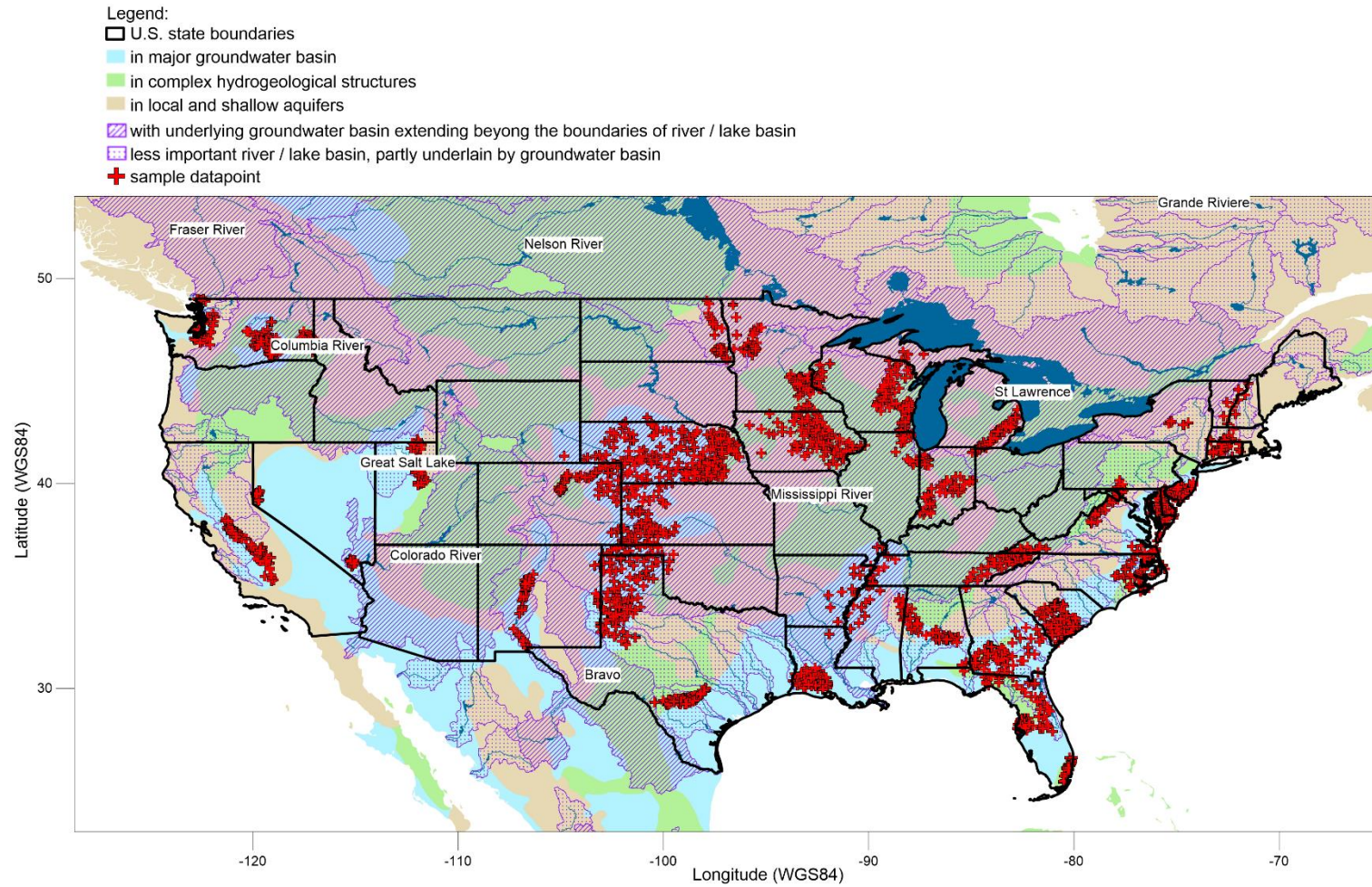


Supplementary Information for:  
Changes in global groundwater organic carbon driven by climate change and urbanization  
by  
McDonough et al.

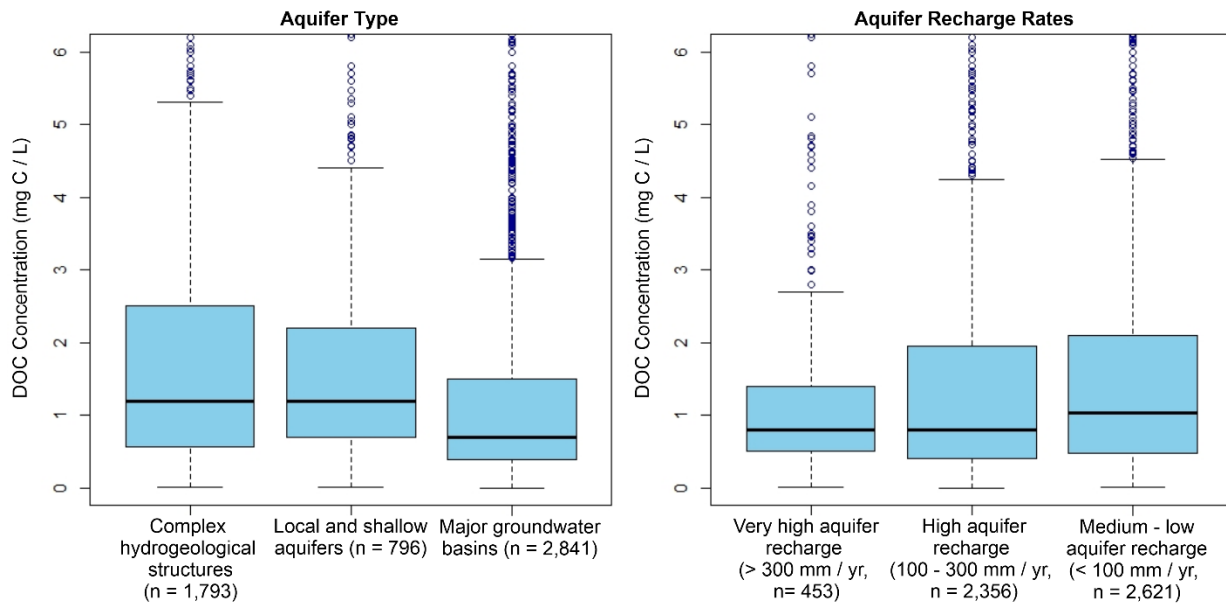
Correspondence to: [lizam@ansto.gov.au](mailto:lizam@ansto.gov.au)

