Chemosphere 364 (2024) 143030

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Managed aquifer recharge as a potential pathway of contaminants of emerging concern into groundwater systems – A systematic review

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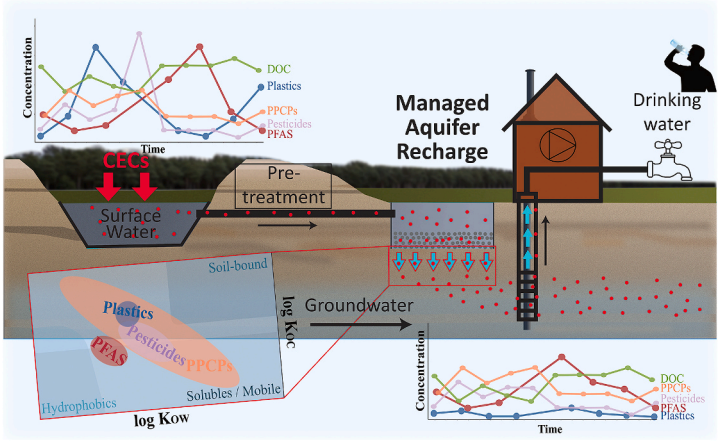
HIGHLIGHTS

- Managed aquifer recharge (MAR) ensures stable and sufficient groundwater systems.
- Contaminants of emerging concern (CECs) in surface waters can pass through MAR.
- (Very) mobile and (very) persistent compounds are of greatest concern in MAR.
- Mostly per- and polyfluoroalkyl substances (PFAS) are likely to pass through MAR.
- Redesign is needed to effectively use MAR systems to even retain CECs.

GRAPHICAL ABSTRACT



ARTICLE INFO

Handling editor: Jerzy Falandysz

Keywords:
Contaminants of emerging concern
Managed aquifer recharge
Groundwater
Infiltration
Surface water
PFAS

ABSTRACT

Groundwater is an often-overlooked resource, while its declining quantity and quality is of global concern. To protect and ensure stable quantity and quality of groundwater systems used as drinking water supplies, a common method is to artificially recharge these groundwater supplies with surface water, a process called managed aquifer recharge (MAR), that has been used globally for decades. However, surface waters used for MAR often contain elevated concentrations of anthropogenic chemicals of emerging concern (CECs), such as plastics, pesticides, pharmaceuticals and personal care products (PPCPs), or per- and polyfluoroalkyl substances (PFAS). When infiltrating this surface water, MAR can thus act as a shortcut for CECs into groundwater systems and eventually drinking water supplies. Especially PFAS are an example of very persistent contaminants showing atypical transport patterns during MAR and thus posing a risk for ground- and drinking water contamination. This systematic review addresses the transport process of CECs through MAR systems by looking at (1) common CEC concentrations in surface waters, (2) factors affecting CEC transport and possible retention during MAR, such as sorption and other physio-chemical mechanisms of CECs, biological and chemical decomposition, or hydrogeological properties of the MAR system, and (3) key contaminants leaching through the MAR systems as

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https://doi.org/10.1016/j.chemosphere.2024.143030
Received 14 June 2024; Accepted 4 August 2024
Available online 8 August 2024

well as possible treatment options to improve the retention of CECs during MAR. Since we are facing increasing needs for high quality drinking water, lower CEC drinking water guidelines as well as an increasing number of identified CECs in surface waters, we conclude with a series of recommendations and future research directions to address these issues. Those include the need for regular monitoring programs specifically addressing CECs and especially not yet regulated, (very) persistent and (very) mobile contaminants, such as PFAS, as well as redesigned MAR systems to ensure stable ground- and drinking water quantity and quality.

List of abbreviations

AFFF	Aqueous film forming foam
AMPA	Aminomethylphosphonic acid
CECs	Contaminants of emerging concern
DDT	Dichlorodiphenyltrichloroethane
DOC	Dissolved organic carbon
EDC	Endocrine disrupting compound
MAR	Managed aquifer recharge
NOM	Natural organic matter
PFAS	Per- and polyfluoroalkyl substances
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctanesulfonic acids
POPs	Persistent organic pollutants
PPCP	Pharmaceuticals and personal care product
SMART	Sequencial managed aquifer recharge technology
WHO	World Health Organization

1. Introduction

Groundwater is an important resource of global concern making up 99% of the global liquid freshwater resources and is essential for nearly 50% of the global drinking water production, 50% of the global river base flow, and supports about 40% of global food production (Cross et al., 2016; Gleeson et al., 2016; Langbein, 1947; Postigo and Barceló, 2015). However, rising populations and climate change lead to increasing risks of groundwater depletion especially in temperate regions or colder areas with low groundwater availability and small or few aquifers. Moreover, in coastal areas, climate change and overexploitation-induced groundwater depletion can trigger saltwater intrusion and thus have adverse effects on groundwater quality. Artificial groundwater recharge or managed aquifer recharge (MAR), that includes the artificial infiltration of surface water from lakes or rivers into groundwater systems (Fig. 1), is a globally implemented strategy designed to mitigate groundwater depletion and to recover, secure, and maintain adequate groundwater volumes with sufficient quality for drinking water supplies (Dillon et al., 2019; Scanlon et al., 2023; Taylor et al., 2013).

MAR has become increasingly relevant since the 1970s and will likely be of utterly importance for future water management strategies ensuring groundwater quality and quantity (Dillon et al., 2019). MAR compromises a variety of different methods of "intentional groundwater replenishment" (Dillon et al., 2019), such as streambed channel modifications, (river)bank filtration, water spreading in infiltration basins, recharge wells and shafts, or reservoir releases (Balke and Zhu, 2008; Dillon et al., 2019). Globally, the highest MAR capacity can be found in India, being strongly implemented during the past 50 years by governmental programs to ensure sufficient water resource quantities and improve water quality (Alam et al., 2021; Dillon et al., 2019; Sprenger et al., 2017). In Europe, surface water spreading is applied in more than half of the MAR systems consisting of either bank filtration, where river- or lake water is used for infiltration followed by extraction via wells, or infiltration basins, where surface water is redirected into specific artificial basins (Balke and Zhu, 2008; Hägg, 2020; Hannappel et al., 2014; Sprenger et al., 2017) and infiltrated through a sandy filtration layer into groundwater systems (Alam et al., 2021).

