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Estimating modern carbon burial rates in lakes using a single sediment sample

William O. Hobbs[1], Daniel R. Engstrom[1], Shawn P. Scottler[1], Kyle D. Zimmer[2], and James B. Cotner[3]*
[1]St. Croix Watershed Research Station, Science Museum of Minnesota, 16910 152 St. North, Marine on St. Croix, MN 55047, USA
[2]Department of Biology, University of St. Thomas, 2115 Summit Ave., Saint Paul, MN 55105, USA
[3]Department of Ecology, Evolution and Behavior, University of Minnesota, 100 Ecology Building, 1987 Upper Buford Circle, Saint Paul, MN 55108, USA

Abstract

The rate of organic carbon (OC) burial in inland waters is an important flux in the global C-cycle. Here we provide methodological improvements that offer a rapid and accurate assessment of modern OC burial rates in lakes from a single surface-sediment sample. Using a 93 lake dataset of reliably dated sediment cores (OC burial of 9 to 318 g m^{-2} y^{-1}), we demonstrate the applicability of this approach in a variety of lake types. We validate our estimated rates of OC burial against (1) measured whole-lake accumulation from the sum of multiple area-weighted sediment cores, (2) single central-basin cores adjusted for sediment focusing, and (3) duplicate sediment cores taken in multiple locations and at different times (4-10 years apart) in 9 lakes. Our single-sample estimates, which were in good agreement with measured values, suggest a within-lake variability of 4 g m^{-2} y^{-1} and have a small inter-lake error of only 6.5%. The applicability of this approach to other lakes and regions requires knowledge of (1) atmospheric ^{210}Pb flux, (2) an estimate of supported ^{210}Pb activity, and (3) some understanding of typical sedimentation rates in the study lakes. This approach provides an accurate assessment of OC burial, with increased potential for greater spatial coverage in inland waters and improved ability to address questions focused on terrestrial–aquatic exchanges of organic carbon.

Roughly 10% of the carbon that enters inland aquatic ecosystems from the land is permanently buried (Cole et al. 2007), which yields rates of organic carbon (OC) sequestration comparable with or even higher than in marine sediments and terrestrial soils (Gudasz et al. 2010). The spatial extent, rates, and efficiencies at which inland waters bury OC are therefore relevant to the discussion of the global C-cycle. Indeed, there has been considerable attention paid to estimating long-term rates of OC burial and storage in lakes (Dean and Gorham 1998; Einsele et al. 2001; Cole et al. 2007; Downing et al. 2008; Sobek et al. 2009; Anderson et al. 2009; Heathcote and Downing 2012; Mackay et al. 2012). However, considerable spatial heterogeneity exists among lakes and geographic regions, and improvements that refine the rate and magnitude of OC burial across lake types and regions are needed.

Previous estimates of OC burial in aquatic ecosystems have generally relied upon ^{210}Pb-dated lake-sediment cores (e.g., Sobek et al. 2009 and Heathcote and Downing 2012). As a result, the characterization of OC burial in inland waters is limited by the number of lake cores that are reliably dated, constraining large spatial surveys of lakes. In addition to the effort and cost required to date individual sediment cores, accurately assessing the lake-wide rate of OC burial is limited by sediment focusing—the spatial redistribution of fine-grained sediments by wave and current action. Focusing contributes a great deal of spatial heterogeneity to the accumulation of geochemical constituents across the depositional basin (Likens and Davis 1975; Lehman 1975; Blais and Kalff 1995; Mackay et al. 2012). As a result, proper estimation of whole-lake sedimentation rates generally requires multiple sediment cores covering the entire depositional basin (Swain et al. 1992; Rippey et al. 2008; Engstrom and Rose 2013) or the adjustment of a single central core based on the atmospheric flux of

*Corresponding author: E-mail: whobbs@smm.org,
Tel: +1 651 433-5953

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^{210}Pb (Rippey and Douglas 2004; Engstrom 2005). Methods that improve our ability to rapidly and accurately estimate OC burial rates over large spatial scales therefore are particularly important to understanding the role inland waters play in the global C-cycle.

Here, we present a method that allows for the rapid and accurate assessment of modern (last ~ 10 y) whole-lake OC-burial rates using a single sediment sample from the near-center of a lake basin and which accounts for sediment focusing to the sample site. We incorporate the earlier work of Binford and Brenner (1986), who formulated an approach to estimate the trophic state of a lake based similarly on the degree of ^{210}Pb dilution in the surface sediments. We demonstrate—by validation against conventional measures of whole-lake OC burial—the applicability of this method in a variety of lake types covering a large range of size and depth, and show how this approach furthers our understanding of the transport, focusing, and burial of OC and atmospheric ^{210}Pb.

Materials and procedures

Study sites

This method was developed using sediment cores from 93 lakes located throughout Minnesota and northern Wisconsin and upper Michigan, USA over the period of 1996–2010 (Table 1). The lakes are situated across several ecoregions as defined by land use, soil type, landforms, and natural vegeta-

Table 1. Study sites across Minnesota and northern Wisconsin and Michigan used in the analysis. Ecoregions are WCBP–Western Corn Belt Plains, NGP–Northern Glaciated Plains, NCHF–North Central Hardwood Forest, and NLF– Northern Lakes and Forests.

