


## Safe water reuse through a quasi-natural water cycle

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### ABSTRACT

Safe water reuse usually means the provision of reclaimed water with its quality meeting the criteria for safeguarding human health and aquatic ecology. Regarding this, people often overstress the technological magic in terms of hazardous substances removal by engineered processes while ignoring the utilization of nature's power to provide an additional barrier for safety control. Based on an understanding of the function of the hydrological cycle to secure water quality and the fact that water reuse is through a water loop, the author presented his viewpoint on creating a quasi-natural water cycle for water reuse. Existing data showed the remarkable effects of emerging pollutants removal and associated reduction of biotoxicities from the reclaimed water during open-storage under a quasi-natural condition. Adsorption by soil particles, sunlight-induced photolysis and purification by aquatic life were thought to be the major actions contributing to effective water quality improvement in a quasi-natural manner. Safe water reuse through a quasi-natural water cycle can thus be a recommendable strategy for system design in accordance with the purposes of water use.

**Key words:** biotoxicity, emerging pollutant, natural purification, quasi-natural, water cycle

### HIGHLIGHTS

- A quasi-natural water cycle is proposed for securing reclaimed water quality.
- Natural actions contribute much to emerging pollutants removal and biotoxicity reduction.
- System design with integration of engineered and natural elements is recommendable.

### INTRODUCTION

Safe water reuse usually means a secured provision of the reclaimed water with quality not only meeting the standards put forward by authority agencies but also confirming the requirements for human health and ecological risk control. There are, therefore, two categories of water quality criteria: legislative criteria in accordance with the societal needs for various purposes of water reuse, and scientific criteria in accordance with the targets of hazardous effects control. These criteria can be within a unified framework if sufficient scientific information has been available to support the legislation when it is enacted. However, this may not always be the case because other factors related to technological and/or economic feasibility may also constrain the legislation. In addition to this, new scientific information may be continuously obtained on the hazardous effects of emerging pollutants newly identified. Consequently, there are always long-lasting discussions on how safe water reuse can be secured and to what level the reclaimed water quality criteria can be set (Paranychianakis *et al.* 2015).

On the other hand, wide attention has also been paid to the processes employed for reclaimed water production. The rationale for this is that, to meet the quality requirement based on any criteria, we have to select the most feasible treatment process or a system combining several processes for effective removal of the target pollutants. It is known that by conventional secondary treatment, most of the suspended solids, bulk organic matter in terms of chemical oxygen demand (COD) or biological oxygen demand (BOD), and part of the nutrients (in terms of total nitrogen and phosphorus) can be effectively removed from domestic wastewater which is the most common source for reclaimed water production. Furthermore, for the removal of emerging pollutants that may result in significant health and ecological hazards, an array of advanced

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treatment processes may need to be employed. We fully understand that adsorption using activated carbon or equivalent adsorbents can cope with most hydrophobic substances, advanced oxidation such as ozonation catalyzed by UV and/or  $H_2O_2$  can destroy refractory organics, and by membrane filtration including nanofiltration and osmosis, the pollutants with dimensions down to ion scale can be effectively removed (Angelakis *et al.* 2018). It is true that if the economic factor is ignored, we can achieve safe water reuse in accordance with any of the strictest water quality criteria. A number of successful cases of direct potable reuse have already provided strong evidence (Keller *et al.* 2022).

However, to meet the needs of water reuse in the whole world covering the developed and developing regions for various purposes, we may need to seriously consider another factor that should have been realized for long but seldom purposely practiced in planning water reuse. In a number of studies, researchers pointed out that water reuse is not a new technique or concept, and ‘unplanned reuse’ or ‘de facto reuse’ has long been practiced in almost all river basins over the world (Rice *et al.* 2016; Švecová *et al.* 2021). As a matter of fact, people living downstream of a long river seldom worry about whether they are drinking part of the ‘urine’ discharged from upstream because though various pollutants may enter the river from somewhere, the water is believed to be as ‘clean’ as it looks. In fact, it is true from a scientific viewpoint that a flowing stream has the capability of ‘self-purification’ which provides a natural barrier for safeguarding the quality of the water source (Ngraha *et al.* 2020).

With this in mind, the author would like to present his viewpoint on the utilization of the natural function of a so-called ‘quasi-natural water cycle’ which may be woven into a reclaimed water production and supply system to assist in safe water reuse.

