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A Review of Threats to Groundwater Quality in the Anthropocene

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Abstract:

Awareness concerning sustainable groundwater consumption under the context of land use and climate change is gaining traction, raising the bar for adequate understanding of the complexities of natural and anthropogenic processes and how they affect groundwater quality. The heterogeneous characteristics of aquifers have hampered comprehensive source, transport and contaminant identification. As questions remain about the behavior and prediction of well-known groundwater contaminants, new concerns around emerging contaminants are on the increase. This review highlights some of the key contaminants that originate from anthropogenic activities, organized based on land use categories namely agricultural, urban and industrial. It further highlights the extensive overlap, in terms of both provenance as well as contaminant type, between the different land use sectors. A selection of case studies from literature that describe the continued concern of established contaminants, as well as new and emerging compounds, are presented to illustrate the many qualitative threats to global groundwater resources. In some cases, the risk of groundwater contamination lacks adequate gravity, while in others the underlying physical and societal processes are not fully understood and activities may commence without adequately considering potential impacts. In the agricultural context, the historic and current application of fertilizers and plant protectants, use of veterinary pharmaceuticals and hormones, strives to safeguard the growing food demands. In the context of a sprawling urban environment, waste, human pharmaceuticals, and urban pesticide outputs are increasing, with adequate runoff and sanitation infrastructure often lagging. Finally, industrial activities are associated with accidental leaks and spills, while the large-scale storage of industrial byproducts has led to legacy contaminants such as those stemming from raw mineral extraction. With this review paper, we aim to underscore the need for transdisciplinary research, along with transboundary communication, using sound science and adaptive policy and management practice in order to procure sustainable groundwater quality.

Key words: Water resources, Groundwater quality, anthropogenic activity, contamination, sustainable, transdisciplinary.

1. Introduction

Groundwater quality degradation is a well-recognized phenomenon and has received considerable attention since the industrial revolution (Arias-Estévez et al., 2008; Spalding and Exner, 1993; Von Der Heyden and New, 2004). In spite of this, many aspects concerning the understanding and management of groundwater as a resource remain complex, and adequate information, in many cases, remains elusive (Famiglietti, 2014). In addition to natural heterogeneities, anthropogenic activities in all sectors have been shown to impact and alter the natural water cycle and subsequent groundwater quality (Figure 1). These changes can have wide-ranging impacts on ecosystem functioning and human health.

The vulnerability of an aquifer and its risk of contamination in the anthropogenic environment arises from the complex interplay of the natural dynamics of the hydrological cycle with the physical alterations of the earth surface, water resource exploitation, and waste emissions from anthropogenic activities. Physical changes to the landscape that can lead to increased risk include changes in surface roughness, surface sealing, topographic alterations, river canalization, or construction of artificial water bodies (Bhaskar et al., 2016; Han et al., 2017). In addition to changing land cover, anthropogenic activities include the extensive application of natural and synthetic chemical products. The use of such products has ensured high crop yields, improved human and animal health, sufficient energy, material production, and functional infrastructure.

However, many substances that are widely used today have been shown to be soluble, mobile, and persistent in groundwater, as well as toxic to environmental or human health (Wakida and Lerner, 2005). Many more products are of yet unknown risk, and the risk of chronic exposure to a cocktail of products in any environmental compartment has been thus far difficult to characterize (Kim et al., 2011; Musolff et al., 2010).

Figure 1: Sources of potential groundwater contaminants, as diffuse and point source inputs, from agricultural, urban and industrial settings and with respect to the dominant flow direction (arrows).

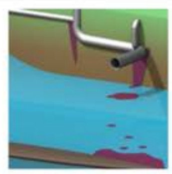
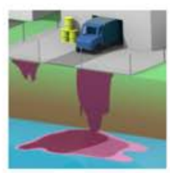
When an aquifer becomes contaminated with harmful chemical products, it may become unusable for decades. The residence time of contaminants within groundwater bodies can be anywhere from weeks to decades, depending on physico-chemical properties of compound and environmental conditions (Chapman and Parker, 2005; Freitas et al., 2015; Moeck et al., 2017a). In addition, the effects of groundwater contamination do not end with the loss of well-water supplies. Several studies have documented the migration of contaminants from disposal or spill sites to nearby lakes and rivers as groundwater passes through the hydrologic cycle (Conant et al., 2004).

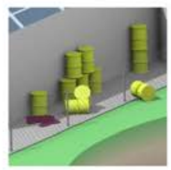
These issues are widely recognized in the scientific community. In many areas of the world, regulatory bodies have implemented concentration limits for a large number of contaminants in drinking water bodies, with nitrate likely being the most notable example. Still, groundwater contamination stemming directly from anthropogenic activities remains a persistent and ever-increasing concern (Lapworth et al., 2017; Wakida and Lerner, 2005). The current expansion of anthropogenic activities is often in direct contrast with what is needed to protect groundwater resources for future use (Howard, 2015; Khan et al., 2016).

In this article, we review some of the major controls on groundwater contamination from anthropogenic activity, and discuss several relevant contaminant classes from these activities. We follow by highlighting a number of case studies in literature that address historic and emerging issues in contaminant hydrogeology, as well as the diversity of issues that arise in different regions of the world. The effect of practices such as land development and waste production on groundwater in terms of pollutant class, sources, and pathways are discussed in order to underscore the myriad of issues that can arise when anthropogenic controls are present on top of natural controls. We focus on dynamics in three major land use types: 1) agricultural, 2) urban, and 3) industrial (Table 1), and the diffuse and point source manner in which they can introduce contaminants to groundwater sources (Figure 1). Due to their relevance in these environments, the following contaminant classes are included: nitrates, pesticides and biocides, pharmaceuticals and hormones, non-aqueous phase liquids (NAPLs), and acid mine drainage (AMD).

The urgent need for an improved understanding of contaminant dynamics in the built environment has been outlined in scientific literature and government directives alike (Bhaskar et al., 2016; Howard, 2015). This review attempts to underscore the complex interaction of groundwater contaminants, sources, and transport processes in heterogeneous environments. The pressing need to secure sustainable groundwater quality, and the call for transdisciplinary and transboundary actions as we move into 2020, are paramount (Montanari et al., 2013; UNDP Sustainable Development Goal 6, 2018).

Table 1: Major contaminants associated with the agricultural, urban and industrial land use categories and their threats to groundwater quality. Images of conceptual contamination pathways refer to Figure 1

Land use Category	Potential contaminating compounds	Remarks on potential pathways and processes	Conceptual contamination pathways
Agricultural	Fertilizer (nitrates and phosphates) application as potential diffuse contamination	When nutrient load exceeds uptake capacity of plants, runoff or infiltration into groundwater bodies may result.	
	Crop protection (pesticides, herbicides etc.) as potential diffuse contamination	Agrochemicals, and their degradation products, can runoff and/or infiltrate into and remain in groundwater bodies for substantial amounts of times.	
	Use of veterinary products (antibiotics, hormones, etc.) as potential diffuse and point source contaminants	Antibiotics and hormones, although currently only detected in low concentrations groundwater, may pose a significant risk to the receiving environment.	
Urban	Wastewater as potential vector for point source and diffuse nitrate and pharmaceutical contaminants	Wastewater may enter groundwater bodies through sewer leakage, poor infrastructure, or via receiving streams down-gradient of wastewater treatment plants.	
	Runoff as potential vector for point source and diffuse nitrate, pesticide, road salts, etc. contamination	Products applied to urban surfaces readily leach into storm water and can reach groundwater at localized or diffuse infiltration points. In combined storm – sewer systems, untreated wastewater can also overflow as runoff.	

	Leachate from solid and liquid waste as potential diffuse contamination	Lack of adequate lining of landfills or inadequate sanitation infrastructure can threaten aquifers through the introduction of solid and/or liquid anthropogenic waste as leachate.	
	Managed aquifer recharge (MAR) and wastewater irrigation as vectors for diffuse nitrate and pharmaceutical contaminants	In many water-scarce regions, wastewater presents a valuable resource. However there is increasing concern regarding aquifers vulnerability underlying such systems, especially when these aquifers are considered as potential drinking water sources.	
Industry	Spills and leaks as potential point source for BTEX (benzene, toluene, ethylbenzene, xylene) contaminants	Subsurface heterogeneities, even in relatively simple porous aquifers, with horizontal bedding features overlying aquitards, compose complex transport processes, posing a significant challenge for the remediation of BTEX spills.	
	Runoff, leachate and precipitate as source and vector for diffuse contamination, and sever water chemistry alteration	Mining exposes minerals sulphates to surface reactions, resulting in radically altered pH conditions in associated waters . This phenomenon, known as AMD, may result in the precipitation or increased solubility of heavy metals.	

2. Natural controls on groundwater contamination

In the context of understanding the propagation of solute fluxes in the subsurface, it is worthwhile to briefly recall the major controls on groundwater storage and quality in the natural environment. Groundwater quantity and quality is controlled by a water balance that can be characterized at several scales (local, regional, and global). Those controls deemed most prevalent to the water balance are discussed here. These underlying factors are important to keep in mind, as they will be built upon and frequently referred to in the construction of conceptual models for anthropogenic groundwater dynamics.

Natural groundwater occurrence and vulnerability depends first and foremost on regional weather conditions and climate oscillations (Gurdak et al., 2007; Randall and Mulla, 2001). In the case of fossil groundwater, this includes historical climatic conditions. The most important climatic parameters are precipitation, humidity and evapotranspiration (O'Driscoll et al., 2005). Precipitation or high humidity serve as major recharge for most aquifers, either through direct infiltration or through indirect runoff regimes (Jones, 2010). Evapotranspiration encompasses further climatic variables such as radiation, temperature, wind dynamics, and heat exchanges, all of which affect groundwater quantity and quality. For example in semi-arid and arid regions, rainfall is erratic and evapotranspiration is often high, resulting in reduced recharge and a concentrating effect of solutes within the unsaturated zone (McMahon et al., 2006).

Topography, vegetation cover, soil types and the underlying geology also play a major role in groundwater dynamics. Steeper slopes tend to increase surface runoff, and decrease groundwater residence time, whereas flat terrains tend to have decreased surface runoff accommodating increased infiltration. Soil types can vary dramatically within and across landscapes, depending on the underlying geology and land surface processes, and dampen or enhance recharge rates (e.g. O'Driscoll et al., 2005). Land cover, including flora (both living and decaying) and surface water bodies, are an additional determining factor for how much water is exchanged between surface and groundwater bodies. In the vadose zone, plant root systems can create preferential flow pathways, while dense floral cover can result in high evapotranspiration rates and reduced surface runoff.