Lake	Ecoregion	County	Latitude	Longitude	Lake maximum depth (m)	Lake area (km²)	Cumulative dry mass (g/cm²)*	210Pb activity (Bq g⁻¹)†	% organic matter (10 y mean)	Lake sediment focus factor	Measured whole-lake OC burial (g C m⁻² y⁻¹)	Predicted single-sample OC burial (gC m⁻² yr⁻¹)
Bass	WCBP	Faribault, Minn.	43.81666667	–94.08083333	6.1	0.80	1.80	0.24	13.3	1.99	48	44
Dunns	NCHF	Meeker, Minn.	45.15555556	–94.42944444	6.1	0.63	2.99	0.15	19.2	3.73	86	105
Fish	NCHF	Dakota, Minn.	44.82222222	–93.16416667	10.2	0.13	3.26	0.28	13.2	2.70	56	38
Hook	WCBP	McLeod, Minn.	44.95465	–94.340176	5.5	1.33	3.18	0.20	26.7	4.04	133	107
Bean	NLF	Lake, Minn.	44.0713241	–95.37360592	7.9	0.13	1.49	0.14	17.0	1.54	75	98
Emily	WCBP	McLeod, Minn.	44.957383	–94.325687	1.6	0.40	1.00	0.23	31.3	1.16	114	101
Emily Peter	NCHF	Le Sueur, Minn.	44.311021	–93.922272	11.3	1.21	4.54	0.19	21.5	5.27	85	94
Fox	NGP	Martin, Minn.	43.67549896	–94.6989975	6.1	2.11	2.77	0.13	14.4	1.47	98	90
Greenleaf	NCHF	Le Sueur, Minn.	44.3972	–93.6267	5.8	1.22	6.30	0.15	19.7	4.78	110	106
Luce	NCHF	Carver, Minn.	44.964807	–93.781129	2.0	0.09	0.83	0.26	21.0	1.19	55	58
Lady Slipper	WCBP	Lyon, Minn.	44.5714	–95.629	3.4	1.16	1.48	0.16	14.5	1.59	55	71
Round	NLF	Itasca, Minn.	47.213207	–93.35848	4.9	0.40	5.36	0.22	20.2	5.21	102	77
Smith	NCHF	Wright, Minn.	45.079034	–94.126167	1.3	0.91	0.90	0.22	37.8	1.01	132	127
E.Bah	NGP	Douglas, Minn.	46.00365	–95.7635	3.0	0.16	0.93	0.25	20.8	1.17	59	59
Little Turtle	NGP	Grant, Minn.	45.88441667	–95.84591667	5.2	0.10	1.09	0.40	28.4	2.54	51	53
Beaver	WCBP	Steele, Minn.	43.89194444	–93.34911111	8.2	0.39	2.68	0.28	14.9	3.14	45	43
Diamond	NCHF	Kandiyohi, Minn.	45.18333	–94.8389	8.2	6.50	0.71	0.38	20.5	1.81	21	34
Duck	NCHF	Blue Earth, Minn.	44.21805556	–93.81555556	7.6	1.18	1.18	0.18	18.7	1.05	101	81
George B.E.	NCHF	Blue Earth, Minn.	44.98472222	–92.88361111	8.5	0.35	1.50	0.27	18.2	2.35	48	53
George Kandi	NCHF	Kandiyohi, Minn.	45.23333333	–94.98361111	9.8	0.92	0.80	0.41	18.8	1.99	28	32
Henderson	NCHF	Kandiyohi, Minn.	45.2307	–94.9929	17.4	0.30	0.40	0.76	30.0	2.11	27	25
Kreighle	NCHF	Stearns, Minn.	45.57916667	–94.47802778	20.1	0.51	0.21	0.60	47.1	1.03	36	33
Long	NCHF	Kandiyohi, Minn.	45.32583333	–94.87	13.7	1.33	0.38	0.53	20.6	1.60	21	23
