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Review

Pharmaceuticals in soils of lower income countries: Physico-chemical fate and risks from wastewater irrigation

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ABSTRACT

Population growth, increasing affluence, and greater access to medicines have led to an increase in active pharmaceutical ingredients (APIs) entering sewerage networks. In areas with high wastewater reuse, residual quantities of APIs may enter soils via irrigation with treated, partially treated, or untreated wastewater and sludge. Wastewater used for irrigation is currently not included in chemical environmental risk assessments and requires further consideration in areas with high water reuse. This study critically assesses the contemporary understanding of the occurrence and fate of APIs in soils of low and lower-middle income countries (LLMIC) in order to contribute to the development of risk assessments for APIs in LLMIC. The physico-chemical properties of APIs and soils vary greatly globally, impacting on API fate, bioaccumulation and toxicity. The impact of pH, clay and organic matter on the fate of organic ionisable compounds is discussed in detail. This study highlights the occurrence and the partitioning and degradation coefficients for APIs in soil:porewater systems, API usage data in LLMICS and removal rates (where used) within sewage treatment plants as key areas where data are required in order to inform robust environmental risk assessment methodologies.

Contents

Abbreviations: AEC, anion exchange capacity; API, active pharmaceutical ingredient; CDOM, colloidal dissolved organic matter; CEC, cation exchange capacity; LLMIC, lower and lower middle income countries; PEC, predicted environmental concentration; PNEC, predicted no effect concentration; POM, particulate organic matter.

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1. Introduction

There has been a global increase in the use of active pharmaceutical ingredients (APIs) in recent decades due to population growth, increasing affluence, changes in disease burdens and easier access to medication. In the low and lower-middle income countries (LLMIC) of Asia, Africa and Central and South America, the use of human pharmaceuticals increased by 23–29% between 2000 and 2011 (WHO, 2011). As a consequence, the loadings of residual APIs and other down the drain chemicals (including personal care products) to soils, surface and ground waters of these countries are likely to have increased. The major vector of this loading is wastewater (Corcoran et al., 2010; WHO, 2006b, 2006c, 2006d). Wastewater is defined as a combination of one or more of blackwater (excreta, urine, faecal sludge), greywater (kitchen and bathing wastewater), commercial and industrial effluent (including hospitals), stormwater and other urban run-off, and agricultural, horticultural and aquacultural effluent. Each may be treated, partially treated or untreated (Corcoran et al., 2010; Jiménez et al., 2010). Difficulties in quantifying the magnitude of wastewater loads, in tandem with a paucity of environmental monitoring data of APIs in LLMIC, makes accurate and precise predictions of temporal trends in API loadings uncertain (Jiménez et al., 2010; Kookana et al., 2014).

Many LLMIC are experiencing physical or economic water scarcity (Fig. 1) with the former particularly important in northern and southern Africa and southern Asia. To counter-act shortages of good quality water in arid and semi-arid regions and to conserve its use, many LLMIC use the wastewater they generate for irrigation of agricultural and horticultural land. The water stressed areas of southern Asia produce wastewater in excess of 10×10^9 m^3 $year^{-1}$ (Fig. S1) with up to 20% being used for irrigation. Other countries such as Israel, Jordan, Syria, Iraq and Mexico use >40% of their municipal wastewater for this purpose (Fig. 2(a)). Globally, about 20 million ha of agricultural land is irrigated with wastewater (Scott et al., 2004), with the highest proportions of cultivated areas equipped for irrigation found in the Middle East, southern Asia and western South America, as shown in Fig. 2(b). Per capita daily food consumption requires 2–5 m^3 of water (Corcoran et al., 2010), making agriculture a significant requirement for water, particularly in the extensively irrigated regions noted above (Fig. S2). Irrigation is dominated by untreated and untreated-diluted wastewater, notably in China (>3.6 million ha), India (>1 million ha) and Mexico (ca. 190,000 ha), while treated water is extensively used in Chile, Mexico and Egypt (238, 000 ha) (Lautze et al., 2014). Across a range of LLMIC, 80% of cities use mainly untreated and untreated-diluted wastewater for irrigation (Jiménez et al., 2010). In arid areas, cities such as Dakar (Senegal), Accra (Ghana) and Tamale (Ghana) produce 60–100% of the consumed leafy vegetables within the city using wastewater irrigation, while 60–80% of the perishable food for local markets in Hanoi (Vietnam) is produced using diluted wastewater (Corcoran et al., 2010; Drechsel et al., 2006). Water shortages are predicted to become more widespread and acute as human populations increase in number and urbanisation and industrialisation expand, food consumption patterns change, and rainfall distribution and volume alter as a result of climate change (Corcoran et al., 2010; Hanjra and Qureshi, 2010). Nevertheless, there appears to be the potential to markedly increase the recovery and re-use of wastewater in many LLMIC, particularly for agricultural use close to highly urbanised areas, given the appropriate incentives (Jiménez et al., 2010; Lautze et al., 2014; WHO, 2006c, 2006d).

