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Medical geology in the framework of the sustainable development goals

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HIGHLIGHTS

- Medical geology is of increasing significance and a global challenge.
- Human impacts of geogenic contaminants (GCs) are underestimated or not recognized.
- Not including toxic GCs in standard water analyses exposed millions of people.
- Economic and population growth accelerate GCs release and human exposure.
- The potential role of GCs in diseases with unknown etiology must be considered.

GRAPHICAL ABSTRACT

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ABSTRACT

Exposure to geogenic contaminants (GCs) such as metal(loid)s, radioactive metals and isotopes as well as transuraniums occurring naturally in geogenic sources (rocks, minerals) can negatively impact on environmental and human health. The GCs are released into the environment by natural biogeochemical processes within the near-

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surface environments and/or by anthropogenic activities such as mining and hydrocarbon exploitation as well as exploitation of geothermal resources. They can contaminate soil, water, air and biota and subsequently enter the food chain with often serious health impacts which are mostly underestimated and poorly recognized. Global population explosion and economic growth and the associated increase in demand for water, energy, food, and mineral resources result in accelerated release of GCs globally. The emerging science of "medical geology" assesses the complex relationships between geo-environmental factors and their impacts on humans and environments and is related to the majority of the 17 Sustainable Development Goals in the 2030 Agenda of the United Nations for Sustainable Development. In this paper, we identify multiple lines of evidence for the role of GCs in the incidence of diseases with as yet unknown etiology (causation). Integrated medical geology promises a more holistic understanding of the occurrence, mobility, bioavailability, bio-accessibility, exposure and transfer mechanisms of GCs to the food-chain and humans, and the related ecotoxicological impacts and health effects. Scientific evidence based on this approach will support adaptive solutions for prevention, preparedness and response regarding human and environmental health impacts originating from exposure to GCs.

Contents

1. Background

Humankind lives in close contact with geological and geologically-impacted environments which can have a strong, often underestimated, permanent influence (beneficial or toxic) on human health and well-being. Natural exposure to geogenic contaminants (GCs, e.g. As, F, Mn, Be, Cd, Hg, Pb, Rn, Se and U) and the chemical speciation have distinct impact on human health. Their occurrence in different geoenvironmental ambience is a result of mobilization by different natural and anthropogenic processes from their source, can affect drinking water, food or air (atmospheric dust particle matter or aerosols). GCs (elements and their species occurring naturally in geogenic sources (rocks, minerals)) may affect human health through primary consumption (e.g. drinking water) or by elemental biomagnification through the food chain (Fig. 1). Contact with GCs can occur in a number of ways, including: directly via the skin, as in mud baths or bathing for relaxation in geothermal water; through the ingestion of water or geological solid matter, either intended (geophagy) or unintended (e.g. ingestion of dirt, sand or soil etc. is a significant risk for children); or inhalation (atmospheric dust, aerosols) (Fig. 1) (e.g. Davies et al., 2004; Skinner, 2007).

The field of medical geology, a relatively new interdisciplinary science, deals with the complex relationships between environmental factors relating to the presence of GCs in different geological settings, their mobility, geographical distribution and their effects on human and animal health (Bunnell, 2004; Komatina, 2004; Dissanayake, 2005; Khandare, 2012).

Medical geology researches interactions between natural geological factors and their health effects on animals and humans (Finkelman et al., 2001) and belongs to a group of emerging interdisciplinary disciplines, such as environmental medicine and medical climatology (Bunnell, 2004; Khandare, 2012), which are increasingly of public interest due to a range of factors, including: the expanding health impacts associated with environmental changes (e.g. climate change and related impacts such as weather conditions); increasingly restrictive exposure regulations (e.g. contaminant levels in food, drinking water and atmospheric dust); improvements in laboratory and monitoring techniques; and rising social awareness of the relationship between contaminant exposure and human health. As a consequence, it is expected that the new discipline of medical geology will both evolve and become increasingly important in the future.

Providing freshwater and food, with GCs in concentrations that do not negatively impact human health, to the growing world population are important targets for sustainable development. Therefore transdisciplinary cooperation in medical geology plays a crucial role. Within the 17 Sustainable Development Goals in the 2030 Agenda for Sustainable Development chartered by the United Nations in September 2015,

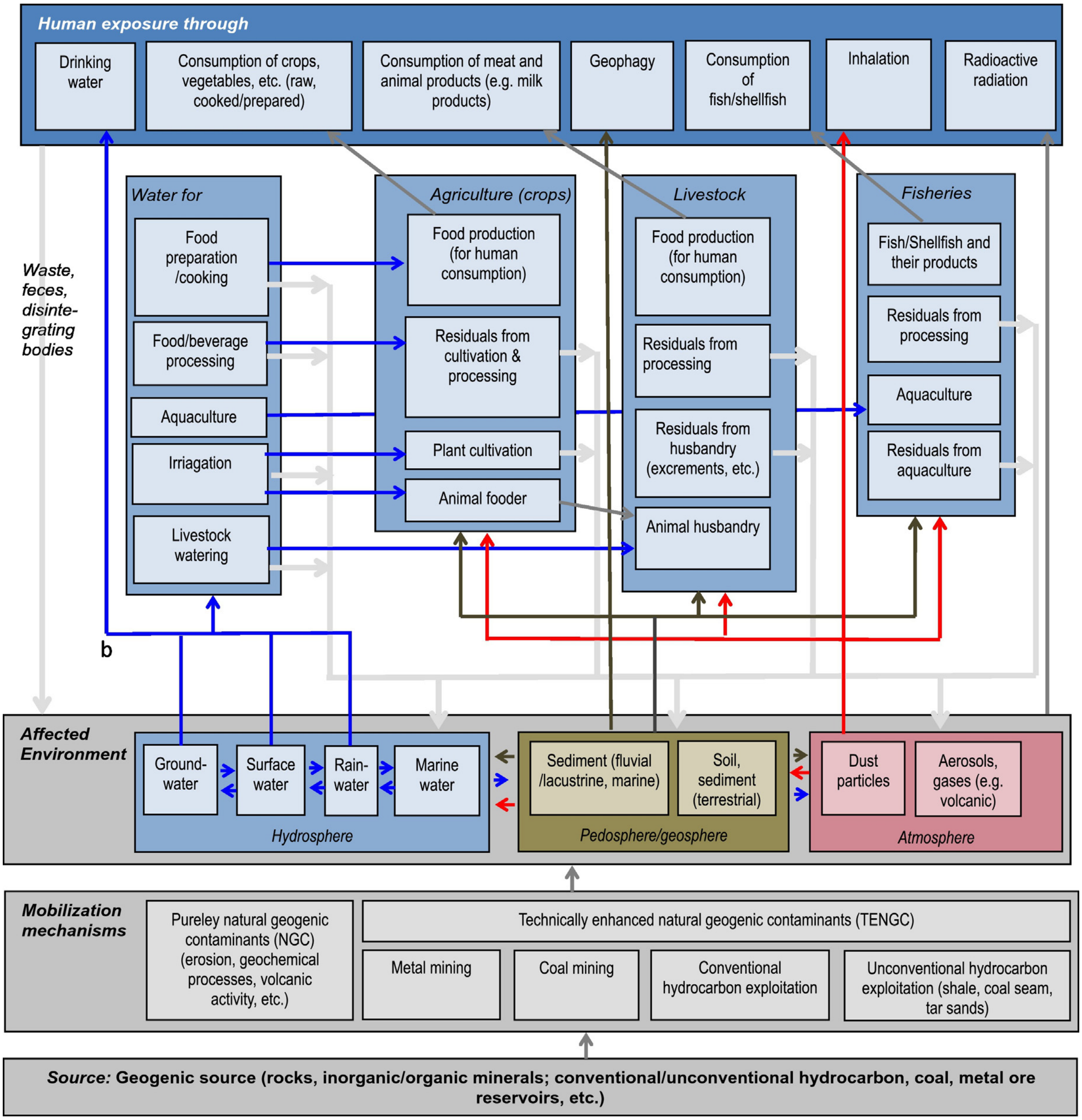

Fig. 1. Geogenic contaminant pathways mobilized by natural and anthropogenic processes affect different environments and potentially impact human health.

in the topic of GCs is included in targets set for achieving Goal 3, Good health; Goal 6, Access to safe water; Goal 10, Fight injustice and inequality; and Goals 14 and 15, Protecting life in aquatic systems and on land. Provision of GC-safe water—by enhancing food security—contributes to Goals 1 and 2, Eradicating or reducing poverty and hunger and to Goal 5, Gender equality—addressing the disadvantages experienced by women walking long distances to obtain GC-safe drinking water. Replacing fossil fuels by locally available, climate-protecting renewable energy sources and technologies, to cover the often high energy demand of removal of GCs e.g. from water or soils links it to Goals 7 and 13 (Energy and Halt climate change, respectively). This further triggers the innovation, creation of jobs and economic growth, which are Goals 8 and 9.

Unfortunately, these Sustainable Development Goals have been formulated as sectoral targets rather than considering that many topics are crosscutting over these fields and should be treated as interconnected. This is also true for the topic of GCs, which requires the consideration of all the respective nexuses within the framework of water, energy, environment, food, climate and public health. This is exactly what the transdisciplinary approach of medical geology, as we describe it in this paper, must aim for in order to help to address the vast array of challenges which the Sustainable Development Goals encompass.

This paper provides a brief overview supported by selected examples of geological biotropic factors and their associated health effects and discusses future directions for this emerging field within the framework of the SDGs. A key focus of the paper is a discussion of “medical geology” in the context of global changes in climate, population and economic growth and related increases in the demand for water, energy, food and minerals. The paper considers inorganic GCs only and is limited to toxic health impacts rather than to elemental deficiencies or the beneficial uses of geological materials such as geothermal muds or geothermal springs for therapeutic treatments. It addresses the importance of medical geology and its role in determining the causes of diseases with unknown etiology.

This paper argues that there is an urgent need to develop medical geology research to build the body of knowledge in this area to have a better understanding of the risks associated with human exposure to GCs. A robust scientific evidence-base will assist in the development of suitable regulations and guidelines for the formulation of adaptive solutions to enhance prevention, preparedness and response to human health impacts originating from GCs.

