water quality parameters like turbidity, chroma and the number of coliform through laboratory column studies.[8,9]investigated the transformations of dissolved organic carbon using lagoon-treated wastewater during laboratory-scale groundwater recharge. However, the research on the clogging...
process and mechanism was few and most of them usually focused on one type of clogging. Previous researchers usually studied aquifer clogging and water quality changes separately during artificial groundwater recharge. However, the two processes occur simultaneously. This study was to investigate the clogging process and water quality changes simultaneously and to analyze the clogging process according to the possible physical, chemical and biological changes in the column during artificial recharge. The results of this study will provide technical support for the implementation of large-scale artificial recharge in the Liaolan groundwater funnel area of Qingdao.

2. Materials and Methods

2.1. Experimental equipment
As is shown in figure 1, a sand column was used to simulate the recharge process. The column material was clear plexiglass. The length of the sand column is 90 cm, and the internal diameter is 15 cm. Water head was kept constant during the experiment with overflow port located at 10 cm from the column orifice. Sampling taps were located respectively at 25, 30, 40, 50, 60, 70 cm from the column orifice. The clogging and the resultant decrease in hydraulic conductivity were monitored with six piezometric tubes located on the other side of the column. Stainless meshes were placed inside the end of the caps to support the sand material and help spread the input solution laterally throughout the columns.

The filling medium sand and injecting water in the sand column were collected from a flood plain of the Changjiang River, in Nanjing city, China. Nanjing city lies in the central part of the inshore areas of China, between northern latitude 30°13′ to 32°36′ and eastern longitude 118°21′ to 119°15′. The flood plain covers an area of about 115 km² with the annual precipitation is 1000-1500 mm. Its geological formations comprise Quaternary alluvial, alluvial-diluvia and residual-slope residual slope deposits. The total thickness of the Quaternary deposits is 63.3 m, most of which are binary structure. The upper layer is composed of sandy loam, silty sand, and the lower sand layer with the thickness of 54 m is composed of fine sand, medium-coarse sand particles and sandy gravels. The hydraulic conductivity of the sand layer ranges from \(1.16 \times 10^{-4} \text{ m/s}\) to \(2.32 \times 10^{-4} \text{ m/s}\) according to pumping tests in the past several years[10], which is close to the aquifer medium of Liaolan groundwater funnel area. The soil sample filled into the column was saturated after being compacted. Particle size
characteristics of the sand were analyzed using screening method. Figure 2 shows the particle distribution and table 1 shows some important hydrogeology parameters of the soil.

Figure 2. Grading Curve of the medium sand.

| Parameters                      | Value      |
|---------------------------------|------------|
| Average particle size (mm)      | 0.34       |
| Porosity                        | 0.31       |
| Hydraulic conductivity (m/s)    | $1.23 \times 10^{-4}$ |
| Specific Yield                  | 0.19       |

2.2. Water quality of the recharge water

Recharge water was obtained from a flood-discharging river in Nanjing city of China. The analysis results of water sample showed that each kind of trace heavy metal content was very low whereas the concentration of trace heavy metal is a very important pollutant factor of water quality problem during artificial groundwater recharge. Copper sulfate solution was added into the river water in order to investigate the change in trace heavy metal during artificial groundwater recharge. The samples were measured by using equipment from State Key Laboratory of Hydrology-water Resources and Hydraulic Engineering. The measuring methods and devices of water quality parameters are listed in Table 2. The water quality analysis result of recharge water is listed in Table 3.

Table 2. Measuring methods and devices of water quality parameters.

| Parameters | Devices             | Type           |
|------------|---------------------|----------------|
| pH         | PH meter            |                |
| TN         | Ultraviolet visible spectrophotometer | Cary50        |
| TP, NH$_4^+$ | Spectrophotometer   | Cary50        |
| TOC, DOC   | TOC device          | Multi N/C2100  |
| Inorganic ions | Ion chromatograph | DIONEX-300    |
Table 3. Water quality analysis result of recharge water.

| Parameters            | Value  |
|-----------------------|--------|
| TOC(mg/l)             | 13.01  |
| DOC(mg/l)             | 8.67   |
| permanganate index    | 10.59  |
| sulfate ion (mg/l)    | 44.95  |
| chloride ion (mg/l)   | 39.73  |
| nitrate ion (mg/l)    | 1.288  |
| total nitrogen (mg/l) | 9.04   |
| pH                    | 8.3    |
| total dissolved solids (mg/l) | 1167  |
| suspended matter (mg/l) | 21.4  |
| copper(mg/l)          | 1.371  |

2.3. Experimental methods and procedures

The aquifer clogging process occurring during the experiment was analyzed according to the hydraulic conductivity changes (K, m/s) [11]:

\[ K = \frac{4Q \cdot \Delta x}{\pi D^2 \cdot \Delta h} \]  

where \( Q \) is the flow rate (m³/s), \( \Delta x \) is the length (m) of the column between any two pressure sensors, \( \Delta h \) is the difference of hydraulic head between the same two points along the column (m), \( D \) is the internal diameter of sand column (m).

