**Changes in global groundwater organic carbon driven by climate change and urbanization**

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**Summary**

Rapid urban growth is increasing demand for potable groundwater. Dissolved organic carbon (DOC) is a major control of groundwater potability and treatment costs. We hypothesize that climate change and urbanization will not only decrease groundwater availability, but also its quality. Here, we summarize global groundwater DOC data (n = 6781) to identify its drivers and predict future changes. Our analysis of 2916 data revealed climate, dissolved inorganic ions and land use explain 31% of DOC variability, whilst aquifer age accounted for an additional 16%. Urban groundwaters were ~19% more enriched in DOC than natural or agricultural lands. We identify hotspots in the United States associated with a groundwater DOC increase of 45% by 2050, which could increase household water costs in some key regions such as Nevada, Georgia and South Carolina. Climate change will make groundwater increasingly difficult and expensive to treat, requiring more efficient and cost-effective methods for groundwater treatment in the future.

**Abstract**

The impact of climate change, urbanisation and changing water chemistry on groundwater dissolved organic carbon (DOC) concentrations remains unquantified. Here, we present an an analysis of approximately 3000 observations to understand the drivers of groundwater DOC concentrations. We show that groundwater chemistry, local climate and land use explain ~ 31% of observed variability in groundwater DOC, whilst aquifer age explains an additional 16%. We identify a 19% increase in DOC associated with urban land. Our results show that up to 1/3rd of the states of Nevada, Georgia and South Carolina are likely to experience major groundwater DOC increases because of changed precipitation and temperature, with increases of up to 45% by 2050 in some areas. For the first time, our results quantify predicted changes in groundwater DOC concentrations and show that in certain locations, future urbanisation and climate change will have significant impacts on groundwater quality and water treatment costs.

**Main**

Groundwater is the largest source of global fresh water. The potability of groundwater is highly dependent upon the concentration of dissolved organic carbon (DOC) due to its ability to alter water chemistry and microbial abundances (Romera-Castillo et al. 2018, Judd et al. 2006). High groundwater DOC concentrations can cause an increase in microbial activity which requires disinfectants such as chlorine to make the water safe to drink. These disinfectants however can react with DOC, resulting in disinfectant by-products (DBPs) such as Trihalomethanes (THMs) which are harmful to human health (Barrett et al. 2000). High DOC concentrations can also increase the mobility of other contaminants in groundwater, by complex association with dissolved or colloidal organic matter (OM), including heavy metals (Dudley et al. 1986, Pohlman and McColl 1988, Driscoll et al. 1995).

Climate variables such as temperature and precipitation impact on net primary production and microbial activity in ecosystems (Nemani et al. 2003, Davidson and Janssens 2006). This consequently drives the availability of vegetation and it’s decomposition to DOC (Jenkinson et al. 1991, Davidson and Janssens 2006). The impact of changed precipitation rates and patterns, and increasing temperatures and evaporation rates under future climate change scenarios are expected to alter biomass, reduce surface water quantity due to decreased precipitation and increased evaporation (Lipczynska-Kochany 2018), and subsequently increase domestic and agricultural reliance on groundwater resources. Increasing reliance on groundwater due to climate change impacts will be compounded by increases in urbanisation and global population. Understanding the impact of climate change on the quality of our freshwater resources are therefore essential, however this research area remains understudied (Lipczynska-Kochany 2018).

Here, we hypothesize that climate change and urbanisation will change not only groundwater quantity, but also the concentration of groundwater DOC and ultimately groundwater quality. We present the largest global dataset of 6,781 published and unpublished groundwater DOC concentrations (Table S1) obtained from aquifers in 15 countries and 4 continents (Figure 1). We provide an analysis of global groundwater DOC concentrations and quantify its key drivers. Specifically, we predict changes in DOC concentrations due to projected changes in temperature and precipitation, as well as increases caused by future urban area expansion.