2. What are the key geological biotropic factors?

The relationship between human diseases and the earth's elements was first predicted by Hippocrates and Aristotle (Bunnell, 2004; NLM, 2014). Subsequently, the famous Renaissance scientist Paracelsus stated that, "All substances are poisons, there is none which is not a poison". The right dosage differentiates a poison and a remedy" (Dissanayake, 2005). Thus, while all elements in the environment are potential poisons, at subtoxic levels they may have beneficial effects and in some cases are even essential for health. Humans, animals and plants all require access to both essential (e.g., Ca, Cl, Cr, Cu, Fe, Fe, P, S, Se, Zn) and non-essential (e.g., As, Cd, F, Hg, Pb) elements which occur in varying concentrations distributed throughout the bio-, atmo-, litho- and hydro-spheres; however, there is still an ongoing controversy whether elements such as As, Pb and F in ultratrace elemental concentrations are beneficial for humans or not (EPA, 2004; Storelli et al., 2010; da Silva et al., 2005; Kirchmann et al., 2005; Cheng et al., 2007; Santamaria and Sulsky, 2010; EC, 2011; Peckham and Awofeso, 2014). However, the occurrence, distribution and source of geogenic elements, their mobility, bioavailability and bioaccessibility, as well as their health effects, depend on a myriad of different factors.

The availability and accessibility of naturally occurring GCs are mainly dependent on bio-hydro-geo-chemical processes involving rocks, soils, the geological setting, geomorphology, exogenic and endogenic geological processes, water bodies, climate, and impacts due to anthropogenic processes such as mining and land use (Finkelman et al., 2001). Biotropic natural geological factors such as volcanic explosions, geochemical anomalies, earth's fluid degassing, geomagnetic activity, natural background radiation, fluid migration and gas emission within fault zones play an important role in the health of man and animals (Dissanayake, 2005).

The contents of GCs within different rocks and minerals may vary by orders of magnitude. While GCs are globally distributed, the pattern of their mobilization into the different environments is highly variable (Fig. 1). However, high contents of GCs in rocks do not necessarily directly correlate with high concentrations of GCs in the surrounding environments such as soil and groundwater. This is exemplified by the 1 million square kilometer Chaco-Pampean plain (Argentina) where concentrations of the GCs such as As, F, Mo, V vary in groundwater regionally but also locally (even over distances of a few meters) by several orders of magnitude (Fig. 2), while the geochemical composition of the aquifer sediments is uniform. Arsenic (As) concentrations in the aquifer sediments are similar to average levels observed in the earth's crust (2.1 mg/kg) (Mandal and Suzuki, 2002); however, in some parts of the plain, groundwater As concentrations can be extremely high (up to hundreds or thousands of μg/L) (Bundschuh et al., 2004; Bhattacharya et al., 2006), far in excess of the World Health Organization (WHO) guideline's permissible value (10 μg/L) (WHO, 2003). The observed differences in concentrations of GCs in groundwater are due to spatial variation in the hydrogeochemical parameters (e.g., pH and redox conditions) which control element mobilization from solid to solution in groundwater (Bundschuh et al., 2004; Bhattacharya et al., 2006). This example illustrates why integrated medical geology approaches are needed in assessing problems of human exposure to GCs.

There are also numerous cases where GCs are released by human activity (Fig. 1). In particular, these include mining and hydrocarbon exploitation (the last may result in release of not only inorganic but also organic GCs) and land use activities (e.g., agriculture) which affect sediments/soils, water (ground, surface, and marine) and atmosphere (Fig. 1). These human activities can accelerate the mobilization of GCs by several orders of magnitude compared to purely natural physicochemical mobilization processes. To distinguish this group of GCs from the Naturally Released GCs (NRGCs), we introduce a new term, the Technical Enhanced Geogenic Contaminants (TEGCs), which is analogous to TENORM (Technical Enhanced Naturally Occurring Radioactive Materials) derived from NORM (Naturally Occurring Radioactive Materials). This is to distinguish between GCs which are released by natural processes such as weathering to the environment (soil, water, biota, atmosphere, etc.) (NRGCs) and those which are of natural (geogenic) origin but their release is by anthropogenic activities such as mining (TEGC).

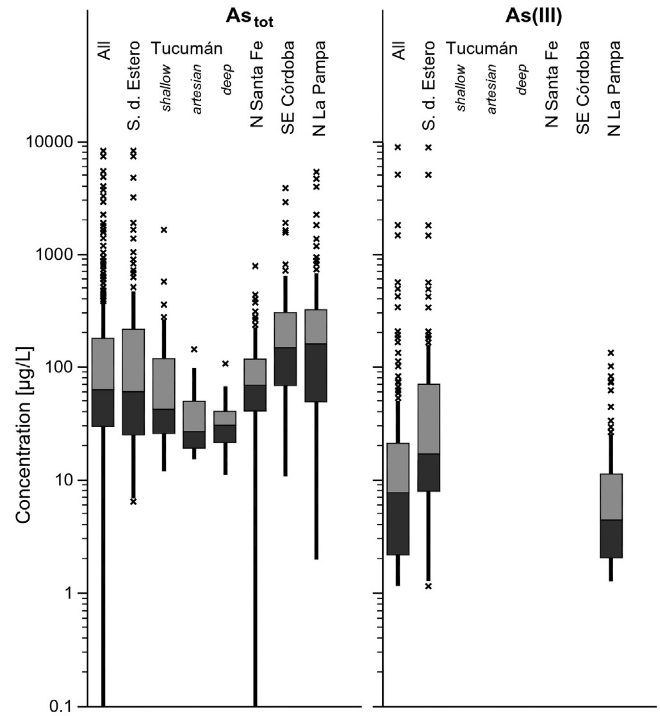

Fig. 2. Local and regional spatial variability of trace element concentrations in groundwater of the Chaco-Pampean plain (Argentina) caused by varying geochemical conditions which control element mobilization from solid to groundwater (450 data sets, ongoing work of authors).

Factors critical to progressing the science of medical geology include: (i) identifying and characterizing the sources and natural and anthropogenic factors associated with exposure to harmful geogenic elements and their species in the environment; (ii) assessing their movement and alteration over time and space; and (iii) developing evidence-based management plans for such materials to minimize or prevent exposure and health risk. In particular, there are two priorities that need to be established within the field of medical geology namely (i) the study of trace elements, especially their bioavailability and bioaccessibility; and (ii) establishing baselines, or background levels of potentially harmful but naturally occurring GCs in water, soil, air, food and animal tissue.

3. How do geological biotropic factors affect human health?

The interaction of different elements in the geological environment and human health is not always clear as there are many naturally occurring elements that are potentially toxic to humans (Dissanayake, 2005; Santamaria and Sulsky, 2010). Studies have shown that biotic life (animals, plants and other organisms) gradually accumulates a range of different elements from the abiotic (water, air, soil and rock) environment through biotic-abiotic interactions which may either result in beneficial or hazardous effects to organisms in the biogeosphere (EPA, 2004; da Silva et al., 2005; Dissanayake, 2005; Kirchmann et al., 2005; Santamaria and Sulsky, 2010). Through these interactions, organisms (including humans) experience the impacts of deficiencies, or excess exposure to one or several chemical elements in the geological materials (Santamaria and Sulsky, 2010). Several metals and metalloids

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(As, Ba, Be, Cd, Cr, Cu, Hg, Ni, Pb, Os, Sb, Se, Ti, V, Zn), inorganic halides (F^-), natural radioactive metals (Ac, Th, U, Ra), transuraniums (Pu, Am, etc.) and metallic radioactive isotopes (^{60}Co and ^{90}Sr) have impact on human and ecosystem health. Many of these elements are released from the geological materials naturally through biogeochemical processes and are potentially bioavailable to enter food chain, and thus pose potential health risk.

Furthermore, various contaminants (e.g. Cd, Co, Hg, Ni, Pb, Se and Sr) in water, sediments, soils and the atmosphere move into the food chain, either directly or indirectly (i.e. via edible plants and animals). These are likely to have a major impact on human health, depending on their bioavailability and/or bioaccessibility (Apostoli et al., 2006; Petruzzelli et al., 2010; Kar et al., 2011). The concentration and exposure time of contaminants are both important factors in determining their toxicological effects (Kar et al., 2011). In addition, contamination affects people not only physiologically but also in terms of their mental health and wellbeing. This is documented for a range of exposures, including: noise, atmospheric dust due to wind erosion of mining sites (in particular open pit mining) and other stressors (Stephens and Ahern, 2002; Zullig and Hendryx, 2010; Moffatt and Baker, 2013).

There are several challenges in medical geology, especially in identifying and quantifying the risks to human health from the geological environment. One of the biggest challenges is delineating the principal responsible element or combinations of elements (i.e. co-exposure), which can have synergistic or antagonistic effects on the toxicity of the GCs. This is because control studies do not provide enough evidence to support major impacts on human health. Instead, realization of a human health impact usually follows either accidental or long-term exposure to GCs through air, water or food. In such cases, exposure is often related to a mixture of compounds or conditions, adding to the complexity of identifying the principal responsible element(s) and chemical species, dose or duration of exposure. Knowing what is in the environment through the mapping of the concentrations, distribution and movement of GCs in relation to human activity would provide toxicologists and epidemiologists accurate exposure maps that detail the major areas to be focused on, in further studies. Knowing the concentrations at which GCs occur, and how and when these concentrations change, would potentially help to develop exposure assessments, maps and criteria which could then be used to develop the relationships on human health effects. A proactive approach of identifying potentially harmful elements without having to wait for adverse health effects to manifest should be adopted.

Key problems in exposure-epidemiological studies (i.e. the identification of GCs as cause for diseases) are, further, due to:

- the short-distance variation of exposure (Wardrop and Le Blond, 2015)—as the examples of arsenic (Bundschuh et al., 2004; Ahmed et al., 2004; Bhattacharya et al., 2006; Bundschuh et al., 2012; Nicolli et al., 2012) and radon (Badr et al., 1993; Oliver and Badr, 1995) show—where contaminant concentrations in water and air, respectively, vary by several orders of magnitude within only tens of meters. This further complicates exposure assessment, necessitating individual level studies (Badr et al., 1993; Oliver and Badr, 1995), as short-distance exposure variation makes ecological (population level) studies for geo-epidemiological assessment unsuitable (Morgenstern, 1982);
- the (often) long time-frame between initial exposure and disease onset. This requires access to retrospective information on potential exposure to GCs based on the movements over time of the individuals studied (Coughlin, 1990; Carling, 2012); this, again, cannot be addressed in ecological studies (Wardrop and Le Blond, 2015);
- the variable exposure to geogenic contaminants over time, requiring a detailed retrospective assessment of exposure (Wardrop and Le Blond, 2015).