Local and regional geology and tectonic deformation define the physical characteristics of an aquifer, and thus the natural dynamics of groundwater recharge and flow in the subsurface. Mineral-water interaction at the rock surface determines much of an aquifer's qualitative characteristics, such as hardness and pH, among others (Naicker et al., 2003). Sediment matrix characteristics such as the presence of confining layers, or grain size and pore spaces are all products of the parent geology combined with the weathering effects of climate. These physical matrix characteristics will influence aquifer storage capacity, including the rate and magnitude of recharge, flow and transport. Along with aquifer depth, these characteristics will also determine the average residence time of groundwater.

Soils and sediment regularly interact with solutes in all phases through chemical processes such as sorption, ion exchange, solute precipitation, and degradation (abiotic and biotic). In an aquifer, many such processes are controlled by pH-dependent redox conditions. In inorganic soils and sediments, redox reactions are generally retarded, while the presence of organic matter or bacteria in these layers tends to speed up reaction rates. As such, organic material in these layers can, to a large degree, regulate the contamination persistence (Jekel et al., 2015). In general confined aquifers tend to be oxygen-deficient, while unconfined, shallow or fissured aquifers tend to be oxygen-rich. Therefore, whether an aquifer is confined or unconfined will affect the cation exchange processes and reduction taking place. These factors determine the ability of the sediment matrix to degrade certain solutes that pass through its pores.

Hyporheic zones are the interface between surface water bodies and groundwater, and tend to be rich in organic matter and oxygen, resulting in an extremely reactive environment. Healthy riparian zones are able to strongly attenuate many potentially harmful solutes. Thick vadose zones and the presence of organic-rich layers are also very reactive and have the potential to sorb and degrade many chemical products, whereas thin vadose zones as well as highly permeable sands, gravels, presence of fissures or karstic features will do little to filter potential contamination (Pitt et al., 1999).

Groundwater recharge in shallow or unconfined aquifers normally occurs in a diffuse manner. Infiltration takes place relatively homogeneously in space, either over the entire wetted area or along preferential pathways. In confined, karstic, or fractured hard-rock aquifers, recharge and flow occur principally within networks of cracks, fissures, and other localized openings, so that both fast and slow recharge is often observed. Some flow pathways in these environments are active only after precipitation events or snow melt (Ballesteros et al., 2015). In fractured aquifers, the direction of flow might not coincide with the hydraulic gradient, as movement is also governed by fracture orientations in these settings (Graf and Therrien, 2007). As a result, in heterogeneous aquifers, water and solutes can spread in complex patterns by following both fast and slow flow pathways (Hunkeler et al., 1999).

Unconfined shallow and karstic aquifers alike are generally subject to the direct vertical inflow of surface waters. The quality of these infiltrating surface waters are therefore extremely consequential in these scenarios, as unconfined and karstic aquifers constitute a considerable amount of global groundwater reserves – approximately 25% of the global population is dependent on karstic groundwater (Ghasemizadeh et al., 2012 (refer to Section 5.6 for contamination risks to karst systems). Contrarily, inflow to confined or deep aquifers is often indirect and may take years or decades. This fact can safeguard confined aquifers from degraded surface waters, but it can also present major remediation challenges once contaminated potentially compromising groundwater quality for decades due to long groundwater residence times (Chapman and Parker, 2005).

It can be seen that detailed knowledge on the nature of flow and chemical reactivity in groundwater bodies has been greatly developed over the last century, but the connection between hydrogeology and solute dynamics is still a developing science. The heterogeneity of most natural environments, combined with a lack of detailed aquifer information, makes a straightforward characterization of many systems nearly impossible (Hakoun et al., 2017). In general, highly complex systems are particularly vulnerable to contamination (Freitas et al., 2015; Wu and Hunkeler, 2013). Karstic aquifers for example are notoriously complex due to high spatial variations in porosity, making the data demand for proper characterization much more intensive (Ghasemizadeh et al., 2012).

3. Anthropogenic controls on groundwater contamination

While the intrinsic vulnerability of an aquifer depends largely on the natural characteristics of its setting, contamination risk from anthropogenic activities depend predominantly on land use (Faye et al., 2004). Anthropogenic alterations are known to have significant impacts on every aspect of the water cycle, both by changing the magnitude of existing terms as well as the addition of new terms (such as leakages, extraction, or irrigation). Land development and land use practices significantly determine contaminant sources and the processes which control the contaminant propagation in the environment.

This review classifies land use into three broad categories (1. agricultural, 2. urban, and 3. industrial), which can be further split in a number of different ways. For example, to account for pastures versus croplands, residential areas versus streets, or primary material extraction sites versus production facilities. Another important example are road networks which connect the three defined land use categories. Although the density and expansion of road networks are generally higher in urban and industrial settings, they account for strong negative environmental impacts as they span across different landscapes (Ledford et al., 2016). Different land use categories rarely exist in isolation: residential areas may interface into pastures and fields, urban agricultural plots are a common feature in many cities, industrial parks may be large and set apart or they may be relatively small and within city limits, and intensive farming can bring elements of industrial activities into the agricultural setting. Current anthropogenic land use processes may also overlay sites of historical activities, including landfills or nuclear waste disposal facilities to name a few. In addition, there are relevant societal factors that define the specifics of land use activity and thus partially define risk of contamination. These include any prevalent human illnesses (refer to Section 5.5 for example) as well as regulations on the use of chemical and pharmaceutical products (see Table 2).

Thus, water quality experts who are looking to assess the risk of anthropogenic contamination to groundwater for a specific region, in addition to considering the natural setting, should not exclude the consideration of current local societal factors in context with existing and historic land use activities such as infrastructure and waste treatment technologies.

3.1 Land development

It is estimated that since 2010, agricultural lands cover 30 - 40% of the world's usable land surface (FAO, 2019). Another 3% is occupied by urban and industrial landscape, including primary material exploitation (FAO, 2009). However, the density and intensity of activities are of equal, if not greater, importance to their spatial extent. For example, because urban areas comprise diverse land use features, high population densities, expansion of road networks, intense resource consumption, and high waste production, cities may pose an equivalent or even higher risk to groundwater contamination compared to adjacent agricultural areas (Han et al., 2017; Marsalek et al., 2007).

Changes to land cover are generally the first step in developing an area. This includes amending vegetation, topography, soil permeability, and surface water body characteristics, all of which feed in to the recharge and groundwater dynamics. Examples of the effect of these changes include thinning or clearing any existing vegetation, altering existing topography, wetland drainage, soil tillage, or addition of exotic vegetation types (usually for cultivation). Modification of vegetation stimulates change in evapotranspiration rates, and can either increase or decrease recharge, depending on the water demands of the dominant plant species and changes in solar radiation absorption or reflection. Deforestation, for example, leads to a decrease in plant transpiration, often stimulating increases in recharge (Brown et al., 2005).

Topographic alterations result from many different land uses. This includes creating artificial depressions or mounds for purposes such as flood control or installation of specific infrastructure. Perhaps one of the most prominent examples is excavation and tunneling in industrial mining operations. These excavations can create artificial surface water bodies, as well as mounds of discarded overburden or tailings. Tunnels can become preferential pathways when conditions become saturated.

Altering the physical characteristics of surface water bodies can change the direction of flow and the dynamics of interactions between water bodies. In catchments where rivers or lakes are present, groundwater-surface water interactions can be intensified, dampened, or reversed by river straightening and other alterations (Kurth et al., 2015). Riverbed alteration also tends to stimulate a loss of riparian zones and cause deeper riverbed incision, which can decrease the capability of this important interface to favorably control mass fluxes from surface to groundwater or vice versa (Groffman et al., 2002).

In addition to changes stimulated by land development, artificial source and sink terms that arise from infrastructure must be added to the conceptual model of an anthropogenic water balance. Infrastructure includes road networks, buildings, pipelines, and drainage networks, among other elements. Heavily modified landscapes that have been built up with such elements change the water balance in significant ways, which in turn has an impact on solute mass fluxes and water quality (Strauch et al., 2008). The specific impact that infrastructure has on the local water balance and groundwater storage is highly variable and depends on the type and upkeep of each element as well as the effect of the ensemble of elements present (Han et al., 2017). A number of studies have found that anthropogenic solutes in groundwater are sourced directly from infrastructure (Bartelt-Hunt et al., 2011; Pitt et al., 1999).

Another class of infrastructure worthy of mention is the extensive networks of gasoline storage tanks and pipelines used in the industrial sector. These elements are potential sources of contaminating non-aqueous phase liquids (NAPLs) through leakages or accidental spills (refer to Section 5.6 for example). The broad distribution of petrol stations, and the abundant networks of above and underground tracks and pipelines carrying petroleum products illustrate a potential significant risk to groundwater quality.

Surface runoff is notably impacted by land development. This is consequential for quantity issues as well as quality, as runoff is a carrier for chemicals present in the atmosphere and leached from surfaces (Lesser et al., 2018; Wakida and Lerner, 2005). In agricultural areas, runoff is activated by storm events as well as by irrigation patterns. In urban and industrial areas, runoff is principally constrained by precipitation events only, and thus is only active during a storm and for a certain lag period following the storm. When surface runoff from the human environment infiltrates the subsurface, it tends to infiltrate in a more localized point- or line-source manner (Han et al., 2017). Because of these dynamics, the behavior of any solutes originating from runoff may be markedly different than the behavior from other sources such as wastewater contamination (discussed further in Section 3.3), which is largely weather-independent (Mutzner et al., 2016). It is sometimes possible to exploit these behaviors in order to detangle sources of contamination in mixed land use areas. In agricultural areas, however, the buildup of solutes in soil can create a nearly constant input over time so that temporal dynamics no longer provide insight on sources (refer to Section 5.3 for example).

Paved or compacted surfaces, including roads, building plots, and parking lots are one of the most common components found in the anthropogenic environment. These surfaces stimulate increases in surface runoff from storm events and snow melt, as well as major decreases in diffuse infiltration and evapotranspiration are common in urban and industrial environments (Marsalek et al., 2007). Increases in groundwater storage is sometimes observed in heavily paved catchments, despite the decreases in diffuse infiltration (Minnig et al., 2018). Decreases in storage may also be observed, and this is dependent on the relative magnitude in changed evapotranspiration, infiltration, as well as the presence of artificial recharge sources (Han et al., 2017). Looking forward, it is assumed that the spatial extent of impermeable surfaces will continue to expand as estimates have projected that nearly half of all urban infrastructure that will exist in 2030 has not yet been developed (Biello, 2012).

