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# Identification of priority organic compounds in groundwater recharge of China

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## HIGHLIGHTS

- We developed a comprehensive ranking system to identify priority organic compounds.
- We developed two different ranking lists of organic compounds.
- 151 OCs were selected as the candidate organic compounds and ranked.
- Nonylphenol, erythromycin and ibuprofen were the highest priority OCs.

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ABSTRACT

Groundwater recharge using reclaimed water is considered a promising method to alleviate groundwater depletion, especially in arid areas. Traditional water treatment systems are inefficient to remove all the types of contaminants that would pose risks to groundwater, so it is crucial to establish a priority list of organic compounds (OCs) that deserve the preferential treatment. In this study, a comprehensive ranking system was developed to determine the list and then applied to China. 151 OCs, for which occurrence data in the wastewater treatment plants were available, were selected as candidate OCs. Based on their occurrence, exposure potential and ecological effects, two different rankings of OCs were established respectively for groundwater recharge by surface infiltration and direct aquifer injection. Thirty-four OCs were regarded as having no risks while the remaining 117 OCs were divided into three groups: high, moderate and low priority OCs. Regardless of the recharge way, nonylphenol, erythromycin and ibuprofen were the highest priority OCs; their removal should be prioritized. Also the database should be updated as detecting technology is developed.



## 1. Introduction

Groundwater recharge using reclaimed water has been rapidly developed around the world in order to replenish decreasing groundwater resources and declining water table. Groundwater recharge can be accomplished by various methods: surface spreading, soil aquifer treatment system, vadose zone injection and direct injection (U S Environmental Protection Agency, 2004). These methods can be categorized as either surface percolation or direct aquifer injection. In China, during 2008 there was $3000 \times 10^4$ t reclaimed water to be recharged. It is, however, difficult to remove all the contaminants in reclaimed water completely (Calderon-Preciado et al., 2011; Matamoros and Salvado, 2012), meaning that some of them such as endocrine disrupting chemicals (EDCs), pharmaceuticals, perfluorochemicals (PFCs) and antibiotics (Karthikeyan and Meyer, 2006; Al-Khashman, 2009; Teijon et al., 2010; Karnjanapiboonwong et al., 2011) were introduced into groundwater, thereby posing risks to groundwater and humans. Different pollutants would impose different degrees of risk to groundwater due to their varied behaviors and fates during recharge. For instance, those pollutants which can be removed by adsorption and degradation, may not pose risks to the safety of the groundwater environment even if the concentration is high in reclaimed water. However, other pollutants that are present at low concentrations in reclaimed water and that do not undergo transformations on entering the aquifer, may also pose greater risk to groundwater (Zhang et al., 2011; Debroux et al., 2012; Lapworth et al., 2012). For these reasons rather priority pollutants should deserve our first concern in wastewater treatment plants (WWTPs) that are supplying reclaimed water to groundwater.

Several researchers have carried out evaluations using various methodologies, of which three methodologies were most commonly adopted for screening. The most popular method is a comprehensive scoring system, for example, Kumar and Xagoraraki (2010) provided a ranking list of 100 OCs in surface water and finished drinking water

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by using this method. The other two most popular methods were step by step screening based on multi-criteria (Boxall et al., 2003; Besse et al., 2008; Eriksson et al., 2008; Perazzolo et al., 2010; Jean et al., 2012) and mathematical simulations (Jonsson et al., 1989; Munoz et al., 2008; Voigt and Bruggemann, 2008). In those studies, the target environment media were surface water (Mitchell et al., 2002; Sanderson et al., 2004; Arnot and Mackay, 2008; Besse et al., 2008; Besse and Garric, 2008; Kumar and Xagoraraki, 2010; Perazzolo et al., 2010; Sui et al., 2012), drinking water (Kumar and Xagoraraki, 2010; Schriks et al., 2010), sludge (Eriksson et al., 2008) and soil (Jeong and An, 2012). However to the best of our knowledge studies on screening for priority chemical substances in groundwater have rarely been reported.

Also, due to the difference of usage amounts, environmental conditions and levels of treatment technology, priority contaminants vary by country. In China only one study (Sui et al., 2012) had focused on priority pollutant screening in the water environment. In that study, 39 pharmaceuticals were ranked based on consumption, removal performance in WWTPs and potential ecological effects. Seventeen pharmaceuticals were screened out as priority pollutants.

In this study, we developed a ranking system based on OCs' occurrence in either reclaimed water or WWTP effluent of China, exposure potential and potential ecological effects. This approach was applied to groundwater recharge using reclaimed water by surface percolation and direct aquifer injection in China.

## 2. Methods

### 2.1. Ranking system

#### 2.1.1. Criteria

The ranking of OCs in this study was based on the overall scores of three different criteria: occurrence in the reclaimed water or WWTP effluent of China, exposure potential and ecological effects.