MAR is a very cost-efficient method that naturally attenuates inorganic and organic contaminants as well as decreases turbidity during the artificial infiltration process. The process is governed by filtration, straining, (ad-)sorption and biological degradation and could possibly act as a (pre-)treatment step in drinking water production (Ahmed and Marhaba, 2017). The natural purification processes during the infiltration process result in complete or partial retention of nutrients, natural organic matter (NOM), metals or organic pollutants within the subsurface and are thus an alternative for chemically treating water during drinking water production (Balke and Zhu, 2008; Fakhreddine et al., 2021; Jokela et al., 2017; Patterson et al., 2010). Removal efficiencies of up to 87% for high molecular weight NOM can be achieved, while MAR is less effective for low-molecular weight NOM (Ahmed & Marhaba, 2017and references therein; Mishra et al., 2021). MAR further acts as a stabilization step reducing variations in temperature as well as pollutant concentrations and making the water more palatable (Ahmed and

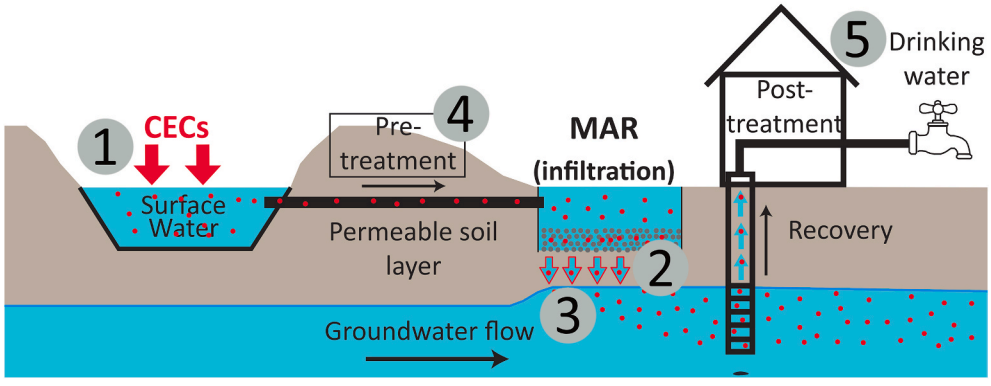

Fig. 1. Conceptual figure of a managed aquifer recharge (MAR) system via basin infiltration. Contaminants of emerging concern (CECs; indicated in red) are moving from point and diffuse sources into surface water through the MAR system during drinking water production. The respective steps (1–5) are discussed in the respective results and discussion sections (3.1-3.5).

Marhaba, 2017; Lee et al., 2009).

However, MAR is not designed for the removal of contaminants of emerging concern (CECs). According to the U.S. Environmental Protection Agency, CECs are "pollutants not currently included in routine monitoring programs and [that] may be candidates for future regulation depending on their (eco)toxicity, potential health effects, public perception, and frequency of occurrence in environmental media" (Smith, 2008) with many of those compounds being recently discovered. However, CECs can also include chemicals that have been present for decades but only recently received attention (Christensen et al., 2022; Smith, 2008). Among the most prominent CECs during the past decade in surface water are plastics (including their plasticizers) (Dalmau-Soler et al., 2021; Re, 2019), pesticides (Díaz-Cruz and Barceló, 2008; Mathys, 1994), pharmaceuticals and personal care products (PPCPs) (Burch et al., 2019; Drewes and Shore, 2001; Heberer, 2002; Malnes et al., 2022; Yang et al., 2017), endocrine disrupting compounds (EDCs) (Ahmed et al., 2021; Ma et al., 2016; Zuehlke et al., 2004), and per- and polyfluoroalkyl substances (PFAS) (Cousins et al., 2022; Evich et al., 2022; Giesy and Kannan, 2001; Skutlarek et al., 2006). They are persistent in the environment and thus often classified as risk in environmental policies and hazard guidelines (Nawaz and Sengupta, 2019; Scheringer et al., 2022). Due to anthropogenic input, CECs are often found at concerning concentrations in surface waters, including those that are used for MAR (Balke and Zhu, 2008; Banzhaf et al., 2015; Christensen et al., 2022), and therefore MAR could act as a pathway for CECs into groundwater systems and drinking water supplies posing a risk to human health and the environment (Díaz-Cruz and Barceló, 2008; Fakhreddine et al., 2021; Lin et al., 2015; Regnery et al., 2017). However, the process of CECs entering groundwater systems during MAR is poorly understood.

A few previous studies (Lin et al., 2015; Sanz-Prat et al., 2020; Schaper et al., 2018) and reviews (Díaz-Cruz and Barceló, 2008; Fakhreddine et al., 2021; Postigo and Barceló, 2015; Regnery et al., 2017) have studied CEC transport and attenuation during MAR as well as occurrence and presence in surface and groundwater. However, CEC fate during MAR is controlled by a large range of additional factors changing groundwater quality, such as flow rate and retention time in the subsurface, redox conditions, mineralogy, temperature, or other physical, chemical, and biological attenuation processes, and therefore challenging to assess or simulate (Drewes, 2003; Fakhreddine et al., 2021; Maeng et al., 2011; Regnery et al., 2017). Additionally, field studies are often challenged by the variety of contaminants, environmental factors affecting CEC transport, and possible non-additive interactions as well as sample contamination, while standardized sampling protocols and monitoring procedures are lacking (Banzhaf and Hebig, 2016; Nagy-Kovács et al., 2018; Yuan et al., 2017). Therefore, a more comprehensive understanding of the factors impacting CEC transport through the MAR system into groundwater and drinking water supplies is critical for regulating CECs to ensure safe drinking water as well as for identifying future research directions. To address this research gap, this review takes a more holistic approach looking at the fate of CECs from their sources to the surface water, through the MAR system, and into groundwater systems and drinking water supplies (Fig. 1).

This includes how CECs are transported through surface- and groundwater systems during MAR and what is needed for future CEC handling during MAR. We are systematically assessing which factors impact the fate of CECs in MAR systems to identify possible pathways of CECs into groundwater systems. Furthermore, we give an overview of the current state of research by investigating different aspects of CEC transport during infiltration as well as identify key contaminants that potentially migrate into groundwater systems during MAR processes and thus might pose a risk to drinking water systems. The specific objectives were to identify (1) common CEC concentrations in surface waters used for MAR and their health limits, (2) current knowledge on subsurface characteristics altering CEC transport and retention as well as their interactions during MAR, (3) the potential of key contaminants possibly passing through MAR systems, (4) pre- or combined treatment solutions, and (5) future research directions to effectively adjust MAR systems for CEC retention (Fig. 1). These insights will aid the understanding and modification of existing or future MAR systems to effectively remove CECs to ensure high water quality for drinking water producers that depend on MAR.

2. Methodology

For this work, we conducted a systematic literature review in the most comprehensive databases Scopus and Web of Science focusing on artificial groundwater recharge, MAR and their contamination as well as specific techniques, such as bank or basin filtration and their contamination, respectively. General searches for the matrixes soil and water were excluded to limit the number of search results. Additionally, specific CECs in connection to surface- and groundwater systems as well as contaminant behavior and characteristics were searched for resulting in 1460 screened papers. The screening was then conducted following the PRISMA flow-chart for systematic reviews using databases and registers (Page et al., 2021). Due to an exorbitant number of papers for certain CECs, especially different pharmaceuticals and personal care products, the number of papers per substance class was limited to 10 and reduced by impact factor and publication day for groups exceeding this limit leading to a total of 83 papers included in this review. An updated search was conducted on April 4th, 2024, resulting in 131 new papers, whereof 6 additional articles were included in this review. The PRISMA diagram (Page et al., 2021) showing the screening process (Fig. S1 in Supporting Information (SI)) as well as the search strings (Fig. S2 in SI) can be found in the supplementary material.

3. Results and discussion

The described results and discussion are structured according to the pathway of the CECs through the MAR system and are connected to the five objectives described in the introduction above (Fig. 1). The respective steps (1–5) in Fig. 1 are discussed consecutively in the following sections (3.1-3.5).

