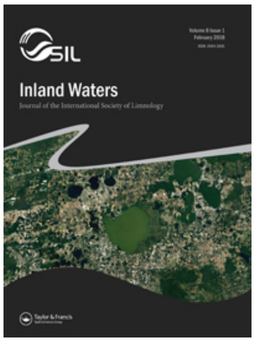

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RESEARCH BRIEF

Size, age, renewal, and discharge of groundwater carbon

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ABSTRACT

Groundwater carbon (C) supply to lakes and streams is important to understanding the role of inland waters in global and regional cycles and in the functioning of aquatic ecosystems. We provide new estimates of the size and discharge of the groundwater C pool using data from a broad survey of groundwater C, information on the depth distribution of groundwater, and data on groundwater age. About 0.25×10^6 km^3 of the 8×10^6 km^3 of groundwater resource is within 100 m of the surface and 4.2×10^6 km^3 is above 2000 m. Ages show an average groundwater turnover time of 10 yr at 25 m, 350 yr at 100 m, increasing to about 100 000 yr at 600 m. Global groundwater discharge is 16 000 km^3 yr^{-1}; >16% of precipitation passes through groundwater. Groundwater dissolved organic C (DOC) can be high in shallow groundwater but stabilizes at ~2–4 mg L^{-1} at 100 m. Average groundwater dissolved inorganic C (DIC) is ~30–43 mg L^{-1}. Groundwater C content to 2000 m is ~145 Pg, about the same as all marine sediments and about one-sixth that of the surface ocean. Groundwater C discharge to continental waters is 0.68 Pg yr^{-1}, or 3.4 times that estimated from river base-flow and submarine groundwater discharge. This discharge is 68 times previous estimates, implying a total C flux from land of 3.6 Pg yr^{-1}; 80% of discharge occurs from above 40 m and 99% from the upper 100 m.

KEYWORDS
carbon; depth; discharge; global; groundwater; renewal

Introduction

Groundwater carbon (C) supply to lakes (e.g., Hanson et al. 2014) and streams (e.g., Oviedo-Vargas et al. 2015) is essential to understanding the role of inland waters in the global C cycle but is poorly constrained in global analyses (e.g., Cole et al. 2007). Groundwater makes up much of the liquid water on the continents and may contain substantial C, yet little is known about the quantity or composition of this C, the global role of groundwater in the C budget, or its rate of discharge to surface waters. Predicting changes in atmospheric carbon dioxide (CO_2) and the course of climate change relies on complete and accurate estimates of the sizes and rates of exchange of all global C pools. In addition to global analyses, groundwater C is increasingly implicated in regional C cycling (Genereux et al. 2013, Olefeldt et al. 2013). Currently, both the size and turnover time of the global groundwater C pool (Cole et al. 2007) and the influence of human domestic, industrial, and agricultural groundwater withdrawal on global C cycling are unknown. Poorly constrained estimates hamper understanding of the role of groundwater and human groundwater withdrawals in inland water functions, global C budgets, and global climate change.

Groundwater volume at a given depth is the product of the total volume of sediments and the fraction of groundwater found in the pore spaces within sediments. No published depth-dependent assessments of groundwater and groundwater C distribution exist from which to determine global groundwater C and C flux. Published estimates of the volume of the global groundwater resource vary between 8 and 330 million km^3 (Garrels and MacKenzie 1971, Gavrilenko and Derpgol'ts 1971, L'vovich 1974, Southam and Hay 1981, Schlesinger 1991, 1997, Clarke 1993, Alley et al. 2002, Shiklomanov and Rodda 2003, Trenberth et al. 2007). Because methods in older publications are vague, unspecified, and imply an impossibly large porosity, we have more confidence in modern estimates that cluster from 8 to 23.4 million km^3 (Clarke 1993, Shiklomanov and Rodda 2003, Trenberth et al. 2007). Groundwater flux and recharge are somewhat better known via GIS, hydrologic models, and independent seepage measurements (Döll and Fiedler 2008) and likely are 12 660 km^3 yr^{-1}. Groundwater C content has not

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been summarized globally, so estimates of the size of the groundwater C pool are conspicuously underrepresented in summaries of global C cycling (IPCC 2013).

The objective of this analysis was to calculate the volume and depth distribution of the groundwater resource beneath continents, estimate the C content of groundwater using a database collected systematically on the diverse land forms across the United States, and, assuming groundwater C content is generally similar across the globe, use groundwater age and depth distributions to approximate the global groundwater C turnover rate and flux to surface waters.