Table 1 presents the information about different attributes of multiple criteria. The "occurrence" (O) is represented by the "prevalence" attribute (O1) and "magnitude" attribute (O2) of an OC. The two attributes are represented as the frequency of detection and the concentration of an OC in reclaimed water or WWTP effluent, respectively. Because the amount of infiltration is mostly affected by adsorption and degradation of pollutants during recharge, the second criterion "exposure potential" (P) is represented by two attributes: (1) persistence (P1) and (2) transportability (P2) of an OC. Assuming that groundwater was extracted from the recharge site, when OCs in groundwater recharged by direct aquifer injection were ranked, the criterion "exposure potential" does not undergo treatment, as pollutants in groundwater recharged by this method aren't adsorbed or degraded in vadose zone. The persistence and transportability of an OC are represented by its degradation half-life and soil organic carbon adsorption coefficients ($K_{OC}$), respectively. The attribute "persistence" only considers biological effect. The third criterion "ecological effects" contains two factors: bioaccumulation (E1) and eco-toxicity (E2). The octanol/water partitioning coefficient ($K_{OW}$), which indicates the lipophilicity of OCs, is used to estimate bioaccumulation, as it has been correlated with bioconcentration factor for different compounds (Schriks et al., 2010). The eco-toxicity is estimated by the lethal concentrations for 50% kill ($LC_{50}$) of the aquatic indicator species (fish, daphnid and green algae which represent three trophic levels) of an OC (He et al., 2014). And in this study, only acute toxicity is considered.

#### 2.1.2. Scoring

For different criteria and attributes, different utility functions were applied (Table 1), some of which were modified from the utility functions used by Kumar and Xagoraraki (2010).

The score of one criterion was calculated using Eq. (1), where $S_i$ was the score of the corresponding criterion, $W_{i,j}$ was the importance weight of each component, and $U_j$ was the value obtained from corresponding utility function of the component. For the three criteria "occurrence", "exposure potential" and "ecological effects", two components were involved and considered to be equally important.

$$S_i = \sum_{j=1}^{n} U_j \times W_{i,j}. \quad (1)$$

The overall score of an OC was calculated using Eq. (2), where $S_{\text{overall}}$ represented the overall score of the OC, and $W_i$ represented the importance weight of each criterion. Similarly, to avoid any judgment bias all the criteria were also considered equally important, so $W_i$ was assigned a value of 1/3 for groundwater recharge through surface percolation. The "exposure potential" criterion is not applicable for groundwater recharge by direct aquifer, so the value of $W_i$ was set as 1/2 for the other two criteria.

$$S_{\text{overall}} = \sum_{i=1}^{3} S_i \times W_i. \quad (2)$$

The illustration of the scoring for one OC (erythromycin) was shown in Table A.1.

### 2.2. Data collection

All the data collected are presented in Table A.2.

**Table 1**
Criteria, attributes and corresponding utility functions used to prioritize OCs in groundwater recharge.

| Criteria | Attributes | Utility functions |
|---|---|---|
| Occurrence (O) | Prevalence(O1)(%) | $U(O1) = \max(f/100)_i$, where $f$ represents frequency of detection of an$i$th chemical in water. |
| | Magnitude(O2)(ng/L) | $U(O2) = (C - C_{\min}) / (C_{\max} - C_{\min})_i$, where $C$ represents concentration of an$i$th chemical in reclaimed water or WWTPs effluent, $C_{\max}$ and $C_{\min}$ represent maximum and minimum concentration values, obtained from the overall list of OCs considered. |
| Exposure potential(P) | Persistence (P1)(1) | $U(P1) = 1$, if $t_{1/2} < 0$(A), "0.8" for category "B" (0–1), "0.6" for category "C"(1–2), "0.4" for category "D"(2–3), and "0.2" for category "X" (>3). |
| | Transportability (P2)(l/mg) | U(P2), which is equals to "1" for category "A" (1–2), "0.8" for category "B"(2–3), "0.6" for category "C"(3–4), "0.4" for category "D" (4–5), and "0.2" for category "X" (>5). |
| Ecological effects(E) | Bioaccumulation (E1)(1) | $U(E1) = 1$, if $\log K_{OW} > 3$;<br>$U(E1) = 0$, if $\log K_{OW} < 3$[a]. |
| | Ecotoxicity (E2) (mg/L) | $U(E2) = \sum_{k=1}^{3} \frac{1}{3} * U(E2)_k$;<br>$U(E2)_k = 1 - \frac{(LC_{50})_i - (LC_{50})_{\min}}{(LC_{50})_{\max} - (LC_{50})_{\min}}$. |

Note: A: lower exposure potential; B: low exposure potential; C: moderate exposure potential; D: high exposure potential; and X: higher exposure potential.
[a] If the pollutant has log $K_{OW}$of more than 3.0, it was indicated to be potentially bioaccumulated (Perazzolo et al., 2010; Schriks et al., 2010).