Two examples of widely distributed GCs are fluoride (F^-) and arsenic (As), which are released and mobilized naturally and anthropogenically into groundwater, soils, plants, atmosphere and other environments from rocks and minerals. Regarding detailed information on geological sources (rocks and minerals) of arsenic and fluorine/fluoride, their release from these sources into different environments, their elemental species and (hydro)geochemistry, their mobility in sediments, soils, water, atmosphere, etc. and their global distribution, we refer to different critical reviews (e.g. for arsenic: Matschullat, 2000; Bhattacharya et al., 2007; Ravenscroft et al., 2009; Jean et al., 2010; Bundschuh et al., 2012, 2015; López et al., 2012; Murcott, 2012; Alarcon-Herrera et al., 2013; Herath et al., 2016 and for fluoride: Vithanage and Bhattacharya (2015).

3.1. The example of fluoride – the problem of human total exposure and the regulatory limits

High concentrations of F^- in groundwater are found globally (e.g., USA, Mexico, Argentina, Scandinavia, Tanzania, Ethiopia, Kenya, Pakistan, Thailand, China, Sri Lanka, and in about half of the states of India (UNICEF, 1999; Fawell et al., 2006; Kut et al., 2016). Globally, more than 200 million people from 25 nations, are currently known to be suffering from dental and/or skeletal fluorosis (Vithanage and Bhattacharya, 2015). It is well-known that chronic exposure to fluoride in drinking water constitutes an increased risk of dental and bone fluorosis in a dose-response manner; however, there is no detectable threshold (EC, 2011; Peckham and Awofeso, 2014), and F^- concentrations in excess of 1.5 mg/L are assumed that they can cause dental and skeletal fluorosis (CDC, 2001; Warren and Levy, 2003; Petersen and Lennon, 2004; Fawell et al., 2006; Selwitz et al., 2007; Peckham and Awofeso, 2014). This value corresponds to the WHO guideline value, which is explicitly stated not to be a fixed value but intended as indicative depending on local conditions). Epidemiological studies show very limited evidence of related adverse health effects due to chronic exposure to fluoride (e.g., carcinogenicity, developmental neurotoxicity and reproductive toxicity) (Fawell et al., 2006; EC, 2011; Tiemann, 2013; Peckham and Awofeso, 2014). Since (i) prevention of dental caries is the only demonstrated positive impact of fluoride on humans, (ii) no fluoride deficiency disease has ever been reported (Peckham and Awofeso, 2014) and (iii) fluoride is not in any natural human metabolic pathway (Cheng et al., 2007), fluoride is not an essential element for humans (EC, 2011; Peckham and Awofeso, 2014). There is no doubt that fluoride reduces caries by remineralization of demineralized enamel (Cheng et al., 2007; Tiemann, 2013; Peckham and Awofeso, 2014) but fluoride uptake, e.g. from drinking water (with fluoride originating from natural sources or from fluoridation) in combination with exposure to fluoride from other sources, can result in adverse effects (Warren and Levy, 2003; Fawell et al., 2006; Peckham and Awofeso, 2014.) which need to be urgently studied and evaluated, particularly in children and with regard to low level exposure (Cheng et al., 2007; Peckham and Awofeso, 2014). Due to these uncertainties, several experts describe fluoridation as a violation of medical ethics and human rights (Cross and Carton, 2003; Cheng et al., 2007; Peckham and Awofeso, 2014) since (i) fluoridation of drinking water (also of cooking salt, toothpaste, etc.) provides uncontrolled doses of fluoride over a lifetime, which may result in unknown total fluoride exposure; and (ii) there is a lack of toxicity testing of silicofluorides. This makes fluoridation a highly controversial and polarized topic, receiving both strong support and opposition within communities (NRC, 1993; Tiemann, 2013; Peckham and Awofeso, 2014), calling for detailed studies on potential benefits (i.e., caries prevention) balanced against uncertainty about the harms and risks due to low-level chronic exposure to fluoride (Cheng et al., 2007; Peckham and Awofeso, 2014). This discussion highlights the potential health impacts of geogenic fluoride in drinking water and the establishment of an appropriate fluoride guideline value for drinking water.

There is disagreement among scientists on the correct dose of F^- for tropical regions as people tend to drink more water in those regions due

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to high temperature and high evaporation rates. Hence, F^- provides an excellent example, discussing evidence of both deficiency and excess accumulation of individual elements having direct and/or indirect consequences on human health.

3.2. The example of arsenic

It has also been known for several decades, As is a common problem in Argentina, Bangladesh, Chile, India, Mexico and Taiwan causing serious health problems affecting several million people as a result of As contamination in drinking water (Kinniburgh and Smedley, 2001; Bhattacharya et al., 2002a, 2002b; Kapaj et al., 2006; Nriagu et al., 2007; Maity et al., 2011a; Maity et al., 2011b; Maity et al., 2012; Bundschuh et al., 2013; Chakraborti et al., 2013; Bundschuh and Maity, 2015) The groundwater arsenic problem is now known to occur in over 70 countries (Jean et al., 2010). Among the diseases, arsenicosis, keratosis, melanosis and other As-related diseases were reported in all six countries as a consequence of high groundwater concentrations of As (1–1300 μg/L). In Bangladesh and India, As was released by reduction of Fe-oxyhydroxides/sulfide oxidation from alluvial sediments (Chen and Ahsan, 2004; Bhattacharya et al., 1997, 2002b; Nickson et al., 2000; Ahmed et al., 2004; Rahman et al., 2010; Argos et al., 2011; Maity et al., 2012; Chakraborti et al., 2013), whereas in the oxidized sedimentary aquifers (normal pH range, $5 < pH < 10$) of the Argentine Chaco-Pampean plain, As is mobilized at high pH (8–9.5) by desorption from Fe, Al, and Mn oxyhydroxides resulting in As concentrations of hundreds to occasionally thousands of μg/L in groundwater (Bundschuh et al., 2004, 2012; Bhattacharya et al., 2006). This is the principal mechanism that also explains high As concentrations in the groundwater of most of the Chilean and Mexican examples as well as in the Appalachian Highlands of NE Ohio, the Interior Plains of South Dakota, the Carson Desert of Nevada, the Pacific Mountain System of NW Washington and Arizona (Bundschuh et al., 2010 and references therein).

Early-life exposure to As is associated with increased risk of cancer of the bladder, lung, liver and urinary tract, as well as cell carcinoma and diabetes. All these diseases have been reported in Taiwan where the As concentration was as high as 1820 μg/L (Chiou et al., 2001; Naidu and Bhattacharya, 2006; Chen et al., 2007; Chen et al., 2010; Su et al., 2011a). Pulmonary tuberculosis, lung cancer and bronchiectasis, lung and bladder cancer and liver and kidney cancers were also observed in Chile due to high concentrations of As in surface and groundwater (100–1000 μg/L) (Naidu and Bhattacharya, 2006; Smith et al., 2006; Marshall et al., 2007; Liaw et al., 2008;).

3.3. Arsenic and fluoride co-exposure in China and India

Environments (biotic and abiotic, water, air and soil) in China are experiencing increasing GCs because of elevated metal discharges from various sources (terrestrial runoff, river input, atmospheric deposition, industrial and municipal discharges, agricultural fertilizers, inferior coal ("bone coal"), indoor burning of arsenic-rich coal, mining tailings, arsenic-bearing minerals mining, etc.) (Li et al., 2012; Luo et al., 2012; Pan and Wang, 2012; Xiao et al., 2012). It is well-known that GCs such as As and F^- can exhibit toxicological effects on human health. Several studies (Centeno et al., 2002; Centeno et al., 2005; Gomes and Silva, 2007) have reported that several million people in Guizhou Province, China, suffer from fluorosis and arsenicosis due to inhalation of indoor air polluted by As and/or F. In this case, atmospheric contamination was attributed to the combustion of local As-rich coals and home-made coal balls made from local F-rich clay soils (An et al., 1997; Lin et al., 2012).

In a recent study Kumar et al. (2016a) investigated the co-contamination of groundwater by As and F^- in the Brahmaputra and Gangetic floodplains in India. Here, As and F^- is found in $Ca\text{-}HCO_3$ type groundwater in the reducing aquifers and the concentrations of Ca^{2+} and F^- are positively correlated which both are explained by the dissolution of carbonate from the neighboring source rocks as well as due to anthropogenic activities. High mobility of these chemical species is explained by high concentrations of PO_4^{3-} and H_4SiO_4. Laboratory experiments by Kumar et al. (2016b) indicate high correlation between Fe (hydr)oxide and total As content of the soils and river sediments which led to the conclusion that Fe (hydr)oxides are the principal source of As in groundwater. The study showed that Fe (hydr)oxides had high affinity for both F^- and negatively charged As oxyanions. Desorption experiments showed that As and F release into the aqueous phase was highest at high Fe (hydr)oxide contents at alkaline pH.

3.4. The example of fluoride in Brazil

Another of the many examples of fluoride contamination and related health effects is from Brazil. A geochemical survey, involving sampling of water, sediment and soil, identified dental fluorosis in children in the Itambaracá region (Parana state: PR) associated with groundwater and bromide and chloride in soil which is possibly related to cancerous diseases that occur in the northern region of this state (Licht, 2005). Geological, hydrogeological and epidemiological studies have also found fluoride anomalies in the waters of São Francisco (northern part of Minas Gerais state: MG) and established the relationship of these anomalies with the prevalence of dental fluorosis in the area. Fluoride minerals originate from the rocks of the Bambuí Group and fluorite, in particular, occurs disseminated in calcite veins and fractures in the limestone. There is a remarkable correspondence between the stratigraphy, rock fracture systems, the flow rates of wells and the fluoride concentrations in groundwater in this region (Ferreira et al., 2010).

3.5. The example of manganese

Manganese is an essential micronutrient for living organisms including plants, humans and animals. Manganese is a transition element ubiquitously found in the earth's crust. Average concentration levels in the upper crust are ca. 600 mg/kg, while in the bulk continental crust concentrations range up to 1400 mg/kg. A number of rock forming silicate minerals such as pyroxene, amphibole, garnet and olivine contain Mn. In sedimentary environments, Mn occurs predominantly as oxides such as pyrolusite, hausmannite and manganite, and is likewise common in carbonate minerals where Mn forms solid solutions with Ca, Fe and other divalent cations. In several aquifers, rhodochrosite is the common Mn carbonate mineral (Ahmed et al., 2004; Hasan et al., 2007; Bhattacharya et al., 2009; von Brömssen et al., 2007).