## Supplementary Figures



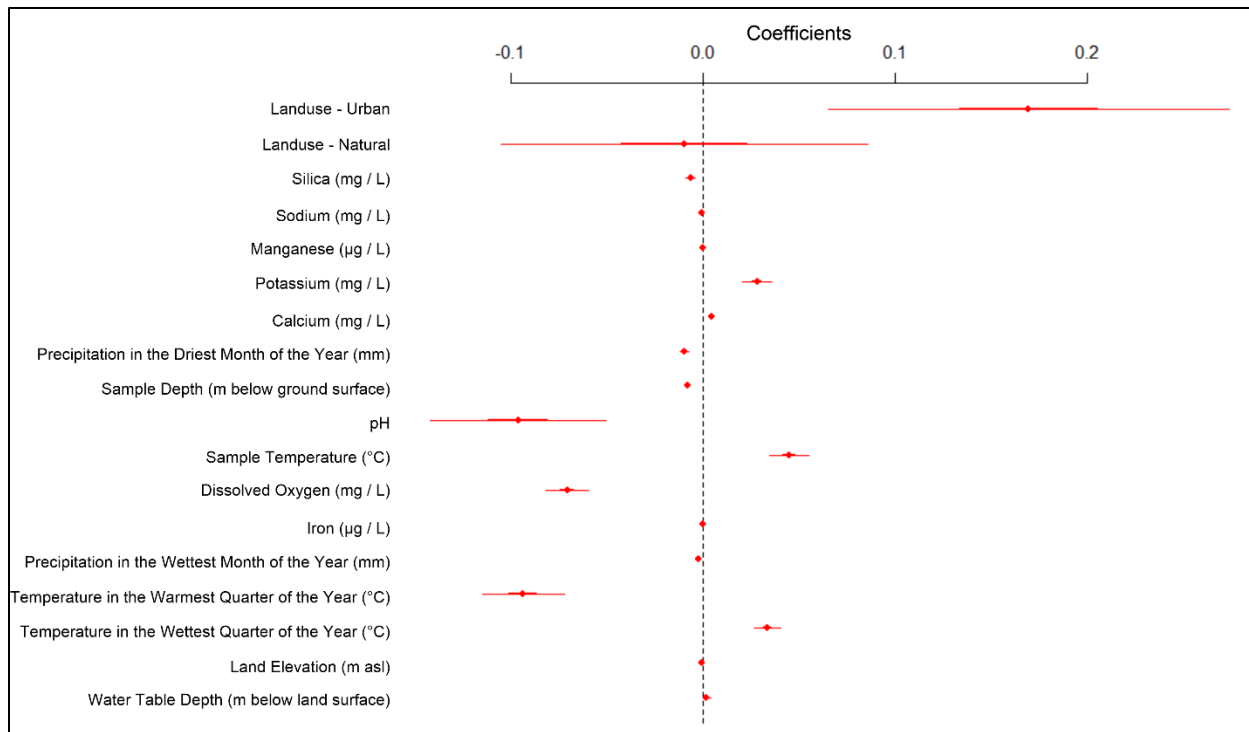
### Supplementary Figure 1

US NWQMC data (red plus symbols) <sup>1</sup> overlain over global aquifer WHYMAP data <sup>2</sup>.



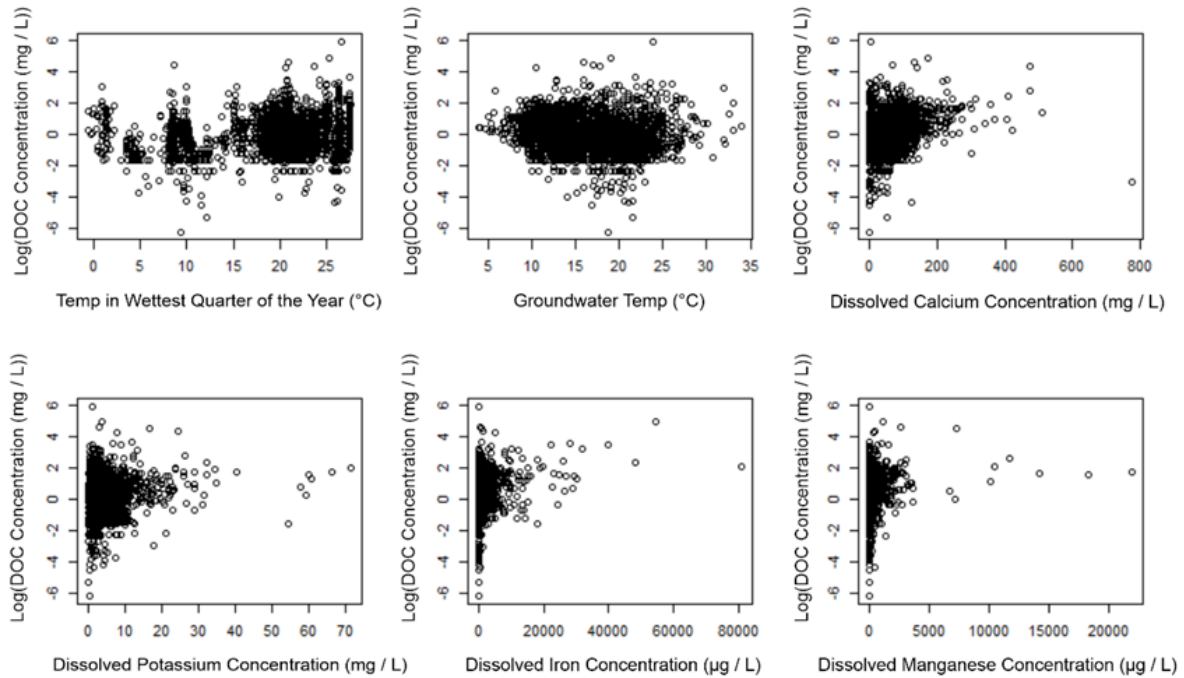
### Supplementary Figure 2

Comparison of groundwater DOC concentrations between aquifer type and recharge rates for US NWQMC data using global aquifer WHYMAP data <sup>2</sup>. Boxes represent the interquartile range, containing median, with whiskers representing the 1.5 times the interquartile range of data. Datapoints beyond this are shown as outliers (dark blue circles). Median DOC concentrations in major groundwater basins were significantly lower than in complex hydrogeological structures or local and shallow aquifers (both  $p < 2.2 \times 10^{-16}$ ). Groundwater DOC concentrations in aquifers with medium – low recharge rates ( $< 100 \text{ mm year}^{-1}$ ) were significantly higher than in aquifers with  $100 - 300 \text{ mm year}^{-1}$  and  $> 300 \text{ mm year}^{-1}$  ( $p = 2.342 \times 10^{-7}$  and  $4.857 \times 10^{-5}$  respectively). Outliers greater than  $6 \text{ mg L}^{-1}$  have been removed for clarity ( $n = 154$  [complex hydrogeological structures],  $n = 26$  [local and shallow aquifers],  $n = 107$  [major groundwater basins],  $n = 15$  [very high aquifer recharge ( $> 300 \text{ mm year}^{-1}$ )],  $n = 153$  [high aquifer recharge ( $100\text{-}300 \text{ mm year}^{-1}$ )] and  $n = 119$  [medium – low aquifer recharge ( $< 100 \text{ mm year}^{-1}$ )]).