3.1. Anthropogenic contaminants of emerging concern in surface waters and their health limits

Most CECs are of anthropogenic origin and often persistent in the environment (Albergamo et al., 2019; Drewes, 2003; Nawaz and Sengupta, 2019; Scheringer et al., 2022). However, surface waters are especially prone to show elevated CECs concentrations since they can drain larger catchments, which are potentially contaminated by CECs and thus, can act as sink for CECs (Ahmed et al., 2021; Malnes et al., 2022). CECs originate from either point or diffuse sources: While ineffectively treated wastewater (Banzhaf and Hebig, 2016; Ma et al., 2016) and other point sources, such as for example contaminant spills or industrial sites (Díaz-Cruz and Barceló, 2008), contribute to the majority of environmental CEC concentrations, diffusive sources such as run off from agricultural activities (e.g. pesticides) or atmospheric deposition (e.g. PFAS) contribute a smaller fraction to the overall CEC contamination in surface waters. Additionally, stormwater and roof runoff resulting from atmospheric input or surface runoff can be a source for CECs, that are transported via surface water catchment drainage (Evich et al., 2022; Happonen et al., 2016) and can further lead to elevated CEC concentrations in surface waters. Due to this variety of diffuse and point sources, surface waters, compared to groundwater reservoirs, are generally more impacted by CEC contamination.

Sampling and assessment of environmental CEC samples is often challenging due to the risk of sample contamination and a variety of additional environmental factors possibly interacting with the studied contaminants, enhancing, or mitigating effects. Moreover, standardized sampling protocols and monitoring procedures for CECs are rarely

established (Banzhaf and Hebig, 2016). This makes the comparison of different studies and CECs challenging. However, in the following sections we will further discuss the major CEC groups regarding their properties, occurrence in surface waters used for MAR (Fig. 1, location 1), as well as health limits and regulations according to current literature.

3.1.1. Plastics

Plastics are a versatile group of polymer materials used abundantly in daily life. They exist in different shapes and sizes and are categorized in macro- (>5 mm), micro- (1 μm–5 mm), and nanoplastics (1 nm–1 μm), even though there is no standard definition of the size ranges (Brewer et al., 2021; Gigault et al., 2018; la Cecilia et al., 2024). Globally, several million tons of plastics are discharged annually as trash directly into surface waters or released from anthropogenic products into waste- and surface water (Brewer et al., 2021; Gigault et al., 2018). While already causing hazardous effects themselves, plastics contain additives, e.g., phthalates, organophosphorus flame retardants, or bisphenol A, which might act as EDCs and can leach into the water (Dalmau-Soler et al., 2021). Once in the environment, plastics can further breakdown into smaller particles eventually reaching nano ranges. The increased surface-to-volume ratio of nanoplastics increases their reactivity and changes the physio-chemical characteristics of the parent material (Nel et al., 2006). Nanoplastics are thus likely to cause more severe effects as well as to surpass biological membranes and treatments more easily than the parent material (Frei et al., 2019; Gigault et al., 2018). However, fate and behavior in the environment, as well as possible hazardous effects, still require further studies for most nanoparticles (Alvarez et al., 2018; Grieger et al., 2019; Hodges et al., 2018; King et al., 2022). Additionally to the effects of the plastic particles and their additives, adsorption of other CECs to the plastics' surface can enhance their toxicity and bioaccumulation effects acting as carrier (Ahmed et al., 2021; Re, 2019).

Abundances of microplastics in aquatic environments, including surface waters used for MAR, are on the magnitude of 10^3 to 10^6 particles per m^2 water surface (Ahmed et al., 2021) and vary widely globally from 10^2 to 10^4 particles per m^3 water (Parashar et al., 2023). Until now, research on plastic pollution is mainly focused on surface waters and stormwater runoff (Bodus et al., 2024), while literature on groundwater (Ahmed et al., 2021; Mintenig et al., 2019; Panno et al., 2019) and drinking water produced from groundwater (Mintenig et al., 2019; Tyree and Morrison, 2017; Wanner, 2020) is scarce (Dalmau-Soler et al., 2021; Re, 2019). To regulate microplastics, and eventually nanoplastics, and mitigate pollution, the United Nations Sustainable Development Goals and especially their Goal 14 "life below water" state since 2015 the urge to address plastic mitigation and many countries are starting to implement regulations accordingly (Gündoğdu et al., 2023). Until the end of 2024, 175 countries agreed to a legally binding resolution to address plastics until the end of 2024 (United Nations, 2022).

3.1.2. Pesticides

Pesticides are primarily used to increase crop quality and quantity during agricultural activities. They comprise several hundred substances that are classified as herbicides, fungicides, and insecticides (Dragon et al., 2019). They are commonly found in surface waters due to stormwater runoff from agricultural fields and diffuse pollution during application of pesticides to crops (Díaz-Cruz and Barceló, 2008). Commonly studied pesticides and their metabolites are atrazine, diuron, and carbofuran (Jaramillo et al., 2019) as well as the herbicide glyphosate (Litz et al., 2011). For glyphosate, 18% of the sampled sites in a study in Germany were above the EU threshold of 0.1 μg/L for glyphosate in drinking water, while more than 70% of the sites exceeded the threshold of 3 μg/L for pesticide metabolites with AMPA (aminomethylphosphonic acid), the metabolite of glyphosate (Litz et al., 2011). The World Health Organization (WHO) on the other hand states that health values for glyphosate and AMPA are usually significantly higher than drinking water concentrations and do thus not require formal guidelines. The Stockholm Convention emphasizes the insecticide DDT (dichlorodiphenyltrichloroethane) and its metabolites among many other legacy pesticides as significant for health-related guidelines in drinking water (World Health Organization, 2022). In contrast, the measured pesticide concentrations in surface water used for MAR and resulting artificial groundwater in Krajkowo, Poland (Dragon et al., 2019), as well as the herbicide isoproturon (Trinh et al., 2012), were below current guideline values and thus not of major concern. But due to seasonal application of pesticides and changing precipitation patterns, concentrations in surface waters vary significantly throughout the year (Dragon et al., 2019; Oberleitner et al., 2020). Furthermore, different studies and sampling methods report varying surface water concentrations in the range of 3 orders of magnitude (Dragon et al., 2019; Page et al., 2014).

Pesticides, as well as their metabolites, are of concern to environmental and human health and regulations are constantly adapted worldwide (Dragon et al., 2019; Trinh et al., 2012) as for example by the Stockholm Convention (World Health Organization, 2022). But even after the stop of application, some pesticides as well as their often more toxic metabolites can pose a long-term risk to below-laying groundwater systems and drinking water reservoirs caused by a time delay during subsurface passage (Suárez et al., 2007). Additionally, different types of pesticides have been used and regulated throughout the last decades, leading to changing composition profiles (Kruisdijk et al., 2022).