Many infrastructural elements have been designed with the sole purpose of controlling surface runoff in order to protect against flooding and water quality. Storm water infrastructure in particular is a decisive factor in controlling the location and infiltration rate of runoff especially in urban and industrial areas (Pitt et al., 1999). Two common infrastructural storm water systems include combined sewers, which are wastewater sewers equipped with drains so that storm runoff is collected into the sewer networks, followed by separated systems, which are more popular for modern city planning as they incorporate drains exclusive for storm runoff that release into nearby surface water bodies (Hensen et al., 2018). Combined sewers run the risk of overflow during heavy storms, releasing untreated wastewater along with storm runoff into the environment (wastewater infrastructure is detailed in Section 3.3). However, separate systems will drain storm runoff directly into surface waters, which in turn may be in direct communication with groundwater bodies). Separated systems may also run the risk of contaminating groundwater with untreated wastewater, either as a result of underground leakages into storm drains or through faulty or illegal pipe connections between storm water and wastewater networks (Panasiuk et al., 2015). Other systems that handle storm runoff include retention basins which are permanent storm water storage ponds, detention basins which are temporary storage ponds that slow-release storm waters, or infiltration infrastructure (including basins, swales, and trenches) which direct storm water into groundwater in areas of high permeability. Contemporary efforts in green infrastructure have popularized further concepts such as rainwater harvesting to control the amount of runoff generated from storm events.

Much of the literature concerning runoff dynamics and pollution is focused on surface water studies (Karn and Harada, 2001; Lee and Bang, 2000). However, groundwater pollution from storm runoff is a known phenomenon and several notable studies exist on the topic (Pitt et al., 1999; Voisin et al., 2018), including discussions on the efficacy of certain storm water infrastructure.

3.2 Land use practices

On top of development, many anthropogenic land use practices have the potential to influence groundwater quality. Chemical treatment of the land and infrastructure is a very common practice for purposes such as plant and infrastructure protection and pest control. The application of plant nutrients and plant protection chemicals is a widespread and ever-growing practice in the agricultural sector, and the resulting groundwater contamination is a ubiquitous and a longstanding issue (Gonçalves et al., 2007; Hakoun et al., 2017). Products used in the agricultural sector can reach groundwater along diffuse pathways through direct infiltration or in a more focalized manner through runoff (Kaczala and Blum, 2016). Alongside liquid chemical applications, the spread of manure in agriculture is also commonly practiced, and also acts as a pathway for a number of chemicals into the environment. The storage and effective pretreatment of manure is expensive so that direct application of untreated product is not an uncommon practice (Boxall et al., 2003).

Urban and industrial activities are known to practice chemical application for the purpose of pest control, or to optimize material properties. Pest control products in these environments are generally referred to as biocides. As opposed to agricultural products which are used almost exclusively outdoors, chemical products in the urban and industrial sector are applied both indoors and outdoors. This diversifies potential sources and pathways of these products into the environment, and has important implications for the biogeochemical processes that a product will undergo. Briefly, outdoor sources of contamination are subject to degradation via photolysis while on surfaces, followed by biodegradation in soils. Indoor sources will follow slightly different degradation pathways, particularly if they pass through a wastewater treatment plant. However, degradation in these environments is often only partial, leading to a multitude of transformation products. In addition, outdoor products may still end up in sewer networks in combined sewer systems which tend to result in focalized infiltration, either as point- or line- sources, into the groundwater environment (Hensen et al., 2018).

Within agricultural areas, the act of irrigation can influence local groundwater flow paths, as the artificial input of water tends to raise the local water tables or create shallow saturated lenses. These shallow saturated layers from irrigation can make soil and groundwater more susceptible to evapotranspiration, sometimes resulting in net losses of water as well as concentrating precipitated salts and contaminants in the uppermost soil layers (Gning et al., 2017). Refer to Section 5.2 for an example of this phenomenon.

Surface and groundwater exploitation can also be a consequential practice for water quality. Water is extracted for irrigation, drinking water supply, electricity generation, and during industrial operations such as mining. Mining and ore refining operations largely extract groundwater either in the process of decanting open pit mines where groundwater regularly infiltrates, or to dewater underground workings. Both abstraction and decanting of groundwater can alter natural groundwater flow and may lead to regional groundwater desiccation (Custodio, 2005). The process of groundwater rebound upon mining and pumping cessation is a well-known process with implications for groundwater quality (Henton, 1981). Indeed, water pumped from any water body has the potential to impact groundwater storage and stimulate changes in groundwater recharge pathways (Khan et al., 2016).

We re-emphasize that historical practices are often of equal importance as contemporary practices that pose a risk of contamination, particularly in the case of chemical applications. Depending on factors such as unsaturated zone thickness, permeability, climate cycles, and storage time, products applied directly at the surface can remain in soils and aquifers for decades (Baillieux et al., 2015). This means that groundwater contamination stemming from chemical application may remain an unsolved issue long after the application practice has ceased.

3.3 Waste production

Because of the sheer magnitude of solid and liquid waste produced in the human environment, these two major contaminant sources are among the most commonly discussed in groundwater literature (Baba and Ayyildiz, 2006; Grimmeisen et al., 2016; Schirmer et al., 2013). The importance of proper waste handling for environmental protection and human health is widely acknowledged, but there are many cases of inadequate or deteriorating waste facilities around the world. Waste and wastewater infrastructure is quite heterogeneous on a global scale, so that risk of contamination is likewise variable (e.g. Laner et al., 2012; Okumu-Okot, 2012). It has been reported that globally, some 80% of wastewater is released into the environment untreated (WWAP, 2017).

Industrial activities collectively produce more waste than agriculture or urban activities combined (Park et al., 2005), with mining activities in particular generating the largest volume of waste handled in the world (ICOLD, 1996). Exponential industrial growth over the last decades within the raw materials, energy production, and engineering sectors has led to the release of an ever-increasing, diverse range of waste products into the environment, which has left its mark on groundwater quality (Manamsa et al., 2016; Musingafi and Tom, 2014). Refer to Section 5.7 for an example.

Concurrently, treatment of urban waste and wastewater remains a persistent issue when we consider that more than half of the world's population currently lives in cities (World Bank, 2018). According to the UN World Urbanization Prospects 2018, this number is expected to increase to nearly 70% by 2050. This brings about an acute risk for urban populations who find themselves in close proximity to poorly managed waste, and creates heightened risks for urban groundwater resources. Refer to Section 5 for an example.

3.3.1 Solid waste

The presence, type, coverage, and upkeep of infrastructure such as landfills (including tailings facilities) may be among the most important factors determining the resulting quality of groundwater and the risk of contamination from solid waste products. Landfills are the most common method of solid waste disposal worldwide, and are still often the cheapest disposal option, especially for the large quantities of industrial waste (Han et al., 2014; Longe and Balogun, 2010). Although landfill leachate is a known source of contaminants in groundwater (e.g. Peng et al., 2014), solid lining of landfills is not enforced everywhere. In addition, landfills with poor or nonexistent lining dating from the industrial revolution are ubiquitous. Currently, areas without access to effective storage or disposal systems may rely on shallow underground disposal for their solid waste.

3.3.2 Liquid waste

For liquid waste, dedicated infrastructure includes drains, sewer networks, septic tanks, and pit latrines. Centralized wastewater collection systems, connected to an adequately functioning wastewater treatment plant (WWTP) with up-to-date treatment technology and regular maintenance are the preferred solution, as they generally have a better track record for keeping wastewater separated from the environment (Munz et al., 2017). Underground and above-ground septic tanks or French-drains are more common in suburban, semi-rural and rural environments, areas that are distanced from dense city networks. Although septic tanks have higher incidence of leakage than more centralized infrastructure, they still perform better than pit latrines, or direct emissions (open, decentralized waste disposal), which generally lack a proper impermeable barrier from soils into the environment. Developing countries are struggling to move away from open defecation (Figure 2), and although improvements are being made, many sanitary solutions are still based on water-dependent technologies, such as the flush system, which are difficult to implement in water scarce countries. The rapid population growth, particularly in India and China, calls for urgent solutions to sanitation deficiencies, as wastewater is often discharged without treatment.

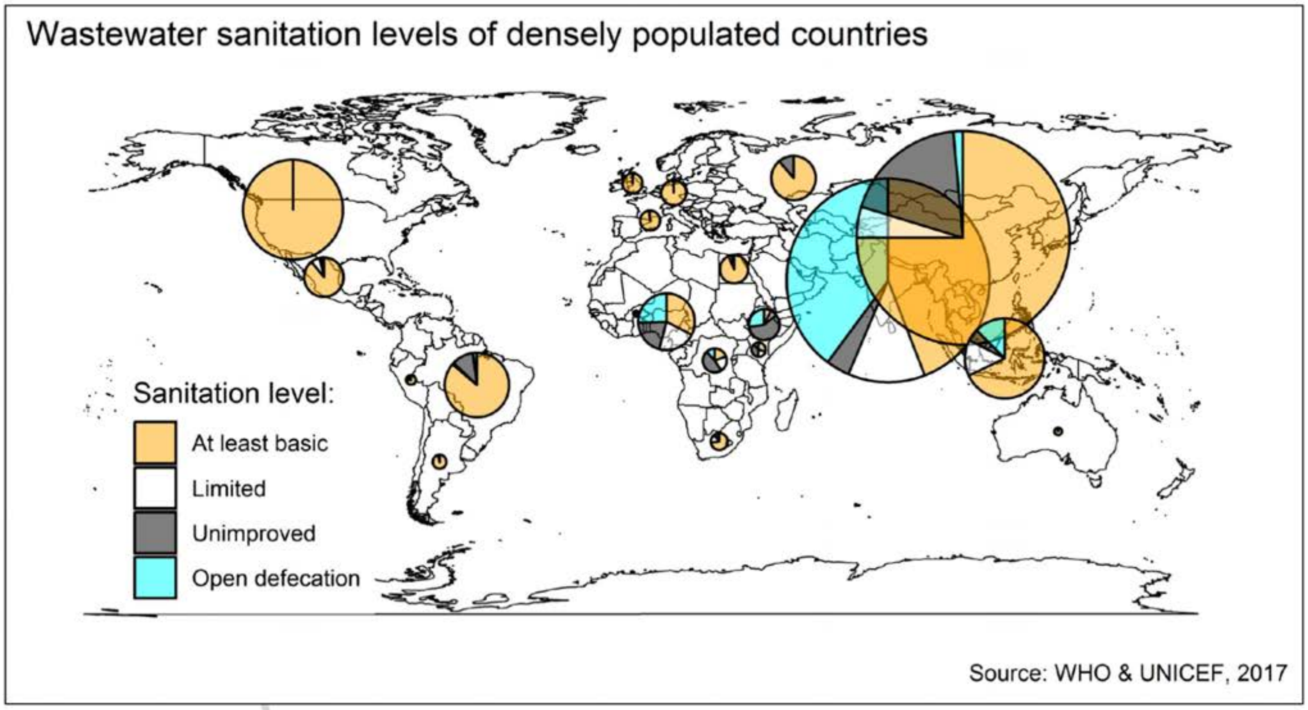


Figure 2: Wastewater sanitation levels of densely populated countries, with pie-sizes relative to populations size (note, the population of India and China is divided by 2 here for scalability), with their levels indicated as ranging from open defecation to at least basic sanitation (WWAP, 2017; https://aashdata.org/data/household).