Richardson	NCHF	Meeker, Minn.	45.15888889	–94.43941667	14.3	0.48	4.85	0.16	12.6	3.96	81	67
Stahls	NCHF	McLeod, Minn.	44.95416667	–94.41833333	11.3	0.55	1.58	0.30	26.3	2.33	71	68
Clear	WCBP	Sibley, Minn.	44.455292	–94.514751	2.4	2.00	0.63	0.11	18.8	0.30	109	109
Lura	WCBP	Blue Earth, Minn.	43.875687	–94.015914	2.8	5.26	1.85	0.20	16.0	1.58	65	64
Buffalo	WCBP	Murray, Minn.	44.07741667	–95.57903333	2.7	0.50	1.93	0.11	15.2	1.17	108	112
Edwards	NCHF	Pope, Minn.	45.50948333	–95.46865	2.8	0.57	0.89	0.27	23.0	1.27	73	63
Gil-Bret	NCHF	Pope, Minn.	45.42958333	–95.35815	2.2	0.10	1.18	0.24	21.4	2.12	49	67
Island	NGP	Lyon, Minn.	44.38283333	–96.00991667	2.3	0.65	1.56	0.20	19.9	1.81	62	76
Little Lower Elk	NCHF	Grant, Minn.	45.93336667	–95.80955	4.0	0.52	0.83	0.29	21.4	1.45	47	52
Lone Tree	WCBP	Yellow Medicine, Minn.	44.68958333	–95.44535	2.7	0.33	1.40	0.16	16.7	1.12	85	83
Malachy	NGP	Swift, Minn.	45.36806667	–95.67901667	1.1	0.13	1.15	0.20	16.4	1.27	62	61
Nelson	NCHF	Pope, Minn.	45.52386667	–95.4443	3.9	1.10	0.95	0.24	19.4	1.46	61	60
Ohsrund	NGP	Grant, Minn.	45.79721667	–96.04708333	2.0	0.76	1.13	0.23	18.3	1.44	63	61
Oak	NGP	Lincoln, Minn.	44.5367	–96.24246667	3.0	0.40	0.82	0.19	16.1	1.13	59	62
Round - Pope	NCHF	Pope, Minn.	45.5571	–95.27426667	3.5	0.79	1.19	0.22	16.0	1.96	70	57
Steep Bank	NGP	Lincoln, Minn.	44.53901667	–96.32761667	1.7	0.76	2.10	0.12	12.7	1.58	81	85
Slotseye	NCHF	Grant, Minn.	46.06308333	–95.84256667	3.8	0.10	1.04	0.28	19.4	1.73	50	52
Solem	NCHF	Douglas, Minn.	45.80981667	–95.63943333	3.7	0.15	0.69	0.40	26.7	1.54	41	45
Turtle A	NGP	Grant, Minn.	45.8859	–95.83581667	4.9	0.25	3.23	0.13	12.8	2.47	68	80
Turtle B	NGP	Grant, Minn.	45.88355	–95.83758333	8.5	1.78	3.82	0.16	14.3	3.38	65	74
Wolf	NLF	Lake, Minn.	43.85691667	–95.08986111	0.7	0.48	3.33	0.08	15.3	1.63	132	162
Hjermsted A	NGP	Murray, Minn.	44.17258333	–95.97130556	0.9	0.28	2.24	0.11	13.8	1.20	106	96
Skunk	NCHF	Grant, Minn.	45.15908	–95.05273	1.2	0.07	0.20	0.21	39.4	0.57	71	93
Murk	NCHF	Douglas, Minn.	45.1201	–95.07285	2.5	0.16	0.66	0.24	22.2	0.92	68	65
Mavis West	NCHF	Otter Tail, Minn.	46.26367	–96.0516	4.3	0.14	0.50	0.29	19.2	1.16	68	52
Mavis East	NCHF	Otter Tail, Minn.	46.0956	–96.04243	3.8	0.22	0.31	0.26	28.5	0.95	63	75
Leverson	NGP	Grant, Minn.	45.01778	–95.06203	1.3	0.08	3.68	0.12	17.2	2.04	120	114