3.1.3. Pharmaceuticals and personal care products

Pharmaceuticals and personal care products (PPCPs) are widely applied in human households as well as animal farming and make up about 60% of the target CECs in surface water screening studies leading to a high detection frequency (Díaz-Cruz and Barceló, 2008; Malnes et al., 2022). PPCPs cover a group of several thousand compounds and are commonly studied resulting in a large number of scientific articles (Candela et al., 2016). Since often not fully metabolized during human passage and wastewater treatment plants not being designed for CEC removal (Lin et al., 2015), wastewater treatment plants are a major entry route of higher concentrations of PPCPs or their possibly toxic transformation products to streams and surface water systems used for MAR (Bade et al., 2015; Kasprzyk-Hordern et al., 2009) if not managed accordingly (World Health Organization, 2022). Even though the parent compounds are often present at concentrations that have no significant effect on humans consuming drinking water, they do impact the environment and especially aquatic life as well as break down into possibly toxic transformation products (Lin et al., 2015) and will thus require further assessment.

In surface waters, a great variety of compounds and their degradation products can be found as for example, caffeine, nicotine, antidepressants (e.g., desvenlafaxine), antihistaminic compounds (e.g., fexofenadine), antiepileptics (e.g., carbamazepine, it's biodegradable alternative gabapentin, primidone), pain killers and anti-inflammatory medication (e.g., ibuprofen, paracetamol, indomethacin, diclofenac), antibiotics (e.g., sulfamethoxazole), blood pressure medication (e.g., losartan, hydrochlorothiazide, metoprolol), as well as sunscreen components (e.g., sulisobenzone) (Díaz-Cruz and Barceló, 2008; Lin et al., 2015; Malnes et al., 2022). They are generally present in nanogram per liter to low microgram per liter concentrations in surface waters, including those used for MAR, groundwater reservoirs as well as drinking water systems (de Carvalho Filho et al., 2022; Lin et al., 2015; Malnes et al., 2022; World Health Organization, 2022). However, due to the large variety in this contaminant class, regulations vary widely with new regulations emerging constantly (Richardson, 2007).

3.1.4. Per- and polyfluoroalkyl substances (PFAS)

Per- and polyfluoroalkyl substances (PFAS) are a group of more than 16,000 compounds consisting of fluorinated carbon chains with a minimum of one fully fluorinated methyl group, attached to a functional

hydrophilic group (Buck et al., 2011; Evich et al., 2022). PFAS occur in a variety of industrially produced products and their input to surface- and groundwater results from atmospheric emissions, wastewater, stormwater runoff, terrestrial application of biosolids, or aqueous film forming foam (AFFF) connected to industrial or other anthropogenic activities (Banzhaf et al., 2017; Evich et al., 2022; Happonen et al., 2016; Sörengård et al., 2022). Due to the strong fluorinated carbon chain, PFAS are extremely persistent in the environment degrading at most to stable, shorter-chained PFAS (Cousins et al., 2020, 2022; Podder et al., 2021; Sims et al., 2022). While they are detected globally in all types of water bodies and even at nanogram per liter levels in extremely remote areas, highest concentrations are found in Europe, US, and Asia with concentrations in the high microgram per liter range at contaminated sites (Ahrens et al., 2009; Dagorn et al., 2023; Sims et al., 2022). Even though more PFAS are added to regulations and awareness is increasing (European Comission, 2020; Evich et al., 2022; United Stated Environmental Protection Agency, 2024), quality control, monitoring, and legislation of PFAS as a group and their precursors by analyzing surface and drinking water is needed (Skutlarek et al., 2006). Groundwater compositions are less impacted by PFAS pollution except in PFAS contamination hotspot areas (Gobelius et al., 2018).

PFAS are carcinogenic among other severe effects and pose a risk to human health in the range of nanograms per liter (Andrews and Walker, 2015; Gobelius et al., 2018). This resulted in the phase-out and listing under the Stockholm Convention of some legacy PFAS such as perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) (Giesy and Kannan, 2001; Hansen et al., 2001; Rich, 2016) and their regulation such as for drinking water approaching low nanogram per liter ranges (Gobelius et al., 2018). In the European Union, current drinking water guidelines regulate the sum of 20 PFAS to 100 ng L^{-1} as well as the total amount of PFAS to 500 ng L^{-1}, while Denmark or the US already restrict the sum of four PFAS to 2 ng L^{-1} or PFOS and PFOA each to 4 ng L^{-1}, respectively (European Comission, 2020; Miljøministeriet, 2024; United Stated Environmental Protection Agency, 2024).

3.1.5. Other contaminants of emerging concern

While the previous sections only address the major groups of CECs, there is a variety of other CECs present in surface waters used for MAR such as industrial chemicals, other nanomaterials in addition to nanoplastics, legacy persistent organic pollutants listed under, for example, the Stockholm Convention (e.g. dioxins, polycyclic aromatic hydrocarbons, polychlorinated biphenyls), flame retardants, algae toxins, cooling agents, or organo-metal compounds (Scheurer et al., 2022; Yu et al., 2022). Further, nanomaterials such as activated carbon or graphene are commonly used in CEC water treatment technologies (Christensen et al., 2022). Nanomaterials are compounds between 1 and 100 nm in at least one dimension, while they classify as nanoparticles if they fulfill this size range in at least two dimensions (Boholm and Arvidsson, 2016). Activated carbon can efficiently adsorb even very mobile CECs, and graphene or other sheet-like nanomaterials are used as membranes for water filtration (King et al., 2022; Wei et al., 2020). However, it needs to be ensured that produced nanoparticles from the membranes are not remaining in the treated water (Nakazawa et al., 2018). Hereby, nanoparticles that form a natural coating, a more likely to be absorbed in the upper infiltration layer, while synthetically coated nanoparticles remain more mobile (Degenkolb et al., 2019).

3.2. Site-specific subsurface characteristics governing the fate of CECs through the MAR infiltration layer

During MAR, water quality is impacted by different physical, chemical, and biological attenuation processes. Different factors such as local hydrogeology, weather and climate, or the subsurface's biogeochemical characteristics strongly influence contaminant removal efficiencies (Díaz-Cruz and Barceló, 2008; Schaper et al., 2018). Therefore, both laboratory and field experiments as well as detailed monitoring programs are necessary for a complete understanding of CEC fate and behavior (Kondor et al., 2024). Those can then be complemented by modelling, e.g. inverse modelling (Jaramillo et al., 2019), groundwater flow and transport models (Barkow et al., 2021; Mustafa et al., 2016; Sanz-Prat et al., 2020) or chemical prediction models (Che Nordin et al., 2021), but data is often scarce – especially for groundwater systems and drinking water supplies (Scheurer et al., 2022). However, migration of the previously described contaminant groups from surface waters into groundwater systems via MAR is complex and several factors and site-specific characteristics impact CEC fate through the infiltration layer during MAR (Fig. 1, location 2). Those include (1) hydrogeological and physio-chemical characteristics of the sandy infiltration layer governing processes such as leaching or further chemical degradation, and (2) biological degradation, chemical decomposition, and redox conditions as well as organic matter interactions and biofilm formation acting as a filtration layer.