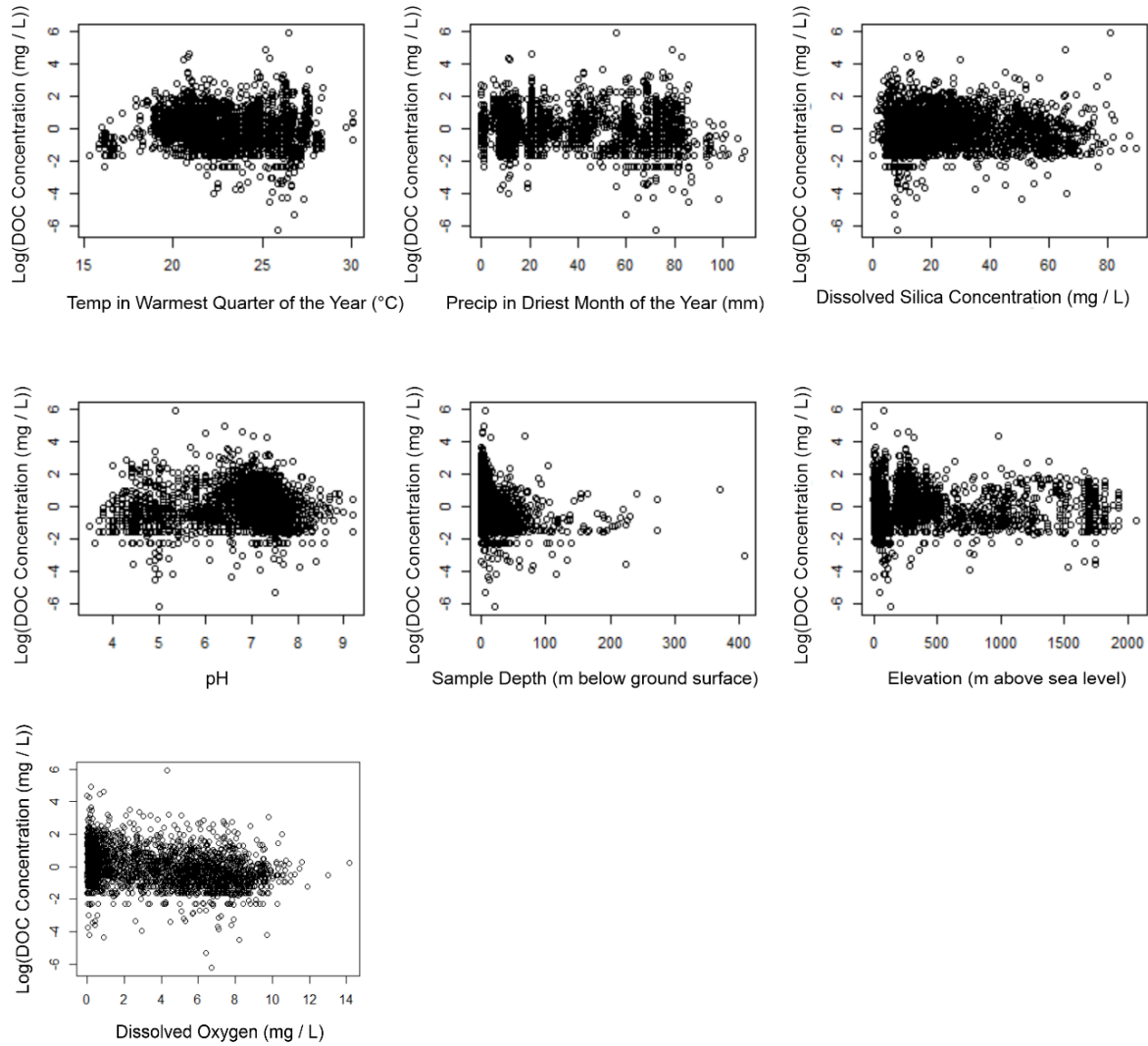
### Supplementary Figure 3

Regression estimates of the effects of model variables on groundwater DOC concentrations. Centre points represent mean regression estimates with inner (thicker) bars representing 50% confidence intervals and outer (thinner) bars representing 95% confidence intervals. Regression estimates from top to bottom are 0.17,  $-9.74 \times 10^{-3}$ ,  $-6.29 \times 10^{-3}$ ,  $-5.66 \times 10^{-4}$ ,  $3.75 \times 10^{-5}$ ,  $2.83 \times 10^{-2}$ ,  $4.45 \times 10^{-3}$ ,  $-9.53 \times 10^{-3}$ ,  $-7.77 \times 10^{-3}$ ,  $-9.61 \times 10^{-2}$ ,  $4.49 \times 10^{-2}$ ,  $-7.06 \times 10^{-2}$ ,  $6.87 \times 10^{-5}$ ,  $-2.45 \times 10^{-3}$ ,  $-9.33 \times 10^{-2}$ ,  $3.35 \times 10^{-2}$ ,  $-2.66 \times 10^{-4}$  and  $1.84 \times 10^{-3}$  (also listed in Supplementary Table 2).



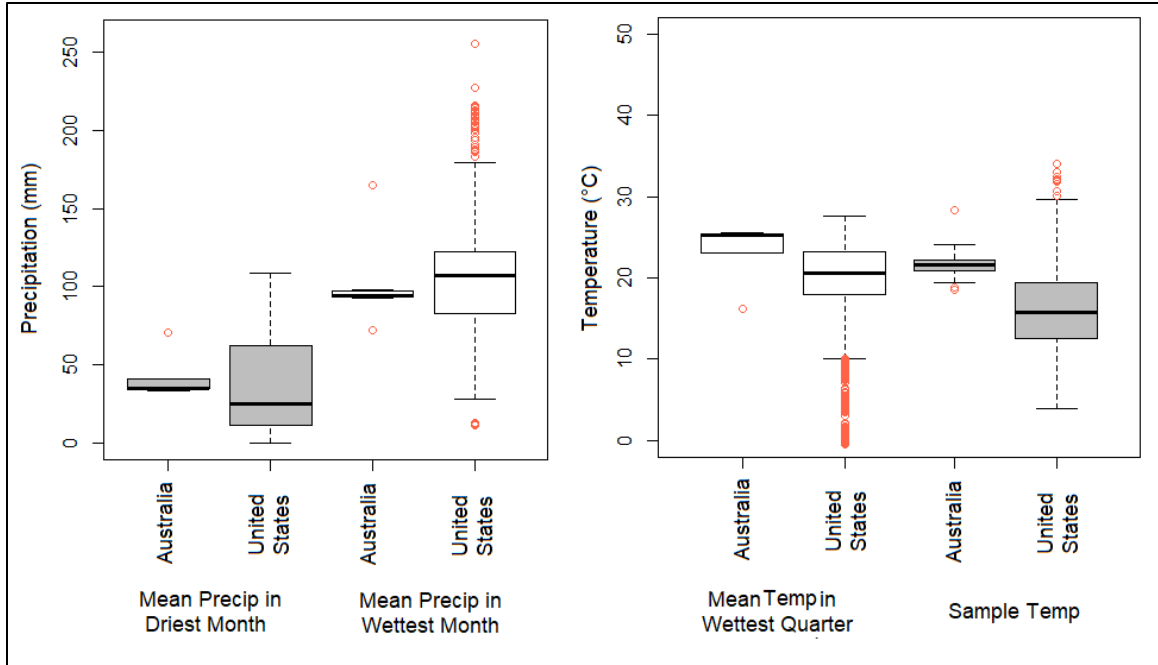
#### Supplementary Figure 4

Simple scatterplots of model variables with positive correlations ( $p < 0.05$ ) with  $\log(\text{DOC concentration (mg / L)})$ . NB: these plots show only the correlation between  $\log(\text{DOC concentration (mg / L)})$  and individual variables. They do not account for the other variables included in the model and do not represent model results.