Manganese is redox sensitive, and its mobility in groundwater environment is influenced by the redox conditions in aquifers. Alluvial sediments are characterized by Mn concentration ranging up to 2000 mg/kg and hence their mobilization, governed by the redox conditions, may impair the quality of drinking water. Under oxidizing conditions, Mn is mostly immobile due to the occurrence of Mn (+III) and Mn (+IV) species and precipitates as hydrous manganese oxides, Mn (+II), which is the predominant species under reducing conditions mobilized in aqueous media. As a consequence, the behavior of Mn in groundwater environments varies significantly with the redox status of the aquifers and may significant impact on drinking water quality. In recent years, the occurrence of Mn in groundwater has received attention due to its neurotoxic effects (Bouchard et al., 2011) including impact on pregnant women and the birth weight and length of the newborns (Rahman et al., 2013; 2015).

Enhanced knowledge of the redox chemistry of hydrogeochemical systems in sedimentary aquifers has highlighted the global synergies of the distributions of Mn, Fe and As in groundwaters. It has been observed that reduced groundwaters from shallow Holocene sedimentary aquifers are characterized by low dissolved Mn concentrations, while oxidized aquifers are enriched in Mn and so considered an important constraint on the safe use of groundwater for drinking water supplies

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(von Brömssen et al., 2007, 2008; Biswas et al., 2012a, 2012b; Hossain et al., 2014, 2016). From the perspective of the drinking water safety plan (WHO, 2004) and its implementation, Mn is considered an important water quality parameter for the assessment of drinking water quality.

3.6. The example of radon

Radon, a radioactive gas originating from radioactive decay of radium—a daughter product of uranium—is another example of a GC. This naturally occurring gas is colorless, odorless and tasteless and is found in soils and rocks in low concentrations but can be highly elevated in soils of/or close to uranium ore deposits, where concentrations can vary significantly over only tens of meters. Outdoors, radon is not a major health risk because it dilutes in the environment. However, a concentration above 4 pCi/L of air in a closed environment, such as in occupational settings (mines) or residential buildings, is dangerous as it can cause lung cancer and other adverse health effects (EPA, 2012). In the 1980s, a new concern involving the accumulation ("build up") of radon in populated areas emerged in the USA. Although radon spreads and accumulates in poorly ventilated cavities (caves, mines), it has also been detected, depending on underground (soil, rock) composition and construction materials, in high concentrations in homes, particularly in basements and ground floors (Silva et al., 2014b). In several areas of Brazil, radon concentrations in buildings exceed the 150 Bq limit of the US Environmental Protection Agency (USEPA) and the International Commission for Radiological Protection (ICRP). Examples of impacts of human health are found in municipalities in the semi-arid uranium mining areas geologically known as the Mining Province of Borborema (Campos et al., 2013a, 2013b), where abnormally high incidence of different cancers (including lung cancer) have been observed (Malanca and Gaidolf, 1996a; Malanca and Gaidolfi, 1996b; Veiga et al., 2003; Silva et al., 2014b).

3.7. Geogenic contaminants related to hydrocarbon exploitation (TENORM)

Increased exploitation of, in particular, unconventional fossil fuel resources results in increased release of TEGCs into water, soils and the food chain, whereas burning of these resources and mine overburden may contaminate the environment and atmosphere. The availability and mobility of GCs associated with the exploitation of unconventional energy resources are geographically variable and practically unknown. Heavy metals, but especially NORM/TENORM, are found at concentrations exceeding background levels in organic-material rich rocks (coal, gas/oil bearing formations) as was reported several decades ago for the extraction and exploitation of conventional coal and hydrocarbon resources (Zielinski and Otton, 1999).

For example, with respect to conventional oil production, Zielinski and Otton (1999) researched TENORM in oil produced water and oil-field equipment, the last of which was recognized as an apparent problem in the 1980s in the USA where unacceptably high radiation was detected in oil-field pipes of scrap metal dealers. A preliminary nationwide reconnaissance of radioactivity of oil-field equipment proved that γ-radiation levels exceeded natural background radiation at 42% of the studied sites (Zielinski and Otton, 1999). Oil-field equipment and sludge can include barite scales, which can incorporate Ra in its lattice and which is documented as primary host for oil-field TENORM (Zielinski and Otton, 1999). The highest reported Ra contents in oil-field waste and scale in the USA are hundreds of Bq/g (several thousand pCi/g) and occasionally as much as 15 kBq/g (400,000 pCi/g) (Zielinski and Otton, 1999). Fisher (1998) found that the concentration of dissolved ^{226}Ra from 215 oil produced waters in the USA correlates positively with water salinity and reported maximum concentrations of dissolved ^{226}Ra of up to 370 Bq/L (10,000 pCi/L) and dissolved ^{228}Ra concentrations of half to twice those of dissolved ^{226}Ra. Zielinski and Budahn (2007) studied the natural radioactivity of produced water from a NE Oklahoma oil field and found similar activity concentrations for dissolved ^{226}Ra and ^{228}Ra (~1500 dpm/L). Hamlat et al. (2001) found ^{226}Ra activities of 0.006–0.02 Bq/g in crude oil, 5.1–14.8 Bq/L (mean 10.4 Bq/L) in produced water and 1–950 Bq/g in hard scale samples (75% of the samples exceeding the 10 Bq/g limit). Godoy and Petinatti da Cruz (2003) reported ^{226}Ra and ^{228}Ra in 36 scale and sludge samples produced at an offshore oil field in Brazil (Campos Basin) and correlated their concentration with the chemical composition. These authors found that the majority (75%) of samples were barium and strontium sulfates, with uniform Ra contents (mean ^{226}Ra and ^{228}Ra content of 106 kBq/kg and 78 kBq/kg, respectively). In the sludge, ^{226}Ra and ^{228}Ra content varied significantly, ranging from 0.36–367 kBq/kg and from 0.25–343 kBq/kg, respectively.

In the case of shale gas (SG) exploitation, recent reports have identified a very wide range of concentrations of TENORM in the environment, including water resources, potentially affecting human health. This has created significant community concern, although this information currently relates only to the USA (EPA, 2010; Vengosh et al., 2014). There, the range of radium activity for samples from Marcellus Shale varies from below the detection limit to 667 Bq/L (18,000 pCi/L), with a median concentration of 91 Bq/L (2460 pCi/L) (Rowan et al., 2011). This range overlaps the range for non-Marcellus reservoirs, which range from below detection limit to 248 Bq/L (6700 pCi/L) with a median of 27 Bq/L (734 pCi/L) (Rowan et al., 2011). At present, worldwide, there are no publicly available reports of TENORM release from coal bed methane (CBM) exploitation. The production of CBM, which requires the dewatering of the coal seams in order to extract gas, involves large volumes of formation water being brought to the earth's surface as a waste product of the process at a methane gas to produced water ratio of 26–78 GL/10^9 m^3. Due to the high volumes of water produced, exploitation of these resources may severely impact groundwater aquifers and other environments. Even if concentrations are moderate (and below regulatory limits), the trace elements present in the waste water, which can include toxic heavy metals and radionuclides, may accumulate with time (biomagnification), leading to serious degradation of the freshwater resource and agricultural soils and long-term environmental and human health impacts.

3.8. Geogenic contaminants related to mining (TEGC) – Brazil as one of the many global examples

The accelerated release of a geogenic contaminants occurs through mining activities. At several sites in the Iron Quadrangle of Minas Gerais state (MG), soils contain anomalous As concentrations. The contribution of mining and metallurgy to As release has been studied by several researchers (Oliveira et al., 1979; Borba et al., 2000; Matschullat et al., 2000; Deschamps et al., 2002; Borba et al., 2003; Borba and Figueiredo, 2004) who identified high As levels in the urine of children living near a gold mining area (n = 126, mean: 25.7 μg/L of total inorganic As), with 20% exceeding the 40 μg/L threshold above which adverse long-term health effects cannot be excluded. Since the As concentration of drinking water was below 10 μg/L, the authors concluded that the As exposure route was via contact with soil and dust. It is likely that climate change will increase such risks in the future. By the end of this century, predicted atmospheric temperature increase of 4–5 °C will cause both an increase in human metabolic processes and also in the movement of As particles in the atmosphere and their reactivity (NIEHS, 2010; USGCRP, 2016). Hence, climate change is likely to exacerbate or increase the prevalence of exposure to high As (and other GCs) in the future.

In the city of Paracatu (northwest of Minas Gerais), which at present has Brazil's largest gold mine and the world's largest open pit, large volumes of liquid and solid As-rich wastes are produced, the last being deposited in the form of sterile waste piles of batteries. Dissolution of As-minerals (e.g. arsenopyrite) is a continuing source of contamination. However, reports indicate that the geology and mining activities have no significant impact on total As exposure in Paracatu (Bidone et al.,

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2014; Egler et al., 2014; Mello et al., 2014; Sabadini-Santos et al., 2014; Silva et al., 2014a). On a regional scale, the concentrations of stream sediments (>1000 mg/kg) around Paracatu are high, while those of surface waters rarely exceed 10 μg/L. However, the risk associated with exposure to As in Paracatu (0.4352 μg/kg/day for adults and 0.3536 μg/kg/day for children) can be considered low compared to the $BMDL_{0.5}$ 3 μg/kg/day (JECFA, 2011; De Capitani et al., 2014; Ng et al., 2014). This conclusion is supported by the results of Ono et al. (2012) and Guilherme et al. (2014) who found very low As bioaccessibility despite the presence of high concentrations of As in the tailings of Paracatu mine. These studies considered the possibility of exposure of children (the most susceptible population group) by involuntary ingestion of As-contaminated soil in the Paracatu mining area, evaluating the As-bioaccessible fraction. Arsenic was predominantly present as As(V) and was mostly associated with poorly crystalline Fe arsenate which explains the low bioaccessibiliy of As and highlights the importance of Fe oxides in immobilizing As in the environment (Freitas et al., 2015). The incidence of diseases, including cancers, in Paracatu is similar to the average values in Minas Gerais state (CEMEA, 2012; De Capitani et al., 2014).