## WATER CYCLE AND ITS FUNCTIONS

### The hydrological cycle

The hydrological cycle is a water cycle up to a global scale involving the continuous circulation of water in the Earth–atmosphere system. The most important processes involved in this cycle are evaporation, transpiration, condensation, precipitation, and runoff. The hydrological cycle has two distinctive functions. One is to secure a dynamically constant quantity of water in each of the hydrological elements (e.g. water bodies) over the world, such as rivers, lakes, and groundwater aquifers, not to mention the vast oceans which are viewed as the original ‘source’ where a water circulation starts by evaporation, and the ultimate ‘sink’ where a water circulation ends; another is to secure the water quality in each waterbody through a series of physical, physicochemical, chemical, and biological actions that naturally occur in the whole water cycle. Since the hydrological cycle is solely driven by solar energy and gravitational energy, and can spontaneously maintain a dynamic equilibrium condition without any unnatural interference, it is virtually a thermodynamically sound system (Wang & Luo 2021).

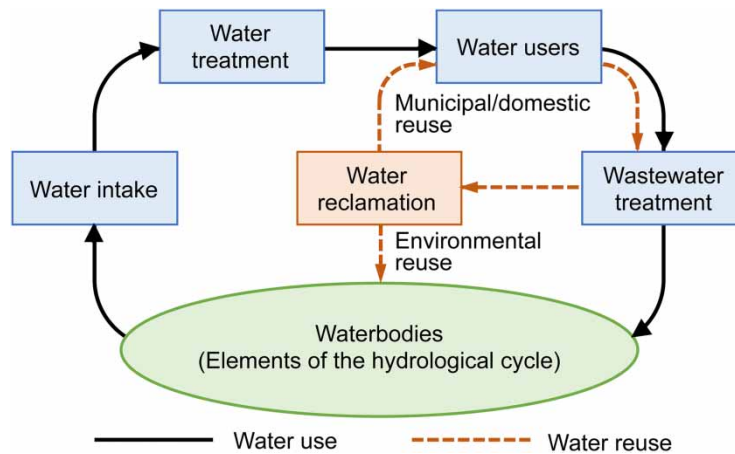
### Local water cycle

Human beings have long depended on the hydrological cycle to obtain source water for supply and consumption. As water molecules cannot be created nor disappear under natural conditions, no matter how water is consumed, it ultimately returns to the hydrological cycle. In this sense, any kind of water use can be viewed as a circulation of water through a ‘loop’ with its starting and ending points attached to the hydrological cycle. Such loops for water use can be of different scales and complexities depending on the requirement for water supply. In most cases, artificial or engineered systems are built for such kinds of water circulation (Figure 1).

In Figure 1, the solid line depicts a local water cycle that starts from water intake (from waterbodies), through water treatment for supply to users, then treatment of collected wastewater, and finally discharge back to waterbodies. When water reclamation and reuse are practiced, another smaller water cycle is added (dotted line in Figure 1). In this framework, the quality of the source water is mainly secured within the natural hydrological cycle, while artificial means are provided through engineered facilities for water quality conversion to meet the requirement of water supply (by water treatment) and that of effluent discharge (by wastewater treatment). Water reclamation for reuse purposes may also depend on more advanced treatment for further water quality improvement (Ma *et al.* 2019).

### Consideration on a quasi-natural water cycle

According to the purpose of water quality conversion, the engineered treatment processes can be classified to the following three categories. The first is drinking water treatment for the removal of impurities, that may be hazardous to human health, from source water; the second is wastewater or sewage treatment for the removal of excessive pollutants, that may damage the water quality of the receiving waterbody, from the used water to be discharged; and the third is the treatment for water



**Figure 1** | Local water cycles for water use and reuse.

reclamation discussed herein. To a certain extent, the treatment for water reclamation can be viewed as ‘wastewater treatment’ + ‘drinking water treatment’ in terms of the types of treatment processes employed. The common objective of the treatments in all these categories can be thought of as meeting the ‘gaps’ between the required water quality and that of the original water (Figure 2). Conceptually, the treatment for water reclamation is to meet the greatest quality gap, because the original water to be treated is the wastewater or sewage.

