Re-evaluation and re-construction of water purification system using soil II. Removal of pollutants from infiltrating water


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Abstract
Presented in this paper are the results of a series of column experiments and a pilot scale test conducted to develop the treatment system of freshwater and wastewater by their percolation through soil and the other materials at the rate of a few metres per day. Materials that had high ability to sorb fulvic acid (FA hereafter) and phosphate ion were selected based on the results of the batch sorption tests and were processed into beads before subjecting them to the column test. The results of the column experiment suggested that the reagents added to increase the strength of the beads reduced the sorption of FA, thus causing the early breakthrough of the FA injected into the column. Less effect from the bead processing was observed for the removal of phosphate ion. Pilot scale test showed that the removal of particulate fraction of phosphorus differed among the materials used. The bead processing technique that can ensure the high water permeability while maintaining the sorption capacity of the material was required for further development of the treatment system.

Keywords
Column test; fulvic acid; soil filtration

Introduction
The aim of our study is to develop a treatment procedure for humic substances (HS) and phosphorus acid in wastewater and environmental water using soil and waste materials as a sorbent. In order to establish the optimal design and operation conditions for such water purification system, we investigated the removal of the pollutants from infiltrating water.

Our work was motivated by the apparent increase in COD relative to BOD in Lake Biwa, Shiga, Japan, in spite of the increasing numbers of sewage treatment plants and the improvement in the wastewater treatment technology. COD there probably consists of recalcitrant organic material such as HS which are the source of disinfection by-products. Because the drinking water in the district depends mostly on the water from Lake Biwa, development of the technology to remove HS and phosphorus from environmental water was considered to be important for the conservation of the water resources there.

In the present study, a series of column experiment and a pilot scale test was conducted to develop and verify the water treatment system using soil. In our preceding report (Fujikawa et al., 2003), sorption of fulvic acid (a form of HS) and phosphate ion to various soil, sludge, mineral and organic materials was tested by the batch experiment to find promising materials to be used for the system. The hydraulically static condition of the batch experiment, however, is different from that in the actual system where pollutants are
removed from the infiltrating water. The column experiment, on the other hand, is conducted under hydraulically dynamic condition and is more similar to the actual water purification system. Verification of the performance of the materials selected by the batch experiment through the column experiment, therefore, was considered necessary.

**Materials and methods**

**Processing of materials**
Among the materials that showed significant sorption of FA and phosphate ion in the batch sorption test, materials were selected for the column experiment based on the cost and marketability. The materials chosen were Andosols (Santo, Shiga), Akadama soil (Kanuma, Tochigi), sludge from a water treatment plant (WTP) in Shiga, limonite, a subsoil rich in goethite mineral (Akamizu, Kumamoto), red sand (Shigaraki, Shiga), and lake sediment (Akanoi Bay, Lake Biwa, Shiga), collected in Japan. In order to maintain the water permeability of the column during the experiment, materials were processed into beads (aggregates) by adding organic material as a binder and using a machine developed by Hitachi Zosen Corp. (Japan).

**Column test**
The column used for the experiment was made from a 5.0 cm diameter acrylic resin pipe, and was 25 cm in length packed with soil or sludge beads. The inlet was polyvinyl chloride funnel fitted to the flange at the end of the column. The filtered river water was pumped into the column at the flow rate of 2.0 mL/min from the lower end and the effluent was collected at the upper end of the column. FA solution (diluted with filtered river water to c. a. 165 mg/L as dissolved organic carbon) and potassium dihydrogenphosphate (diluted with filtered river water to 20 mg/L as total phosphorus) was injected into the column for a short period of time. The effluent was analyzed for DOC, pH, EC and total phosphorus (T-P) to obtain the breakthrough curve. Breakthrough curve of chloride in each column was also obtained.

The retardation of the FA and phosphorus acid due to its sorption to the solid phase was evaluated utilizing an experimental retardation factor \( R \) defined as follows:

\[
R = \frac{T_{FAP}}{T_{Cl}}
\]

where \( T_{FAP} \) is the time from the injection of FA (or phosphorous acid) to the occurrence of maximum DOC (or T-P) concentration in the effluent at the column outlet (i.e., peak of a breakthrough curve), and \( T_{Cl} \) is the time from the injection of chloride ion to the occurrence of maximum chloride concentration in the effluent at the column outlet.

Batch sorption experiment and sorption isotherm experiment was conducted as a follow-up study using the materials processed into beads. The method of the batch test is stated in Fujikawa et al. (2003).

**Pilot scale test**
The monitoring of pilot-scale river water purification system using materials selected based on the results of the column experiment was conducted.