In urban areas, wastewater can reach shallow groundwater via several routes, including direct leakage from infrastructure or infiltration from surface water runoff (Wakida and Lerner, 2005). Streams and rivers are common recipients of both treated and untreated wastewater, in which case they may act as line sources of contamination. Even where cities are serviced largely by centralized pipelines and WWTPs, treatment facilities are unable to completely remove all contaminants, and this is especially true for emerging organic contaminants (covered in more detail in Section 4). The effluent of treated water from these treatment plants will be a common source of these products, regardless of the treatment method (Lapworth et al., 2012). Wastewater infiltration can sometimes be identified as a source of groundwater contamination through the presence of a number of different chemical or biological indicators specific to anthropogenic activity (Panasiuk et al., 2015). Refer to Section 5.3 for an example of disentangling urban versus agricultural wastewater contamination.

3.4 Wastewater reclamation

The reclamation of used waters for a variety of ends (including for irrigation and use in industry) has been practiced to varying degrees for several centuries, especially in arid and semi-arid environments (e.g. Lesser et al., 2018). As issues of water security continue to increase, wastewater reclamation is an appealing practice that can safeguard a region's adequate supply (Tweed et al., 2007). With the variety of techniques in use for treating or storing reclaimed water improving, the risks of contamination stemming from imperfect or no wastewater treatment prior to its reuse is lowered (San-Sebastián-Sauto et al., 2018). This is in part also to the efficacy of soils used to filter out a large portion of contaminants that are present.

One such reclamation practice is that of managed aquifer recharge (MAR) that uses wastewater (MAR practices in general may use a variety of source waters). MAR can involve the intentional infiltration of wastewater into an aquifer via spreading, percolation, or injection for use such as water supply, ecosystem sustenance, or to control flow fields (Dillon et al., 2009; Bonilla Valverde et al., 2018). An example of existing MAR methods that is relevant to the present discussion is the practice of soil aquifer treatment (SAT), which involves soaking wastewater through the soil and vadose zone into (usually) unconfined aquifers, where it can be stored on a seasonal basis (Sharma and Kennedy, 2017). Other MAR methods such as bank filtration are practiced in more humid regions as well, as an answer to the issue of groundwater quality rather than quantity (e.g. Hiemstra et al., 2003). Apart from increasing water availability, MAR can be used to build up a local groundwater mound that serves as a hydraulic barrier to prevent inflow of contaminated water from areas upstream (Hendricks Franssen et al., 2011; Moeck et al., 2016).

Another common water recycling practice is crop irrigation with urban or industrial effluent. Many examples of long-standing wastewater irrigation practices can be found in places such as Asia (Tang et al., 2004), the Middle East and across Latin America (Bonilla Valverde et al., 2018). The arid Mezquital Valley in Mexico claims to be the world's largest and one of the oldest documented untreated wastewater irrigation systems (Lesser et al., 2018). While wastewater irrigation is an appealing practice as a means of water reuse, and is anticipated to become more important and widespread in the future, it is not without risk of contamination (Tang et al., 2004). Biological contamination such as coliforms are mostly filtered from wastewater as it passes through soil, but other products such as heavy metals, nutrients, and organic contaminants can persist for years or decades at these sites (Gallegos et al., 2015).

4. Major contaminants of groundwater

In this section, we offer details on the characteristics of a number of prominent groundwater contaminants. Many chemical compounds with contamination potential are well-characterized and regulated, but the continued development of new products coupled with poor knowledge of aquifer characteristics and groundwater quality is hampering sustainable water management in many areas (Mateo-Sagasta et al., 2017; Sorensen et al., 2015). In addition, many newer products on the market are of yet-unknown toxicity, especially at low concentrations or when found as part of a cocktail of other products (Munz et al., 2017; Musolff et al., 2010). Details on existing regulations (or lack thereof) for concentration limits in groundwater are available for many developed countries, and guidelines for drinking water quality are provided by the World Health Organization (Table 2). In many countries however, contaminants are either not regulated, or regulation limits are difficult to implement and are therefore overlooked and often not enforced (Knüppe, 2011). Table 2 gives an overview of some of the common contaminants regulated by industrialized, chemical producing countries.

Table 2: Contaminant regulations and restrictions from leading chemical producing countries with regard to drinking water

Parameter (µg/l)	WHO[1]	EU[2]	USA[3]	China[4]	Canada[5]	Switzerland[6]
1,2-dichloroethane	50	3.0	5.0		100	
Alachlor	20	Banned (2006)	2.0		7.8	Banned (2012)
Aldicarb	10	Banned (2003)	1.0		1.0	0.1
Aluminium		200			no limit listed	5.0
Antimony	20	5.0	6.0		6.0	
Arsenic	10	10	10	5	10	2
Atrazine	2.0	Banned (2003)	3.0		1.8	Banned (2012)
Barium	700		2000		1000	
Benzo(a)pyrene	0.7	0.01	0.2	0.0028	0.015	
Boron	500	1000			5000	
Bromate	10	10	10			
BTEX	10	1.0	5.0		370	100
Cadmium	3.0	5.0	5.0	5.0	5.0	0.5
Carbamazepine	no limits defined					
Chromium	50	50	100	50 (Cr^{6})	50	10
Copper	2000	2000	1300	1000	1000	20
Cyanide	70	50	200	50	5.0	
Diclorfenac	0.25	No limits defined				
Fluoride	1500	1500	4000	1000		
Ibuprofin	no limits defined					
Iron		200		300	300	50

Lead	10	10	15	10	10l	1.0
Manganese		50			50	20
Mercury	1.0	1.0	2.0	0.05	1.0	0.1
Metholachlor	10				7.8	0.1
Nitrate	50000	50000	10000 (as N)	10000 (as N)	13000	25000
Nitrite	3000	500	1000 (as N)			50
Pesticides – Total		0.5				
Pesticides (individual)		0.1				0.01
PAHs		0.1				0.1
Selenium	10	10	50	10	1.0	1.0
Simazine	2.0	Banned (2003)	4.0	10	10	Banned (2012)
Sulfamethoxazol	no limits defined					
Tetrachloroethene	40	10	5.0		21	0.1
Trichloroethene	20	no limits defined				
Toluene	700		1000		2.0	0.1
Uranium	15	no legal limit	30	30	20	15
Vinyl chloride	0.3	0.5	2.0			0.1
Zinc			7400		7.0	100

1 Guidelines for Drinking-water Quality, Fourth Edition; World Health Organization; 2011

2 EU Commission, 2008

3 Clean Water Act 1972. EPA 1992; Safe Drinking Water Act (SDWA; USEPA, 1996).

4 Ministry of Health, 2007, National standard of the People's Republic of China

5 Canadian or B.C. Health Act Safe Drinking Water Regulation BC Reg 230/92, & 390 Sch 120, 2001. Canadian Council of Ministers of the Environment: Water Quality Guidelines for the Protection of Aquatic Life.

6 BAFU 2015 GSchV; https://www.bafu.admin.ch/bafu/en/home/topics/chemicals/glossary-of-pollutants

Guidelines for drinking-water quality incorporating 1st and 2nd addenda (3rd ed.). World Health Organization. 2008.ISBN 978-92-4-154761-1.

Improving understanding on the extent and behavior of contaminating compounds in the ground, and the cumulative effect of a mixture of products remains a pressing topic for environmental scientists and decision makers (Botkin and Keller, 2011; Kunz et al., 2016; Moschet et al., 2014). Despite the complexity of solute transport, reactivity and potency in the subsurface, these are important factors which need to be understood in order to determine whether the regulation or mitigation of compounds, or the rehabilitation of aquifers, is effective in protecting or improving groundwater quality.

4.1 Inorganic contaminants – Nitrogen

Nitrogen is an inorganic chemical that is naturally present in the environment. At high enough concentrations, it becomes toxic to environmental ecosystems and to human health. Several human activities may entail elevated levels of nitrogen in groundwater, including the application of plant fertilizer and the production of wastewater. Nitrate (NO_3^-) in particular is one of the most common contaminants measured in aquifers globally, and is the most mobile form of nitrogen (Spalding and Exner, 1993).

Persistence of nitrogen in the subsurface is principally governed by the biological reactions of nitrification and denitrification, which are in turn a function of environmental redox conditions. In the absence of oxygen, reducing conditions favor denitrification: the conversion of nitrate into nitrogen gas. In the presence of oxygen, ammonia or nitrite readily oxidize into the highly mobile nitrate. In such oxidizing conditions, nitrate becomes relatively inert due to the fact that it is negatively charged, and is thus unlikely to sorb to substances in the unsaturated zone such as clays (Fetter et al., 2017).

Recorded nitrate contamination of groundwater through agricultural land use has a relatively long history, and agriculture is assumed by-and-large to be the largest source of nitrates in the environment. It is estimated that 50–70% of all nitrogen applied to crops is lost from the soil-plant system through diffuse leaching (Green et al., 2008; Spalding and Exner, 1993). Although fertilizer application is being moderated in many countries, the global fertilizer production rate continues to increase (European Commission, 2008; FAO, 2018) , with 113 million tones consumed in 2014 (Figure 3). Additionally, many places where agricultural nitrogen application has been considerably reduced through regulations (e.g. Switzerland and the European Union), groundwater nitrate values often continue to exceed official drinking water limits (Decrem et al., 2007; European Commission, 2008). In some cases renewed concentration highs were measured post-remediation or regulation (Baillieux et al., 2015), with studies also reporting delays in response of groundwater quality post-regulation (Gutierrez and Baran, 2009; McMahon et al., 2006).