In Amapá (AP), As occurs in arsenopyrite associated with the Precambrian manganiferous formations from which manganese ores have been exploited for >50 years in the Serra do Navio mine. However, in this case, the As contamination source is not located at the mine, but 350 km away in the municipality of Santana, on the banks of the Amazon River, where the ore is processed and shipped. Ores and outdoor-exposed waste contain up to 0.17% As and the groundwater collected from monitoring wells near these deposits contains up to 2000 μg/L As (Figueiredo, et al., 2010). Santos et al. (2003) investigated exposure in the population living near the tailings, analyzing samples of hair and blood, without identifying significant risk to people.

In southern Brazil, coal deposits associated with As and coal mining in the states of Santa Catarina (SC) and Rio Grande do Sul (RS) have caused significant environmental impacts, represented today by giant deposits of waste and sulfurous lakes (Akcil and Koldas, 2006; Blodau, 2006; Campaner and Silva, 2009).

In addition to the concerns and research of As mobilized by mining activities in Brazil, several researchers have studied the release of Pb and its impact on human health (Paoliello et al., 2001; Kuno et al., 2013; Freire et al., 2014; Lopes et al., 2015); most data included children, women or occupationally exposed population groups. Accidental ingestion of soil particles is one of the most important routes of exposure to heavy metals and metalloids, occurring with hand-mouth transfer during outdoor activities, especially in children (Paoliello et al., 2002). USEPA studies estimate a soil ingestion rate of 50 mg/day for children aged 1 to 6 years (EPA, 2008). The Center for Disease Control and Prevention (CDC) recommends public health actions be initiated at a blood lead level of 5 μg/dL, a value derived from the 97.5% percentile of values obtained in children, aged 1 to 5 years, sampled by the National Health and Nutritional Examination Survey (NHANES) (http://www.cdc.gov/nceh/lead/).

It is, however, not easy to estimate the amount of Pb that is absorbed into the systemic circulation (the concept of bioavailability), because this depends on the form and solubility of Pb in the soil matrix and the child's nutritional status (Ruby et al., 1996; Ryan et al., 2004). The way that Pb is connected to solid soil phases can make it unavailable for absorption. To understand the effect of soil properties on the bioavailability of metals, we need knowledge of a metal's bioaccessibility, absorption and metabolism.

The first studies to determine the bioavailable fraction involved in vivo assays with different animal models (Marschner et al., 2006; Juhasz et al., 2009). However, such studies are ethically controversial, expensive and time consuming; cannot be applied to a large number of samples; and are, hence, impractical to use routinely. As the bioaccessibility of a contaminant is a major limiting factor of its bioavailability, this parameter can be used in risk assessments as a conservative measure of bioavailability, with the advantage of being estimated in vitro. The first in vitro method was developed by Ruby et al. (1996) and named Physiologically Based Extraction Test (PBET). Currently, several in vitro methods for estimating bioaccessibility have been reported in the literature and thoroughly reviewed (Wragg and Cave, 2002; Wragg et al., 2011), all of which are based on the PBET developed by Ruby et al. (1996).

The first assessments of children exposed to Pb were conducted in the city of Santo Amaro da Purification (Bahia) in the 1980s and 1990s (Carvalho et al., 1984; Carvalho et al., 1985; Silvany-Neto et al., 1989; Tavares et al., 1989; Carvalho et al., 2003). A cohort of 555 children (1–9 years old), living within radius of 900 m from Pb smelting factory, showed an average blood Pb level of 59.1 ± 25.0 μg/dL (Carvalho et al., 1985). After the final closure of the smelter, sampling children aged one to four years, the same researchers obtained average blood lead levels of one-third (17.1 ± 7.3 μg/dL), but with 88% of children still at >10 μg/dL and 32% at >20 μg/dL (Carvalho et al., 2003). The Pb concentrations in whole blood, serum, saliva and house dust in samples collected at two time points (12 months apart) showed that Pb concentrations decreased from 2010 to 2011 (de Souza Guerra et al., 2015).

From 1945 to 1995 in the Ribeira Valley region on the border between the states of São Paulo (SP) and Paraná (PR), several Pb-Zn-Ag mines operated together with a lead refining plant (Plumbum) in the city of Adrianople (PR). High contents of Pb have been found in the blood of children and adults living in the vicinity of the Plumbum plant (Paoliello et al., 2001; Paoliello et al., 2002; Cunha et al., 2005; Figueiredo, 2005; Paoliello and De Capitani, 2005). The first study (in 1999 and 2000) found adult blood Pb levels of 14.55 μg/L for men (n = 46) and 6.80 μg/dL for women (n = 55) (Paoliello et al., 2001). In another study, Paoliello and De Capitani (2005) found a mean Pb level of 11.25 μg/dL in the blood of 295 children (7–14 years of age) living up to 1.5 km from the contamination site, whereas control children living 40 km upstream of the source had an average of 1.8 μg/dL confirming the environmental liability.

4. Future directions and needs in medical geology

With the aim of contributing to a better understanding and raising awareness of the need to reduce human exposure to GCs, we discuss perspectives of "medical geology" within the framework of global change involving climate, population and economic growth and the consequential related increases in demand for water, energy, food and metallic and industrial minerals. In addition to these "negative" changes, we further consider beneficial developments which may assist in protecting humans against exposure to GCs and highlight the reasons why the role of medical geology will increase. In particular, we highlight recent research progress in the fields of geology/geochemistry, toxicology and epidemiology which has resulted in an exponential increase in evidence and understanding of the interactions between GCs and human health.

4.1. Generating better understanding of the global incidence and impact of GCs

Thanks to advances in instrumentation and analytical chemical techniques, GCs and their species are now capable of being detected at low levels almost routinely in more and more regions where they have not been previously identified. Several procedures and instrumentations have been continuously improved (atomic absorption spectrometry (AAS); atomic emission/fluorescence spectrometry (AES/AFS); high/ultra performance liquid chromatography-atomic fluorescence spectrometry-interfaced via a UV-photoreactor and a hydride generation unit (HPLC/UPLC-UV-HG-AFS). Inductively coupled plasma mass/optical emission spectrometry (ICP-MS/ICP-OES)—which further can be coupled to HPLC/UPLC, neutron activation analysis (NAA), X-ray fluorescence (XRF); energy dispersive X-ray fluorescence (EDXRF) and anodic striping voltammetry (AVS)— are available to determine the GCs in soil, waste or drinking water, bodily fluids, minerals and chemical compounds (Šlejkovec et al., 1999; Helaluddin et al., 2016). Techniques such as NMR, mass spectrometry and chromatographic procedures are also useful for GC determination. Both quantitative and qualitative determination of GCs are important in environmental and health studies. Quantitative measurements of GCs use different techniques such as optical atomic spectroscopy, flame atomic absorption, graphite furnace atomic absorption, and inductively coupled plasma atomic emission spectroscopy. Neutron activation analysis is a process that involves the activation of a contaminate matrix through the process of neutron capture. In this process the radioactive target nuclei of contaminate begin to decay, emitting gamma rays of specific energies that identify the radioisotopes present in the sample and the concentration of GCs determine using an irradiated standard. The qualitative study of GCs is determined by different mass spectrometric atomic spectroscopy techniques (e.g. ICP-MS, ICP-SF-MS, ICP-Q-MS, X-ray fluorescence, particle-induced X-ray emission, X-ray photoelectron spectroscopy, and Auger electron spectroscopy) (Šlejkovec et al., 1999; Helaluddin et al., 2016). Furthermore, synchrotron X-ray microfluorescence (μXRF) and synchrotron X-ray computed microtomography (μCMT) techniques can be used for detection of metal concentration and spatial distributions in biotic and abiotic samples (Feng et al., 2016). A wider global geographic understanding of GC distributions will greatly assist in building better knowledge and awareness of the critical factors that impact human health. The pressing need for better collaboration between geologists, toxicologists and epidemiologists can be demonstrated through the example of groundwater contaminated by geogenic As.

The first scientific descriptions of As contamination of groundwater are from the early 1900s in Argentina, where toxic effects on public health (cardiovascular and cutaneous manifestations) due to groundwater consumption were described in the area of Bell Ville, near Córdoba city. At that time the observed effects were nominated as a new disease, "Bell Ville disease", in several publications in the years between 1913 and 1917 (Ayerza, 1917; Goyenechea, 1917). However, the disease was considered a local issue only.

The lack of collaboration between geologists, toxicologists and epidemiologists, the scarcity of data on medical problems linked to the occurrence of As and its speciation in drinking and irrigation water supplied from groundwater and, to a minor extent, from surface water resources in many countries, especially in the developing world, and the non-inclusion of As as a key parameter in standard drinking water analysis, has led to significant numbers of people being unnecessarily exposed to As. No global research on As has been performed and aquifers with high concentrations of dissolved As have often only been detected by accident (maybe as a consequence of research originally performed for other purposes) or through the observation of a significant increase in As related health lesions in a given region. Over the last 20 years, As has been found as NRGC in the groundwater of over 70 countries and 250 principal regions (Fig. 3a) (Bundschuh and Litter, 2010; Jean et al., 2010) and every year new countries or regions are reported from where this problem has not be known before (the same can be expected for other GCs). This has finally changed the long-lasting opinion that the As groundwater problem was limited to just a few regions (e.g. in Argentina, Bangladesh, Chile, India, Mexico, and Taiwan).