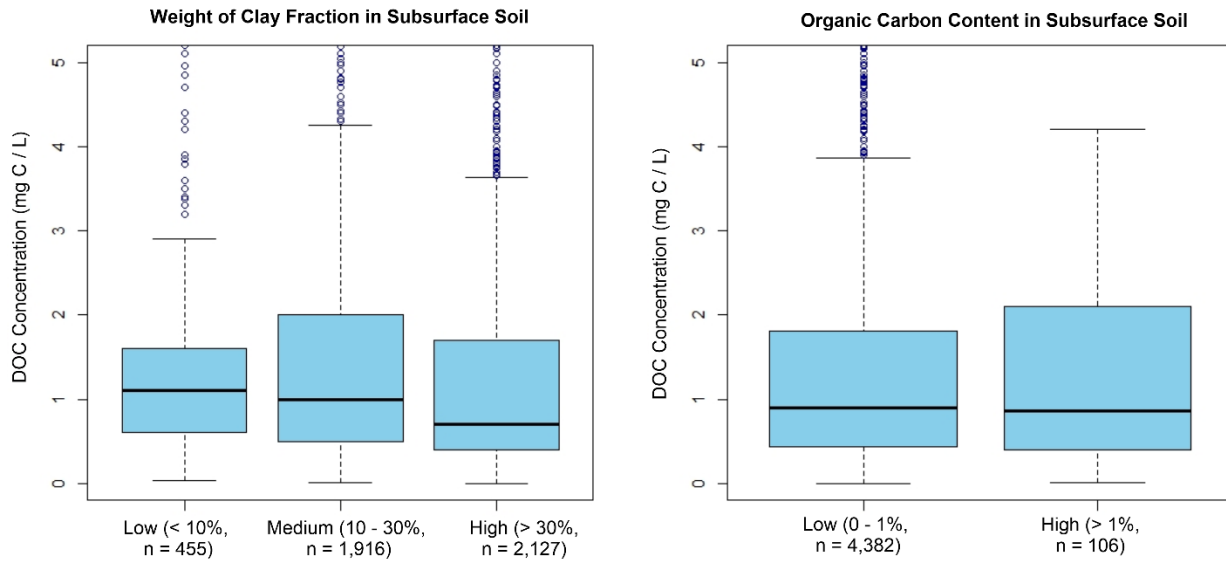
### Supplementary Figure 5

Simple scatterplots of model variables with negative correlations ( $p < 0.05$ ) with  $\log(\text{DOC concentration (mg / L)})$ . NB: these plots show only the correlation between  $\log(\text{DOC concentration (mg / L)})$  and individual variables. They do not account for the other variables included in the model and do not represent model results.



### Supplementary Figure 6

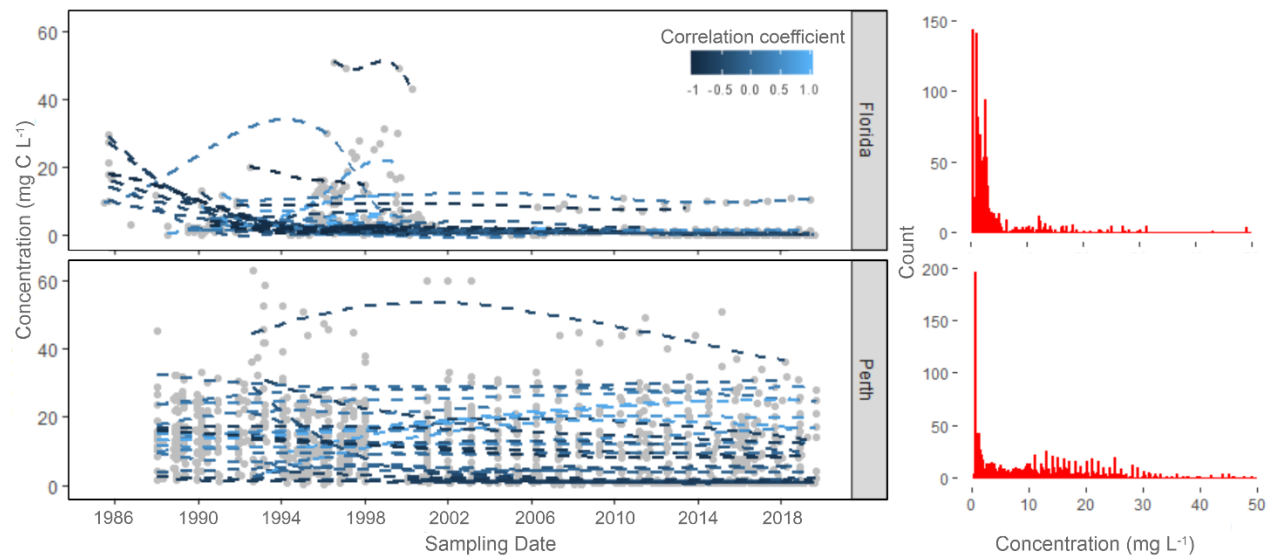
Boxplots comparing variables which show opposing correlations in the Australian and US datasets. Boxes represent the interquartile range, containing median, with whiskers representing the 1.5 times the interquartile range of data. Datapoints beyond this are shown as outliers (red circles).



### Supplementary Figure 7

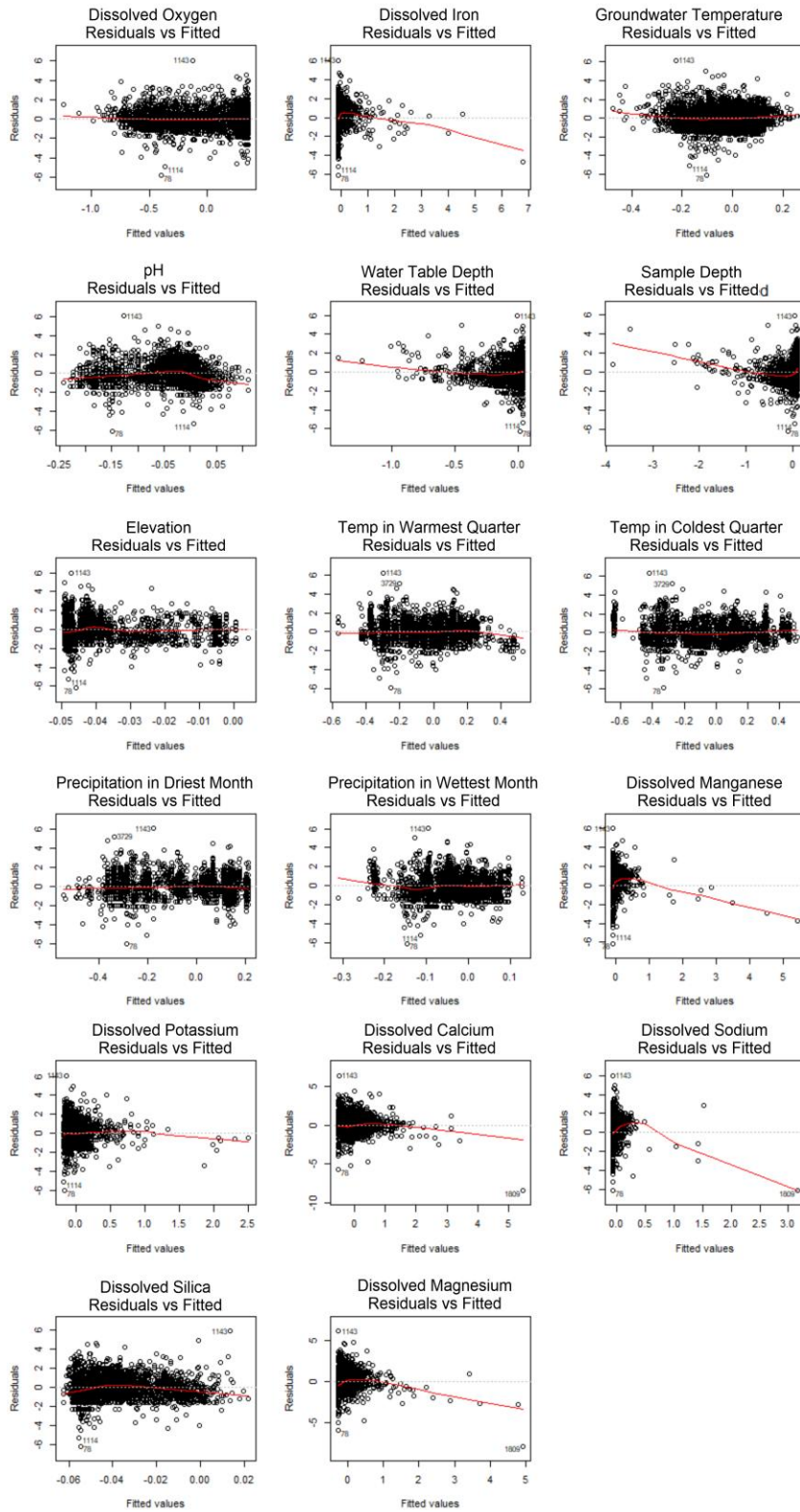
Comparison of groundwater DOC concentrations with varying subsurface percent weight of clay and organic matter content using Regrided Harmonized World Soil Database v1.2<sup>3</sup>. Boxes represent the interquartile range, containing median, with whiskers representing the 1.5 times the interquartile range of data. Datapoints beyond this are shown as outliers (dark blue circles). Median DOC concentrations in soils with high clay percent weight (> 30%) are significantly lower than areas where the subsoil clay percent weight is medium (10 – 30%,  $p = 5.628 \times 10^{-13}$ ) or low (< 10%,  $p = 5.533 \times 10^{-8}$ ). Groundwater DOC concentrations in aquifers with low (0 – 1%) and high (> 1%) soil organic carbon content are not significantly different ( $p = 0.4723$ ). Outliers greater than 5 mg / L have been removed for clarity ( $n = 25$  [low (< 10%) clay fraction in subsoil],  $n = 108$  [medium (10 – 30%) clay fraction in subsoil],  $n = 124$  [high (> 30%) clay fraction in subsoil],  $n = 253$  [low (0 – 1%) organic carbon content in subsoil], and  $n = 4$  [high (> 1%) organic carbon content in subsoil]). N.B. this data represents agricultural and natural areas only due to the potential for paved urban areas to affect infiltration of DOC through the subsoil.