3.2.1. Hydrogeological characteristics of the sandy infiltration layer

The efficiency of MAR for CEC removal is governed by complex biological, biogeochemical, and hydrogeological settings in the infiltration layer and no standards regarding optimization exist making effects and efficiencies very site-specific. During infiltration, hetero atoms of the CECs bind to the electron deficient organic matter in the infiltration layer and thus increase retardation factors, which is the deceleration of chemical transport in the subsurface compared to the water moving through the MAR system (Mishra et al., 2021). Formation of a clogging layer on top of the infiltration layer further increases organic matter concentrations and thus the number of sorption sites for CECs. Especially in column studies, this effect of increasing retardation factors of CECs over time has been observed (Høisæter et al., 2019; Mishra et al., 2021; Roberts and Valocchi, 1981).

In general, greater subsurface residence times increase CEC removal during MAR. Therefore, removal rates of CEC are highly impacted by the infiltration or flow rate (Regnery et al., 2017), which depends on aquifer hydrogeology and especially hydraulic conductivity since it determines infiltration and thus aquifer residence time. Thus, higher flowrates and shorter travel times along with high gradients, coarse aquifers or very heterogenous materials as well as soil types with higher hydraulic conductivity and less surface area reduce the contaminant removal effect as well as the DOC reduction and thereby decrease water quality significantly (Ahmed and Marhaba, 2017; Fox and Makam, 2009; Mustafa et al., 2016). This has been shown by several studies (Ahmed and Marhaba, 2017; Moradnejadi et al., 2018; Pulido-Reyes et al., 2022), where, for instance, the organo-chloride pesticide lindane was removed by more than 85% during sand column studies with removal efficiency increasing with increasing column depth (Moradnejadi et al., 2018) or nanoparticles being retained 3-log-fold in a laboratory column study (Pulido-Reyes et al., 2022). Aged sand including a biofilm seemed to further increase the retention efficiency as well as to ensure a stable long-term removal efficiency (Pulido-Reyes et al., 2022). However, longer transport distances might also allow for further degradation into possibly hazardous transformation products and can therefore show a higher removal rate than present.

3.2.2. Microbial degradation, chemical decomposition, and biogeochemical conditions

The importance of contaminant transport and behavior during infiltration in MAR systems (Fig. 1, location 2) identifying adsorption and microbial degradation as main removal processes was stated already decades ago by Roberts et al. (1980). However, chemical and biological degradation can often also be "transformation to unknown transformation products" (Muntau et al., 2017), since breakdown products are rarely measured and degradation is commonly defined as a difference in removal rates between a matrix with and without active microbial communities (Bertelkamp et al., 2012; Muntau et al., 2017). This leads to removal rates being often overestimated due to not measuring

degradation or transformation products (Díaz-Cruz and Barceló, 2008). First-order rate constants and linear distribution coefficients serve as a good starting point for modelling this phenomenon. Those more general descriptors then aid when comparing different sites with different site-specific settings (Henzler et al., 2014). Reactive transport modelling can then help to identify non-degradable compounds with retardation factors being the major driver of fate and sorption within the subsurface (Henzler et al., 2014; Mishra et al., 2021).

Retention time, redox conditions, mineralogy, and temperature in the infiltration layer, are positively controlling microbial and chemical contaminant depletion and debilitation (Regnery et al., 2017). Degradation by microorganisms requires a redox potential gradient along with longer residence times in the subsurface (Bertelkamp et al., 2012). This, in addition to charged compounds being chemically degradable and removal occurring typically within the upper layers of the subsurface (Bertelkamp et al., 2012), shows that biogeochemical conditions play an important role in contaminant stability and fate during MAR (Albergamo et al., 2019). Even though redox conditions and residence time highly impact different CEC removal rates in the subsurface and are crucial for microbial degradation, the number of representative studies is limited (Postigo and Barceló, 2015; Schaper et al., 2019; Storck et al., 2013).

While aerobic conditions are common during column or batch studies and lead to more effective biodegradation, most MAR systems operate under anaerobic conditions (Schaper et al., 2018; Shareef et al., 2014; Sopilniak et al., 2018). Filtration during MAR commonly occurs at low infiltration rates leading to dissolved oxygen concentrations being quickly depleted and turning the system anoxic that in turn causes denitrification as well as sulfate, iron(III), and manganese(VI) reduction into nitrite, sulfide, iron(II), and manganese(II). Since those are rather unwanted byproducts, controlling oxygen concentrations is crucial (Ahmed and Marhaba, 2017; Massmann et al., 2008). Higher temperatures (above 20°C) can further enhance this effect (Rudolf von Rohr et al., 2014). Different redox conditions have further shown to result in different metabolites when herbicides were degraded, leading for example to enhanced glyphosate removal during increased oxygen concentrations and residence time in the soil as well presence of a biofilm layer (Litz et al., 2011), and thus emphasizing the relevance of assessing local MAR conditions (Shareef et al., 2014; Wiese et al., 2011). Additionally, laboratory studies often use significantly higher concentrations than naturally occurring contaminant concentrations (Bertelkamp et al., 2012) and the commonly assumed first-order rate constants vary in the range of three orders of magnitude with high uncertainties for soil and groundwater. Similar trends for surface waters are likely (Greskowiak et al., 2017).

Other factors such as organic matter content, other contaminants present, aquifer hydrogeology or pH and temperature further impact CEC removal rates during MAR. For example, higher pH values and temperatures during summer led to reduced removal rates of EDCs used as plastic additives (e.g. bisphenol A), even though those conditions were likely related to sorption processes and chemical decomposition rather than microbial degradation (Ma et al., 2016; Takagi et al., 2011). Generally, MAR is a complex process and CEC removal via microbial degradation during surface passage remains highly dependent on site-specific features (Greskowiak et al., 2017).

3.3. Contaminants of emerging concern passing through MAR systems

Despite the widespread application of MAR worldwide and the elevated CECs concentrations in surface water used for MAR, our knowledge regarding CEC transport during MAR into groundwater system and drinking water supplies is limited. When CECs enter the subsurface, different physio-chemical characteristics such as persistency or mobility lead to a different behavior of most CECs compared to other persistent organic contaminants, such as classical persistent organic pollutants (POPs) including dioxins, polycyclic aromatic hydrocarbons, polychlorinated biphenyls, or flame retardants. POPs are more hydrophobic, less soluble, and thus more likely retained in the sandy infiltration layer, while most CECs can potentially pass through the MAR system (Fig. 1, location 3). The biological degradation rates depend hereby additionally on functional group for some organic micropollutants such as PFAS or PPCPs (Ahmed and Marhaba, 2017; Bertelkamp et al., 2014; Filter et al., 2021). Additionally, CECs often show different transport and adsorption patterns due to their polarity or containing ionic functional groups compared to the commonly less-polar POPs.

The fate and transport of CECs through MAR systems therefore requires further investigation as a basis for models and risk assessments. (Barkow et al., 2021; Handl et al., 2023; Ma et al., 2016; Mishra et al., 2021; Page et al., 2019; Sanz-Prat et al., 2020). Unintentional desorption or dissolution from the infiltration layer can further cause the opposite effect introducing toxic metals, pesticides, industrially produced compounds, microorganisms, natural toxins or other concerning micropollutants to groundwater systems and drinking water supplies (Fakhreddine et al., 2021; Mathys, 1994; Yu et al., 2022) or leading to accumulation within the aquifer (Díaz-Cruz and Barceló, 2008). When used for drinking water production, those CECs then pose a risk to human health (Banzhaf and Hebig, 2016; Malnes et al., 2022).