Today groundwater As is recognized as a global problem. Fig. 3b shows the modeled probabilities of finding As in groundwater at concentrations > 10 μg/L based on geological/geochemical conditions: As source availability; and favorable hydrogeochemical conditions for mobilization into groundwater (Amini et al., 2008). Comparing the actual observed global distribution of As in water resources (Fig. 3a) with the modeled probabilities indicates numerous areas with a high probability that As would be detected in groundwater in concentrations > 10 μg/L. In these places, ongoing lack of detection

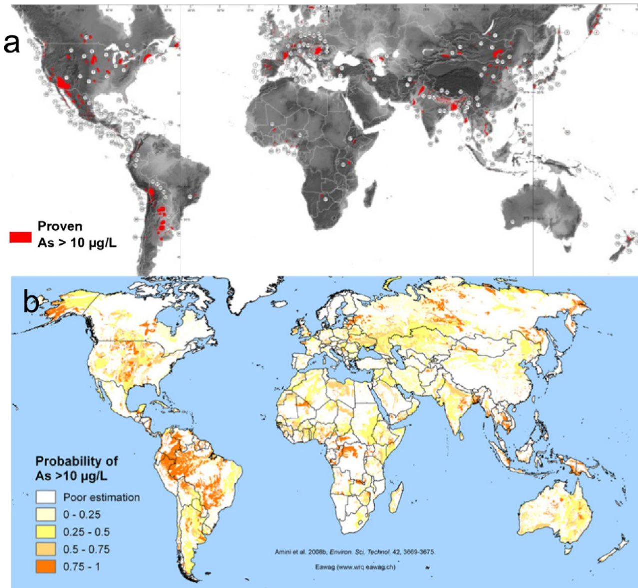

Fig. 3. (a) Global distribution of As (>10 μg/L) in water resources (mostly groundwater) (adapted from (Bundschuh and Litter, 2010; Jean et al., 2010); for details see supplementary material); (b) Modeled probability of groundwater contamination by As for reducing groundwater environment (reductive mobilization of As into groundwater) and oxidizing/high-pH environment (desorption of As into groundwater) (adapted from Amini et al., 2008).

may be due to lack of research and/or non-inclusion of As as a parameter in standard drinking water analysis.

Food, in turn, can be a source of exposure to As. Foods are known to contain organic As (fish, crustaceans and other seafood) (Hsueh et al., 2002) but also inorganic arsenic which in many cases is more toxic than organic species. In 1999, a study on the concentration of As in food consumed in the United States drew attention to high concentrations of inorganic As (As-i) in rice (Schoof et al., 1999), starting numerous As and rice studies to improve our understanding of the metabolism and mitigation of such contamination, since this cereal is of great importance in the global food system.

Rice, in contrast to other cereals, is usually cultivated under flooded conditions where excess water can lead to increased mobilization of As (especially As-i), and consequently to an increase in the accumulation of As by the plant, mainly in the grain. As(III), the more toxic inorganic species found in foods, has high solubility in water; this increases its mobility in soil where it is efficiently absorbed by the roots (Ma et al., 2008; Xu et al., 2008). Hence, chemical speciation of As-i in grain studies is essential, because the total As concentration may not provide the best measure of risk; the grain may contain high concentrations of total As-i but low concentrations of A(III) (Cullen and Reimer, 1989).

Different cultivation techniques affect the interaction of As with other elements such as silicon (Si), phosphorus (P), iron (Fe) and sulfur (S). Agricultural practices, such intermittent irrigation (Arao et al., 2009; Li et al., 2009; Rahaman et al., 2011; Hu et al., 2013; Rahaman and Sinha, 2013); the use of microorganisms of the natural microflora that adsorb (Sharples et al., 2000; Srivastava et al., 2011; Su et al., 2011b), accumulate and/or metabolize the As present in the soil; control of gene expression to increase plant resistance (Meng et al., 2011); grain polishing (Sun et al., 2008); and preparation for consumption (Raab et al., 2009; Sun et al., 2012), seem to be viable strategies to reduce the concentration of As in rice and its transfer to the final consumer.

Given the range of reported toxic effects from exposure to arsenic and its presence in rice, various agencies such as the Food and Agriculture Organization (FAO), the World Health Organization (WHO), the United States Food and Drug Administration (FDA), the Food Standards Agency (FSA) and the European Food Safety Authority (EFSA) are in

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intensive discussions on the concentration of arsenic safe for human health.

The FDA has been monitoring the concentration of toxic elements, including As, in a variety of foods including rice and juices since 1991 (FDA, 2016). In 2012, the FDA issued a statement that the As concentrations found in an assessment of more than 1300 samples were not high enough to cause immediate damage to human health. The results do not report, however, the health effects of chronic exposure or what can be done to reduce As concentrations. Still, this analysis is the first step in a major effort to ensure the safety of the consumption of rice and rice products (FDA, 2013).

On 17 July 2014, in Geneva, the Codex Alimentarius Committee, together with the FAO/WHO, adopted new standards to protect the health of consumers around the world, including the establishment of a maximum acceptable concentration of 0.2 mg/kg of As-i in rice. This committee has the task of establishing international standards for the safety and quality of food for consumers worldwide, which are widely used as a basis for national legislation (FAO, 2014).

These examples demonstrate the importance of including potentially harmful chemical species in water quality testing and highlight the need for closer collaboration between geologists, toxicologists and epidemiologists. Testing for many parameters, which in the past often had financial limitations or in some cases technical restrictions, is today much easier and more affordable, thanks to continual improvements in analytical instrumentation and techniques that allow rapid multi-species GC speciation and quantification at trace levels. Still, there is critical demand for increased investment in research activities to better understand aquifer systems and in the monitoring and reporting of the presence and concentrations of potential geogenic contaminants.

4.2. The role of geogenic contaminants (GCs) in diseases with unknown etiology and exposure to multiple GCs

In addition to the proven health impacts of GCs, there are cases where diseases with unknown etiology (causation) have been found. In some cases, these may be due to exposure to specific contaminants (or combinations of contaminants) of geogenic or anthropogenic origin found in water resources (drinking or irrigation), soils used for crop production for human consumption or in the atmosphere. Also of significance are the interactive or additive effects of individual potential geogenic or anthropogenic toxins (or combinations of these). For example, cadmium and arsenic have additive effects on the kidney (Jayatilake et al., 2013). Such effects will be missed if exposures to single compounds are examined. Relevant examples include endemic levels of the chronic kidney disease of uncertain etiology (CKDu) in Sri Lanka (Jayatilake et al., 2013), which recently achieved much interest, and blackfoot disease (BFD) in coastal areas of southwestern Taiwan before 1990 (Tseng, 2005). BDF, for which the NRGC As could be clearly identified as one—but not the only—cause, provides an excellent example of an unknown etiology and how additive effects can lead to specific health effects. This severe type of peripheral vascular disease, which is caused by As in drinking water combined with one or more other unknown environmental co-factors, has attracted global research. The combination of As and fluorescent substances, humic acids, and silica in artesian well water has been hypothesized as the cause of BFD. However, due to the absence of integral geomedical approaches at that time, there has never been any epidemiological study to assess the association between exposure to these chemicals in well water and the occurrence of the regionally-endemic BFD. Neither hazard identification nor dose-response assessment has been done for these chemicals (Jean et al., 2010). The fact that BFD could be correlated to exposure to As, but its cause could not be determined, highlights the importance of distinguishing between correlation of contaminant exposure and causation of GC induced diseases.

Over the past few decades, several researchers have studied the CKDu problem in Sri Lanka and put forward hypotheses for possible causative factors (Jayatilake et al., 2013; Dharma-wardana et al., 2015; Wimalawansa, 2015; Wasana et al., 2016). Possible causes include (i) hardness and/or high content of fluorides and/or high iconicity in drinking water; (ii) use of cheap aluminum cookware which is easily solubilized by fluoride in water; (iii) excessive use of agrochemicals containing nephrotoxic chemicals, such as compounds of heavy metals and metalloids like cadmium and arsenic or phosphate (hyperphosphatemia); (iv) consumption of food items such as lotus roots and smoking tobacco, both of which have high cadmium (Cd) levels; (v) algal toxins in drinking water supply; (vi) nephrotoxic ingredients (e.g. Sapsanda) in widely used ayurvedic herbal medicines; (vii) excessive dehydration in the work environment of farmers; (viii) genetic susceptibility to nephrotoxins; and (ix) multi-factorial causes (i.e. all or most of the above). Among the hypotheses, heavy metals/metalloids such as Cd and As and pesticide residues (Jayatilake et al., 2013; Jayasumana et al., 2014; Jayasumana et al., 2015) and fluoride, high iconicity, aluminum (Dharma-wardana, 2015) have significant attention. Since high levels of Cd were detected in people living in CKDu-prevalent areas, geological and spatial mapping of the distribution of As, Cd and other potentially toxic heavy metal(loid)s in soil, water and food crops, overlaid with the distribution of CKDu, would help determine the link with As or Cd of geological origin (Jayatilake et al., 2013). With regard to causative agents, fluoride and cadmium ions seem to be most appropriate but the latter species, i.e., cadmium, is reported non-significant, in soluble forms in the waters the CKDU vulnerable people drink (Rango et al., 2015; Diyabalanage et al., 2017). These differences in results may be related to the analytical techniques employed for As and Cd analysis; in many cases, analytical techniques used are outdated, or not validated, or not verified with environmental standards, and, consequently, results are incomparable. However, to date, the attention is focused on high iconicity and fluoride in groundwater as causative factors for CKDu in Sri Lanka. Well-designed experiments, together with the use of correct methodology, are needed to address the CKDu problem globally. It is estimated that Sri Lanka has a CKDu affected population of 400,000 people and resulted in a death toll of around 20,000 people during the last 10–12 years (Gunawardena, 2012). Similar cause-unknown kidney diseases have been found in the Srikakulam District in Andhra Pradesh, India (Machiraju et al., 2009), Egypt (Kamel and El-Minshawy, 2010) and Mesoamerica (Correa-Rotter et al., 2014).

In Sri Lanka, fluoride distribution in groundwater wells may be linked to the presence of high dental fluorosis in the dry zone (Dissanayake, 1991; Jayawardana et al., 2010; Young et al., 2011; Chandrajith et al., 2012). High fluoride levels in groundwater are suspected as the causative factor for the CKDu (Chandrajith et al., 2010; Wanigasuriya, 2012). Anomalous concentrations of fluoride in groundwater wells in Sri Lanka are due to the basement rocks of hornblende biotite gneiss, biotite gneiss, and granitic gneiss (Young et al., 2011). The most recent groundwater fluoride map of Sri Lanka indicates that over 25% of wells in the dry zone have fluoride concentrations of >2 mg/L with some wells reaching 5 mg/L (Chandrajith et al., 2012). Longer groundwater residence time in fractured crystalline bedrocks may enhance fluoride levels in these areas and those of other ions, which are the major suspected causative factor for CKDu. Furthermore, irrigation practices may induce leaching of fluoride from soils which corroborates the high fluoride levels found in shallow groundwater in the intensive agricultural areas in the dry zone of Sri Lanka (Young et al., 2011).