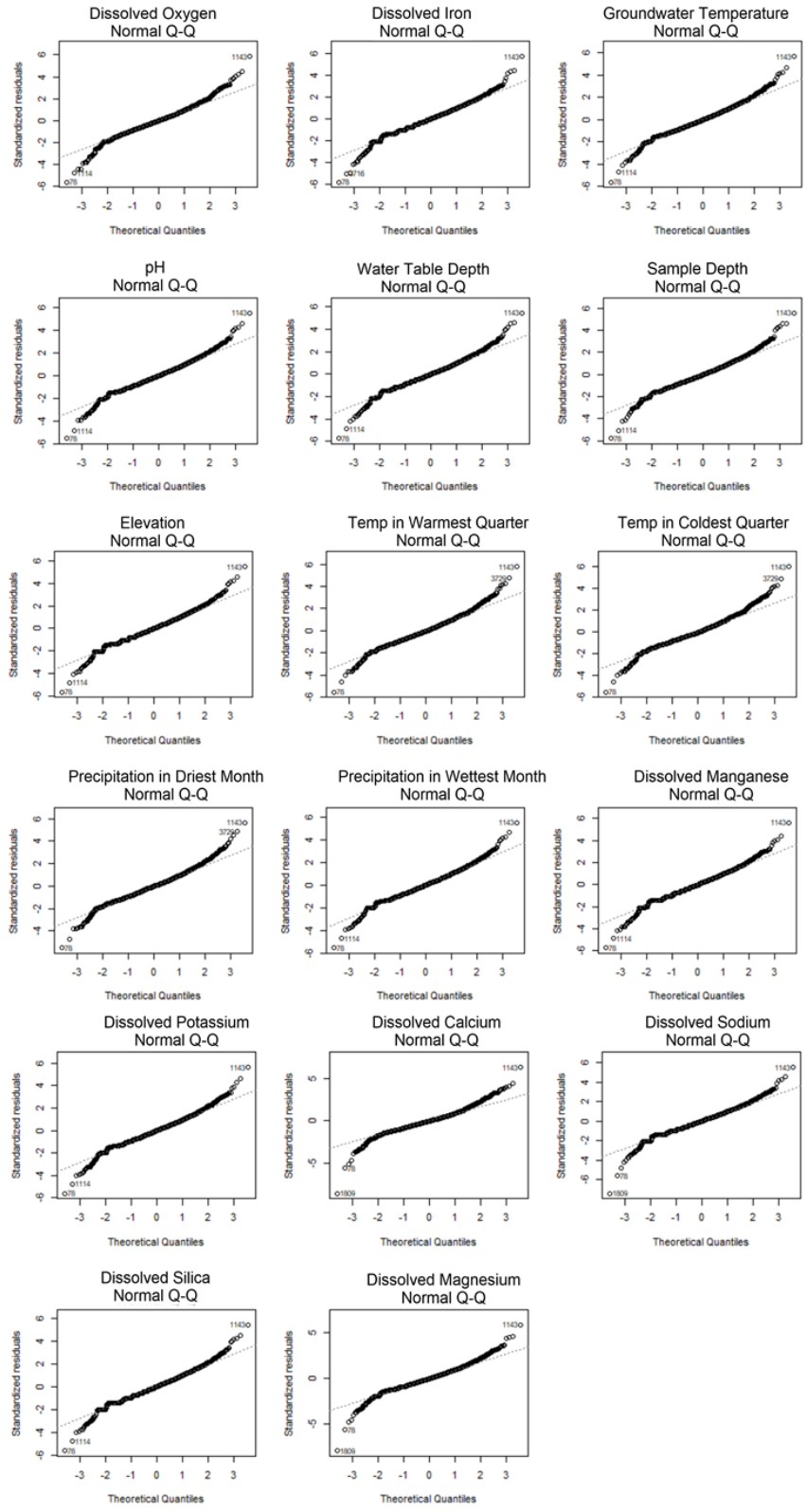
### Supplementary Figure 8

Timeseries of groundwater TOC concentrations in south-west Florida, United States (upper left plot) with corresponding histogram shown on the upper right. Timeseries of groundwater DOC concentration data in Perth, Australia (lower plot) with corresponding histogram shown on the lower right). TOC data used for Florida due to the paucity of groundwater DOC datasets available. Here we assume that majority of the TOC in groundwater is dissolved. Both datasets represent currently residential areas. Grey dots represent individual concentration data with dashed lines representing locally estimated scatterplot smoothing (LOESS) colored by correlation coefficient for individual bores ( $n = 45$  bores and  $n = 51$  bores for Perth and Florida, respectively). LOESS smoothing used as many datasets are non-linear. The data suggests a mix of trends including increasing concentrations, decreasing concentrations and no change in concentrations over time. Florida and Perth data were provided by the Southwest Florida Water Management District and Water Corporation (Western Australia) respectively.