**Global groundwater DOC distributions**

Groundwater DOC concentrations vary spatially and are usually lower than surface water concentrations. The global average, median and standard deviation of groundwater DOC concentrations is 2.7 mg C / L, 1.0 mg C / L and 15.8 mg C / L respectively. There is a significant difference between the median groundwater DOC concentrations between samples collected in North America, Oceania, Asia and those from European countries (p = 0.000) at the 95% confidence level, with Asian concentrations significantly higher than Europe, North America and Oceania. In Europe, DOC concentrations are significantly higher than Oceania and North America, whilst DOC concentrations in North America are significantly lower than in Oceania.

To determine the drivers of global DOC concentrations in groundwater, we generated a linear mixed model (Table S2) for a large dataset (n = 2196) collected by the National Water Quality Assessment (NAWQA) program of the U.S. Geological Survey (USGS). This dataset was selected as it contained supplementary data including chemical parameters unavailable for other samples. This allowed us to extract supplementary climatic data (Hijmans et al 2005), water table depth (Fan et al 2013) and land use data (Friedl et al 2010) for analysis in the model.

Figure 1: Median groundwater DOC concentrations (mg C / L). Boxes represent the interquartile range, containing median, with whiskers representing the upper and lower 25% of data. The red circles represent outliers. Note: outliers above 20 mg C / L are not included in the graph for visual clarity (n outliers < 20 mg C/L = 30 [Australia], 14 [U.S.], 2 [Denmark and Portugal respectively] and 1 [U.K.]).

Overall, the model explained 47.7% of the variation in DOC concentrations, with 31.3% explained by the fixed factors alone (all factors excluding aquifer age), and 16.3% explained by the random factor aquifer age. Our analysis (Table S2) shows positive relationships between DOC and temperature in the wettest quarter of the year (p < 2e-16), groundwater temperature (p < 2e-16), and dissolved calcium (Ca) (p < 2e-16 ), potassium (K) (p = 1.69e-13) and iron (Fe) (p < 2e-16). There was also a weaker relationship between DOC and manganese (Mn) (p < 0.039). We also found negative relationships between DOC and temperature in the warmest quarter of the year (p < 2e-16), precipitation in the driest month of the year (p = 0.001), silica (Si) (p = 1.97e-06), pH (p = 4.06e-05), sample depth below land surface (p < 2e-16), land elevation (p = 1.34e-06) and dissolved oxygen (DO) (p < 2e-16). Our analysis also shows negative relationships (p < 0.01) between DOC and sodium (Na) (p = 0.001), and DOC and precipitation in the wettest month of the year (p = 0.001). Water table depth as a variable improved the overall model fit, however it is not a significant predictor of DOC concentrations (p = 0.071).

The results suggest that groundwater DOC concentrations are determined by the interaction of four major controlling factors: (1) climatic controls affecting surface vegetation production and decomposition, (2) land-use, (3) redox controls affecting DOC biodegradation rates, and (4) aquifer age and sedimentary OM as a source of DOC within the aquifer (Figure 2).



Figure 2: Factors and processes leading to decreasing (A) or increasing (B) groundwater DOC concentrations in the surface and vadose zones including climatic controls affecting surface production and availability of OM, land use type, redox and climatic controls affecting biodegradation and sorption of OM in the subsurface and, the influence of aquifer age and sedimentary OM as a source of DOC within the aquifer.

**Climate controls**

Temperature and precipitation play an important role in predicting groundwater DOC concentrations. DOC decreases by 9.5% for every 10 mm increase in precipitation in the driest month of the year and decreases by 2.5% for every 10 mm increase in precipitation in the wettest month of the year. This is likely due to a dilution effect whereby accumulated soil DOM infiltrates the saturated zone during initial rainfall and is later diluted by additional rainfall (Smith et al 2014). In arid climates, some of these trends may be reversed. For example in an extremely dry climate, an increase in precipitation would increase groundwater DOC concentration since the precipitation in the wettest month of the year is likely to be too low to cause significant dilution. Furthermore, in arid climates, DOC concentrations would decrease due to limited vegetation cover and bioavailable DOM (Chapelle et al 2013). We observed some of these trend reversals in simple linear analyses for a smaller Australian dataset (n = 79 after removing incomplete data).