If we consider again the de facto water reuse which can be viewed as a quality conversion from the discharged effluent (upstream) to natural water (downstream) through natural actions (including dilution), then we may conceptually depict a gap between the effluent quality and the natural water quality as that circled in Figure 2 to indicate the utilizable natural action for water purification.

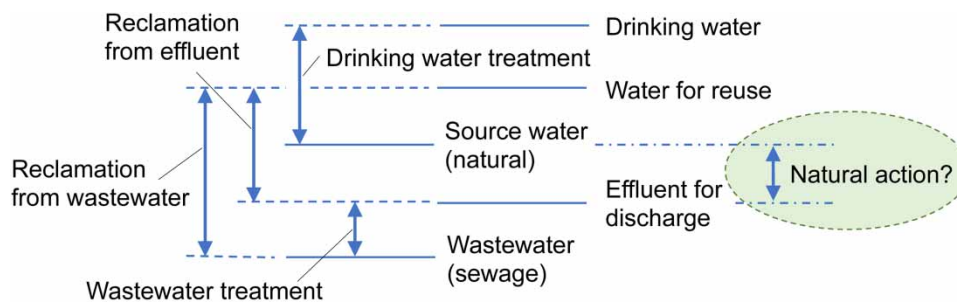
This is thus the author’s consideration on the configuration of a quasi-natural water cycle where natural elements are purposely introduced to the water reclamation and reuse system.

## EFFECTS OF QUASI-NATURAL ELEMENTS IN A WATER CYCLE TO IMPROVE WATER QUALITY FOR SAFE WATER REUSE

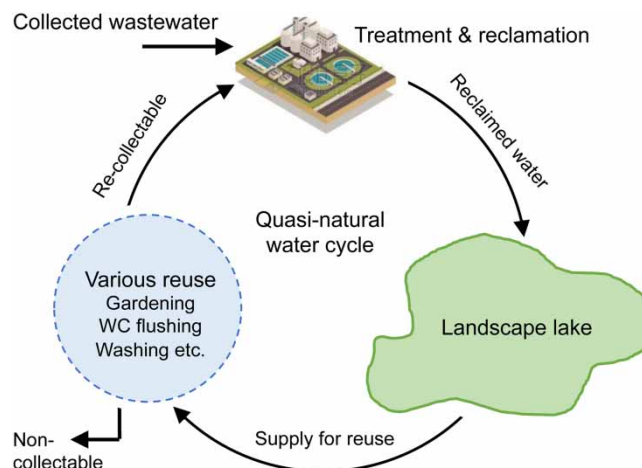
Figure 3 is one example of the so-called quasi-natural water cycle formulated for water reclamation and non-potable reuse (Wang *et al.* 2015). The landscape lake introduced into such a system is the most important hydrological element of quasi-natural feature, which performs the functions of the reclaimed water reservoir, and a buffer zone between reclaimed water supply and ultimate use, in addition to its primary role of aquatic landscaping using the reclaimed water source.

### Trace pollutants removal through the landscape lake

For safe water reuse, there are wide concerns regarding the health and ecological risk of exposure to trace pollutants that may not be easily removed by treatment processes commonly applied for water reclamation, compared with conventional



**Figure 2** | Water quality gap to be met by various treatments (gaps are arbitrarily plotted but not to quantify the real quality differences).



**Figure 3** | An example of water reclamation and reuse through a quasi-natural water cycle (location: Xi'an, China).

pollutants such as bulk organic matter, nutrients, etc. Table 1 summarizes the occurrence of a series of trace pollutants in the wastewater, reclaimed water, and the lake water (Ma *et al.* 2016).

Of the four categories of trace pollutants, pharmaceuticals and personal care products (PPCPs) are believed to be solely from wastewater rather than any other nonpoint source. Using PPCPs as an index to briefly assess the efficiency of trace pollutants removal, it can be seen that many chemicals in this category may not be easily removable by biological processes (membrane bioreactor in the referred case, average PPCPs removal evaluated as about 57%). However, after open storage in the landscape lake (depth as 0.8–1.0 m, hydraulic retention time as about 5 days), about 86% PPCPs removal has been achieved with regard to the average concentration in the reclaimed water while the overall PPCPs removal with the average concentration in the wastewater becomes as high as about 94%.

