

Potable reuse: Which chemicals to be concerned about

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Abstract

The growth of potable reuse as a planned water supply strategy has led to an increased focus on the presence and significance of trace chemical contaminants. By the use of reclaimed wastewater as a source, potable reuse projects require serious consideration to be given to the range and character of chemicals which may be present and may pose unacceptable risks to public health if not properly managed. The first step, required to assess and manage risks, is to consider the range of chemical contaminants which may be present. Although it is impossible to derive an exhaustive list, it is useful to consider broad categories and the specific types of challenges that may be posed by the chemicals within those categories. Such a broad categorisation of chemical contaminants is presented in this review and provides the basis for initial consideration by those tasked with assessing the water quality and treatment requirements of a potable reuse project. Chemicals, which may potentially be of concern in potable reuse projects, are diverse in terms of their (anthropogenic or natural) source, chemical characteristics and their likely human toxicity. Public health risk assessments are further complicated by the inevitable presence of unidentified chemicals and potential impacts of 'mixture effects' on the overall toxicity of complex, low-concentration chemical mixtures.

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Introduction

Growing water scarcity and the threat of drought-related water shortages are leading many cities to consider the

opportunities associated with supplementing conventional drinking water supplies with appropriately treated reclaimed water [1,2]. This practice, known as 'potable reuse', has been formally adopted on a large scale in countries including the USA, Australia, South Africa and Namibia. Furthermore, potable reuse occurs informally in thousands of cities, where raw drinking water is sourced from surface waters impacted by effluent discharges from the wastewater treatment plants upstream [3]. Such practices are increasingly being recognised and referred to as 'de facto potable reuse' [4,5].

Despite the global prevalence of de facto potable reuse, it is generally considered that traditional drinking water standards identifying 'safe' water quality are insufficient for potable reuse of reclaimed water [6]. Compared with conventional water sources, reclaimed water and potable reuse may introduce a number of additional risks related to chemical contaminants, which need to be considered [6]. These include chemicals that may be associated with the wastewater source [7], chemicals which may be formed during conventional and advanced treatment processes (e.g. disinfection byproducts) and chemicals that may be released from aquifer storage or distribution system materials as a consequence of treated reclaimed water quality.

Chemicals which may be of concern in potable reuse scenarios include a wide range of naturally occurring and synthetic, organic and inorganic species. Some key classes of chemical hazards include heavy metals, synthetic industrial organic chemicals, pesticides or their metabolites, algal toxins, radionuclides, pharmaceuticals, oestrogenic and androgenic hormones, perfluoroalkyl substances, nanoparticles and disinfection byproducts. Attempts, such as this, to categorise water quality contaminants are always imperfect because the assigned categories are never mutually exclusive. Nonetheless, this categorisation of potable water chemical contaminant classes is adopted here to provide discussion and insights regarding important chemical hazards.

Heavy metals

Heavy metals may be present in municipal wastewater at concentrations reaching the μg per litre level as a result of industrial discharges to sewers. Some heavy metals such as lead, cadmium, chromium and mercury have been associated with human health concerns at μg per litre concentrations in drinking water. These

chemicals have been subjects of regulatory control in public drinking water supplies for many decades, and these same regulatory requirements are equally applicable to potable reuse scenarios. Most heavy metals tend to be lipophilic and therefore partition extensively to sludge during wastewater treatment [8]. Consequently, most concerns regarding heavy metals have been in relation to disposal and reuse of wastewater sludges [9]. Concerns relating specifically to residual concentrations in reclaimed water have been focused primarily on risks associated with irrigation of edible crops, which may lead to elevated human exposure due to bio-concentration [10,11]. Recent work has allayed fears that heavy metals present in (non-potable) reclaimed water may induce detrimental effects on fabrics during household machine clothes washing [12]. Some potable reuse practices involve storage of reclaimed water by aquifer recharge [13,14]. In such cases, risks of potential contamination of treated water, by dissolution of natural geochemical substances, should be considered.