Methods

We calculated the exchangeable groundwater volume using a conservative global estimate of total groundwater beneath continents (Clarke 1993) and estimates of the depth distribution of groundwater derived from spherical geometry and relationships between porosity and depth. The depth distribution profile of groundwater was estimated by calculating the volume of concentric hollow spheres with 1 m thickness with Earth's radius (r) of 6371 km. For example, the volume (V) of the first 1 m of the Earth is calculated as:

$$V = (4/3\pi r^3) - (4/3\pi(r - 1)^3).$$

Concentric sphere volumes were calculated to a depth of 8000 m below ground and then reduced to 29.2% to represent the area beneath landmasses. The volumes of Earth at each descending depth (V_p) were adjusted for porosity (p) or its inverse, "solidity," following known depth–solidity relationships (Baldwin and Butler 1985). Therefore, the volume of the potential pore space or groundwater volume in the first 1 m of the Earth is estimated by the following formula:

$$0.292V_p.$$

Estimated total groundwater volume in the pore-space of sediments down to about 8000 m range from 8.0 × 10^6 to 330 × 10^6 km^3 (Hay and Leslie 1990, Clarke 1993, Trenberth et al. 2007). A depth of 8000 m was used as a practical limit because the temperature of the Earth would make liquid water unlikely at greater depth, and porosity would be near zero (Hay and Leslie 1990). To make the most conservative estimate of groundwater and groundwater C, we weighted the smallest of these estimates (Clarke 1993) by the calculated distribution of groundwater from pore space and global geometry, down to 2000 m assuming water below that depth would exchange at negligible rates.

We assessed groundwater ages at different depths (Fig. 1a) and annual rates of renewal. Groundwater age-at-depth was characterized from multiple published sources (Fig. 1 legend) and included observations from several countries and conditions. These ages were determined using a variety of isotopes appropriate to the age ranges analyzed (Suckow 2014). We determined the average age at various depths by smoothing the depth versus groundwater age relationship using locally weighted sequential smoothing (LOWESS; Cleveland 1979; Fig. 1a). Predicted LOWESS fits were averaged across ranges of depths for which C measures could also be averaged. The annual renewal rate and thus the rate of discharge of groundwater from each 1 m stratum in a given year was calculated, assuming groundwater equilibrium, as the inverse of groundwater age. This fraction, multiplied by the volume of groundwater in each stratum, yielded an estimate of the rate of annual renewal by precipitation (Table 1).

Estimates of water volume and mean age of groundwater at depth permit the equilibrium calculation of annual groundwater discharge to seeps, streams, lakes, and oceans. Data on dissolved organic (DOC) and inorganic (DIC) C were derived from a wide-ranging survey across the diverse landforms of North America (http://waterdata.usgs.gov/nwis/qw). Because of the diversity of landforms and geology covered, we believe these groundwater C values are likely substantially similar to the concentrations and distribution beneath other continents. Measurements of groundwater DOC (e.g., Huang et al. 2015, Thayalakumaran et al. 2015, Weigand et al. 2017) and DIC (e.g., Chaillou et al. 2014, Samanta et al. 2015, Cao et al. 2016) under other countries and continents, although rare, are in the same ranges as those in our survey. We determined the average C content at various depths by smoothing the depth versus groundwater age relationship using locally weighted sequential smoothing (Cleveland 1979) as described earlier. Predicted LOWESS fits were averaged across ranges of depths.

Results and discussion

The calculated volume of groundwater to a depth of 100 m is about 0.25 × 10^6 km^3 and to 2000 m is 4.2 × 10^6 km^3 (Table 1). Groundwater has an average turnover time of about 10 yr at 25 m, 350 yr at 100 m, 10 000 yr at 200 m, and about 100 000 yr at 600 m. Average near-surface groundwater age matches published short turnover times (<10 yr; Alley et al. 2002), and the volume-weighted average age of groundwater to <800 m depth (29 000 yr) is within 30% of published values (Smith and Wheatcraft 1993). Groundwater involved in hydrologic cycles and inland waters on the scale of years to centuries is generally shallower than ~200 m (Fig. 1a).