Continued…

Table 1. Continued

Lake	Ecoregion	County	Latitude	Longitude	Lake maximum depth (m)	Lake area (km^2)	Cumulative dry mass (g/cm^2)*	210Pb activity ($Bq\ g^{-1}$)†	% organic matter (10 y mean)	Lake sediment focus factor	Measured whole-lake OC burial ($g\ C\ m^{-2}\ y^{-1}$)	Predicted single-sample OC burial ($gC\ m^{-2}\ yr^{-1}$)
Grandokken	NCHF	Douglas, Minn.	46.027	–95.14308	1.9	0.02	0.23	0.37	40.4	0.87	59	59
Frolund	NCHF	Pope, Minn.	45.08212	–95.53333	1.9	0.06	0.91	0.30	27.9	1.66	63	67
Blakesley	NCHF	Grant, Minn.	45.14057	–95.04053	2.0	0.03	1.11	0.27	38.8	1.42	110	108
Morrison	NCHF	Otter Tail, Minn.	46.131115	–95.892983	2.5	0.20	0.95	0.20	25.7	1.12	93	95
Org	NCHF	Douglas, Minn.	45.05857	–95.09078	3.3	0.03	1.43	0.21	24.4	1.76	67	87
Pisa	NGP	Stevens, Minn.	45.04808	–95.05515	1.3	0.10	0.80	0.31	28.5	0.96	83	64
Bellevue	NCHF	Grant, Minn.	45.09948	–95.02497	3.0	0.12	0.44	0.33	19.9	1.25	43	41
Christina E	NCHF	Douglas, Minn.	46.084006	–95.690649	4.3	2.00	0.87	0.40	27.0	1.61	46	51
Christina W	NCHF	Douglas, Minn.	46.098365	–95.742881	1.3	16.19	0.28	0.25	21.2	0.49	54	45
Tettegouche	NLF	Lake, Minn.	47.3449135	–91.2686615	4.6	0.27	0.25	2.04	44.8	2.25	19	19
Siskiwit	NLF	Keweenaw, Mich.	48.0005271	–88.7956283	45.1	16.35	0.27	2.06	23.4	1.89	13	10
Rainy	NLF	St. Louis/Koochiching, Minn.	48.539183	–92.8291833	49.1	233.80	0.34	0.82	13.3	1.15	14	15
Kabetogama	NLF	St. Louis, Minn.	48.4557667	–92.95295	24.4	97.26	0.33	0.85	24.7	1.27	29	26
Lac La Croix	NLF	St. Louis, Minn.	48.3611226	–92.1751029	51.2	55.47	0.14	1.50	20.6	1.14	13	11
Namakan	NLF	St. Louis, Minn.	48.4338	–92.702267	45.7	48.24	0.60	0.78	15.3	2.49	15	18
Moskey Basin, Lake Superior	NLF	Keweenaw, Mich.	48.068972	–88.5663986	NA	NA	0.84	0.98	13.8	2.40	19	13
Ahmik	NLF	Keweenaw, Mich.	48.14787	–88.54153	2.6	0.10	0.45	0.90	43.3	1.96	44	45
Harvey	NLF	Keweenaw, Mich.	48.05067	–88.79602	4.3	0.55	0.21	0.46	39.7	0.50	79	76
Richie	NLF	Keweenaw, Mich.	48.04092	–88.70236	10.5	2.16	0.32	1.21	24.1	1.76	19	18
Outer	NLF	Ashland, Wisc.	47.004173	–90.4597519	0.8	0.22	0.51	2.47	49.4	3.62	24	18
Beaver	NLF	Alger	46.56524	–86.34362	9.2	3.10	0.23	1.93	45.6	1.73	23	20
Grand Sable	NLF	Alger	46.641305	–86.0357166	19.2	2.55	0.52	1.27	11.6	2.51	12	9
Florence	NCHF	Leelanau, Mich.	45.010527	–86.1198853	7.3	0.26	0.09	2.15	56.5	1.60	21	21
Manitou	NCHF	Leelanau, Mich.	45.12693	–86.0237	13.1	1.04	0.46	1.25	34.5	2.05	30	25
Shell	NCHF	Leelanau, Mich.	44.9477137	–85.9000603	4.0	0.41	0.12	0.86	47.2	1.65	30	48
Bass	NCHF	Leelanau, Mich.	44.9231008	–85.884445	7.9	0.35	0.55	2.26	66.8	5.83	25	28
Peary	NLF	St. Louis, Minn.	48.52423	–92.77164	4.6	0.45	0.16	0.60	28.3	0.48	29	36
Ek	NLF	St. Louis, Minn.	48.46975	–92.836	5.8	0.37	0.17	1.62	45.1	1.21	25	23
Cruiser	NLF	St. Louis, Minn.	48.49753	–92.80225	27.7	0.47	0.24	1.28	31.8	1.16	26	22
Swamp	NLF	St. Louis, Minn.	47.951333	–89.858083	5.8	1.44	0.12	1.07	52.7	0.88	38	40
Speckled Trout	NLF	Cook, Minn.	47.95	–89.8463	6.4	0.26	0.24	1.89	51.7	2.39	22	24
August	NLF	Lake, Minn.	47.762531	–91.608573	5.8	0.90	0.23	0.89	29.5	1.06	31	29
Intermediate	NLF	Keweenaw, Mich.	48.0304239	–88.7283577	6.7	70.80	0.44	0.97	26.8	1.22	32	25
Whittlesey	NLF	Keweenaw, Mich.	48.0058915	–88.707158	7.6	65.00	0.32	1.09	27.8	1.45	26	23
Little Trout	NLF	St. Louis, Minn.	48.396615	–92.522264	29.0	1.10	0.19	1.84	24.2	1.52	9	11
Locator	NLF	St. Louis, Minn.	48.540272	–93.005386	15.8	0.54	0.06	2.91	44.9	1.41	9	10
Nipisiquit	NLF	Lake, Minn.	47.355569	–91.247845	5.5	0.24	0.12	1.57	38.1	1.33	21	20
Tooth	NLF	St. Louis, Minn.	48.397123	–92.642813	13.1	0.24	0.08	2.71	49.2	1.45	12	12
Wallace	NLF	Keweenaw, Mich.	48.057148	–88.627924	3.0	0.05	0.18	0.63	39.9	0.64	47	53
Mukooda	NLF	St. Louis, Minn.	48.334024	–92.488719	23.8	3.13	0.15	2.31	32.4	1.55	11	11
Ryan	NLF	St. Louis, Minn.	48.518566	–92.706795	5.2	0.15	0.20	1.24	43.6	1.24	29	30
Kjostad	NLF	St. Louis, Minn.	48.109206	–92.606389	15.2	1.68	0.13	2.37	41.6	1.97	14	15
Dunnigan	NLF	Lake, Minn.	47.70722	–91.630521	4.3	0.33	0.25	2.02	58.3	1.01	24	24

*Cumulative dry mass of lake sediment equivalent to ~ 10 years accumulation.
†Total unsupported decay corrected ^{210}Pb activity corresponding to regional cumulative dry mass values (0.2 g cm^{-2}, northern Minn. Lakes, and 0.9 g cm^{-2}, southern Minn. lakes).