Synthetic industrial organic chemicals

Depending on the catchment area and the extent of the trade waste program to control chemicals at the wastewater source, a very wide range of synthetic industrial chemicals and byproducts are often measurable in urban municipal wastewater treatment plant effluents at ng per litre to µg per litre levels. Examples include plasticisers [15,16], biocides [17,18], surfactants [19], dioxins [20], flame retardants [7,21], dyes [22], polychlorinated biphenyls [23] and phthalates [24,25]. For many of these chemicals, prolonged exposure is known to present chronic health risks including cancer [26–28], and many are environmentally persistent [29]. The ability of advanced treatment processes to control a range of pollutants which can be at transiently elevated concentrations is a current area of research interest. One such chemical which has presented particular challenges in some circumstances is 1,4-dioxane, which is used both as a solvent and as a stabiliser for other organic solvents. 1,4-Dioxane is a known groundwater contaminant in some regions [30,31], but it also presents particular difficulties for control in some potable reuse projects because of its poor removal by reverse osmosis treatment [32]. Consequently, 1,4-dioxane has been a major driver for the increased application of advanced oxidation processes for potable reuse [33,34].

Pesticides

A variety of chemical pesticides, including herbicides and insecticides, have been detected in municipal wastewater effluents at ng per litre concentrations [35,36]. Pesticides may enter municipal wastewater systems by a variety of means, including stormwater influx and illegal direct disposal to sewage systems. Washing domestic pet dogs with insecticide solutions is a known pathway for these chemicals to be discharged to municipal sewers

[37]. Additional routes of unknown significance include washing fruits and vegetables before household consumption, insect repellents washed from human skin and/or washing clothes and equipment used for applying pesticides. Because pesticides have been specifically designed or selected for their detrimental effects on a range of biological species, they present an obvious focus for concern regarding health risks from human exposure and efficacy of biological treatment processes.

Cyanotoxins

Cyanotoxins such as microcystins, nodularins, cylindrospermopsin and saxitoxins are all produced by freshwater cyanobacteria (blue-green algae). Under suitable conditions, cyanobacteria may grow in untreated or partially treated wastewaters, producing these and other toxins [38]. Numerous cyanotoxins have been implicated as having serious impacts on human and animal health by the consumption of contaminated water. Many of these toxins are hepatotoxic, and some are neurotoxic.

Radionuclides

Radionuclides, such as radioactive isotopes of strontium, cobalt, caesium, uranium and selenium, may enter sewage by natural runoff or as a result of medical or industrial usage [39–41]. In the treatment of some public water supplies, radium is removed from drinking water by coagulation, and the concentrated sludge may be transferred to sewage systems. Commercial laundry detergents may also be a source of radiological contamination of wastewaters [42]. Radionuclides are carcinogenic and mutagenic substances.

Natural and synthetic steroidal hormones

Natural steroidal hormones such as oestradiol, oestrone and testosterone are excreted to sewage by people. During the last three decades, natural steroidal hormones have been widely implicated in a range of endocrinological abnormalities in aquatic species that are affected by sewage effluent [43,44]. Related impacts via human endocrinological pathways have been widely postulated and are sources of public concern regarding drinking water quality [45,46]. However, human health risk assessments have generally concluded that these chemicals are unlikely to cause public health impacts at concentrations normally encountered in reclaimed wastewaters [46–48]. Reported removals of steroidal hormones by conventional sewage treatment plants have been variable, but removal to below current analytical detection limits (approx. 1 ng/L) is commonly observed in well-performing plants [49,50]. Further removal may be achieved by advanced treatment [51].

Pharmaceuticals

Dozens of pharmaceutical substances (and their active metabolites), along with illicit drugs, have been detected in treated and untreated municipal wastewaters

globally [52–54]. These chemicals are excreted to sewage by people and direct disposal of unused drugs by households [55,56]. Because pharmaceuticals are designed to instigate biological responses, their inherent biological activity and the diverse range of compounds identified in sewages (and the environment) have been cause for considerable concern. Specific public health concerns have not been identified for most classes of drugs at ambient drinking water concentrations, but issues regarding the presence of antibiotics and the role they may play in antibacterial resistance proliferation have received much attention [57,58]. A broad range of pharmaceutically active compounds have been reported in drinking waters as a consequence of unplanned indirect potable reuse [59].