The groundwater content of DIC and DOC at depth were calculated via a large groundwater database covering the diverse landforms of North America and therefore

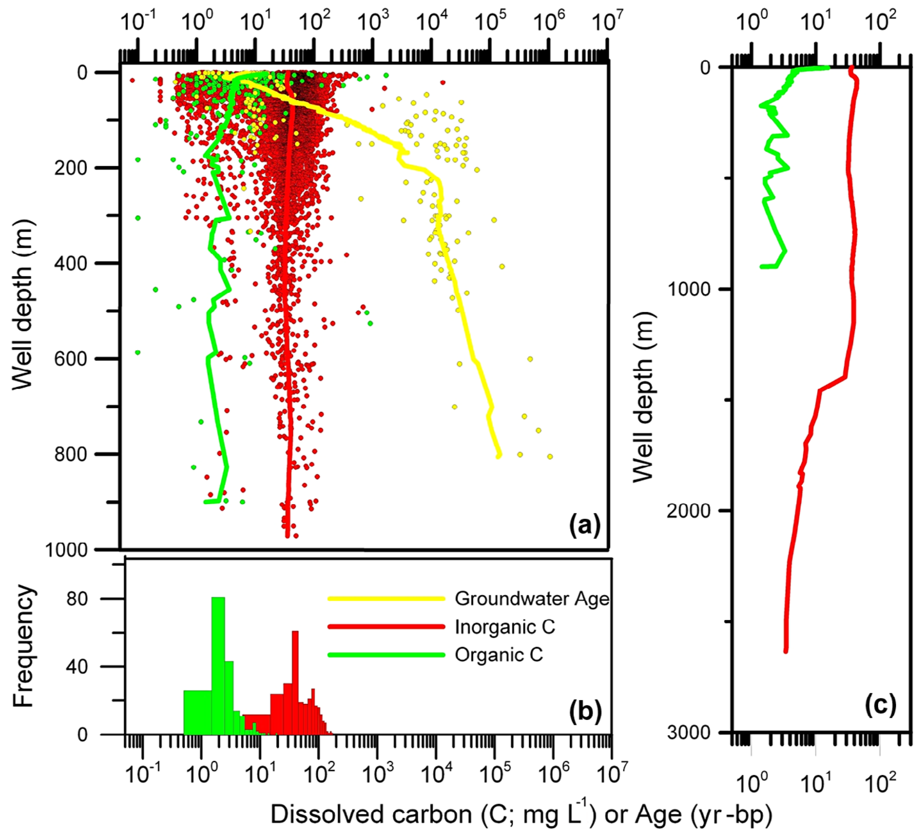

Figure 1. Depth distribution of groundwater age and DIC and DOC concentrations: (a) groundwater <1000 m from the surface; (b) histogram of DIC and DOC concentrations; and (c) to greater depths. Lines are fitted using LOWESS. Age estimates (yr) are from multiple published sources (Elliott 1990, 2000, Elliott et al. 1999, Manga 1999, Edmunds and Smedley 2000, Iwatsuki et al. 2001, Katz et al. 2001, Plummer et al. 2001, 2000, Swanson et al. 2001, Swarzenski et al. 2001, Dowling et al. 2003, Chen et al. 2003, Sturchio et al. 2004).

Table 1. Estimates of groundwater volume, annual water renewal and turnover, and concentrations and annual discharge of DIC and DOC.

Depth (m)	Mean age (yr)	Renewal (%/yr)	Volume (km^3)	Discharge (km^3 yr^{-1})	DIC (mg L^{-1})	DOC (mg L^{-1})	C discharge (Tg yr^{-1})
0–10	6	16.173	28 646	4633	35.5	8.8	205.3
10–20	6	15.812	25 961	4105	34.9	4.9	163.4
20–30	10	9.610	25 885	2488	35.1	4.7	99.0
30–40	14	7.046	25 808	1819	36.8	4.4	74.9
40–50	24	4.210	25 732	1083	41.0	4.0	48.7
50–60	34	2.939	25 656	754	42.9	3.9	35.3
60–70	57	1.768	25 581	452	43.6	4.0	21.6
70–80	120	0.831	25 505	212	43.4	3.7	10.0
80–90	143	0.699	25 430	178	43.2	3.4	8.3
90–100	325	0.307	25 355	78	42.9	3.3	3.6
100–110	499	0.201	25 280	51	41.8	3.2	2.3
110–120	722	0.139	25 206	35	41.2	2.8	1.5
120–130	927	0.108	25 132	27	40.5	2.5	1.2
130–140	1478	0.068	25 057	17	39.7	2.5	0.7
140–150	2597	0.039	24 984	10	39.0	2.0	0.4
150–160	2625	0.038	24 910	9	38.7	2.0	0.4
160–170	3276	0.031	24 836	8	38.2	2.0	0.3
170–200	2984	0.034	74 071	25	37.6	2.0	1.0
200–250	12 516	0.008	122 002	10	36.1	2.1	0.4
250–300	14 441	0.007	384 363	27	34.6	2.2	1.0
300–350	13 867	0.007	118 452	9	33.5	2.2	0.3
350–400	17 477	0.006	116 716	7	32.7	2.2	0.2
400–450	23 142	0.004	115 006	5	32.3	2.5	0.2
450–500	30 083	0.003	113 320	4	32.3	2.5	0.1
500–800	96 506	0.001	645 883	7	36.8	1.8	0.3
800–2000	1 444 965	0.000	2 076 113	1	28.2	2.4	0.0
Total				16 051			680.3