### Associated biotoxicity reduction

Biotoxicity evaluation by bioassays can assist in a general assessment of the possible hazardous effects of chemicals (including known and unknown ones) existing in water on aquatic life and human health. Table 2 shows the results of an array of bioassays regarding the reclaimed water and lake water and indicates the remarkable action of the landscape lake, which is equivalent to open storage of the reclaimed water under natural conditions, for a substantial reduction of the biological toxicity from the residual hazardous substances (Ma *et al.* 2018).

By estimating the hazard quotient (HQ), which is the ratio of the potential exposure to a substance and the level at which no adverse effects are expected, it has been noticed that of all the typical trace pollutants detected from the reclaimed water,

**Table 1** | Occurrence of typical trace pollutants at different locations

| Category   | Number of chemicals detected | Detected concentration (ng/L in total) |                               |                               |
|------------|------------------------------|--|-------------------------------|-------------------------------|
|            |                              | Wastewater                             | Reclaimed water               | Lake water                    |
| PPCPs      | 26                           | $1.0\text{--}2.4 \times 10^5$          | $0.1\text{--}1.0 \times 10^3$ | $0.3\text{--}1.3 \times 10^2$ |
| Phenols    | 16                           | $3.3\text{--}6.5 \times 10^3$          | $0.1\text{--}1.8 \times 10^2$ | $0.6\text{--}1.6 \times 10^2$ |
| Pesticides | 3                            | $0.8\text{--}8.7 \times 10^1$          | $0.4\text{--}1.2 \times 10^1$ | $2.4\text{--}2.7 \times 10^0$ |
| PAHs       | 13                           | $1.3\text{--}3.2 \times 10^1$          | $1.6\text{--}2.7 \times 10^1$ | $2.6\text{--}4.6 \times 10^1$ |

Notes: (1) The lake mainly for landscaping is operated under a condition shown in Figure 3, where water quality is maintained by continuous replenishment using reclaimed water (average hydraulic retention time as about 5 days). (2) Samples were collected during a 7-month period after the whole system had been put into continuous operation for 5 years. (3) PPCPs detected mainly include tetracycline, sulfapyridine, sulfamethoxazole, sulfadiazine, roxithromycin, oxytetracycline, ofloxacin, norfloxacin, lamotrigine, cotinine, cimetidine, etc. (4) Phenols detected mainly include o-phenylphenol, 2-Nitrophenol, 4-methylphenol, 3-methylphenol, 2,4-dimethylphenol, 4-chloro-3-methylphenol, 4-chloro-3,5-dimethylphenol, bisphenol A, etc. (5) Pesticides detected include dichlorophos, chlorpyrifos, and atrazine. (6) PAHs detected include pyrene, phenanthrene, fluorene, fluoranthene, chrysene, anthracene, acenaphthylene, acenaphthene, etc.

**Table 2** | Biototoxicity comparison of the reclaimed water and lake water

| Bioassay        | Luminescent bacteria   |                            | Alga <i>Chlorella vulgaris</i> | Larva zebrafish | SOS/Umu              |
|-----------------|------------------------|----------------------------|--------------------------------|-----------------|----------------------|
|                 | <i>Vibrio fischeri</i> | <i>Vibrio qinghaiensis</i> |                                |                 |                      |
| Index           | TEQ <sub>phenol</sub>  | TEQ <sub>phenol</sub>      | Inhibition                     | Mortality       | TEQ <sub>4-NQO</sub> |
| Reclaimed water | 7.1 ± 1.1 mg/L         | 18.9 ± 0.8 mg/L            | 19.1 ± 0.4%                    | 14.6 ± 2.1%     | 4.6 ± 0.1 µg/L       |
| Lake water      | 4.1 ± 0.5 mg/L         | 6.3 ± 0.4 mg/L             | 8.9 ± 1.0%                     | ND              | 0.5 ± 0.1 µg/L       |

Notes: (1) Samples were collected during a 7-month period after the whole system (shown in Figure 3) had been put into continuous operation for 5 years. (2) TEQ: toxic equivalent concentration with phenol as a reference compound for luminescent bacteria assays and 4-nitroquinoline-N-oxide (4-NQO) as a reference compound for the SOS/Umu assay.

PPCPs account for 92–97% of the total HQs, and their significant reduction after storage in the lake (about one-order reduction as shown in Table 1) may have contributed much to biototoxicity control.