Positively charged functional groups of contaminants such as some pharmaceuticals and antimicrobials can get attracted to the negatively charged biofilm, which can facilitate microbial degradation and formation of a biological filtration or clogging layer, whereas the biodegradation process of negatively charged compounds is not as well understood. Generally, negatively charged CECs as well as charged compounds with higher distribution coefficients (e.g. gemfibrozil) are often more biodegradable, since they cannot sorb to the negatively charged sand and are therefore available in solution (Bertelkamp et al., 2014; Mishra et al., 2021). Degradation of neutral compounds (e.g. caffeine, atrazine, sulfadiazine), however, is likely steered by other processes than electrostatic interactions or hydrophobicity (Bertelkamp et al., 2012). Other factors, such as ionic strength in the soil-water mixture, amount of clay, and pH can additionally positively affect dissolved organic matter (DOM) sorption to the infiltration layer (Ahmed and Marhaba, 2017; Mishra et al., 2021). CECs can then sorb to organic matter in the subsurface, a process governed by either hydrophobic partitioning through attraction to non-polar organic compounds or physical sorption through electrostatic forces (e.g., Du et al. (2014)). While sorption is one factor retaining pollutants during MAR, ion-exchange capacity can additionally retain cations. However, several studies suggest hydrophobic partitioning as the dominant sorption process for polar compounds (Patterson et al., 2010, 2011; Schaper et al., 2019), especially for the CECs presented in this review, which mostly are (super-)polar or small ionic compounds (Henzler et al., 2014; Mishra et al., 2021) with low K_{OW} values (Fig. 2). Generally, higher hydrophobicity positively affects adsorption due to lower solubility and mobility. However, if polar compounds are soluble or highly mobile (Fig. 2), they pose a risk to travel through the MAR system and enter below-laying groundwater systems (Mishra et al., 2021).

In a field-scale study testing the efficiency of MAR systems to remove CECs, MAR has been shown to be effective for removal of dissolved organic carbon (DOC), as well as some EDCs and PPCPs (alkylphenol polyethoxycarboxylates, caffeine, analgesic or ant-inflammatory drugs, and blood lipid regulators) at sufficient residence times of several weeks (Drewes, 2003; Page et al., 2014). However, carbamazepine and primidone, both antiepileptic drugs, could not be removed sufficiently (Drewes, 2003). Due to the great variety of PPCPs (Table S3 in SI) and their versatility in properties spanning over a wide range of chemical properties (Fig. 2), a great variety of removal efficiencies of PPCPs can be found (Fig. 3).

For microplastics, MAR and conventional drinking water treatment can effectively remove plastics with sand and membrane filters and using surface water with low pollutant concentrations (Gündoğdu et al.,

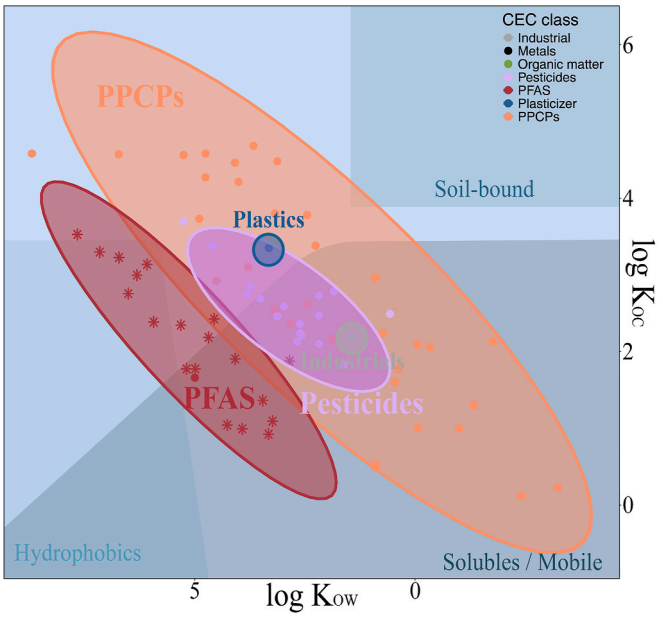

Fig. 2. Partitioning diagram of the major CEC groups based on the reviewed literature. No values were found for metals and organic matter, while each one paper reported partitioning coefficients for PFAS, plastics, and industrial contaminants. PPCPs span a wide variety of reported values in the literature in both the soil-water partitioning coefficients K_{OC} as well as the octanol-water coefficients K_{OW}. The references can be found in Table S3 in the SI. Due to the versatility of chemical properties of PFAS, additional reported partitioning coefficients from papers not included in the search string of this review are marked with stars.

2023). It is usually an easy to implement and cheap method to even out contaminant peeks during high discharge seasons and to ensure safer water supplies in countries with less developed water treatment systems (Dragon et al., 2019; Jaramillo et al., 2019). However, quantitative investigations on the effectiveness of MAR to be a sink for microplastics or shortcut to groundwater systems are lacking (la Cecilia et al., 2024) (Fig. 3). Generally, MAR and conventional drinking water treatment can remove larger microplastics (≥6.6 μm) leading to low estimated uptake concentrations from drinking water of 1 microplastic particle per year per person (Kirstein et al., 2021), even though the relevance of the different steps in the drinking water treatment train is under debate (la Cecilia et al., 2024). The larger particles (>50 μm) tend to primarily accumulate through interaction with organisms and sedimentation and are thus more easily retained, while smaller particles (at or below pore scale <50 μm) might be transported via water exchanges through the sediment (Frei et al., 2019). However, microplastic concentrations generally increase with decreasing particle size (Frei et al., 2019) emphasizing the relevance of assessing the lower microplastics size fractions during MAR. Additionally, standardized sampling protocols and monitoring procedures for micro- and nanoplastic analysis (Dalmau-Soler et al., 2021; Pulido-Reyes et al., 2022; Re, 2019) as well as studies reporting exact removal efficiencies during MAR (Fig. 3) are often lacking, which makes the comparison and assessment challenging.

Mobile CECs with low adsorption coefficients (low K_{OC}) generally have a low retardation in MAR systems (Fig. 2) and get transported to greater depth possibly contaminating underlying groundwater compared to less mobile or easily degradable CECs (Collard et al., 2023). However, CECs cover a wide range of chemical properties and variable partitioning coefficients are reported (Fig. 2). Retardation and degradation of glyphosate with depth, for example, showed a linear pattern and a removal rate of 70–80% during the similar process of slow sand infiltration, while adsorption depended on contaminant concentration (Litz et al., 2011). The pesticides bentazon and cycloxydim were shown to be especially mobile with retardation factors below 1.1, while desphenyl chloridazon, methyl desphenyl chloridazon, and imidacloprid showed intermediate mobility with retardation factors below 1.5 (Kruisdijk et al., 2022). Most other pesticides are easily degradable in MAR systems, even though transformation products are potentially toxic. Retardation is further regulated by interactions between soil organic matter and organic contaminants (Roberts and Valocchi, 1981; Sopilniak et al., 2018). Contaminant transport through water and soil during MAR therefore further depends on particle properties, such as shape, size, density, or surface characteristics, especially for microplastics (Frei et al., 2019). Additionally, aquifer properties, soil organic matter content, recharge and precipitation volumes as well as assessment methods further impact measured concentrations of pesticides by altering dilution factors and water quality (Kruisdijk et al., 2022; Page et al., 2014).