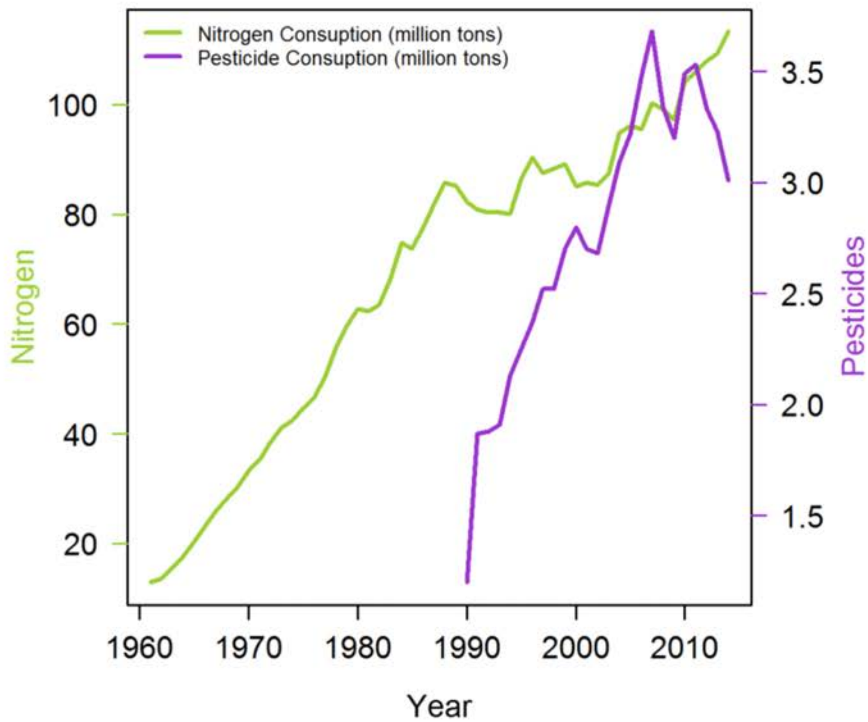


Figure 3: Total fertilizer and pesticide used globally in million tons per year from 1961-2014 (FAO, 2018, 2009). Note that although pesticides were already in widespread use, global monitoring data for pesticide consumption is only available post 1989.

Nitrogen contamination of groundwater is also a relevant issue in many cities (e.g. Appleyard, 1995; Tang et al., 2004). Globally, the magnitude of urban nitrogen contamination is smaller than its counterpart in agriculture, though the variety of urban sources are much higher. Nitrogen sources in urban areas include wastewater, solid waste, water supply, construction zones, urban parks and gardens, storm water, and atmospheric deposition (Wakida and Lerner, 2005). Groffman et al. (2004) hypothesize that this variety of sources and types of nitrogen released in urban environments will have an impact on the microbial communities involved in nitrogen transformation processes, potentially rendering them less efficient. Wastewater in particular is one of the most commonly discussed emitters of urban nitrogen. Wastewater nitrogen occurs most often in the form of ammonia, though it can be readily transformed into nitrates in the oxic conditions of some aquifers (Fukada et al., 2004; Musgrove et al., 2016).

4.2 Organic contaminants

Organic contaminants (OCs) are a diverse class of products that have widespread use in agriculture, urban, and industrial environments. They include a range of hydrocarbons, pesticides, and pharmaceuticals, among others. These products can occur in groundwater either as pure compounds or a mixture of compounds, some of which are readily metabolized into stable transformation products (equally known as degradation products). Some OCs are partially or fully hydrophobic, therefore, they may occur dissolved in water or in a separate phase (Bhatt et al., 2007; Fetter et al., 2017). A large number of OCs have been linked to environmental degradation and risk to human health by researchers as well as policy makers (Fent et al., 2006; Gavrilescu et al., 2015).

Concentrations of measured organic contaminants in groundwater range from nanograms per liter to milligrams per liter (Conant et al., 2004; Hunkeler et al., 2004; Moeck et al., 2017b). Observable concentrations and attenuation rates of many organic contaminants can be attributed either to transformation products, to sorption, or to dilution (Schreglmann et al., 2013). Some compounds are easily degradable and therefore short-lived, while others are persistent and either become mobilized in the aqueous phase (the case for uncharged species), or sorb to soils (the case for charged species). Additionally, some degradation products have been found to have a higher toxicity than their parent compound (Banaszkiewicz, 2010; Sinclair and Boxall, 2003).

4.2.1 Pesticides

The recognition of pesticides as environmental contaminants was brought to people's attention in the early 1960s. Chemical pest control has become a staple of agricultural land development and is also widespread in urban and industrial development. Global monitoring data (starting from 1989) has shown a continued increase in pesticide consumption (Figure 3), with a maximum of 3 million tons consumed in 2007. However, as pesticide use has become more heavily regulated over the past two decades, in particular within Europe and North America (Table 2; European Commission, 2008; FQPA, 1996), a decline in global pesticide consumption is noticeable (see Figure 3). Despite regulations, pesticides remain a persistent issue for global groundwater resources (refer to Section 5.2 for example).

Although groundwater pesticide contamination has been widely reported, current approaches in tracing the environmental degradation and making relevant predictions on their persistence are nonetheless limited (Elsner and Imfeld, 2016; Kunz et al., 2016). Characterizing the dynamics of these products in the subsurface is complicated due to the differences in degradation and sorption rates (the two most important processes governing pesticide persistence), which are a function of individual pesticide compounds as well as sediment and aquifer matrix characteristics (Arias-Estévez et al., 2008). Porous aquifers are generally better at filtering pesticides from the groundwater, while karstic aquifers are more prone to long-term pesticide contamination issues due to rapid flow and low sediment reactivity (Andreo et al., 2006).

Pesticide products are diffuse and ubiquitous in agricultural landscapes, and most major groundwater contamination studies for pesticides are carried out in these areas (e.g. Baran et al., 2008; Gonçalves et al., 2007; Hakoun et al., 2017). In addition, pesticides are the most commonly detected organic contaminants in urban groundwater (Stuart et al., 2011). In cities, pesticides are used in places such as urban parks and gardens, urban agricultural plots, on roads, rail tracks, and sidewalks, in building material (including insulation and paint) and in household protection products (Mutzner et al., 2016). It is known that storm water runoff is a major pathway of pesticides from agriculture, and the same is true of cities (Hensen et al., 2018).

There is a large amount of overlap in product use across environments – many products available on the market today are used outdoors in both agricultural and urban environments. This creates a challenging task of source identification in aquifers impacted by mixed land uses (refer to Section 5.3 for example). The detection of more specific products used in building materials may aid in defining urban versus agricultural sources in groundwater. For example, in many areas of the world, the herbicide *prometon* is generally only applicable to urban areas (Capel et al., 1999), and can therefore be used to clarify an urban signature. In addition, in many cases pesticides that reach environmental waters through sewer leakage and wastewater effluent – i.e. indoor sources – often follow slightly different degradation pathways and can thus be classified as largely urban in origin (Wittmer et al., 2011).

Historically utilized pesticides that are now banned (see Table 2) continue to be measured in groundwater bodies as persistent compounds, and it is not always clear precisely why this is the case (Gonçalves et al., 2007). Current approaches in tracing the environmental degradation and making relevant predictions of their persistence within groundwater bodies are limited (Elsner and Imfeld, 2016; Kunz et al., 2016). Concentration time series, parent-to-metabolite compound ratios, and compound-specific isotope analysis are some of the current approaches to determining pesticide contaminant sources and pathways in groundwater bodies (Schreglmann et al., 2013).

4.2.2 Pharmaceuticals

Pharmaceuticals as contaminants have become a primary concern in recent decades due to their prevalence in modern society, and are increasingly found in anthropogenically impacted surface and groundwater bodies in many regions (e.g. Barnes et al., 2008; K'oreje et al., 2016; Lesser et al., 2018; Loos et al., 2010; Peng et al., 2014). Pharmaceuticals pose a particular risk, as they are purposefully designed to be bioavailable, and have effects which could go beyond the intended recipient once they enter the environment (Kaczala and Blum, 2016).There are few concepts as to what it might take to remediate existing and future pharmaceutical problems in groundwater bodies (Horvat et al., 2012). And indeed, to date there are no, or only very limited, legislative limits on pharmaceutical concentrations in groundwater (Küster and Adler, 2014; see Table 2).

For reasons similar to pesticides, characterizing the propagation of pharmaceutical products in the subsurface is not a straightforward task. Laboratory-scale experimentation has shown that individual product's sorption coefficient will be a major determiner of persistence and mobility (Scheytt et al., 2005), and some product degradation rates have been shown to be highly dependent on soil and aquifer redox conditions as well (Banzhaf et al., 2012). Studies have suggested that characteristics of the soil-aquifer matrix are more important than the characteristics of individual pharmaceuticals in determining their degradation (Hebig et al., 2017). This would explain why many field-based experiments have come up with contradictory results on the persistence of specific products in the environment.

The discussion on pharmaceuticals in groundwater often revolves around urban-sourced contamination. Urban pharmaceuticals reach groundwater bodies in a point-source manner via wastewater emission, combined sewer overflow, or from leakages in wastewater infrastructure (Christoffels et al., 2016; Kolpin et al., 2002). Reclamation practices of municipal wastewater, such as MAR or wastewater irrigation (see Section 3.4), may present an additional risk of urban-sourced pharmaceuticals entering groundwater bodies. The presence of pharmaceuticals in urban environmental waters is highly variable on the global scale, and in addition to physico-chemical controls, is a function of wastewater infrastructure and local pharmaceutical consumption patterns (refer to Section 5.5 for example). Relevant societal factors include illnesses prevalent in local society, as well as product availability (i.e. prescription versus over-the-counter) (Mutiyar & Mittal, 2013).

Veterinary compounds in agriculture are another major source of pharmaceuticals into the environment, although they tend to be underrepresented in the literature (Bottoni et al., 2010; Kim et al., 2011). An abundant range of veterinary pharmaceutical compounds are used in the agricultural sector including antimicrobials, anthelmintics and ectoparasiticides, antifungals, hormones, anaesthetics, tranquilisers, euthanasia and anti-inflammatory products (Boxall et al., 2004). It is estimated that 50 - 90% of doses administered to livestock is excreted (largely unmetabolized) into the environment (García-Galán et al., 2010; Kaczala and Blum, 2016). Veterinary antibacterials, have been repeatedly detected in groundwater wells associated with animal feed lots (Batt et al., 2006). Hormones are detected less frequently than other veterinary pharmaceuticals, though they have been measured both in surface and groundwater samples (Bartelt-Hunt et al., 2011; García-Galán et al., 2010; Vulliet and Cren-Olivé, 2011).