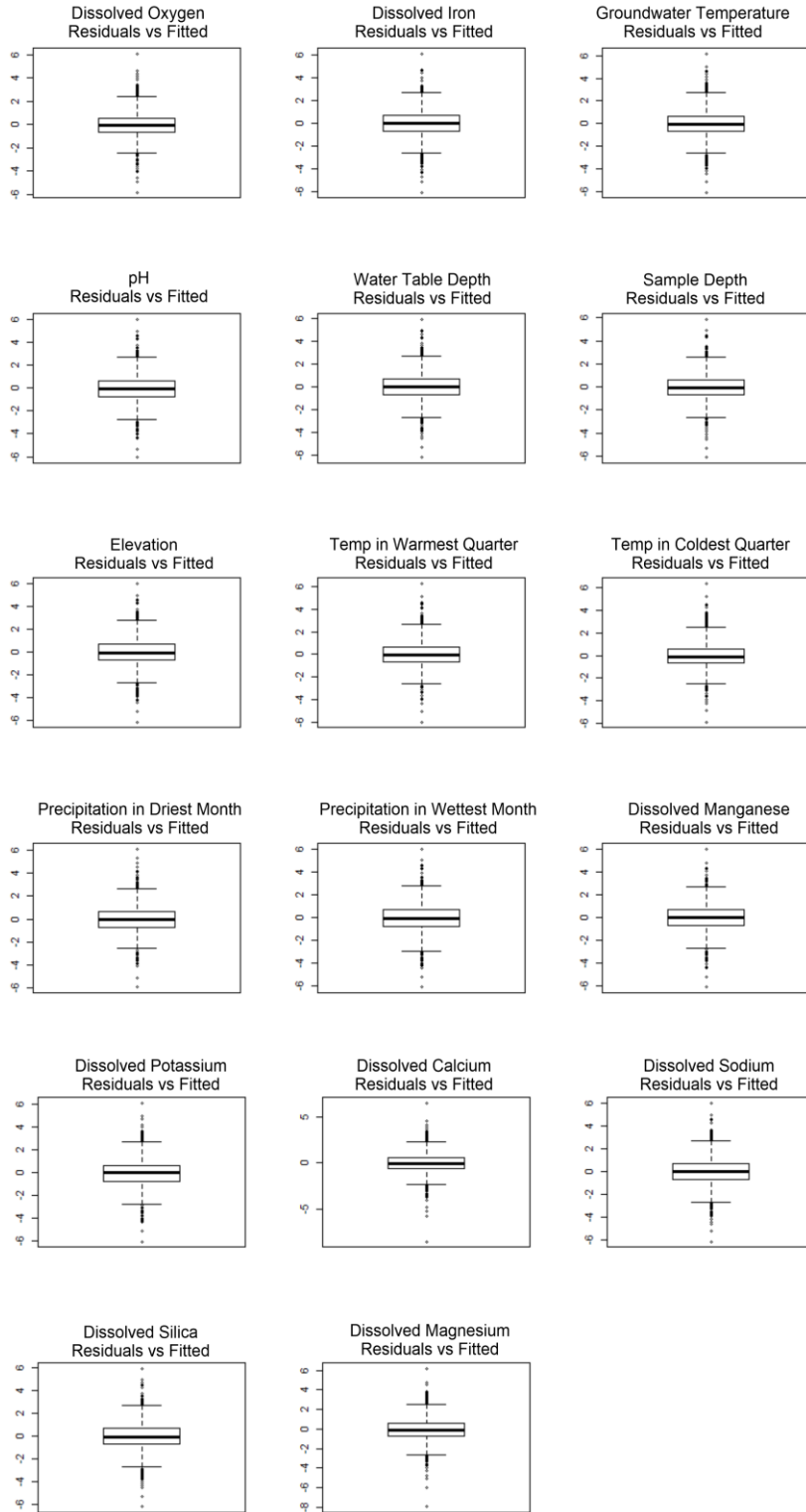


**Supplementary Figure 9**

Residuals vs Fitted plots for all quantitative variables used in the model.



**Supplementary Figure 10**  
 Q-Q plots for all quantitative variables used in the model.



### Supplementary Figure 11

Box plots of residuals for all quantitative variables used in the model. Boxes represent the interquartile range, containing median, with whiskers representing the upper and lower 25% of data. The circles shown outside of the whiskers represent outliers.

## Supplementary Tables

### Supplementary Table 1

Summary of compiled data for groundwater DOC concentration comparison between countries.

Source	Country	n	Data location	DOC filtration size (µm)	
4	Algeria	5	Data provided by authors of published source	0.45	
5	Australia	29	<a href="https://www.sciencedirect.com/science/article/pii/S0048969717314961?via%3Dihub#t0005">https://www.sciencedirect.com/science/article/pii/S0048969717314961?via%3Dihub#t0005</a>	0.2	
6		8	Data provided by authors of published source	0.7	
7		10	Data provided by authors of published source	0.7	
8		15	Data provided by authors of published source	0.7	
9		17	Data provided by authors of published source	0.45	
10		33	Data provided by authors of published source	0.45	
11		2	Data provided by authors of published source	0.45	
12		5	Data provided by authors of published source	0.7	
13		74	Data provided by authors of published source	0.7	
14		10	Data provided by authors of published source	0.7	
15		14	Data provided by authors of published source	0.7	
16		10	Data provided by authors of published source	0.7	
17		38	Data provided by authors of published source	0.7	
18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33		1,909	Data provided by co-authors to this paper, Water NSW and Water Corporation W.A.	0.45	
34		Argentina	15	Data provided by co-authors to this paper	0.7
35		Bangladesh	13	Data provided by co-authors to this paper	0.45
34		Brazil	30	Data provided by co-authors to this paper	0.7
36	China	17	<a href="https://www.sciencedirect.com/science/article/pii/S0048969715301091#t0005">https://www.sciencedirect.com/science/article/pii/S0048969715301091#t0005</a>	0.45	
37		17	<a href="http://www.swdzgcdz.com/oa/pdfdown.aspx?Sid=201303">www.swdzgcdz.com/oa/pdfdown.aspx?Sid=201303</a>	0.45	
31	Cook Island	17	Data provided by co-authors to this paper	0.7	
38	France	4	<a href="https://www.sciencedirect.com/science/article/pii/S004896970400155X?via%3Dihub">https://www.sciencedirect.com/science/article/pii/S004896970400155X?via%3Dihub</a>	0.45	
39	Germany	34	<a href="https://www.sciencedirect.com/science/article/pii/S0883292799000219">https://www.sciencedirect.com/science/article/pii/S0883292799000219</a>	0.45	
40	Canada	52	Data provided by authors of published source	0.45	
41		3	<a href="https://agupubs.onlinelibrary.wiley.com/doi/epdf/10.1029/WR026i012p02949">https://agupubs.onlinelibrary.wiley.com/doi/epdf/10.1029/WR026i012p02949</a>	0.45	
42		12	<a href="https://www.sciencedirect.com/science/article/pii/016896229190032R">https://www.sciencedirect.com/science/article/pii/016896229190032R</a>	0.45	
43	Belgium	18	Data provided by authors of published source	0.45	
	Czech Republic	104	Data provided by authors of published source	0.45	