The pilot-scale treatment system was constructed using four different combinations of soil and sludge ((1) Akadama soil, (2) limonite combined with sludge from the WTP in Shiga, (3) sludge from the WTP, and (4) red sand). The influent was river water (Hayama River; flowing into Lake Biwa, Shiga, Japan) after removal of coarse suspended particles in a sedimentation pond. Both the influent and the percolate from the soil layer were analyzed for T-P, and dissolved (DT-P) and particulate (P-P) forms of phosphorus.
Results and discussion

Column test

The results of the column experiment are shown as the experimental retardation factor $R$ plotted versus $K_d$ obtained for the original material in the batch experiment 1 hr after the addition of the solute (Figure 1). Theoretically, $K_d$ of the batch sorption experiment should have linear relationship with $R$ but the actual correlation was low especially for FA. Namely, limonite, which consists mostly from goethite, showed a relatively significant retardation of FA while Andosols (Santo, Shiga), for which also significant sorption of FA was observed in the batch experiment, showed very little retardation of FA loaded to the column.

The sorption isotherm experiment conducted later for Andosols (Santo, Shiga) and limonite beads showed that the $K_d$ values of the materials processed into beads were decreased compared to the original material for sorption of FA (Figure 2). The reason that limonite showed some retardation of FA in the column test while Andosols in the column experiment showed no retardation at all is understandable from the $K_d$ values obtained for the soil beads (13.7 mL/g for limonite beads while 0 for beads of Andosols).

The probable reason that the sorption of fulvic acid was much affected by processing of the materials into beads was the addition of organic binder. The addition of the binder resulted in significant leaching of the organic carbon from the solid phase especially for Andosols (20 mg/L in DOC without processing while 150 mg/L DOC after processing, at the solid to liquid ration of 1 to 2.5). Competition between the added FA and the leached components of the binder for sorption sites could have reduced the sorption of FA. Batch sorption test showed that some reduction also occurred in the case of sorption of phosphate ion after the processing.

The measurement of the apparent $K_d$ values was also done for Andosols from Mt. Sanbe, Tottori, that was processed into beads by different techniques ((1) no processing, (2)

![Figure 1](https://iwaponline.com/wst/article-pdf/50/5/369/419983/369.pdf)

**Figure 1** Apparent $K_d$ of each materials plotted versus experimental retardation factor of column test (retardation factor of $PO_4^{3-}$ for Andosols is higher than 350 as the peak of the breakthrough curve was not observed during the experiment)

![Figure 2](https://iwaponline.com/wst/article-pdf/50/5/369/419983/369.pdf)

**Figure 2** Sorption isotherm of FA for processed and unprocessed materials: (a) Limonite, (b) Andosols (Santo, Shiga)
slightly heated but without adding organic binder, (3) adding organic binder). While the $K_d$ value of FA was decreased from 21.7 (mL/g) to 10.5 (mL/g) by heating but without adding organic binder, a more significant decrease in $K_d$ (0.7 mL/g) was observed by processing with the organic binder.

**Pilot scale test**

Results of the monitoring are shown in Table 1 as removal ratio (in percentage) of T-P, DT-P and P-P from influent river water. Apparently removal of T-P (ca. 70%) was not much different among the four materials, but the removal of P-P and DT-P differed significantly depending on the material used. Namely, while the WTP sludge had higher removal ratio of dissolved phosphorus (ca. 90%), its ability to remove particulate phosphorus was lower than Akadama soil and red sand. Because filtration of SS also plays an important role in water purification using soil, more effort is required on the development of a simple experimental procedure to evaluate the filtration ability of materials. The removal of the HS from the river water was not monitored in the present test because of the leaching of organic materials from the soil during the test period.

**Conclusions**

A series of column experiment and a pilot scale test was conducted to verify the removal of FA and phosphorus acid from freshwater and wastewater by percolation of the water through soil and other materials. The removal of FA in the column test was much affected by the reduction in sorption induced probably by the binder used to process the original materials into beads. In the pilot scale experiment, both the filtration of particulate form of phosphorus and sorption of dissolved phosphorus were shown to differ between the materials. The development of the bead processing technique that can maintain the original sorption capacity of the materials while ensuring the water permeability is now under investigation.

**References**


### Table 1

Removal (in %) of phosphorus from river water by various soil (average of 5 week monitoring)

<table>
<thead>
<tr>
<th>Forms of phosphorus</th>
<th>(1) Akadama soil</th>
<th>(2) Limonite combined with sludge from WTP</th>
<th>(3) Sludge from WTP</th>
<th>(4) Red sand</th>
</tr>
</thead>
<tbody>
<tr>
<td>T-P</td>
<td>71 ± 10</td>
<td>70 ± 5</td>
<td>73 ± 4</td>
<td>74 ± 13</td>
</tr>
<tr>
<td>P-P</td>
<td>74 ± 12</td>
<td>65 ± 8</td>
<td>66 ± 8</td>
<td>71 ± 18</td>
</tr>
<tr>
<td>DT-P</td>
<td>63 ± 6</td>
<td>82 ± 12</td>
<td>91 ± 13</td>
<td>81 ± 11</td>
</tr>
</tbody>
</table>