In addition to direct excretion, the extensive application of animal manure to crop fields (see Section 3.2) is also considered a major route through which veterinary pharmaceuticals can enter groundwater systems (Boxall et al., 2003). Additionally, leachates from livestock burial pits is poorly described in association with groundwater contamination, though it may pose a threat to environmental and public health (Kaczala and Blum, 2016; Yuan et al., 2013).

4.2.3 NAPLs

Non-aqueous phase liquids (NAPLs), which are either slightly soluble or completely insoluble, make up a major subset of organic contaminants that have been detected frequently in anthropogenically-impacted groundwater. NAPLs can be classified as light (L) and dense (D), according to their density relative to water. Benzene, toluene, ethylbenzene, and xylene (BTEX) are a prominent example of LNAPLs, while chlorinated solvents and heavy crude oil are examples of DNAPLs.

The majority of NAPL pollution is a product of industrial environments, though issues are present in urban and agricultural environments as well, as some chlorinated compounds are used in pesticides or cleaning agents, and gasoline transport and storage networks are present in all anthropogenic environments (Baker et al., 2016; Bhatt et al., 2007). Landfills, leaking gasoline storage tanks and pipes, leaking septic tanks, and accidental spills are all potential sources of NAPLs in groundwater.

When infiltration to the saturated zone occurs, LNAPLs will accumulate on top of the groundwater table and generally flow in the direction of the hydraulic gradient. On the other hand, DNAPLs migrate vertically until the impermeable base of the aquifer is reached. This vertical migration tends to leave behind residual phase DNAPLs along its pathway. The direction of flow in this case may not coincide with the hydraulic gradient (Kueper and McWhorter, 1991; Parker and Park, 2004). The slow dissolution of lightly soluble NAPLs can result in contamination plumes that are larger and longer-lived than the measured spread of the pure product, further complicating characterization and remediation efforts (Soga et al., 2004).

Chlorinated solvents including tetrachloroethene (PCE), trichloroethene (TCE) are among the most common NAPL pollutants, found both in association with industrial and urban activities (Matteucci et al., 2015; Stroo et al., 2003). Highly chlorinated solvents such as PCE are persistent or degrade only slowly under aerobic conditions in aquifers (Hunkeler et al., 1999), while under reducing conditions partial or complete microbial dechlorination occurs (Fennell and Gossett, 1998; Picardal et al., 1995). Less chlorinated solvents such as TCE can also be biodegraded under aerobic conditions (Enzien et al., 1994; Palau et al., 2016). Dechlorination of PCE leads to the formation of TCE, cis-1,2 dicholorethene (cDCE), and vinyl chloride (VC) as intermediate products (Hunkeler et al., 1999). Typically, ethene (ethylene) is the end product of dechlorination (Fetter et al., 2017).

Natural subsurface heterogeneities, even in relatively simple porous aquifers, make the remediation of NAPLs a challenging task (Schnarr et al., 1998). Compared to porous aquifers, studies on the spatial and temporal distributions of NAPLs in karst aquifers are notably less prevalent yet significantly more complex (Yu et al., 2015). Depressions in karstic conduits may act as traps for DNAPLs (Wu and Hunkeler, 2013; Xu et al., 2018), making detection and remediation extremely difficult (Field, 2018).

4.3 Raw mineral extraction

Acid mine drainage (AMD) is a significant contaminant arising from mine waste such as tailings facilities. Tailings facilities compose dumps of crushed waste rock and liquid, and generally contain high proportions of so-called 'byproducts' of the host rock, such as pyrite (FeS_2). Leachate from tailings is often enriched with salts and heavy metals due to low pH and interactions between the solid and liquid phase, leading to the phenomenon of AMD. In more arid regions, where rivers derive their base flow from groundwater seepage, stream flow resulting from elevated groundwater levels can be composed almost entirely of AMD (Tutu et al., 2008).

Very low pH values can cause heavy metals to become soluble. This is a known environmental hazard emanating from many active and abandoned mine workings (Rösner, 1998). However, some studies do suggest that shallow or fractured aquifers can be remarkably effective at sorbing heavy metals from mine leachate, depending on pH, soil conditions, and leachate concentrations. For example, studies by Schwartz and Kgomanyane (2008) and Von Der Heyden and New (2004) showed that sorption of heavy metals from mine leachate occurred within 700 m down gradient from the polluting source due to ideal physicochemical conditions within the groundwater. In these cases, relatively high pH conditions of the groundwater (greater than 6) and H^+ surface complexation onto hydroxides and oxides of iron are believed to have facilitated the adsorption of heavy metals to the clays and organic material of the shallow unconfined groundwater.

However, the absence of carbonate or calcite in the underlying geology, or the cessation of lime addition to tailings facilities could reduce the groundwater's ability to buffer mine leachate. Just a small decrease in pH has been shown to result in an increase in soluble metal concentrations. Should leachate and groundwater pH decrease to below 5, markedly increased concentrations of aluminium and copper within the groundwater can be expected, adversely affecting the surface and groundwater quality and their related ecosystems (Ashton, 2010; Musingafi and Tom, 2014).

5. Case studies highlighting current threats to groundwater quality

5.1 Nitrate in unsewered cities

Nitrate pollution from wastewater is a particular threat in cities that overlie shallow aquifers and lack adequate underground sewer networks, and where urban wastewater is able to infiltrate in a relatively diffuse manner. A study by Faye et al. (2004) presents an analysis of the impact of various land occupations on the quality of the unconfined quaternary sand aquifer underlying the suburban area of Thiaroye, located on the Dakar peninsula in Senegal. According to the authors, land use is highly variable in Thiaroye and includes densely populated areas that are lacking centralized sewer networks, depending instead on septic systems that are not always properly constructed. As groundwater levels are very shallow in the Thiaroye region, consistent communication between surface activity and groundwater has been evidenced. Nitrate contamination in the regional groundwater is a longstanding issue, with values in excess of 500 mg/l having been recorded in groundwater at drinking water extraction points.

The authors used a GIS approach to combine data on the geospatial distributions of measured nitrate concentrations with information about soil type, groundwater characteristics, and land use. Three classes of spatial vulnerability were first assigned based on intrinsic aquifer characteristics. On top of vulnerability, spatial potential for pollution was also assigned to one of three classes based on land use features. Ratings for both steps were determined using a Boolean logic operation to avoid subjective assignment of classes. From their analysis, the authors deduce that nitrate concentrations are a good proxy for both aquifer vulnerability as well as other anthropogenic contamination on a regional scale. In evaluating their constructed vulnerability map, the authors point out that observed nitrate levels also depend intimately on contaminant loading and contaminant characteristics, which is more of a function of land use and layout rather than any climatic or hydrogeological factors.

5.2 Nitrate accumulation and long-term leaching

In a study conducted on the High Plains aquifer of the United States, isotopic tracers of nitrate (^{15}N) and water (^{2}H, ^{3}H, ^{18}O) were used to assess the storage and transit time of nitrates in the subsurface (Figure 4). Sites were selected to compare unsaturated zones of rangelands (Figure 4a) with those associated with irrigated cropland sites (Figure 4b) where chemical where applied (Gurdak et al., 2007). McMahon et al. (2006) demonstrated how the mobilization of natural salts, as a result of irrigation return flow and long-term evaporative (Et) concentration near the base of the root zone, resulted in a larger accumulation of nitrate within thick unsaturated zones beneath irrigated sites. This accumulation accounted for as much as 60% of the nitrate found in groundwater reservoirs beneath irrigated croplands.

Although advective transit times in the unsaturated zone were estimated to range from 50 to 375 years (longer than any of the sites had been irrigated for), agrochemicals were detected in groundwater at 66% of the sites associated with irrigated crop sites. McMahon et al., (2006), suggests that transport occurs along multiple flow paths ranging from slow paths (associated with fine-grained sediments with little or no flow) to fast paths (associated with areas of focused recharge such as depressions, streams or playas). The authors concluded therefore that the amount of contaminants reaching the unconfined aquifer from the irrigated sites could increase, even if input was completely stopped, as the mass of historically accumulated compounds under irrigated fields continue to slowly travel downwards through the thick unsaturated zone. This storage of contaminants in sediments is one explanation for the continued excessive nitrate load measured in many groundwater bodies.

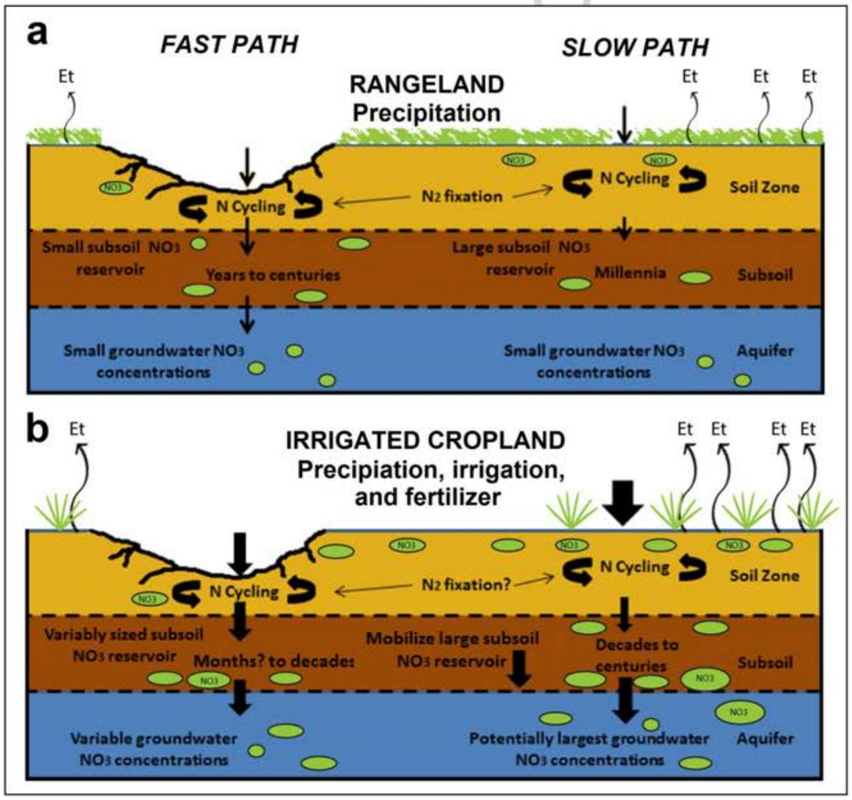


Figure 4: Conceptual models for the evaporative (Et) concentration and transport of NO_3^- from the land surface to the water table through the unsaturated subsoil to the aquifer, under a) rangeland conditions

and b) irrigated cropland conditions in the High Plains of the United States (abbreviated from McMahon et al., 2006).