## NATURAL ACTIONS CONTRIBUTED TO TRACE POLLUTANTS REMOVAL AND BIOTOXICITY REDUCTION

### Adsorption by soil particles

Adsorption of various pollutants by soil particles contained in the water is believed to be the most important for trace pollutants removal. Fine soil particles of colloidal dimension ( $<10^{-1}\mu\text{m}$ ) are usually suspended in bulk water, and larger soil particles ( $10^1\text{--}10^2\mu\text{m}$ ) may be scored from the bottom sediment to bulk water by wind current. They provide considerable surface areas to perform adsorption. The decisive factors for the adsorptive removal are usually the organic carbon-water partition coefficient ( $K_{oc}$ ) and octanol-water partition coefficient ( $K_{ow}$ ) of the organic pollutant. It has been found that among PPCPs, those with  $\log K_{oc} < 2.2$  and  $\log K_{ow} > 2.7$  tend to be more removable by adsorption (Ma *et al.* 2018).

### Sunlight-induced photolysis

By radiation under sunlight, direct photolysis and/or other light-mediated reactions may also contribute much to the decomposition of organic pollutants. It has been identified that sunlight exposure has stimulated the generation of photochemically reactive intermediates such as singlet oxygen, and triplet-excited hydroxyl radicals from dissolved organic matter (DOM) in water. Both the chromophoric DOM and fluorescent DOM tend to be much more easily removable in the summer season when sufficient sunlight radiation is received (Wang *et al.* 2021a, 2021b).

### Purification by aquatic life

As surface water can provide primary habitats for various aquatic life including plants and animals, these life forms may assist the pollutant decomposition and/or assimilation in their lifecycle. It has been revealed that the growth of the *Acorus calamus* (a flowering plant growing at the edges of waters and wetlands) may effectively enhance the removal of emerging pollutants, such as sulfamethoxazole, from water (Zheng *et al.* 2022).

### Other actions

A waterbody under a quasi-natural condition in fact has a wide range of self-purification abilities through other physical, chemical and biological processes. For example, although most pollutants residual in the reclaimed water are believed to be refractory to biological degradation, during longer hydraulic retention (e.g. several days), some organic chemicals with biodegradation constant  $K_{biol} > 10 \text{ L/g}_{SS}/\text{day}$  can still be effectively degraded (Ma *et al.* 2018).

## QUASI-NATURAL ELEMENTS IN THE WATER CYCLE FOR WATER REUSE

Although there are successful cases of planned potable reuse (indirect or direct augmentation of drinking water supplies), unplanned potable reuse (addition of reclaimed water to drinking water sources), and groundwater recharge, using reclaimed water for recreational and environmental purposes is gaining popularity nowadays in many countries and regions. In this case, the reclaimed water is to be used for replenishing landscaping lakes or ponds or augmenting streamflow. Such kinds of surface waters are not only the end-users of the reclaimed water but also the important hydrological elements which can be integrated into the so-called quasi-natural water cycle discussed herein. Such a water cycle may be primarily

formulated for facilitating water reclamation, storage of the reclaimed water, and its multifunctional use for various reuse purposes (environmental, municipal, and/or domestic) rather than specially designed to target pollutants removal. However, it consequently results in remarkable water quality improvement as indicated by trace pollutants removal and associated biotoxicity reduction. All these are due to the introduction of quasi-natural elements where a series of natural actions are performed.

Even for the piped supply of reclaimed water for other non-potable urban uses, it may also be recommendable that a quasi-natural open-storage tank or reservoir be introduced in between reclaimed water production and supply for both quantitative regulation and qualitative stabilization.

## CONCLUDING REMARKS

This article presented the author's viewpoint on safe water reuse through a quasi-natural water cycle. In terms of water treatment, we cannot deny the fact that almost all the technologies widely used by us can find their prototypes in nature. In other words, human beings have learned a lot from nature in developing water treatment technologies. Unfortunately, when we discuss and practice water reuse, we often ignore nature's example of water quality purification through the hydrological cycle but put too much stress on technological magic. Therefore, the author would like to call for full consideration of the utilization of nature's power as far as possible through a sophisticated system, such as the quasi-natural water cycle proposed herein.

## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

## CONFLICT OF INTEREST

The authors declare there is no conflict.

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