



Mapping human health risks from exposure to trace metal contamination of drinking water sources in Pakistan

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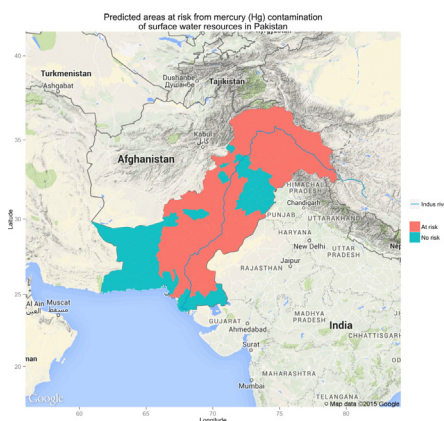
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HIGHLIGHTS

- Predictions of trace metal concentration use geographically weighted regression
- Human health risk mapping on the predicted levels of trace metals
- Drinking water was predicted to be at risk from studied trace metals
- 53% of the total area of Pakistan found to be contaminated with trace metals

GRAPHICAL ABSTRACT



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ABSTRACT

The consumption of contaminated drinking water is one of the major causes of mortality and many severe diseases in developing countries. The principal drinking water sources in Pakistan, i.e. ground and surface water, are subject to geogenic and anthropogenic trace metal contamination. However, water quality monitoring activities have been limited to a few administrative areas and a nationwide human health risk assessment from trace metal exposure is lacking. Using geographically weighted regression (GWR) and eight relevant spatial predictors, we calculated nationwide human health risk maps by predicting the concentration of 10 trace metals in the drinking water sources of Pakistan and comparing them to guideline values. GWR incorporated local variations of trace metal concentrations into prediction models and hence mitigated effects of large distances between sampled districts due to data scarcity. Predicted concentrations mostly exhibited high accuracy and low uncertainty, and were in good agreement with observed concentrations. Concentrations for Central Pakistan were predicted with higher accuracy than for the North and South. A maximum 150–200 fold exceedance of guideline values was observed for predicted cadmium concentrations in ground water and arsenic concentrations in surface water. In more than 53% (4 and 100% for the lower and upper boundaries of 95% confidence interval (CI)) of the total area of Pakistan, the drinking water was predicted to be at risk of contamination from arsenic, chromium, iron, nickel

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and lead. The area with elevated risks is inhabited by more than 74 million (8 and 172 million for the lower and upper boundaries of 95% CI) people. Although these predictions require further validation by field monitoring, the results can inform disease mitigation and water resources management regarding potential hot spots.

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1. Introduction

Inland ground and surface water are globally important drinking water sources, which are influenced by natural and anthropogenic processes. These can result in elevated levels of different contaminants in drinking water (Winkel et al., 2008; Shah et al., 2012). Several pathogens as well as organic and inorganic components occur in drinking water sources of many regions of the world and may have acute and chronic effects on consumers' health (Srinivasa and Govil, 2007; US-EPA, IRIS, 2014). For example, one-third of annual mortality in Pakistan has been attributed to drinking water contaminated by microbial and/or chemical components (Azizullah et al., 2011).

Trace metals represent a major group of contaminants of drinking water sources that can have severe implications for human health, e.g., cardiovascular and skeletal diseases, infertility and neurotoxicity (World Health Organization, 2011). In developing countries, contamination of drinking water sources by trace metals have been triggered by rapid industrialization and excessive usage of pesticides and chemical fertilizers in agriculture during the last decades (Srinivasa and Govil, 2007; Farooqi et al., 2008; Eqani et al., 2012). In addition, geogenic sources contribute to the wide occurrence of trace metals such as arsenic in ground and surface water (Winkel et al., 2008). Given inadequate water purification and remediation measures, trace-metal-contaminated water is regularly consumed by the population of developing countries, especially in rural areas (Ullah et al., 2009; Khan et al., 2012).