Perfluoroalkyl substances

Perfluoroalkyl substances (PFAS) such as perfluorooctanoic acid, perfluorohexane sulfonate and perfluorooctane sulfonate are persistent and toxic chemicals that have recently emerged as drinking water contaminants of concern. Although much of the focus has been directed to these three examples, more than 400 PHASs have been identified in the aquatic environment [60]. They also arise from the breakdown of fluorotelomer alcohols, which are widely used in consumer products such as greaseproof food wrappers and stain-resistant carpet treatments. A range of perfluorinated chemicals have been widely reported in municipal wastewater effluents [61,62]. Owing to the presence of precursor compounds in wastewater treatment plant influents, additional PFAS is known to be produced during biological wastewater treatment [63,64]. Water treatment trials have shown that conventional carbon adsorbents are only partially effective for the removal of PFAS in a potable reuse scenario [65].

Nanoparticles

An important group of emerging environmental contaminants of concern is nanoparticles or nanomaterials [66]. These are commonly defined as particles between about 1 and 100 nm in diameter that show properties that are not found in bulk samples of the same material. Nanoparticles isolated from wastewater treatment plants have been found to be composed of 70%–85% carbon and low amounts of oxygen, heavy metals and other elements [67]. It is apparent that the municipal wastewater loads of some nanoparticles, such as TiO₂ and ZnO, may exhibit seasonal variability [68]. This is assumed to be partially a consequence of these nanoparticles being used in functionalised products, such as sunscreens and moisturising lotions, which have seasonal use patterns [68]. The toxicological concerns for nanoparticles are related not only to their chemical composition but also to physical parameters including particle size, shape, surface area, surface chemistry, porosity, aggregation tendency and homogeneity of

dispersions [66]. Furthermore, nanoparticle bioavailability and toxicity may be influenced by transformation processes caused by other substances, such as humic acids or sulphides, present in the wastewater treatment plant [69].

Disinfection and oxidative byproducts

Disinfection and chemical oxidation by ultraviolet (UV) radiation and chemical oxidants is practiced to ensure pathogen inactivation and to reduce concentrations of some target chemical contaminants in reclaimed water. These processes induce chemical reactions and transformations and thus are well-known to produce chemical byproducts [70,71]. As chlorine-based disinfection processes are used in potable reuse, well-known chlorination disinfection byproducts such as trihalomethanes and haloacetic acids also commonly occur [72].

In a potable reuse scenario, the combination of UV and hydrogen peroxide is particularly relevant because it is now used as an important contaminant barrier in a number of large potable water reuse projects [73]. Other UV-based advanced oxidation processes, such as UV/free chlorine and UV/persulfate, are also rapidly emerging as attractive approaches for the removal of trace organic contaminants. Each of these processes produces distinctly different concentrations and characters of byproducts, as a consequence of the different reactive intermediates formed [74,75]. Alternative potable reuse treatment trains, such those that involve ozonation followed by biological activated carbon, are also prone to the formation of disinfection byproducts [76,77].

As a disinfection byproduct, N-nitrosodimethylamine (NDMA) presents a number of significant challenges for potable reuse. These stem partially from this contaminant's status as a suspected human carcinogen with a very high potency (i.e., a high cancer slope factor) [78,79]. This has led to variable water quality objectives, which in some jurisdictions require NDMA concentrations <10 ng/L. Although NDMA has been associated with many water and wastewater systems, potable reuse systems may be particularly problematic if elevated levels of ammonia are present. Chloramination is the most common process that results in formation of NDMA during water and wastewater treatment [80]. However, ozonation of wastewater can also produce high concentrations [80]. Furthermore, advanced oxidation processes may degrade larger molecules to smaller substances, which can subsequently act as precursors for additional NDMA production during advanced water treatment [77]. Challenges encountered by NDMA, in particular, have led to efforts towards the development of an inline detection system for this compound and other N-Nitrosamines [81,82]. It has been observed that, as potable reuse becomes increasingly important

for drinking water supply, NDMA formation and mitigation strategies will become increasingly more important [80].