There is currently a lack of public usage data for the amount and type of APIs used in many LLMIC due to poor record keeping, extensive self-medication and the use of non-prescribed APIs over large population numbers (Kookana et al., 2014; Kotwani et al., 2012; Rehman et al., 2015). This knowledge gap is further confounded by inconsistent adherence to therapeutic treatments, particularly for longer-term prescribing (Kookana et al., 2014). For some groups of APIs, per capita use may be similar between LLMIC and higher income countries, but owing to larger populations in LLMIC (40% of the global human population live in China, India, Bangladesh and Pakistan; Rehman et al., 2015) the actual tonnage used is much greater (Kookana et al., 2014). Usage data are often commercially sensitive and thus unavailable to the wider scientific community; however, projected spending patterns indicate continued expansion of API use in LLMIC (Fig. S3). In addition, there has been a marked relocation of pharmaceutical manufacturing from high income countries to LLMIC in recent years, with an annual growth of 10–15%, resulting in ca. 13,000 industrial production units in India and China alone (Cardoso et al., 2014; Rehman et al., 2015). The effluents from these generally poorly regulated sites have been identified as a significant source of APIs to adjacent surface waters and sewage treatment works (Larsson, 2014; Liu and Wong, 2013; Rehman et al., 2015). This can lead to localised 'hot spots' which are manageable if, inter alia, site

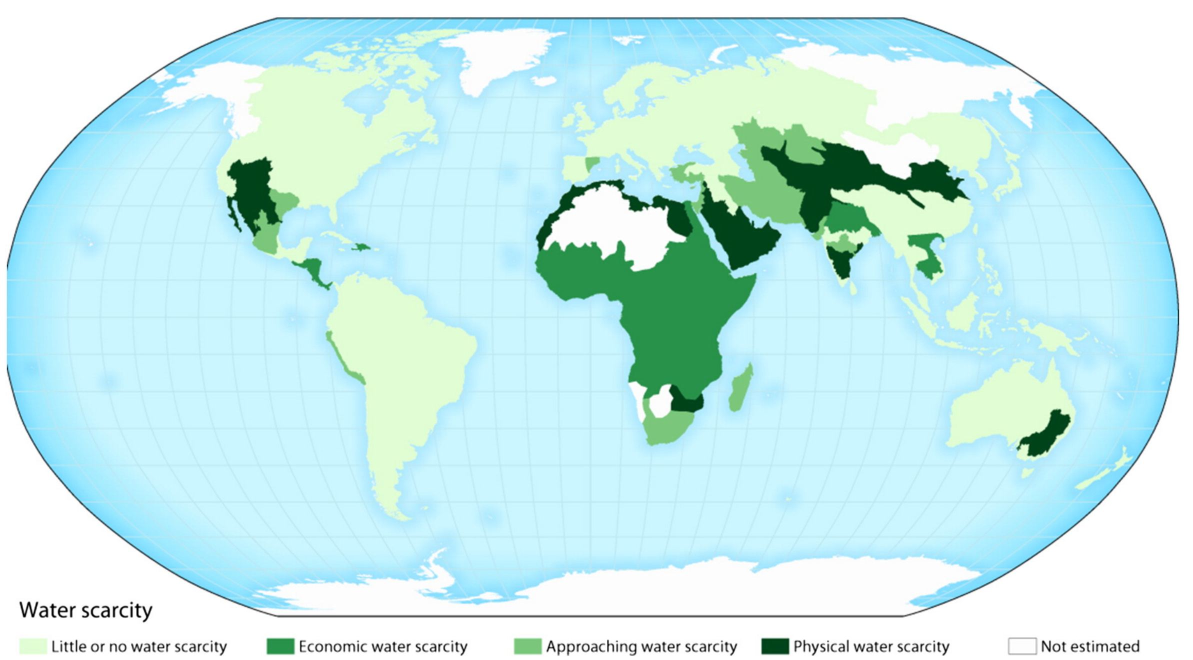

Fig. 1. Regions of physical and economic water scarcity (International Water Management Institute, 2006).