Risk assessments for trace metals in drinking water sources are crucial to estimate the total population at risk, to identify hot spots and to develop management strategies to reduce the anthropogenic input and to remediate contaminated areas (Srinivasa and Govil, 2007). However, in developing countries such as Pakistan, limited technical expertise, inadequate laboratory facilities and resource constraints often limit water quality monitoring activities to a few locations and/or administrative areas (Azizullah et al., 2011). Moreover, the monitoring often exclude rural and remote areas, where drinking water contamination may be more severe (Khan et al., 2012). Consequently, regional scale (i.e. nationwide) risk assessments are often not available and information on the extent of trace contamination and the total population at risk is largely unknown (Törnqvist et al., 2011).

Numerous spatial prediction techniques supported by geographic information systems (GIS), i.e. spatial interpolation and spatial regression models, allow for the prediction of trace metal concentration in water at unsampled locations based on the values from sampled locations (Javi et al., 2014; Pebesma and de Kwaadsteniet, 1997; Nas and Berkay, 2010). The origin and transport of trace metals through water mainly depends on the speciation form of metals, and the physical and chemical processes within the aquatic environment and sediments therein (Huang et al., 2015; Winkel et al., 2008). As these physical and chemical processes are highly influenced by the soil properties, land use characteristics, and different climate and environmental variables, the transport and distribution of trace metals usually show spatial continuity and a close association with these variables (Amini et al., 2008; Winkel et al., 2008; Rodríguez-Lado et al., 2013). Therefore, these variables may serve as covariates in the spatial prediction of trace metals at unsampled locations (Rodríguez-Lado et al., 2013).

We present the first nationwide human health risk maps (approximated) for Pakistan from exposure to 10 trace metals in surface and groundwater sources. Trace metal concentration data was compiled

from previously published studies. Concentrations of trace metals at unsampled locations were predicted using geographically weighted regression (GWR) models with soil properties, land cover and elevation as covariates (spatial predictors) (Harris et al., 2010). Thereafter, risk quotients (RQ) were computed by comparing exposure concentrations with the World Health Organization (WHO) guideline values to identify the fraction of area at risk and total inhabitants in risky areas. We discuss the relevance of these risk maps for water resources management in Pakistan.

2. Materials and methods

2.1. Study area

Pakistan is situated in South Asia within the coastal belt of the Arabian Sea and consists of 137 administrative districts (second order administrative division) with a total area of 796,095 km² (Fig. 1a and b). Approximately one-third of the country is covered by desert, whereas the rest is mostly covered by grassland and agricultural land (Fig. 1c). The geography is characterized by the flat-lying Indus Plain in the East, the mountains of the Himalayas, Karakoram and Hindukush in the North and the upland Baluchistan plateau in the West (Fig. 1d). The climate of this region is mostly arid to semi-arid and exceptionally temperate in the Northwest (Farooqi et al., 2008). The Indus river delta and its tributaries feed the inland surface and groundwater system of the region, which is the major source of drinking water for 172.3 million people (Fig. 1d; Pakistan Bureau of Statistics, 2010).

2.2. Data compilation and processing

We compiled data on the concentrations of 10 trace metals, i.e. arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn) in ground and surface water of Pakistan, measured in multiple sites within 26 districts during 1991–2014, from previously published studies (Tables S1; S2). The individual studies provided concentration values that were typically corrected for sampling and processing errors (see cited studies in Tables S1 and S2). In 82% of the studies, concentration data were reported as summary statistics (e.g. mean, max and standard deviation, see Tables S1 and S2 for details on the number of ground and surface water samples in each district used to compute the district mean) for the districts. The trace metal concentrations of samples from each district exhibited a low variation (all coefficient of variation (CV) ≤ 20% and relative homogeneity of CV across districts) and no outlier value was detected in the data (cited studies in Tables S1 and S2). Hence, given that in 96% of the studies sampling sites were not georeferenced, we regarded the mean as a sufficiently robust parameter and assigned district mean values to the georeferenced districts as the representative trace metal concentrations in the surface and ground water (country-level summary statistics are presented in Table S3). In 5% and 32% of districts for ground and surface water, respectively, we had mean concentrations from two years for a few trace metals (Tables S1; S2). In these cases, we used the latest concentrations to avoid heterogeneity in the uncertainty of predicted concentrations across trace metals and districts, and thus in risk assessment. The district level aggregation of trace metal concentrations may result in uncertainties regarding variability within the

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