tion (Omernik 1987). From south to north, the lakes are in the Western Corn Belt Plains (WCBP), Northern Glaciated Plains (NGP), North Central Hardwood Forest (NCHF), and Northern Lakes and Forests (NLF). The WCBP and NGP are cultivated, heavily agricultural landscapes with NGP having marginally more pasture. The NCHF is a transitional landscape with cultivated, pasture, urban, and forested regions, whereas NLF is overwhelmingly forested with coniferous and deciduous vegetation. The region's many glacial lakes follow a gradient of nutrient enrichment from north to south, because of differences in land use, vegetation, soil conditions, and lake size (Heiskary and Wilson 2008). In general, lakes in the south (WCBP and NGP) are shallower and eutrophic to hypereutrophic, whereas lakes in NLF are deep and oligotrophic to mesotrophic. In our study, maximum lake depths ranged from 0.65 m to 51.2 m, and lake surface-areas range from 0.02 km^2 to 234 km^2. Two additional lakes, Dunnigan and Kjostad, were cored at multiple locations (12 and 14 sites, respectively) as part of an earlier study of atmospheric mercury deposition to northern Minnesota (Swain et al. 1992; Engstrom et al. 1994). The latter two lakes were chosen for analysis of carbon burial based on their contrasting morphometry and size (Fig. 1). Given the range of lakes used in validating this method, we feel it is broadly applicable to many temperate lake types and sizes.

Sediment core collection

This method relies on the retrieval of undisturbed lake sediment cores with an intact sediment-water interface. In our validation of the method, we use a piston-type corer (Wright 1991) with a 2.75-cm polycarbonate core barrel operated from the lake surface by Mg-alloy drive rods. Cores were sectioned vertically in the field at 0.5 or 1 cm increments for the uppermost sediments (and more coarsely at depth), and stored in polypropylene jars for subsequent analysis of water and organic content and ^{210}Pb dating.

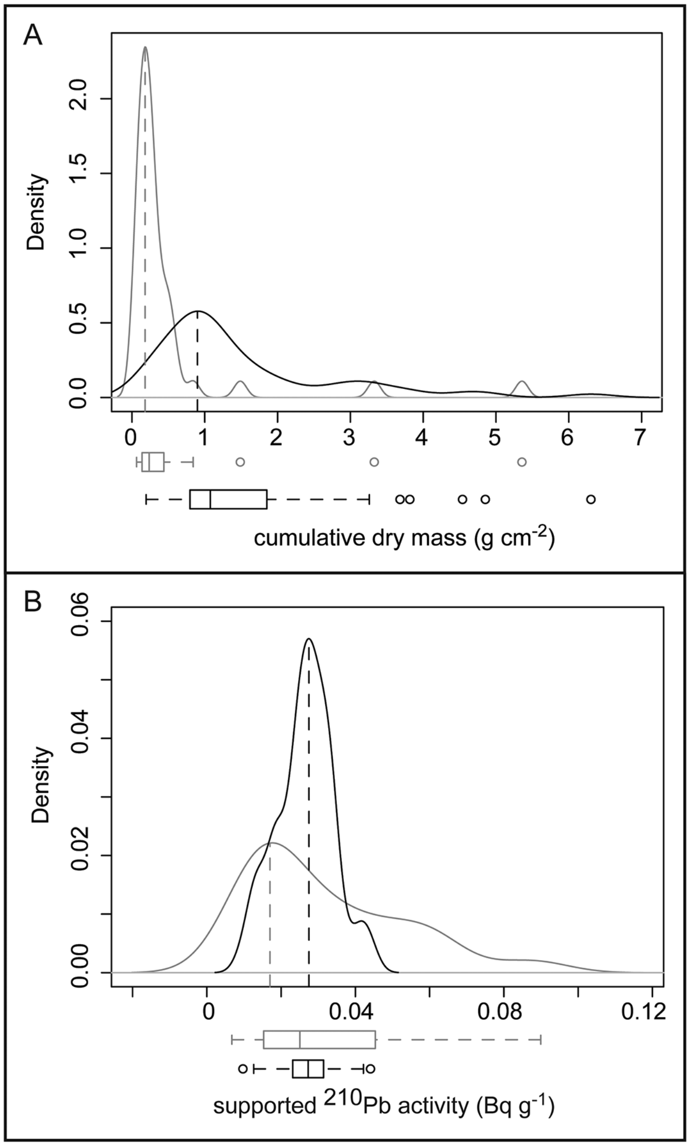


Fig. 1. The probability density functions of northern forested (gray) and southern agricultural (black) Minnesota lakes for (A) the cumulative dry mass of sediment, which corresponds to ~10-years accumulation, and (B) the supported ^{210}Pb activity in the lakes used in our dataset. Dashed lines represent the most probable values for northern and southern lakes, respectively: 0.2 g cm^{-2} and 0.9 g cm^{-2} cumulative dry mass, and 0.019 Bq g^{-1} and 0.028 Bq g^{-1} supported ^{210}Pb. Data are also summarized below each density plot with a boxplot showing the median and quartiles.

Gravity coring methods (Glew et al. 2001; Renberg and Hansson 2008) would be equally suitable for retrieval of short cores with an intact sediment-water interface, and operationally superior to piston coring if only surface sediments are needed. If the study goal is solely to quantify recent OC burial, an integrated surface interval representing ~10 years can be homogenized in the field (see methods below). However, we recommend subsampling at 0.5-1.0 cm, in the eventuality that greater temporal resolution or further analysis may be warranted in the future.