Compared to plastic particles, pesticides, and other CECs or persistent organic pollutants, PFAS behave especially different, since they are poorly photo- or biodegradable, and many PFAS are very mobile as well as highly water soluble (Ahrens et al., 2011; Liou et al., 2010; Page et al., 2019; Post et al., 2012; Vaalgamaa et al., 2011), but studies investigating partitioning of PFAS in MAR are scarce (Fig. 2). Therefore, additional studies reporting PFAS soil-water and octanol-water partitioning coefficients have been overlayed in Fig. 2 to show the variety of chemical properties of this versatile class (Geosyntec Consultants NC PC, 2019; Gomis et al., 2015). However, reported values vary between the different isomers and some studies argue to only include the octanol-water coefficients (log K_{OC}), since PFAS are, due to their simultaneous hydrophobic chain and hydrophilic functional group, more likely to sorb to interfaces instead of partitioning into one phase (Guelfo and Higgins, 2013; Higgins and Luthy, 2006; Nguyen et al., 2020). Additionally, different contaminants may compete for sorption sites and less soluble compound sorb stronger and thus release at first retained more soluble compounds (for example short-chained PFAS) after a certain operation time (McCleaf et al., 2017). This leads to PFAS being generally not effectively removed during MAR or other conventional drinking water treatment (Eschauzier et al., 2010; Pramanik, 2015), but again, quantitative studies of the fate of PFAS in MAR systems are lacking (Fig. 3). Additionally, longer-chained PFAS (and other very persistent CECS such as the herbicides simazine and atrazine) can be retained in the unsaturated zone accumulating to several orders of magnitude of the groundwater concentrations and posing a risk to the groundwater below for up to decades (Cáñez et al., 2021; Høisæter et al., 2019; Sörengård et al., 2022; Suárez et al., 2007). This is mostly governed by two processes, namely sorption to soil particles or adsorption at air-water interfaces (Brusseau, 2018). Furthermore, the sum of PFOS and PFOA concentrations in the vadose zone seem to positively correlate with changes in groundwater levels due to lateral recharge (Cáñez et al., 2021). Further studies are necessary to fully understand fate and bioaccumulation patterns (Banzhaf et al., 2017; Sims et al., 2022). Persistent and mobile chemicals such as PFAS have recently been receiving more attention in regulatory processes, but there still is a lack of studies identifying these persistent and mobile CECs.

3.4. Modification of MAR systems to increase contaminant attenuation

It is often mentioned that MAR can be an easy and cost-effective step in the water treatment chain, with the potential to be further improved for CECs by increasing residence times through biofilm formation (Høisæter et al., 2019; Pulido-Reyes et al., 2022), adjusting pH and temperature (Ma et al., 2016), or mixing additional adsorbing materials into the sand such as biochar or granular activated carbon (Hsieh et al., 2022; Takagi et al., 2011; Valhondo et al., 2020). While sand filtration alone removes about half of the microplastics, addition of biochar can remove up to 100% of the microplastics even at high flow rates that usually lower contaminant retention (Hsieh et al., 2022). Especially a thin layer of biochar (i.e. produced at 700 °C from woodchips) resulted

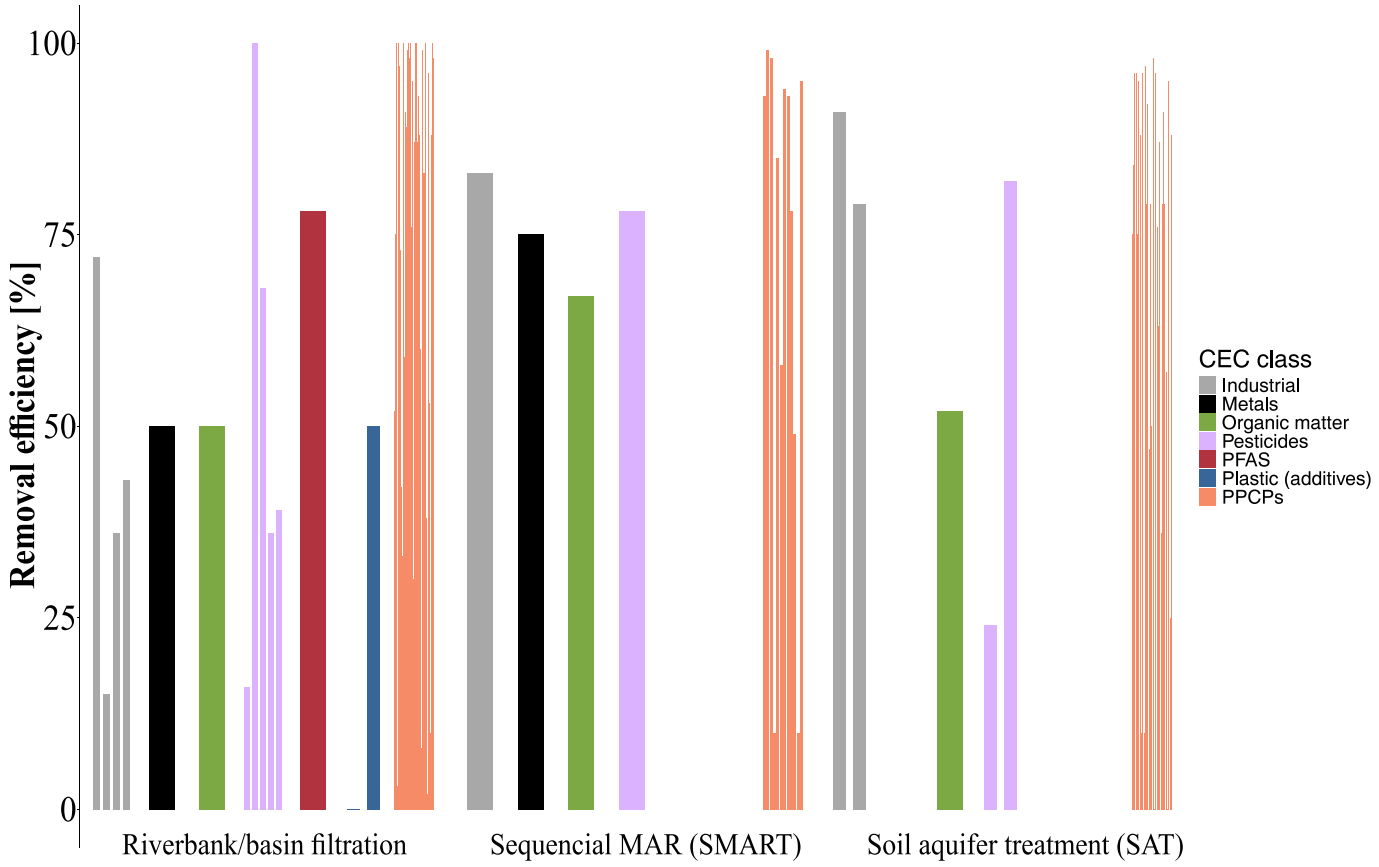

Fig. 3. Reported removal efficiencies in different MAR systems from the reviewed literature. Most studies report a variety of PPCPS as well as some pesticides and industrial contaminants. PFAS and plastics are clearly under-represented, with one or two studies, respectively, reporting exact removal efficiencies. The references and below-laying data can be found in Table S3 in the SI.

in complete microplastic retention during filtration (Hsieh et al., 2022). Recycling and effective removal during multiple filtration cycle further increase cost-effectiveness (Hsieh et al., 2022).