To obtain reliable results, the sand column was injected with distilled water continuously until the hydraulic conductivity sustained stable in couple of minutes. In the first 48 hours, the hydraulic conductivity decreased gradually, then kept stable value of 1.23×10⁻⁴ m/s. After that, the recharge water was supported by BT300 peristaltic pump, which is an economical speed regulation pump. The hydraulic conductivity changes were measured every 8 hours during artificial recharge, whilst water quality parameters, at different sampling places, were measured shortly after sampling at regular intervals. Water quality parameters included the suspended solids (SS), total bacteria count, DO, total organic carbon (TOC), permanganate index, copper, total nitrogen, total phosphorus etc. All water sample analysis was carried out on State Key Laboratory of Hydrology-water Resources and Hydraulic Engineering. Measuring methods and devices of water quality parameters used in this study are shown in table 2.

3. Results and discussion

3.1. Changes in hydraulic conductivity

In order to display where the most serious clogging occurred, the changes in hydraulic conductivity of the upper (10-15cm) and the lower column (15-55cm) were calculated respectively using Darcy’s law. Figure 3 showed that the decline of the permeability was obvious after 80 hours of continuous recharge. The hydraulic conductivity of the upper column (0-10cm) reduced from 1.23×10⁻⁴ to 9.84×10⁻⁵ m/s, while the lower one decreased from 1.23×10⁻⁴ to 1.06×10⁻⁴ m/s. During the first 56 hours, the hydraulic conductivity over the entire length of the column dropped steeply. with hydraulic conductivity of the upper column (10-15cm) and the lower (15-55 cm) decreasing from 1.23×10⁻⁴ m/s to 9.76×10⁻⁵ m/s and from 1.23×10⁻⁴ m/s to 1.05×10⁻⁴ m/s, respectively (Figure 3).
Figure 3. Decrease in hydraulic conductivity after passage of recharge water.

The hydraulic conductivity tended to be stable from the 56th hour to the 80th hour after the experiment began. Compared with the hydraulic conductivity decrease of the lower column (15-55cm), the hydraulic conductivity of the upper column was larger and quicker. Noteworthily, the hydraulic conductivity of the lower column increased slightly after the 56th hour, which could be caused by calcite dissolution in the sand column.

3.2. Clogging mechanism and water quality changes during recharge
Some important water quality parameters including SS, total bacteria count, DO, TOC, permanganate index, copper, total nitrogen, total phosphorus at different sampling places along the column were measured every 8 hours in this experiment. The results showed that the sand column had significant self-purification effect on the water quality parameters such as suspended matter, bacteria, TOC, copper, total nitrogen, total phosphorus etc, and that the concentration of inorganic anions changed inconspicuously in different sampling ports.

3.3. Physical clogging and changes in suspended matter
This research provided a good indication of physical clogging in the column to monitor the changes in suspended matter in the sampling places. The concentration of suspended matter at the 56th hour, changing with the sampling places (Figure 4), proved that the sand column had obvious removal effect on suspended matter mainly because of the effect of physical adsorption and filtration to the porous media. The concentration of suspended matter of the outlet sample decreased to less than 1mg/l in the first 48 hours. The removal effects were much stronger in the upper column, which showed that the main cause of the decrease of permeability was physical clogging, that is, suspended matter filled the void volume of the sand. The self-purification of the upper column was stronger, so the clogging was more severe, and the hydraulic conductivity decrement was larger. Figure 5 shows the suspended matter changed with time at 10cm water samples, which decreased gradually from 12mg/l to approximately 7mg/l between the 8th hour and the 32nd hour, and then started to increase gradually until the 24th hour. This result is consistent in the previous researches [2, 12], which demonstrated that suspended matter intercepted by porous media was beneficial to the absorption and filtration effect at first and that the filtration capacity of sand column decreased along with the porous in the sand column was filled gradually.
3.4. Biological clogging and changes in bacteria, TOC