	Denmark	20	Data provided by authors of published source	0.45
	Estonia	19	Data provided by authors of published source	0.45
	France	7	Data provided by authors of published source	0.45
	Malta	8	Data provided by authors of published source	0.45
	Poland	40	Data provided by authors of published source	0.45
	Portugal	28	Data provided by authors of published source	0.45
	Spain	10	Data provided by authors of published source	0.45
	United Kingdom	113	Data provided by authors of published source	0.45
44	Ethiopia	44	Data provided by co-authors to this paper	0.45
45	Iceland	24	Data provided by co-authors to this paper	0.45
35	India	79	Data provided by authors of published source	0.45
46	Kenya	36	Data provided by co-authors to this paper	0.45
47	Mali	12	Data provided by co-authors to this paper	0.45
48, 49	Malawi	41	Data provided by co-authors to this paper	0.45
		48	Data provided by co-authors to this paper	0.45
50	Nepal	40	Data provided by co-authors to this paper	0.45
51	Nigeria	35	Data provided by co-authors to this paper	0.45
52	Scotland	270	Data provided by authors of published source	0.45
53	Senegal	22	Data provided by co-authors to this paper	0.45
54, 55	Uganda	20	Data provided by co-authors to this paper	0.45
		51	Data provided by co-authors to this paper	0.45
56	United States	9	<a href="https://esajournals.onlinelibrary.wiley.com/doi/10.1890/0012-9658%282000%29081%5B3133%3AOCSSAMI%5D2.0.CO%3B2">https://esajournals.onlinelibrary.wiley.com/doi/10.1890/0012-9658%282000%29081%5B3133%3AOCSSAMI%5D2.0.CO%3B2</a>	0.22
40		5459 <sub>1</sub>	Data provided by authors of published source	0.45
57		99	<a href="https://pubs.usgs.gov/journal/1974/vol2issue3/report.pdf">https://pubs.usgs.gov/journal/1974/vol2issue3/report.pdf</a>	0.45
58		89	Data provided by authors of published source	0.7
59		156	Data provided by authors of published source	0.7
60	Zambia	110	Data provided by authors of published source	0.45
Notes	<sup>(1)</sup> Sample number after removing samples known to be contaminated.			

## Supplementary Table 2

Model parameters, intercepts, confidence intervals and significance levels for US dataset. P value tests that the intercept and slopes > 0 using Satterthwaite approximations.

	<i>Estimates</i>	<i>Standard Error</i>	<i>Degrees of Freedom</i>	<i>T statistic</i>	<i>P-value (&gt; t )</i>
<b>Fixed Parts</b>					
(Intercept)	2.02	0.32	530.4	6.28	7.28x10 <sup>-10</sup> (***)
Water table depth (m below land surface)	1.84x10 <sup>-3</sup>	1.02x10 <sup>-3</sup>	2913	1.81	0.071 (.)
Land Elevation (m asl)	-2.66x10 <sup>-4</sup>	5.49x10 <sup>-5</sup>	2915	-4.84	1.34x10 <sup>-6</sup> (***)
Temperature in the Wettest Quarter of the Year (°C)	3.35x10 <sup>-2</sup>	3.49x10 <sup>-3</sup>	2888	9.61	< 2.00x10 <sup>-16</sup> (***)
Temperature in the Warmest Quarter of the Year (°C)	-9.33x10 <sup>-2</sup>	1.09x10 <sup>-2</sup>	2906	-8.53	< 2.00x10 <sup>-16</sup> (***)
Precipitation in the Wettest Month of the Year (mm)	-2.45x10 <sup>-3</sup>	7.51x10 <sup>-4</sup>	2905	-3.27	0.001 (**)
Iron (µg / L)	6.87x10 <sup>-5</sup>	4.97x10 <sup>-6</sup>	2903	13.80	< 2.00x10 <sup>-16</sup> (***)
Dissolved Oxygen (mg / L)	-7.06x10 <sup>-2</sup>	5.53x10 <sup>-3</sup>	2915	-12.80	< 2.00x10 <sup>-16</sup> (***)
Sample Temperature (°C)	4.49 x10 <sup>-2</sup>	5.28 x10 <sup>-3</sup>	2908	8.50	< 2.00x10 <sup>-16</sup> (***)
pH	-9.61x10 <sup>-2</sup>	2.34x10 <sup>-2</sup>	2907	-4.11	4.06x10 <sup>-5</sup> (***)
Sample Depth (m below ground surface)	-7.77x10 <sup>-3</sup>	6.30 x10 <sup>-4</sup>	2899	-12.30	< 2.00x10 <sup>-16</sup> (***)
Precipitation in the Driest Month of the Year (mm)	-9.53x10 <sup>-3</sup>	1.05x10 <sup>-3</sup>	2899	-9.07	< 2.00x10 <sup>-16</sup> (***)
Calcium (mg / L)	4.45x10 <sup>-3</sup>	4.08 x10 <sup>-4</sup>	2915	10.90	< 2.00x10 <sup>-16</sup> (***)
Potassium (mg / L)	2.83x10 <sup>-2</sup>	3.82x10 <sup>-3</sup>	2902	7.41	1.69x10 <sup>-13</sup> (***)
Manganese (µg / L)	3.75x10 <sup>-5</sup>	1.81x10 <sup>-5</sup>	2899	2.07	0.039 (*)
Sodium (mg / L)	-5.66x10 <sup>-4</sup>	1.73x10 <sup>-4</sup>	2904	-3.27	0.001 (**)
Silica (mg / L)	-6.29x10 <sup>-3</sup>	1.32x10 <sup>-3</sup>	2914	-4.77	1.97x10 <sup>-6</sup> (***)
Landuse- Natural	-9.74x10 <sup>-3</sup>	4.87x10 <sup>-2</sup>	2911	-0.20	0.841 (.)
Landuse - Urban	0.17	5.31x10 <sup>-2</sup>	2915	3.19	0.001 (**)
<b>Random Parts</b>					
σ <sup>2</sup> (1)				0.689	
T00, Aquifer Age (2)				0.215	
ICC <sub>Aquifer Age</sub> (3)				0.238	
Observations				2916	
R <sup>2</sup> / Ω <sub>0</sub> <sup>2</sup> (4)				.429 / .429	
AIC (5)				7282.231	
Notes	<p>* p&lt;.05 ** p&lt;.01 *** p&lt;.001 . p&gt;.05            (1) Within group variance            (2) Between group variance (Aquifer Age)            (3) Intraclass correlation coefficient            (4) Random slope intercept correlation            (5) Akaike information criteria</p>				

**Supplementary Table 3**

Summary table of annual average temperature and precipitation in the US NWQMC data dataset.

	Annual average temperature (°C)	Annual average precipitation (mm)
Min.	2.8	94.0
1st Qu.	9.2	480.2
Median	11.5	812.0
Mean	12.6	809.1
3rd Qu.	16.5	1133.0
Max.	24.2	1798.0



#### Supplementary Table 4

Model coefficient estimates, standard error, t value and p values for simple linear models of  $\log(\text{DOC concentration (mg / L)})$  vs significant individual quantitative variables used in the US model shown for available Australian data. P value is testing that the slope of the linear model  $\log(\text{DOC concentration (mg / L)})$  vs variable is significantly different from 0.