may be similar to the concentrations found beneath other continents. When a broad, global data collection is eventually compiled that allows testing this assumption, it may be necessary to revise our estimates. Across the United States, average DOC ($n = 1263$) is as high as 20 mg L^{-1} in shallow groundwater, with a few extreme values into the hundreds, perhaps in some cases because of surface water contamination. Average groundwater DOC stabilizes at ~2–4 mg L^{-1} at a depth of 100 m (Fig. 1a and b). Average groundwater DIC is considerably higher, averaging 30–43 mg L^{-1} ($n > 30\,000$), and does not vary substantially with groundwater depth (Table 1), although shallower DIC estimates showed wide variations among sites (Fig. 1a and b). DIC concentrations may decline substantially below 1500 m (Fig. 1c), but our dataset only included 20 observations at these depths.

Our calculations (Table 1) suggest a conservative estimate of annual global groundwater discharge is 16 000 $km^3\ yr^{-1}$, a value within 25% of the global groundwater recharge estimated using GIS, hydrologic models, and independent seepage measurements (Döll and Fiedler 2008). This calculation indicates at least 16% of annual precipitation (Berner and Berner 1995) passes through the groundwater system before discharge, a value equivalent to ~60% of the volume released to the sea annually by rivers (Wetzel 2001), implying that 40% of groundwater discharge may be lost to evaporation or discharged to marine systems. Because the average of submarine discharge estimates (Taniguchi et al. 2002) seems to be ~9% of river discharge, evaporation may consume 30% of annual discharge of groundwater.

According to these calculations, total global groundwater C content to 2000 m (Table 1) is likely to contain about 145 Pg of dissolved inorganic plus organic C, about the same as the C content of all marine sediments and about one-sixth that of the surface ocean (IPCC 2001). Approximately 30% of this C is in the organic form in shallow groundwater, with a negligible quantity of DOC at greater depth (Table 1). Calculated annual groundwater C discharge is ~0.68 Pg yr^{-1} or about 3.4-times that estimated from river base-flow and submarine groundwater discharge (IPCC 2001). This amount of groundwater C export is 68 times that of a previous estimate (Cole et al. 2007), bringing the back-calculated estimate of C influx to inland waters to 3.6 Pg yr^{-1} (Tranvik et al. 2009); 80% of this discharge occurs from the upper 40 m of groundwater and 99% from the upper 100 m (Table 1).

Groundwater flow and storage is being altered by human stresses (Alley et al. 2002). The current rate of water withdrawal from groundwater for human use is ~600 $km^3\ yr^{-1}$, or about 4% of annual groundwater discharge. At 0.03 Tg km^{-3} of C, groundwater withdrawals for agricultural, domestic, and industrial uses bring about 19 Tg of C to surface waters annually. Unless this growing human withdrawal for domestic, industrial, and agricultural use is compensated for by reduced natural groundwater discharge, human use already releases additional C equivalent to 10% of annual marine C sequestration (IPCC 2013). Human withdrawal of water might therefore be considered another aspect of global change that could potentially alter the size of the groundwater pool and atmospheric CO_2.

Groundwater is a pool that stores and releases substantial amounts of C and thus is valuable to consider in global budgets and analyses of inland water functions. Here we believe are the first estimates of the size of the global groundwater pool and the rate of discharge of groundwater DOC and DIC. As noted elsewhere (Downing 2009), these first-order global estimates are often made with an unestimated degree of error. We have tried to be conservative in the estimate of groundwater volume to avoid upscaling to unrealistically high estimates of groundwater C and C discharge. This new estimate of global groundwater C and C discharge will be improved when updated assessments of the global size of the whole groundwater pool have been made and science verifies that the global concentrations of groundwater DOC and DIC are similar to those found across our region.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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