Conclusions

The various classes of chemical contaminants identified in this short review represent those for which most attention is currently paid in regard to potable water reuse projects. They include some reclaimed water contaminants (eg, many pharmaceuticals and hormones) that are now reasonably well understood and others for which a working understanding is still emerging (eg, nanoparticles and some disinfection and oxidative byproducts). Nonetheless, hundreds of new contaminants continue to be identified, and new sensitive analytical methods are developed each year [83]. The rate of such new developments far exceeds the rate at which any regulatory regime or potable reuse project operator could hope to adapt.

In 2017, the World Health Organization published 'Potable Reuse: Guidance for Producing Safe Drinking Water' [84]. These WHO potable reuse guidelines state that the management of potable reuse schemes should be based on the framework for safe drinking water, including water safety plans. These guidelines do not provide any new guideline concentrations for chemical contaminants, beyond what are already provided in the WHO Guidelines for Drinking-water Quality. This is based on the fact that chemicals of emerging concern, such as pharmaceuticals and personal care products, tend to be present at concentrations, which are generally low and generally do not warrant setting of new guideline values. Nonetheless, in specific circumstances, where a chemical with no guideline value is identified as a concern, approaches for developing screening values are identified to support investigations into potential risks and the need for implementation of additional control measures. The principal reference is to the framework presented in the Australian Guidelines for Water Recycling [85]. Arguably, this concept should also be one that is generally applied to drinking water, beyond just planned potable reuse. This would be particularly appropriate in situations of known, or suspected, de facto potable reuse.

A conspicuously absent topic in this review, and almost all others of its nature, is a detailed discussion of toxicology. The vast majority of chemical contaminants, considered to be of relevance to potable reuse projects, have very limited bases upon which to draw conclusions that ambient concentrations present meaningful risks to public health. A few have been assessed to present little or no risk to public health, whereas most suffer from a lack of relevant information necessary to draw such conclusions. It is highly possible, perhaps likely, that the chemical contaminants responsible for imparting the

greatest public health risks are contaminants or transformation products, for which molecular identities are as yet unknown or their significance not yet appreciated. Furthermore, there are important acknowledged gaps in our toxicological understanding of complex low-concentration mixtures, as drinking water contaminants are inevitably presented as.

Based on current knowledge, summarised in this short review, the chemical contaminants most warranting concern in potable reuse projects are dependent upon system-specific characteristics, most notably, the advanced water treatment processes included in the treatment train. For potable reuse treatment trains, which include very broadly effective processes such as reverse osmosis, concern will be focused on the relatively small subset of chemicals, known to be poorly or unreliably removed by these processes. For reverse osmosis, this tends to include small (low molecular weight), uncharged molecules such as trihalomethanes, NDMA and 1,4-dioxane. Consequently, these chemicals are often targeted for monitoring.

Owing to current limitations in the ability to meaningfully assess public health risks associated with individual and mixture chemical contaminants in drinking water, many researchers and practitioners working on potable reuse risk management have advocated alternatives to direct monitoring of contaminants of concern. One such approach has been the identification of a limited number of measurable chemical contaminants, such as those highlighted in this review, to be used as 'indicators' for the potential presence and effective removal of a much wider range of unidentified contaminants [86,87]. Another widely advocated approach involves the assessment of water quality by bioassay effect-based measurements [88–90]. Although both approaches require further research and development, they both have the potential to play an important role in monitoring the performance of chemical contaminant removal for potable reuse projects.

Conflict of interest statement

Nothing declared.

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