5.3 Pesticide contamination in urban versus agricultural settings

Due to the overlapping application practice of pesticides in the urban and agricultural settings, it can be difficult to detangle the exact sources of products detected in groundwater, particularly in cities with dense parks, gardens, and in close proximity to agricultural zones. Many products used in urban outdoor areas are the same products used in proximal agricultural zones. According to Sinclair and Boxall (2003), considering pesticides transformation products can be important in detangling pesticide sources in groundwater studies.

As an illustration of the similarities and differences between urban and agricultural pesticides, Barlow et al. (2012) measured 38 pesticides and their degradates in the shallow, largely unconfined, aquifer associated with urbanized areas in and around Memphis, Tennessee, USA. The authors made use of local criterion and usage patterns to identify products as 'primarily urban', 'urban and agricultural' and 'primarily agricultural'. In the Memphis aquifer, for example, the herbicide *simazine* was the most often detected primarily urban product, and its occurrence and concentration was reported to have increased through the duration of the study. *Atrazine* was the most often detected product of both urban and agricultural use. Other pesticides deemed to be primarily of urban use included *tebuthiuron*, *prometon*, *diuron*, *bromacil*, and *dieldrin*. The authors compared detected values from measurements in groundwater between 1994 and 2009 to outline changes in land use and product use patterns, thus highlighting the importance of obtaining knowledge on local usage patterns for stronger interpretation. For example, while *simazine* is primarily used in urban environments in Memphis, it is also known to be used in many agricultural contexts such as berry fields, orchards, and vineyards in other places in the US, as well as around the world. Further, the use of both *atrazine* and *simazine* was banned in the EU over a decade ago (refer to Table 2). In many countries today, including the United States, Australia, Brazil, and India, *atrazine* remains one of the most-used herbicides (Singh et al., 2018).

5.4 Veterinary pharmaceuticals in groundwater

Sulfonamides are commonly used veterinary antibiotics, usually allied in mixtures, that have low soil sorption tendency, are highly soluble, and have half-lives ranging between 5 - 40 hours (in serum). Sulfonamides have been repeatedly detected groundwater wells located down gradient from confined animal feeding operations (Batt et al., 2006; Karthikeyan and Meyer, 2006; Lindsey et al., 2001; Sacher et al., 2001). An investigation by García-Galán et al. (2010) in Catalonia (Spain) of 39 groundwater sites representing a variety of aquifers, including the fractured multilayer confined sedimentary Plana de Vic aquifer and the unconfined alluvial La Selva aquifer, found a 90% detection frequency of sulphonamides. Although the average concentration of sulfonamides detected was generally below 50 ng/l, highs of 3,461 ng/l were measured for *sulfacetamine* and 745 ng/l for *sulfamerazine*. These sulfonamides were measured predominantly in an aquifer from which 68% of the water was abstracted for agricultural purposes, but from which 20 % of the water was also designated for drinking water purposes. In the García-Galán et al. (2010) study, a strong correlation between sulfonamides and nitrates was established, justifying the consideration of this coupling of compounds an indicator of groundwater pollution stemming from animal origin.

5.5 Production and consumption patterns of human pharmaceuticals

As an example of a relatively extreme case of urban groundwater contamination from human pharmaceuticals, we highlight the publication by K'oreje et al. (2016). The authors carried out a study in the cities of Nairobi and Kisumu, Kenya to characterize the presence of pharmaceutical products in wastewater, surface water, and groundwater in what were deemed to be at-risk zones. The authors differentiated between multiple classes of pharmaceuticals, and made special mention of commonly used products at the city scale. In their study, they were able to identify major stresses on groundwater, due not only to WWTP effluent, but also untreated wastewater discharges from septic tanks and pit latrines used in informal settlements along with effluent from pharmaceutical production facilities. Although concentrations measured in shallow groundwater wells in Kisumu were magnitudes lower than those of associated surface waters, anti-inflammatory, antibiotics and psychiatric drugs were measured (Figure 5). In particular, high concentrations of *nevirapine* (an anti(retro)viral drug) were measured in two out of the three groundwater wells assessed in Kisumu (1200 ng/l and 1600 ng/l for Well 1 and Well 2 respectively). This study is one of the first in identifying the persistence of anti(retro)viral drugs in groundwater. The authors note that HIV/AIDS is the leading cause of death in Kenya, hence a widespread consumption of anti(retro)viral drugs would lead to its prevalence in effluent and, under these circumstances, may find its way into shallow groundwater bodies.

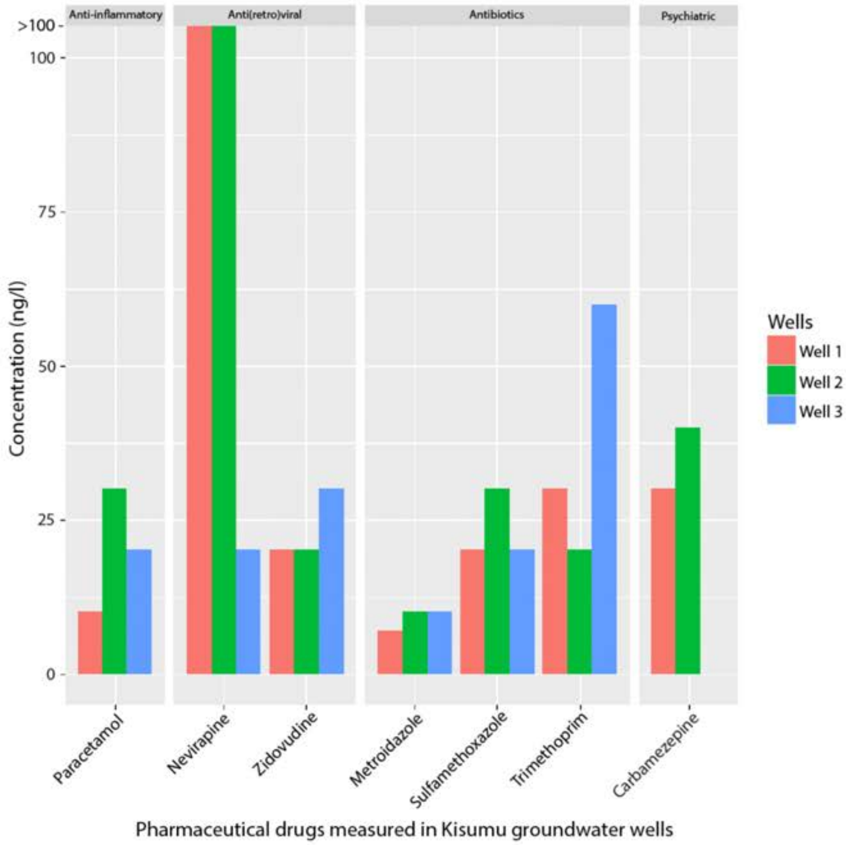


Figure 5: Concentration of pharmaceuticals detected in groundwater three wells in Kisumu, Kenya. Note that in Well 1 and Well 2, *nevirapine* concentrations were 1200 and 1600 ng/l respectively (adapted from K'oreje et al., 2016).

This conclusion highlights the dependence of pharmaceutical presence on local consumption patterns. To emphasize this point further, in another study Rehman et al. (2015) discuss an important large-scale contribution of pharmaceutical products in the aquatic environment, one that is rarely discussed in western literature: pharmaceutical and precursor production facilities. The authors here point out that as a result of international economic relocation, some 20% of generic products on the global market are produced in

India alone. Similarly, more than half of pharmaceutically active precursor ingredients are made in China. The authors coin the term 'EPMC' – emerging pharmaceutical manufacturing country, to refer particularly to Bangladesh, China, India, and Pakistan, where a proportionally large amount of pharmaceutical precursors and finished products are manufactured. For this reason, as well as for the fact that sanitation levels are limited and that nearly half of the global population resides in these regions (refer to Figure 2), these EPMCs stand out. The authors claim that one pharmaceutical production park in India, the Patancheru estate located in Hyderabad, produces more than 1500 m^3 of wastewater every day. Further, these countries possess low wastewater treatment capacities; the authors claim that only 2% of wastewater in Pakistan is treated, while the number rises to 24% in India. Due to these factors, urban groundwater in EPMCs are some of the most at-risk in the world for pharmaceutical contamination.

5.6 Chlorinated organic compounds in karstic systems

Yu et al. (2015) investigated the distribution patterns of chlorinated organic compounds in a karst aquifer in northern Puerto Rico, where PCE and TCE were some of the most commonly detected and persistent contaminants. They identified a decreasing trend in contaminant concentrations with depth and distance from source, but also a spreading of contaminants beyond the extent of known sources. They concluded that either unidentified waste disposal sites must exit, or that the transport of the contaminants through the karst system took place along unknown flow paths. A detailed description of these preferential pathways is typically impossible (Andreo et al., 2006; Kaufmann, 2016). When attempted, geophysical techniques (Al-fares et al., 2002; Chalikakis et al., 2011) and/or artificial and intrinsic tracer tests (Hillebrand et al., 2014; Maloszewski et al., 2002; Reh et al., 2015) are common methods to identify such pathways. However, even with the best methods, generally only a fraction of the flow and transport pathways can be identified (Göppert and Goldscheider, 2008; Robert et al., 2012).

Wu and Hunkeler (2013) studied the hyporheic flow in sediment filled karst conduits. They demonstrated that hyporheic flow is induced by conduit bends, even in the case of flat sediment surfaces. This can have strong implication for the storage and slow release of chlorinated solvents, but could also have a positive effect on water quality by filtration and due to biogeochemical transformation of solutes. They also noted that this hyporheic flow in karst sediments has received little attention so far.