Organic carbon

The organic fraction of the lake sediments (%OM) was quantified through combustion at 550°C (Heiri et al. 2001) and the percent OC calculated using the equation, %OC = %OM • (12/30) from Rosén et al. (2010). Sediment organic carbon can also be measured directly with a dedicated carbon analyzer, which avoids interferences from other hydrated sedimentary materials (e.g., clays) and is more accurate.

Lead-210 dating

This method, for use on surface sediments, was developed using fully detailed chronologies from 93 sediment cores, established by conventional ^{210}Pb dating. Selected sediment samples (15-25 per core) were analyzed for ^{210}Po, the granddaughter product of ^{210}Pb, by α spectrometry and isotope dilution with ^{209}Po. Samples were pretreated with concentrated HCl and the Po isotopes distilled at high temperature (500°C) and plated directly onto silver planchets (Eakins and Morrison 1978). Activity was measured for 1-7 days with an Ortec α spectrometry system. The mean supported (background) activity for each core was derived from the asymptotic activity at depth and subtracted from total activity to calculated unsupported (excess) ^{210}Pb. Dates and sediment accumulation rates were established for each core using the constant rate of supply (c.r.s.) model (Appleby and Oldfield 1978).

For validation of the surface-sample method, we numerically integrated data representing the most recent ~10 years of accumulation post hoc based on the full-core dating and cumulative sediment dry mass. The decay-corrected ^{210}Pb activity of this integrated interval ($^{210}Pb_{site}'$) was calculated according to

$$^{210}Pb_{site}' = {}^{210}Pb_{site} \bullet e^{(k \bullet \frac{t}{2})} \qquad \text{(Eq. 1)}$$

where $^{210}Pb_{site}$ is the measured unsupported ^{210}Pb, k is the ^{210}Pb decay constant (0.03114 y^{-1}), and t is the estimated age at the base of the integrated sediment interval, calculated using modeled sediment accumulation rates. We chose a 10-year integration period in part to overcome short-term variability in burial rates and an undue influence of early diagenetic alteration of recently deposited sediments (OC mineralization). This diagenetic loss of OC occurs largely within the first 5 years of deposition (Gälman et al. 2008). Conversely, the 10-year integration period is short enough that the lakes have likely not experienced dramatic changes affecting OC production (e.g., eutrophication; Heathcote and Downing 2012). It also necessitates a relatively small decay correction (17%), which limits the error associated with estimating sediment age for undated surface cores.

Our use of fully dated cores to validate the surface-sample method provided information that would not normally be available in any subsequent study applying this method to undated lakes. In particular, we knew a priori sediment cumulative dry mass corresponding to 10 years as well as supported ^{210}Pb established from core depths > 150 years of age. In the absence of sedimentation rates for the lakes under investigation, dating from a small number of similar lakes from the same region could be used to estimate a suitable cumulative dry mass corresponding to ~ 10 years. We emulate this approach in our validation of the method by establishing a representative 10-year cumulative dry mass for northern Minnesota lakes (Northern Lakes and Forest ecoregion) and southern lakes (Northern Great Plains, Western Corn Belt Plains, and North Central Hardwood Forest ecoregions). Using a probability density function to describe the cumulative dry mass for the two regions yielded a value of 0.2 g cm^{-2} for northern lakes and 0.9 g cm^{-2} for southern lakes (Fig. 1). In doing so, the resulting error associated with ^{210}Pb decay correction for 90% of the lakes in our dataset was < 23% for northern lakes and < 14% for southern lakes. We suggest that this information can be used for small glacially formed lakes in similar ecoregions.

Regional estimates of supported ^{210}Pb can also come from other lakes if ^{210}Pb is measured by α spectrometry. Supported ^{210}Pb is typically a small portion of total ^{210}Pb in recently deposited sediments, 3 ± 2% for northern lakes and 14 ± 6% for southern lakes in our dataset. Using the probability density function to describe regional supported ^{210}Pb gave activities of 0.02 Bq g^{-1} for northern lakes and 0.03 Bq g^{-1} for southern lakes (Fig. 1). Alternatively, supported ^{210}Pb (as well as total ^{210}Pb) can be measured directly by γ spectrometry (as ^{214}Pb or ^{214}Bi) (Appleby 2001).

Calculations

The mean lake-wide burial (flux) of organic carbon in the sediments of a lake, $F(OC)_L$ (g m^{-2} y^{-1}) is equal to the concentration of organic carbon in the surface sediments $[OC_S]$ (g • g^{-1}) multiplied by the sediment mass accumulation rate at the sampling site R_S (g m^{-2} y^{-1}) and a correction term for sediment focusing ($1/ff_{OC}$):

$$F(OC_L) = [OC_S] \bullet R_S \bullet \frac{1}{ff_{OC}} \qquad \text{(Eq. 2)}$$

Focusing is defined here as the ratio of the site-specific rate of OC burial to the rate for the lake as a whole and can be approximated by the focusing factor for ^{210}Pb,

$$ff_{^{210}Pb} = \frac{F(^{210}Pb_{site})}{F(^{210}Pb_{atm})} \qquad \text{(Eq. 3)}$$