For determining the possible biological clogging in the sand column, the total bacteria count in different sampling places along the column was quantified using plate counting method. Figure 6 shows that the total bacteria count decrease rapidly from the upper to the lower along the column and the highest number of bacteria was observed at 10cm sampling place. Consistent with previous research [13, 14], this result showed that the sand column had significant removal effect on microorganism during recharge and most of the bacterial cells adhere or grow at the upper column. The change of dissolved oxygen (DO) at different sampling places along the columns was a good indicator of bacterial activity within the columns. The average DO value of the sampling at 10 cm, 25 cm, 55 cm were presented in Figure 7, respectively. As shown in Figure 7, the DO of the upper column was much larger than the lower, so the bacteria at the upper column grows faster because of providing enough oxygen and the bacteria clogging was much more serious there. With the continued recharge, DO decreased gradually due to the oxygen consumption in the process of the organic compounds oxidative decomposition. TOC changing with the sampling places at the 56th hour showed that TOC decreased from the upper to the lower along the column, too (Figure 8). TOC of outlet and 10cm sampling place was 4.25mg/l and 8.45mg/l and the removal rate on TOC was about 67% and 35%, respectively, which demonstrated that the removal effect of the upper column (0-10cm) was the strongest.

Okubo T, Matsumoto J and Rubol S [7, 15] found that the main mechanisms of aquifer removing organic compounds were adsorption, chemical reaction and biodegradation that usually takes a relatively long time. The decrease of both organic compounds and DO in the column inhibited the growing and propagation speed of microorganism and leading to the weakening of biological clogging[16].

3.5. The removal effects on copper

Copper concentration was measured shortly after sampling by atomic absorption spectrophotometer (A320CRT). Atomic absorption spectrometer, also known as atomic absorption spectrophotometer, is used to analyze metal elements according to the effect of ground state atomic vapor on characteristic radiation absorption. It is sensitive and reliable for the determination of trace elements. Copper of outlet sample decreased to only 0.021mg/l at the 56th hour and the removal rate reached 98.5% (Figure 9). Copper of 10cm samples decreased sharply from 1.371 to 0.188mg/l in the first 8 hours, and then increased gradually to 1.222mg/l until the 24th hour (Figure 10). The reason for this phenomenon might be that the capacity for medium sand adopting copper was limited. Water in the column would be polluted again after adsorption saturation. Previous researches showed that the removal effect of aquifer on trace heavy metals was a complicated process, which was composed of dilution effect,
adsorption, convection and dispersion, and that the different aquifer medium had different adsorption capacity.

**Figure 6.** Total bacteria count changed with sampling places at the 56th hour.

**Figure 7.** DO changed with time.

**Figure 8.** TOC changed with sampling places at the 56th hour.

**Figure 9.** Copper concentration changed with sampling places.

**Figure 10.** Copper concentration changed with sampling places.
3.6. Changes in total nitrogen and total phosphorus
Nitrogen and phosphorus contamination of ground water, which typically results from leaching of fertilizer, animal waste, or human waste, become a global problem in recent years [17]. Total nitrogen and total phosphorus changes with time and sampling places were measured in this study. The result shown in Figure 11 indicated that the sand column had obvious removal effect on both of them, however, the removal effect of the upper column (0-10 cm) was stronger. The removal effect of the sand column on phosphorus was much larger than nitrogen and the removal rate was about 90%. Total nitrogen of outlet sample decreased to 5.8mg/l and the removal rate was about 30% (Figure 11).

Figure 11. Total nitrogen and total phosphorus changed with sampling places.

4. Conclusions and Recommendation

4.1. Conclusions
Physical clogging and biological clogging are two main process of aquifer clogging. To the colloidal clogging existed, and the pollutants easily adhere to the colloidal surface and block the pores of the aquifer. The two type of clogging cause the drop in hydraulic conductivity rapidly in the first 56 hours. With the extension of recharge, the suspended solids and bacteria retained by sand gradually decreased and organic carbon were consumed, therefore, the degree of clogging gradually decreased, and the hydraulic conductivity tended to be stable. The self-purification of the upper column was much stronger than that of the lower, so the clogging of former was more severe. This experiment did not analyze the possible chemical clogging in the sand column for the limited experimental conditions. The aquifer had significant removal effects on some water quality parameters such as suspended matter, bacteria, TOC, copper, total nitrogen, total phosphorus through complicated physical, chemical and biotic process. The concentration of inorganic anions in different water samples, however, had no obvious changes during the recharge. The removal effect of the sand column was limited, so groundwater would be polluted sustainingly when the concentration of the pollutants was high.

4.2. Recommendation
Future research will focus on the three types of clogging (which three types), and investigate the relationship between the time and influence degree for each clogging process. To do the comparison, we plan to use undisturbed soil samples from different sites of the flood plain to do the same experiment, furthermore, we also want to expand our research scale from laboratory scale to Liaolan groundwater funnel area of Qingdao in the future.
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