5.7 Long-term impacts of mining

The Highveld coal fields in South Africa have been exploited since 1894, and provide insight into the long-term impacts of mining (McCarthy, 2011). The extensive historical mining in the Highveld has left a scarred landscape of abandoned and collapsing mines (Drebenstedt and Singhal, 2014), and has leached acidic water into the groundwater system for decades. In lieu of long-term open cast mining, surface water quality in parts of the Highveld has deteriorated to such a degree, with recorded SO_4^{2-} values exceeding 7,000 mg/l and pH < 2, that local coal-powered electricity supply utilities are forced to import water for use in their power stations. This process of AMD decanting from the groundwater to the surface water is expected to continue until all the pyrite (FeS_2) hosted in the bedrock of the exposed coal fields has been completely oxidized, with little or no effective mode of cessation or remediation currently at hand (McCarthy, 2011). As mining licenses continue to be issued in the Highveld, farmers downstream (an area famous for its fruit production), risk losing their license for agricultural export as surface and groundwater quality worsen.

In addition, AMD can have a negative effect on karstic systems. The Proterozoic Dolomites of the Malmani Subgroup in South Africa (a UNESCO World Heritage site known as the Cradle of Humankind) overlies the infamous gold- and uranium-rich Witwatersrand Supergroup; from which more than one third of the world's gold has been produced and which still contains six times more gold than the world's second largest gold field (Tucker et al., 2016). The Malmani karst system has experienced repeated long-term dewatering (as far back as 1903) as the underlying gold mines were pumped to keep the stopes and shafts dry, followed by rewatering as mines were abandoned and pumping ceased (Durand, 2012; Naicker et al., 2003). The massive discharge of contaminated groundwater (up to 80,000 m^3/day) has altered surface stream characteristics in the surrounding areas from non-perennial to sizable rivers and swamps contaminated with mine effluent containing aluminum, iron, nickel, zinc, cobalt, copper, lead, radium, thorium and uranium (Durand, 2012; Naicker et al., 2003). As AMD water continues to rewater ground- and surface water, the structural stability of the region is threatened, and the archaeological and palaeontological heritage site risks losing its UNESCO status. The exact number of abandoned mines and resultant AMD as a result of historic mining activities in South Africa remains poorly quantified and monitored (Fourie et al., 2010).

5.8 Managed aquifer recharge and pesticides

When urban storm water or wastewater is intentionally used for recharge during managed aquifer recharge (MAR), the risk of contamination cannot be disregarded and should be assessed regularly. A study conducted by Shareef et al. (2014) does exactly this for the Parafield storm water harvesting site in Adelaide, Australia. The authors investigated the question of biogeochemical degradation capacity of select pesticides – *simazine* and *diuron* – under aerobic vs anoxic conditions, in order to investigate the behavior of these products as they move from their source waters into deeper, confined aquifers. The Parafield system harvests rain water from two urban storm water catchments, where it is pre-treated and then stored either in an aquifer storage and recovery (ASR) well or in an aquifer storage transfer and recovery (ASTR) system within a confined limestone aquifer (please refer to the original publication for more details on ASR and ASTR systems). The authors added either *diuron* or *simazine* to a simulated system under either aerobic vs anoxic conditions, and then monitored the concentration evolution of the two compounds over a series of two months. Comparing their results with other literature results, Shareef et al. (2014) concluded that a decrease in both product concentrations was due to biodegradation, with little degradation occurring from abiotic processes or hydrolysis. This identifies potential risk of persistence of select pesticides and other organic pollutants in anoxic aquifer systems.

6. Discussion

Over the past decades, many advances have been made towards better groundwater characterization, due in large parts to technological advances. Numerical modelling of flow and transport, isotope analysis, better analytical power for synthetic compounds, and improved geophysical methods are all greatly increasing our understanding of aquifer contamination (Baillieux et al., 2015; Botter et al., 2011; Brunner and Simmons, 2012; Clements and Denolle, 2018; Levison and Novakowski, 2012; McMahon et al., 2006).

Still, great uncertainties persist in these methods and in our understanding (Montanari et al., 2013). Due to variability in the spatio-temporal scale of given processes, the comprehensive characterization of the links between the surface, unsaturated and saturated zones in response to land use changes and the associated contamination risk remains a challenge (Scanlon et al., 2005). Currently no single measure is able to describe the water quality for any one water body (Rickwood and Carr, 2009).

Uncertainty also results from a poor understanding of how our activities will impact groundwater, and this is particularly true for newer practices. For example, groundwater vulnerability to pharmaceutical loading in response to temperature change or disease outbreaks is poorly understood. With increasing numbers of studies detecting pharmaceuticals in groundwater bodies (e.g. Bu et al., 2013; García-Galán et al., 2010; Kaczala and Blum, 2016), the question concerning antibiotic resistance and proliferation of compounds in the aqueous environment should concern us. Further studies are needed concerning the consequence of these compounds, both in their individual concentrations and as cocktails, in the groundwater environment.

There exist many classes of anthropogenic products with contamination potential that were not discussed in-depth in this review. This includes plastics and plastic-associated chemicals (Teuten et al., 2009), musk fragrances (Teijon et al., 2010), deicing agents (Ledford et al., 2016), and flame retardants (Stepien et al., 2013), to name a few. In addition, processes such as shale gas exploitation (hydraulic fracturing), are among those which have come under scrutiny as a potential source of groundwater contamination (Gordalla et al., 2013). While the risk from such practices is gaining increasing attention in the environmental sciences, there are as yet few publications on this topic. Future studies are needed to illicit the impact of products used in emerging technologies in a more comprehensive way. In light of this, there is also a global need for effective early warning systems that are capable of anticipating risks associated with compounds used before they become “contaminants of emerging concern” (Dulio et al., 2018).

Furthermore, the scarcity of centralized and easily accessible data for most products is an issue, in particular for emerging contaminants. Information along the lines of product consumption by geographical region or by sector is not readily available to environmental researchers, stakeholders, or law makers. When planning a monitoring campaign, for example of certain products in groundwater, having prior knowledge on what products, and how much, are commonly used in a local or regional areas of interest, could be equally as useful as having knowledge of hydraulic conductivity or groundwater-surface water interactions. Emerging monitoring technologies including wireless technology, automated sensors, and new tracers have all added to the massive growth in data availability (Lovett et al., 2007), so that high quantity and quality data collection and data management should continue to improve the ability to monitor and exchange information in hydrogeology with related fields (Staudinger et al., 2019).

Although there is a general consensus among many experts that the threat of groundwater contamination is important to address, continued political indifference, social stigma, and disregard of ecosystem services results in an undervaluation of groundwater as a renewable resource (Knüppe, 2011). For example, in many developing countries, areas immediately surrounding and downstream of mining operations are often inhabited by poor communities with little access to service provision. These communities are dependent on local streams, wetlands and groundwater sources for their water supply, and many are at risk of acute metal toxicity from heavy metals (Dhakate and Singh, 2008; El Khalil et al., 2008; Hobbs et al., 2008). In order to address these and similar shortcomings, the link between groundwater, groundwater-dependent sectors and groundwater governance needs to be understood and communicated, both in policy and in practice (Montanari et al., 2013). However, it is important to note that in some cases, especially in the developing world, countries are cutting back on their monitoring investments (Harmancioglu et al., 2003).

In a world where water consumption is predicted to increase, water scarcity will continue to intensify and a dependence on water reuse will become common practice, the monitoring of pollutants in the effected environment will become imperative (Elsner and Imfeld, 2016; Friedler, 2001). It is up to the scientific community to clearly impress the importance of monitoring networks and the upkeep and development of long-term data sets on decision makers, while prioritizing the need for installation and maintenance of measuring systems in the face of resource constraints (Lovett et al., 2007). With conditions often changing faster than scientists or policy makers can anticipate, adaptive management strategies and interdisciplinary research provide a means to address sustainable resource governance under uncertain conditions.

7. Summary and Conclusion

This review highlights some of the key groundwater contamination issues that are sourced from anthropogenic activities. A selection of major contaminants are discussed, including established products as well as emerging contaminants. Case studies addressing unique issues regarding groundwater quality, contaminating sources, and highlighted contaminants are presented. The diversity of cases illustrates the variability of qualitative threat to groundwater in terms of a sustainable resource for the human population and to safeguard the environmental integrity. Intensive agriculture, urban sprawl, globalized pharmaceutical production and consumption, insufficient wastewater infrastructure, dwindling empirical data on water quality, and in some cases the insufficient emphasis of groundwater as a renewable resource, are all hampering the complex process of managing groundwater quality. Although advances in measuring, monitoring, and modelling groundwater are astride, the rate at which new contaminants and contaminating issues are entering the scene may outpace current progress. Transdisciplinary research and action may provide an opportunity in developing a comprehensive understanding of contamination dynamics and their effects on the groundwater system. This presents great opportunities for merged fields of research and transboundary communication.

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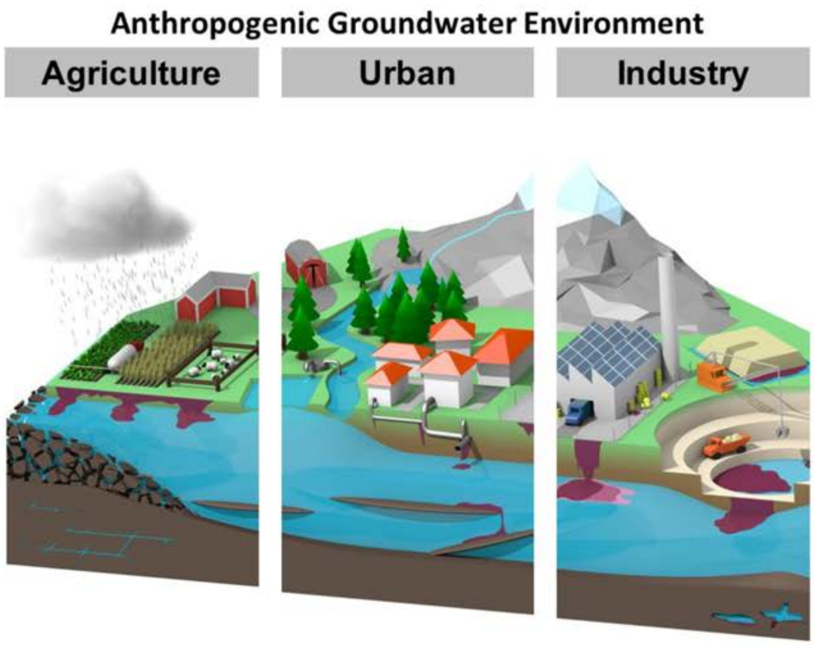
Graphical abstract



Highlights

- A review of anthropogenic impacts on groundwater quality is presented
- Groundwater contamination risks accompany all types of human land use activities
- Major groundwater contaminants of modern relevance are detailed
- Case studies displaying diverse global contamination issues are highlighted
- The importance of transdisciplinary groundwater management is emphasized