Parameter	Estimate	Std. Error	T value	P value (> t )	Change in DOC concentration with one unit increase in parameter for Australian data (US results shown in brackets)
Dissolved Oxygen (mg / L)	-0.05883	0.07011	-0.839	0.40400	-5.713% (-6.812%)
Iron ( $\mu\text{g}$ / L)	0.00026	0.00004	6.249	<0.00001	0.026% (0.007%)
Sample Temperature ( $^{\circ}\text{C}$ )	-0.22571	0.09605	-2.350	0.02130	-20.205% (4.593%)
pH	-0.39960	0.47140	-0.848	0.39900	-32.941% (-9.162%)
Land Elevation (m above sea level)	-0.00513	0.00131	-3.911	0.00020	-0.512% (-0.027%)
Sample Depth (m below ground surface)	-0.00265	0.00121	-2.183	0.03210	-0.264% (-0.774%)
Temperature in the Wettest Quarter of the Year ( $^{\circ}\text{C}$ )	-0.17840	0.03508	-5.085	<0.00001	-16.339% (3.407%)
Temperature in the Warmest Quarter of the Year ( $^{\circ}\text{C}$ )	-0.44833	0.09439	-4.750	0.00001	-36.131% (-8.908%)
Precipitation in the Wettest Month of the Year (mm)	0.01606	0.00408	3.938	0.00018	1.619% (-0.245%)
Precipitation in the Driest Month of the Year (mm)	0.04462	0.00887	5.031	<0.00001	4.563% (-0.949%)
Calcium (mg / L)	0.01317	0.00362	3.643	0.00049	1.326% (0.446%)
Potassium (mg / L)	0.18690	0.23410	0.798	0.42700	20.551% (2.869%)
Sodium (mg / L)	-0.00891	0.00447	-1.992	0.04990	-0.887% (-0.057%)
Manganese ( $\mu\text{g}$ / L)	0.00015	0.00015	1.052	0.29600	0.015% (0.004%)
Silica (mg / L)	-0.04096	0.01308	-3.131	0.00246	-4.013% (-0.627%)

### Supplementary Table 5

Comparison between US and Australian data for climate variables. P-values based on Welch's Two Sample t-test for differences in means for variables with equal variance (temperature in the wettest quarter of the year, precipitation in the driest month of the year and sample temperature) and standard t-test used for variables with equal variance (precipitation in the wettest month). P values test whether the means for each variable are significantly different from 0.

	Temperature in the wettest quarter of the year		Precipitation in the driest month of the year		Precipitation in the wettest month of the year		Sample temperature	
	Australia	United States	Australia	United States	Australia	United States	Australia	United States
Min.	16.2	-0.5	33.0	0.0	72.0	11.0	18.6	4.0
1st Quarter	23.1	18.0	35.0	11.75	93.5	83.0	20.9	12.6
Median	25.3	20.6	35.0	25.0	94.0	107.0	21.6	15.78
Mean	23.2	19.2	42.6	36.3	101.9	104.7	21.57	16.15
3rd Quarter	25.4	23.3	41.0	62.0	97.0	122.0	22.25	19.5
Max.	25.6	27.6	71.0	109.0	165.0	255.0	28.4	34.0
St. Dev.	3.4	6.1	13.5	27.7	31.0	36.0	1.4	4.7
n	79	2916	79	2916	79.0	2916	79	2916
p-value for differences in mean	< 2.2 x 10 <sup>-16</sup>		1.54 x 10 <sup>-4</sup>		0.507		2.2 x 10 <sup>-16</sup>	

**Supplementary Table 6**

Comparison of linear model ( $\log(\text{DOC concentration (mg / L)})$  vs variable) slopes between US and Australian data for climate variables after confining US data to the min and max ranges available in the Australian dataset for the same variables. P values test whether the slope is significantly different from 0.

	Temperature in the wettest quarter of the year		Precipitation in the driest month of the year		Precipitation in the wettest month of the year		Sample temperature	
	Australia	United States	Australia	United States	Australia	United States	Australia	United States
Estimate	-0.178	-0.275	0.045	0.016	0.016	0.007	-0.226	-0.099
Std. Error	0.035	0.041	0.009	0.008	0.004	0.008	0.096	0.045
T value	-5.085	-6.764	5.031	2.014	3.938	0.896	-2.350	-2.200
P value (> t )	2.52 x 10 <sup>-6</sup>	1.483 x 10 <sup>-9</sup>	3.11 x 10 <sup>-6</sup>	0.047	1.79 x 10 <sup>-4</sup>	0.373	0.021	0.030
Adjusted R <sup>2</sup>	0.242	0.337	0.238	0.034	0.157	-0.002	0.055	0.042

### Supplementary Table 7

Cost of construction, annual operation and household water cost increases resulting from GAC filtration implementation and operation as determined using the US EPA work breakdown structure model for cost estimation of GAC <sup>61</sup>. Calculations assume a system of gravity-fed concrete GAC contactors with an empty bed contact time of 20 minutes, treating a design flow of 6.6 MGD (25 MI/d).

	\$US*	\$US/gallon (\$US/litre)
Total Capital Cost	4,344,568	0.66 (0.17)
Annual O&M Cost	1,049,545	0.16 (0.04)
Annualized Cost (35.4 years at 7%)	1,384,170	
Annualized cost per 1,000 gallons (3785 litres) average flow	0.57	
Annualized cost per household per year	134	
* Prices at 2014 base date.		

## Supplementary Notes

### Supplementary Note 1

#### Comparison to Australian groundwater DOC data

The results for each significant ( $p < 0.05$ ) quantitative (chemical and climatic) model variable in the US dataset were compared to simple linear regression models of  $\log(\text{DOC concentration})$  vs. the same variables from the Australian dataset (sample  $n = 79$  after removing any row containing one or more missing value of any variable). The results (Supplementary Table 4) show that the slope directions for all chemical variables match the slope directions in the US model, suggesting that the effect of water chemistry on DOC appears to be globally consistent. The regression analyses also however show that some climate specific variables exhibit an opposite trend in the Australian dataset compared to the US dataset. These include precipitation in the wettest and driest months of the year, temperature in the wettest quarter of the year and sample temperature. Welch's Two Sample t-test for differences in means reveal that the average temperature in the wettest quarter of the year, precipitation in the driest month of the year and sample temperature are significantly higher with lower standard deviations (44%, 51% and 70% lower respectively) in the Australian dataset than the US dataset ( $p < 0.001$ , Supplementary Table 5 and Supplementary Figure 6). This implies that different climate types have different effects on groundwater DOC concentrations. This was further confirmed by constraining the US dataset to the minimum and maximum ranges available in the Australian dataset for each of the four climate related variables. Simple linear regression models were performed for  $\log(\text{DOC concentration (mg / L)})$  vs. the four individual variables and it was confirmed that the direction of correlation shown by the four simple linear models (Supplementary Table 5) are equivalent to the direction of correlation in the Australian dataset (Supplementary Table 6). Increased temperatures in arid climates such as Australia, limits rather than primes biological activity due to water limitation and low variability in precipitation rates. In these climate types, groundwater DOC is more likely to be sourced by river recharge than diffuse rainfall recharge.

### Supplementary Note 2

#### Calculation of costs associated with implementation of GAC treatment for DOC removal from groundwater

A USA Environment Protection Agency (EPA) costing tool for GAC <sup>61</sup> was used to determine the costs associated with the construction and implementation of GAC (<https://www.epa.gov/dwregdev/drinking-water-treatment-technology-unit-cost-models-and-overview-technologies>). This was applied to a 25 Megalitre per day (ML/d) plant (Supplementary Table 7) which was determined to be large enough to achieve economies of scale.

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