The example of the mysterious kidney disease in Mesoamerica is a good example for a chronic health disaster requiring a fast response. Here, the CKDu epidemic occurs for about 1000 km along the Pacific coast expanding from southern Mexico through El Salvador (Peraza et al., 2012; VanDervort et al., 2014) and Nicaragua (O'Donnell et al., 2011) to Costa Rica (Cerdas, 2005). This currently unexplained new disease has emerged in the last decade and has been named Mesoamerican nephropathy (Correa-Rotter et al., 2014). A recent study reports the involvement of infectious etiology in the Mesoamerican epidemic and indicated the need for strong and quick actions in diagnosing, treating and preventing the disease (Murray et al., 2015; Kupferman et al., 2016). Data from El Salvador and Nicaragua indicate a five-fold increase in fatalities in young men in the last 20 years; however, the observed increased incidence may be related to improved case search/diagnostics. In El Salvador, CKDu is the second leading cause of death among adult men, and has caused the premature death of at least 20,000 men in the region (Wesseling et al., 2013). In El Salvador alone, CKDu of all causes kills at least 2500 people each year (Cohen, 2014).

A high-level meeting held in San Salvador (El Salvador) in April 2013 has led the Panamerican Health Organization (PAHO) to finally declare CKDu “a pressing and extremely serious health problem in the region” (PAHO, 2013). The Ministers of Health of the Central American Integration System (SICA, the Spanish) declared their commitment to comprehensively address CKDu and to “strengthening scientific research in the framework of the prevention and control of chronic non-communicable diseases” (Health Ministry El Salvador, 2013).

While numerous risk factors have been proposed so far, the causes of the CKDu remain uncertain and controversial. There is some consensus that this is a multifactorial disease, and the involvement of pesticides, environmental toxins, well water contamination, heavy metals and arsenic of geogenic or anthropogenic origin, high temperatures, insufficient water intake and other factors have been speculated. However, agrochemicals and heavy metals, for example, are ubiquitous in both endemic and non-endemic areas. The Mesoamerican Pacific coast is rich in young and recent volcanic activities and the environment (e.g. soil, water resources, atmospheric dust, irrigation water, food chain) is consequently rich in arsenic, cadmium and other toxic elements; however, no studies of these in relation to CKDu and other chronic diseases have been conducted so far.

Determining the etiology of endemic diseases such as CKDu requires an integrated approach which links the occurrence/prevalence area of the respective disease with potential anthropogenic, biological and geogenic contaminants. The need to develop such environmental matrices highlights the importance of a geomedical science that integrates research among geologists, toxicologists, epidemiologists, etc. Standard epidemiological methods, such as observational studies combined with individual level exposure data, are not practical in this context for several reasons. First, there is little or no information available on exposure(s). Second, the population numbers and diversity of occurrences across countries are enormous. Third, the costs of conducting an epidemiological study in even one country will be huge. Therefore, tools such as a regional cloud-based GIS with data upload, processing and sharing of secondary health, geological, environmental and risk factor (Multi Hazard Vulnerability Mapping, MHVM) data would provide an initial basis for designing epidemiological investigations and monitoring the change of health outcomes with risk factors in this vast area of Mesoamerica.

At present, Mesoamerican nephropathy is a medical enigma yet to be solved. A medical geology approach is needed to determine whether or not geogenic contaminants or combinations of them (multiple stressors) are principal cause(s) of CKDu. If affirmative, this will allow mitigation measures to be developed to prevent continuation of the Mesoamerican CKDu disaster. If negative, it will limit, and so simplify, the research to other non-geogenic stressors.

Another example of (partly) unknown disease causation is the occurrence of podoconiosis, a non-lymphatic elephantiasis, which could be correlated to long-term (>10 years), bare-foot exposure to specific soil types (red soils of volcanic origin) in tropical climates (Davey et al., 2007; Molla et al., 2014). However, no specific (or combination of) individual elements or other characteristic in these soils have yet been identified (Wardrop and Le Blond, 2015), despite of several studies proposing a correlation of podoconiosis with silicon and aluminum (Price and Henderson, 1978), zirconium (Frommel et al., 1993), smectite, mica and quartz (Molla et al., 2014).

4.3. The role of accelerated exploitation of geological resources in GC release

Population and economic growth, together with the uncertain impacts of climate change, are forcing energy, water and food security issues to the forefront. Such growth drives increased demand for and exploitation of groundwater and mineral resources, as well as conventional but increasingly more unconventional fossil fuel resources such as tight gas (TG), shale gas (SG), shale oil (SO), coal bed methane (CBM), and tar sands (TS) in order to replace conventional declining nonrenewable resources. The wide global distributions of SG and SO highlights the importance of understanding the implications of increasing exploitation of unconventional fossil fuel resources to the presence of TENGCs and increased potential for human and ecosystem exposure (EIA, 2013).

Energy demand is predicted to increase rapidly over the next few decades (EIA, 2014) with associated increased mobilization of GCs and potential health impacts. Increased exploitation of geogenic energy resources commonly results in increased release of TEGCs into water, soils and the food chain, whereas erosion or burning of these resources and mine overburden primarily contaminates the environment and atmosphere. Since unconventional fossil fuel resources are forecasted to fill the energy gap between the ever increasing energy demand and the declining availability of conventional fossil fuels, special consideration of unconventional fossil fuels is needed. Indeed, while SG and CBM extraction industries are mature in the USA, other countries rich in these resources have only recently commenced exploitation or are still in the exploration phase.

4.4. The role of accelerated expansion of agricultural areas, soil degradation, desertification erosion and climate change

Global food demand is forecast to increase by 70% to feed the 9.2 billion people expected to populate the earth by 2050 (Alexandratos and Bruinsma, 2012). Since about 70% of current freshwater demand is for agriculture, this sector is likely to be the main consumer responsible for the predicted increase in freshwater withdrawal, which is expected to increase by about 10–12% per decade, corresponding to an increase of 38% from 1995 to 2025 (World Water Assessment Programme, 2009).

Comparison of the maps in Figs. 4a–d indicates that areas of highest population density, water scarcity and soil degradation coincide. This is also true for projected adverse climate change impacts on agriculture in several regions worldwide. However, a main concern for groundwater quality governance, in most cases, is not climate change but global change, which depends on population, human activity, living standards, land-use and other factors related to development. While the impact of these global changes is likely to be significant in the coming decades, this is not always duly considered in water resource or food production systems planning, research and knowledge transfer.

Unsustainable irrigation and intensive farming practices have already resulted in significant environmental damage by soil degradation (e.g. salinization, desertification) in many areas of the world, a process that is difficult to reverse. Every year about 10 million hectares of arable land are lost due to desertification or, to a lesser extent, through leaching and erosion of soil associated with intensive agriculture (Fig. 4c) (den Biggelaar et al., 2003). Increased erosion through wind and water activity results in the mobilization and transport of GCs to streams, sediments and atmosphere, where they can be transported over long distances and affect water resources and the food chain, and be directly ingested or inhaled by humans. The same is true for more conventional agricultural areas and other areas without permanent vegetation cover (e.g. mining areas).

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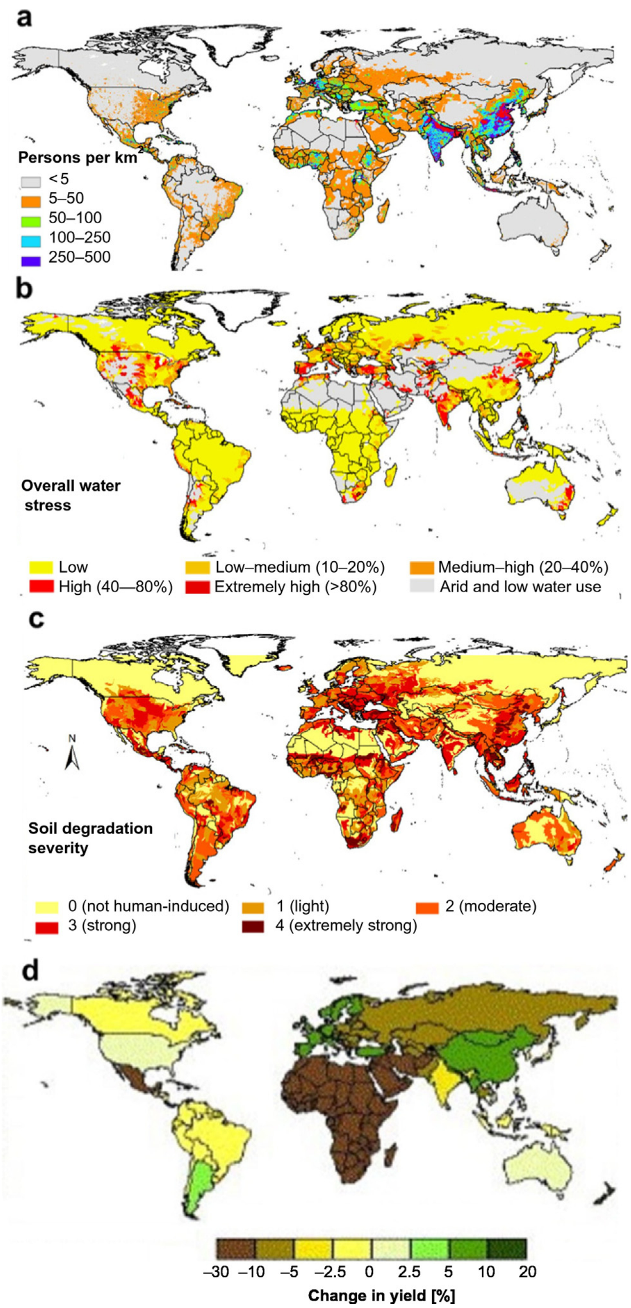

Fig. 4. (a) Global population density in 2000 (Source: CIESIN, Columbia University, and CIAT, 2005); (b) Global water stress, expressed as the ratio total annual water withdrawals (municipal, industrial, and agricultural) by total annual available blue water (Source: Aqueduct global maps 2.0 - World Resources Institute); (c) Human-induced soil degradation severity (ISRIC-World Soil Information; http://www.isric.org/data/global-assessment-human-induced-soil-degradation-glasod); (d) Global warming and Agriculture: Potential change in national cereal yields for the 2080s (compared with 1990) under the HadCM3 SRES A1FI with CO_2 effects (Parry et al., 2004).