Similarly to MAR, observations of interactions between soil organic carbon content and organic CECs can be made during coagulation and slow sand filtration, which appear to be effective in removing plastic particles. Straining let to especially larger microplastics i.e., above maximum pore size, being retained in the sand column. Smaller particles where additionally retained by attachment, even though not as effective (Na et al., 2021). Thus, if modified accordingly, MAR can be an effective step in removing CECs from infiltrated waters instead of acting as a contaminant pathway into groundwater systems, especially if coupled to other (non-)destructive pre-treatment processes of water (Fig. 1, location 4) such as coagulation, slow sand-filtration, photolysis, sono-chemical degradation, biodegradation, advanced oxidation or reduction, electrochemical oxidation, incineration, activated carbon, ion exchange resins, ozo-fractionation, foam fractionation, or polymeric adsorption (Page et al., 2019; Yoon et al., 2013).

However, water quality during MAR is impacted by different physical, chemical, and biological attenuation processes (Regnery et al., 2017) that must be tuned and monitored. Another approach is sequential MAR technology (SMART), that can be run as a combination of two MAR systems with aeration in between for improved removal rates of CECs compared to conventional bank filtration. Especially using ozone as oxidant during aeration resulted in an increase of 47% for DOC removal and a general trend of increased removal efficiencies of CECs as well as formation of transformation products that are easier to biodegrade (Hellauer et al., 2017, 2018). Finally, carbon-based materials such as activated carbon are commonly used in CEC water treatment technologies (Christensen et al., 2022), since they can efficiently adsorb even difficult-to-treat compounds such as PFAS during water filtration (King et al., 2022; Wei et al., 2020). A possible combination with MAR could further increase treatment efficiency and lower operation costs.

3.5. *MAR as a pathway for contaminants of emerging concern into groundwater systems?*

MAR might act as a pathway of CECs leaching into groundwater systems with PFAS, caffeine, and sulfonamide antibiotics being detected within the nanogram per liter range in more than half of the sampled groundwater in Taiwan (Lin et al., 2015). Minimum travel time criteria for MAR in Europe and the US are often derived from virus survival rates and vary between 50 days to half a year (Fox and Makam, 2009), but pathogens or biological contaminants often show different behavior during subsurface passage than anthropogenic CECs (Bertelkamp et al., 2012; Hamann et al., 2016). While a positive correlation between storage time in the aquifer and pollution risk mitigation seems likely, fate and behavior of those pollutants still require further studies (Regnery et al., 2017; Zheng et al., 2022). To be able to model contaminant behavior, high resolution spatial and temporal data considering a variety of factors is important. However, since this data is often lacking, less accurate first-order rate constants are commonly applied for modelling contaminant transport during MAR (Greskowiak et al., 2017; Sanz-Prat et al., 2020). Detailed understanding of the fate of CECs during MAR, however, is crucial for designing efficient pre- or post-treatment steps (Fig. 1, location 5) to complement natural attenuation processes and ensure sufficient water quality (Patterson et al., 2011). Especially persistent and mobile CECs that enter groundwater systems are of relevance, as shown in this review.

This is especially important to assess for PFAS, since drinking water

regulations are approaching low nanogram per liter ranges (Miljøministeriet, 2024; United Stated Environmental Protection Agency, 2024) and therefore further treatment of the water of modification of the MAR system could be required. PFAS are a prominent example of CECs showing atypical characteristics. Being poorly photo- or biodegradable, soluble in water, and short chain PFAS having low sediment partitioning coefficients, PFAS contrast from other organic CECs in physio-chemical, transport, and retention behavior during MAR (Ahrens et al., 2011; Evich et al., 2022; Page et al., 2019). Furthermore, PFAS comprise a large contaminant class (>16 000 compounds) with different properties and behavior depending on chain length and functional groups (Banzhaf et al., 2017; Evich et al., 2022; Xing et al., 2023) and very little studies report exact properties (Fig. 2) and removal efficiencies (Fig. 3) for PFAS (Alam et al., 2021; Regnery et al., 2017). Thus, PFAS can enter groundwater systems during MAR posing a risk to environment and human health already at nanogram per liter levels (Andrews and Walker, 2015; Gobelius et al., 2018; Skutlarek et al., 2006). Moreover, the varying results on mobility, retention, and occurrence of CECs during MAR presented above emphasize the need for further studies on the transport through and modifications of MAR systems for different CECs as well as standardized sampling and analysis protocols together with regular monitoring.

4. Conclusion and future research directions

Looking at the occurrence of CECs in surface waters, concentrations are often near or above current guideline levels if implemented, even though analytical challenges impede comparison. However, for many CECs presented here, MAR systems can possibly act as a pathway into groundwater systems and drinking water supplies. Especially CECs differing from other persistent organic contaminants in properties and behavior in the subsurface, such as PFAS, are recommended to be studied urgently and thoroughly.

Since application of MAR and the variety of CECs are likely to increase in coming years, effective and adjusted MAR systems are urgently needed to remove the broad variety of CECs. We thus consider following studies and modifications as crucial to ensure safe and clean drinking water in the future.

- Implement regular monitoring and source water tracing to ensure early actions and a good understanding of the site-specific MAR system. This includes a good understanding of the local aquifer characteristics and respective infiltration rates to be able to counteract seasonal peaks in CEC concentrations by utilizing different water sources and operating at respective infiltration rates.
- Focus not only on regulated CECs but also on hazardous CECs which have not been identified yet is crucial for future risk assessments of MAR systems. One important group of CECs are (very) persistent and (very) mobile chemicals (Hale et al., 2020), which need to be characterized on their hazardous potential to humans and the ecosystem.
- Redesign MAR systems to remove CECs by adding pre-treatment steps, coupling different treatment options, or incorporating additional sorption layers while keeping the system as cost-efficient as possible to avoid CECs entering groundwater systems and potential drinking water sources via MAR.

CRediT authorship contribution statement

Tabea Mumberg: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Visualization, Writing – original draft, Writing – review & editing. **Lutz Ahrens:** Conceptualization, Funding acquisition, Methodology, Resources, Supervision, Writing – review & editing. **Philipp Wanner:** Conceptualization, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data used in this review paper and the corresponding references can be found in the supplementary material.

Acknowledgments

This research was supported by the FORMAS Early Career Grant awarded to Dr. Philipp Wanner (2021-00510). We appreciate the help with setting up and guidance on conducting the systematic literature review of Linda Hammarbäck, Librarian at the University of Gothenburg, Sweden. We thank the two anonymous reviewers for their positive feedback.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2024.143030.

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