where $F(^{210}Pb_{site})$ is the site-specific flux of unsupported (excess) ^{210}Pb and $F(^{210}Pb_{atm})$ is the atmospheric flux of excess ^{210}Pb (both Bq m^{-2} y^{-1}). Combining Eqs. 2 and 3, and decay-correcting for 10 years of accumulation using Eq. 1, then rearranging terms,

$$F(OC_L) = [OC_S] \bullet F(^{210}Pb_{atm}) \Big/ \frac{F(^{210}Pb_{site}')}{R_S} \qquad \text{(Eq. 4)}$$

we note that $F(^{210}Pb_{site}')/R_S$ equals the decay-corrected activity (concentration) of excess ^{210}Pb in surface sediments at the sample site $[^{210}Pb_S]$ (Bq g^{-1}), and thus

$$F(OC_L) = \frac{[OC_S]}{[^{210}Pb_S]} \bullet F(^{210}Pb_{atm}) \qquad \text{(Eq. 5)}$$

In this derivation, the mean lake-wide burial of organic carbon is simply the ratio of organic carbon to ^{210}Pb measured in a surface sediment sample multiplied by the atmospheric flux of ^{210}Pb for the region in which the lake is located.

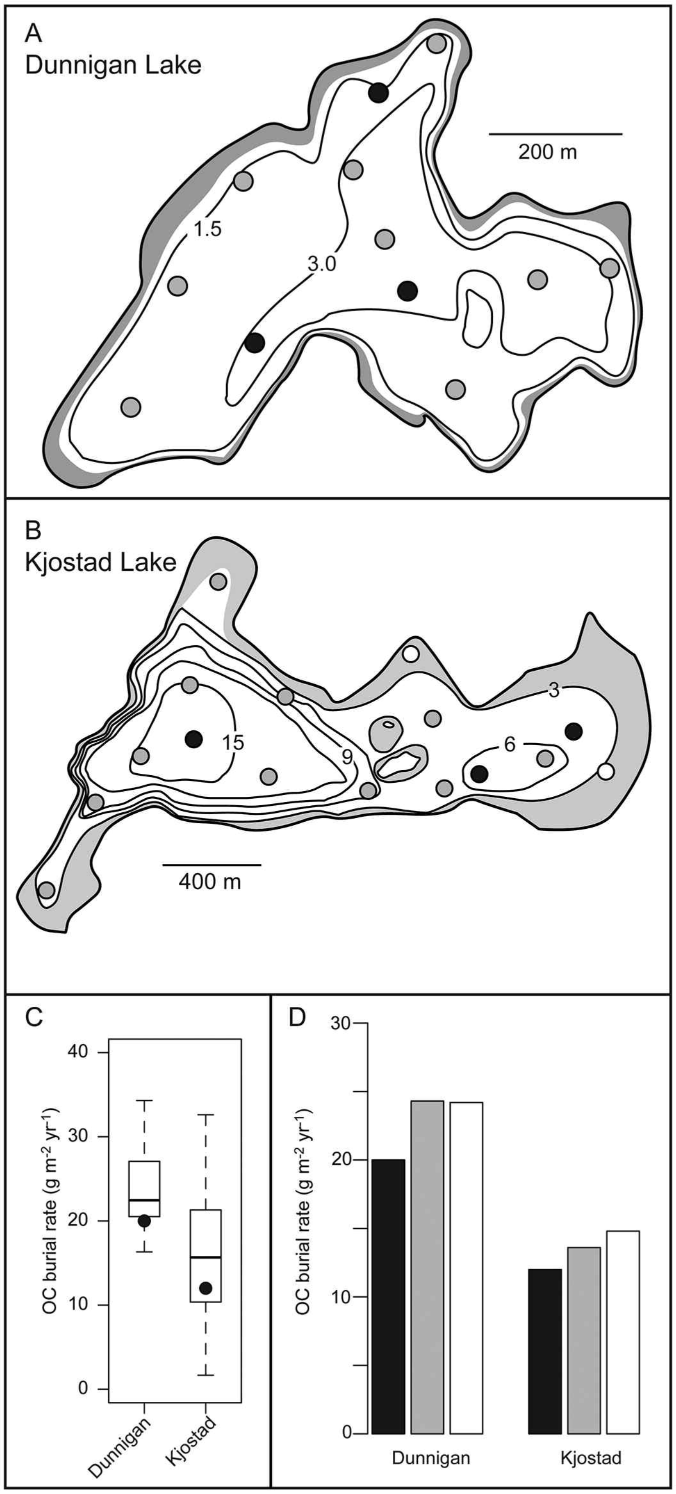
The same equation (Eq. 5) was originally proposed by Binford and Brenner (1986) to estimate the trophic state of a lake based on the degree of ^{210}Pb dilution by organic matter in surface sediments. What these authors did not highlight at the time is that this simple model actually accounts for sediment focusing and that the resulting flux approximates a lake-wide average, rather than a core-specific value. This outcome is incredibly powerful because it removes a major impediment to comparing sediment fluxes among lakes based on the analysis of a single-core—that is, the spatial variation in sediment deposition across a lake basin.