Groundwater DOC concentrations increase by 3.4% for every 1 °C increase in average temperatures in the wettest quarter of the year and 4.6% for every 1 °C increase in sample groundwater temperature. In contrast, groundwater DOC concentrations decrease by 8.9% for every 1 °C increase in temperatures in the warmest quarter of the year. The source of DOC is dependent upon availability of water. In non-arid climates, increases in mean surface temperature in the wettest quarter of the year and increased groundwater temperatures are likely to cause increased temperatures in the soil zone, which under conditions of high soil moisture, are likely to stimulate biological activity and DOC breakdown (Grieve 1990).

**Water quality**

In the saturated zone, redox conditions and pH are strongly related to DOC concentration, with DOC concentrations 9.2% lower for each unit increase in pH, and 6.8% lower with every 1 mg / L increase in DO. The smaller Australian dataset (n = 79) also was consistent with the larger US dataset (n = 2916) (see supplementary material). The mineralisation of DOC via biodegradation produces CO2 which causes a decrease in pH levels in high DOC environments. Reduced dissolved species of Fe(II) and Mn(II) are associated with a DOC increase of 6.9% per 1 mg / L increase in Fe concentrations and 0.4% increase in DOC for every 100 µg / L increase in Mn concentrations. This trend can be explained by microbial community use of Mn- and Fe-oxides as alternative electron acceptors to DO in the presence of DOC under anoxic conditions (Apello and Postma 2005). Aerobic microbes metabolise carbon at a faster rate than other microbes, therefore a lack of DO limits DOC biodegradation rates. Fe can accumulate within oxic sediment layers due to oxidation and precipitation of dissolved Fe in young sediments (Schwertmann 1966). This Fe can become coated with OM and re-dissolve under reduced conditions releasing the OM.

**Aquifer age and groundwater evolution**

The age of the geological formation, or aquifer age, explained 16.3% of variability in groundwater DOC. Groundwater in younger aquifers of Cenozoic sediments contained 41% higher DOC concentrations than older Mesozoic and Paleozoic Era aquifers which support previous observations in smaller datasets (Jakobsen and Postma, 1994). Sedimentary OM in young aquifers is more likely to be accessible to microbial degradation than in older, lithified aquifers.

We also observed a decrease in groundwater DOC concentrations of 7.7% for every 10 m increase in sample depth. Increases in sample depth are correlated with longer groundwater residence times (Weissmann et al 2002). This is implied by a positive relationship between the weathering product Si and sample depth (p < 1.97e-06). We found a negative correlation between groundwater DOC and Si, with DOC decreasing by 6.3% for every 10 mg / L increase in Si. The main source of dissolved Si is silicate mineral dissolution (Appelo and Postma 2005). The negative relationship between DOC and Si is explained by the accumulation of water-rock interaction in older groundwaters (Peters et al 2014). These older groundwaters are more likely to be depleted in DOC due oxidation processes and adsorption to aquifer mineral surfaces.

**Land use**

There is a significant increase of 19% in groundwater DOC concentrations in urban areas compared to natural land. This may be explained largely by increased potential for DOC contamination in urban areas due to leaking sewage systems, landfill leaching, animal waste, fertiliser run-off and industrial and residential waste washed into stormwater drains (Pitt et al. 1999, Foley et al. 2005, Lapworth et al. 2008, Utton 1982). Therefore, our global dataset supports earlier local scale observations of correlations between urban land use and DOC in surface waters (Johnson et al 1997, Chang 2008, Meierdiercks et al. 2017) and shows that the impact of urban land use also extends to groundwater systems on a broader scale. However, there is no significant difference (p = 0.841) between groundwater DOC concentrations between areas of natural land use and areas of agricultural land use.