4.4.1. Uncertain impacts on groundwater, surface water and seawater bodies

Soil degradation, and its extreme form desertification, results in increased risk of erosion by water and/or wind. Erosion by water, increases sediment (and so GCs) loads in rivers and sediments discharge into oceans affect sediments and water quality in both freshwater and marine environments. Erosion in mining areas together with other mining related releases of GCs can have particularly severe impacts, contributing high concentrations of GCs to rivers (dissolved or as suspended sediment) and the sea where they can contaminate biota and enter the food chain.

Since groundwater is the most important component of global freshwater supply, estimated at 983 km^3/year (Margat and van der Gun, 2013), knowledge of the species and concentrations of GCs and factors influencing their spatial and temporal variation in groundwater are crucial for understanding the impacts on human exposure. Under natural conditions, aquifers are complex dynamic systems influenced by past climate variability, land erosion, land use changes and other changes, which impact groundwater flow and geochemical conditions. Even excluding climate change, it is likely that future availability of groundwater of sufficient quantity and quality will be severely impacted by increased water demand for agricultural, energy, industrial and domestic use (Brekke et al., 2009; Döll et al., 2012). Such changes will have implications for the security of food, water and energy supplies, and impact mitigation and adaptation pathways (World Bank, 2013; IPCC, 2014).

Most groundwater is used for irrigation (Fig. 5) and increasingly deeper groundwater resources are being exploited. Increased groundwater use requires greater consideration of the GCs contained within aquifers, their temporal changes and potential for release through irrigation into the food chain. For example, in Izmir, the third largest city of Turkey, increased pumping of groundwater to supply metropolitan drinking water has changed the natural groundwater flow and groundwater sourced from ever deeper aquifers was found to contain elevated heavy metal(loid) concentrations, which in the case of Ni and As exceed the drinking water permissible limit in 20% and 58% of the samples, respectively (Kavcar et al., 2009). Consequently, Izmir has installed a two-phase As removal plant to treat 178 ML/day (completed in 2010).

4.4.2. Climate change

Climate change is one of the greatest ecological, economical and social challenges facing civilization today. The Intergovernmental Panel on

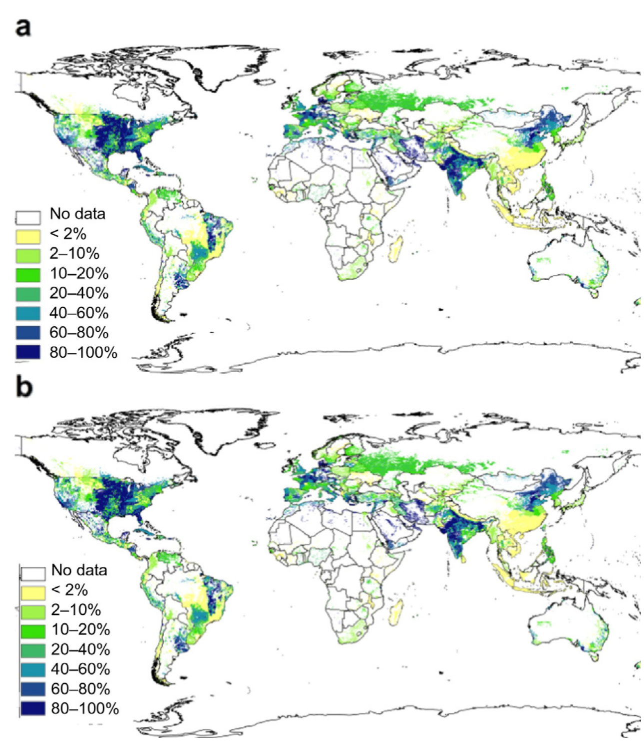

Fig. 5. (a) Area equipped for irrigation as a percentage of land area; (b) Percentage of irrigated area serviced by groundwater (Source: http://www.fao.org/nr/water/aquastat/irrigationmap/index.stm). For the majority of countries the base period of statistics is 2000–2008.

Climate Change (IPCC) (IPCC, 2013) estimates that if emissions continue to track at the top of IPCC scenarios, global temperature could rise by between 3.2 and 5.4 °C by the end of the century (relative to a 1850–1900 baseline). This increase will have implications for vegetation cover, particularly in arid and semi-arid regions. Increased temperatures and a decrease in rainfall will influence soil moisture and therefore reduce the amount of vegetation cover on the soil surface. The consequences are an increased risk of wind erosion and dust storm events (Stefanski and Sivakumar, 2009; Munson et al., 2011).

Climate variability and climate change can affect the quantity and quality of various components in the global hydrological cycle, including groundwater resources (Loaiciga et al., 1996; Sherif and Singh, 1999; Alley et al., 2002; Milly et al., 2005; Taylor et al., 2013; IPCC, 2014). However, the extent and direction of future climate change is very unpredictable and impacts on the availability of freshwater for humans and ecosystems may vary from one region to another. Changes in groundwater recharge affect groundwater availability in aquifers and modify biogeochemical conditions and the mobilization of GCs along the flow path of rainwater moving through the soil, the unsaturated zone and the aquifer. The impact of a changing climate on groundwater has been assessed (Holman, 2006; Taylor et al., 2013) and is quantifiable in terms of recharge and mineral quality, but there is still considerable uncertainty regarding components such as GCs and their mobilization. The (bio)geochemical processes that determine GC solubility and mobility are related to available geogenic sources and influenced by climatic conditions (evapotranspiration, precipitation and temperature). Thus, global climate change and climate variability are likely to interact with, and potentially exacerbate, factors that may contribute to increased concentrations of GCs in groundwater. However, other impacts such as land use changes, soil degradation, erosion and desertification are more likely to be the primary reasons for increasing health impacts of GCs from drinking water, food and the atmosphere.

4.4.3. Increasing exposure to atmospheric dust, aerosols and gases of geogenic origin

Increasing desertification, land degradation and frequent drought are challenges of a global dimension. Associated with these conditions, reduced vegetation cover leaves soils susceptible to wind erosion. Atmospheric dust has the potential to alter the fertility and edaphic characteristics of soils in areas downwind (Reynolds et al., 2001; McTainsh and Strong, 2007). Dust enriches surface soils with nutrients (Ca, Cu, Fe, K, Mg, Na, Mn, P and Mo), but soils are also a major sink for heavy metals released into the environment as a result of geogenic and anthropogenic processes (Akubugwo et al., 2013). Soils contaminated with heavy metals pose increased risks to humans and other organisms through indirect ingestion or contact, uptake through the food chain, decline in food quality and decrease in land usability for agricultural production. Mineral dust has also been suspected in the spread of meningococcal meningitis in sub-Saharan Africa during the dry season, a period when the region is affected by severe dust storms from the Sahara to the Gulf of Guinea (Cuevas et al., 2011; Pérez García-Pando et al., 2014). Also implicated in human health impacts are air pollutants, such as carbon monoxide (CO), sulfur dioxide (SO_2), nitrogen oxides (NOx), and volatile organic compounds (VOCs) and hydrochloric acid (Kampa and Castanas, 2008).

4.5. The need for improvements in analytical instrumentation and stronger legislation for GCs

Continuous improvements in both techniques and analytical instrumentation (as described in Section 4.1) and technologies for sampling, sample preservation and analytical instrumentation are required to facilitate monitoring of GC distribution. Critically, this should lead to improvement (i.e. decreases) in detection limits and allow more chemical species to be analyzed (e.g. redox species, radioisotopes). Despite stronger regulation, the absence of long-term studies, in particular studies which investigate exposure to low contaminant concentrations, often do not allow precise exposure limits to be established. As a result, in such cases the WHO only provides guideline values and individual governments may use these as a base for their legislation. Often guideline levels contain a precautionary margin. As exposure to GCs is often long-term and at low dose levels (and not just to one but multiple elements), more research into chronic health effects is needed; this should involve close cooperation between geologists, toxicologists and epidemiologists. Such new epidemiological studies of the health impacts of GC exposure will result in ever stronger and more reliable regulation and improved public health mitigation measures.

5. Conclusions and future directions: from here to where?

This work identifies multiple levels of evidence indicating the increasing significance of medical geology as a transdisciplinary research discipline. Much of this evidence is related to the growth of the global population and increasing living standards, which in turn result in increasing demand for water, food and energy demonstrating the close linkage of medical geology to most of the 17 Sustainable Development Goals in the 2030 Agenda for Sustainable Development. The integrated medical geology approach proposed in this paper crosscuts many of these Goals, incorporating the nexuses between GCs and water, energy, environment, food, climate and public health. Clearly, a better understanding of the risks of exposure of humans to GCs and the ecotoxicological effects of GCs (and combinations of GCs) in different environments is needed to facilitate both a reduction in these risks while, at the same time, meeting increasing demand for drinking and irrigation water, needed to ensure food security.

To minimize such threats and allow the co-existence of agriculture, mineral resource exploitation, environment and society requires an optimized integrated management of all these sectors, involving regulatory bodies and other state or federal entities. Decisions must be made based on strong scientific evidence and will result in trade-offs which must be acceptable to all sectors involved. The interdisciplinary research needed in the wide field of medical geology is fundamental to this principle.

Critical requirements for the development of this field include:

a) The development of a global transdisciplinary platform and forum in the area of medical geology to improve our understanding of the occurrence of GCs in rocks, their release into water and soils, their geochemical, microbiological and other mobility controls, and factors affecting their transport into the biosphere and to humans in order to better manage these environments to reduce human GCs exposure to human population.
b) Interdisciplinary information exchange to stimulate scientific research in the fields of medical geology involving groundwater, surface water, soil, plant related sciences and social and health sciences to achieve the United Nations Sustainable Development Goals (SDGs) for sustainable development and to provide the population safe drinking water, food and air; for healthy living.
c) Information on the bioavailability and bioaccessibility of GCs from water, food and soil to different organisms and their subsequent metabolism in plants, fish, animals and humans to better understand the transfer of GCs to humans through the food chain;
d) Identification of effective and sustainable monitoring, regulation and management options of human exposure to and health effects of GCs, considering simultaneous exposure to multiple elements;
e) Improvements in human access to water and food in adequate quantity and good quality, considering economic and social limitations and benefits;
f) Training to improve and broaden our understanding of the relationship between GCs and health outcomes and how factors affecting this relationship might be mitigated;
g) Continual improvement in analytical instrumentation and

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techniques that allow for GC speciation and quantification at trace levels; and

h) Consideration of the potential role of geogenic contaminants (GCs) in diseases with unknown etiology.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2016.11.208.

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