**Implications**

Continental-scale changes to groundwater DOC concentrations are caused by changing temperature and precipitation patterns. Using our results and IPPC5 (CMIP5) climate projections (www.worldclim.org), we identify more extreme groundwater DOC concentration changes associated with changing temperatures modulated by changing precipitation rates and patterns (Figure 3). A large portion of the U.S such as New York, Indiana, Illinois, Tennessee and Mississippi will experience a decrease in groundwater DOC concentrations due to increased temperatures in the warmest quarter resulting in aridity. In contrast, we also identify hotspots of high groundwater DOC concentration (increases of up to 45%) largely associated with increased temperatures in the wettest quarter of the year for up to 1/3rd of a number of south eastern U.S states including Louisiana, Alabama, Georgia and South Carolina, as well as central-western states including Nevada, Utah, Colorado, and Wyoming under the “business-as-usual” Intergovernmental Panel on Climate Change (IPCC) climate change prediction scenario RCP85 (figure 3). Higher increases in temperatures in the wettest quarter compared to the warmest quarter are also predicted in areas such as the east coast of Madagascar, south-east China, central Chile, north-eastern Peru and along the east coast of Venezuela (Worldclim v1.4; Hijmans et al. 2005). We predict that these areas may also be potential hotspots for groundwater DOC increases associated with climate change in the future.

The areas most at risk of future groundwater DOC concentration increases are those facing increased DOC due to climate change and where urbanisation is also predicted to increase. This will be a particularly significant issue for impacted areas which have a large or increasing reliance on groundwater as a source of fresh water. For example areas in the U.S predicted to be impacted by DOC increases associated with climate have between 13% to 87% reliance on groundwater as their source of fresh water (Figure 3). Some of those regions such as Atlanta, Georgia are within the top 10 U.S locations projected to have the largest increases in urbanisation by 2020 (Bounoua et al. 2018), which will further increase groundwater DOC concentrations. On a global scale, 54% of the worlds’ population live in urban areas. By 2050 the world’s urban population will increase to 66% (UN, 2014) due to both the reclassification of rural land to urban land, as well as an increase in migration. Seto et al (2012) predict a high probability (> 75%) of urban extent exceeding 185% that of circa 2000 levels by 2030, which equates to an increase in urban area of 1.2 million km2. Areas most likely to see urban expansion and population growth including eastern China and India. These areas are already facing high urban growth and severe groundwater contamination issues (Roderiguez-Lado et al. 2013, Sahu & Saha 2015). Groundwater quality issues in south-eastern China may be further compounded by groundwater DOC increases associated with large predicted increases in temperature (up to 10 °C) in the wettest quarter of the year (Worldclim v1.4; Hijmans et al. 2005).



Figure 3: Changes in groundwater DOC concentrations by year 2050 due to temperature and precipitation changes. Panel A shows the two areas in the U.S predicted to experience the largest increases in DOC concentration due to temperature and precipitation changes by 2050. Panels B and C show changes in DOC concentrations in these areas caused by temperature variables (temperature in the wettest and warmest quarters of the year) and precipitation variables (precipitation in the wettest and driest months of the year) alone. Groundwater DOC concentrations changes are calculated using model results and IPPC5 (CMIP5) climate projections from the end of the 20th century (average of values from the period 1960 – 1990) to year 2050 (average of predicted values for the period 2041 – 2060) (Hijmans et al. 2005) for a “business-as-usual” climate change scenario (RCP8.5) as outlined in IPCC (2014). Lowermost map shows U.S. state reliance on groundwater as for total water use (Dieter et al 2018) overlain with areas predicted to experience increases in groundwater DOC concentrations due to climate change variables and urbanisation.

Overall, our investigation reveals that imminent climate change and urbanization will impact on groundwater quality and quantity. These impacts on groundwater DOC will not be evenly distributed. Increases in temperatures in the warmest quarter of the year will decrease groundwater DOC concentrations due to aridity, whilst increased temperatures in the wettest quarter of the year will increase groundwater DOC concentrations due to the stimulation of microbial activity. We identify hotspots of high groundwater DOC concentrations, particularly in areas that will undergo future urbanisation and population growth. This will substantially increase the treatment costs to remove high concentrations of DOC from groundwater in many locations, including parts of the U.S (Nevada, Georgia and South Carolina) and south-eastern China.

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