There are several critical assumptions inherent in this model, which are discussed at length in a subsequent critique (Benoit and Hemond 1988) and response (Binford and Brenner 1988) to the original publication. Most important among these are that (1) the atmospheric flux of ^{210}Pb is known with some level of certainty, (2) organic matter (or any sediment constituent of interest) is focused to about the same degree as ^{210}Pb, and (3) direct atmospheric deposition is the primary source of ^{210}Pb to the lake. We review these assumptions in detail, and present study results supporting them elsewhere in the paper.

Assessment and discussion

Multiple-core validation, whole-lake accumulation

Whole-lake OC burial rates were assessed in detail for two lakes (Dunnigan and Kjostad; Fig. 2) using an area-weighted approach based on multiple sediment cores (12 and 14 cores, respectively). The cores, collected as part of an earlier study on mercury loading to northern Minnesota lakes (Swain et al. 1992; Engstrom et al 1994), were assigned depositional areas of the lake basin based on nearest-neighbor topology and summed to the OC accumulation (the product of OC concentration and sedimentation rate) for the whole lake (Fig. 2). The resulting OC burial rates were significantly different between the lakes (P = 0.01; Fig. 2C), with Dunnigan showing higher lake-wide values than Kjostad, but lower within-lake variability. These differences were largely explicable in terms of basin size and morphometry. Dunnigan Lake occupies a small (0.33 km^2), shallow, flat-bottomed basin with a single area of sediment deposition and a proportionally small catchment (0.46 km^2). The sediment focus factors for the core sites range from



0.62 to 1.24, whereas the range of OC accumulation rates was 16 to 34 g m^{-2} y^{-1}. In contrast, Kjostad Lake has a more complex morphology, with two depositional basins separated by an island, and a large catchment (9.85 km^2). The sediment focus factors range from 0.28 to 1.98, and the range of OC accumulation for the core sites was 1.8 to 33 g m^{-2} y^{-1}. The whole-lake accumulation of OC was 20 g m^{-2} y^{-1} for Dunnigan and 12 g m^{-2} y^{-1} for Kjostad (Fig. 2D).

These whole lake, multiple-core rates can be used to assess the accuracy of OC burial rates determined from a single dated core in the center of the main depositional basin corrected for sediment focusing to the core site. As discussed previously, the focusing correction for each lake is calculated as the ratio of unsupported ^{210}Pb flux measured at the core site to the measured atmospheric fallout for the region. In the case of Minnesota lakes, there are two National Atmospheric Deposition Program (NADP) sites where the annual atmospheric ^{210}Pb flux is known for a 27-month period (2003-2005), one in the northeastern part of the state (MN16, Marcell Experimental Forest; 211 Bq m^{-2} y^{-1}; 47.531°N, 93.469°W) and another in the southwest (MN27, Lamberton; 181 Bq m^{-2} y^{-1}; 44.237°N, 95.301°W) (Lamborg et al. 2012). Using a single core from the central location, the resulting OC burial rates were very comparable to the whole-lake rates calculated using multiple cores (Fig. 2D; Dunnigan, 24 g m^{-2} y^{-1}; Kjostad, 14 g m^{-2} y^{-1}). Thus a single, well-dated sediment core from the central area of the lake basin can, when focusing-corrected, provide a reliable measure of whole-lake OC accumulation.

Finally, the OC burial data from the multiple-core lakes provided a first-order appraisal of the accuracy of calculating modern OC burial rates from single surface samples. Using the same core locations as the focus-corrected single-core sites and integrating surface samples representing a cumulative dry mass of 0.2 g cm^{-2} (Minn. northern lakes) or approximately 10 years of sediment accumulation, we found very similar OC burial rates compared with the measured whole-lake values (Fig. 2D; Dunnigan, 24 g m^{-2} y^{-1}; Kjostad, 15 g m^{-2} y^{-1}). This result is also encouraging as it supports our assumption that sediment focusing is accounted for, because of the fact that OC and ^{210}Pb are focused in a similar manner. Indeed, there was a similar ratio of OC : ^{210}Pb (g Bq^{-1}) at multiple sites across the main depositional area of the two lake basins. We found that this ratio varied (expressed as 2 standard errors relative to the mean) by 9% in Dunnigan Lake and by 26% in Kjostad Lake, the latter being higher as it reflects two depositional basins.

Fig. 2. Multiple sediment-core locations in two morphologically distinct lake basins in the Superior National Forest, Minnesota: (A) Dunnigan Lake with a simple, single basin and (B) Kjostad Lake, Minnesota, with two depositional basins and an island. In both (A) and (B), black circles show the locations of cores dated in detail, gray circles are coarsely dated cores, and white circles are cores not included as they fall in the erosional zone of the lake (gray shaded area) (from Engstrom et al. 1994). Bathymetry contours are in meters. (C) A box plot of OC burial rates for each lake showing the median (black line), quartiles, and a significant difference in mean (black circle) burial rates between the lakes. (D) A bar plot of whole lake OC burial rates calculated by area-weighted accumulation of multiple cores (black), a single sediment core from the central part of the basin, dated in detail and adjusted for sediment focusing (gray), and the predicted burial using the surface-sample approach (white).

Single core validation, whole-core accumulation

Using our dataset of 93 lakes, we compared OC burial rates in fully dated single cores with burial rates estimated by the surface-sample approach. The modern OC burial rate across the lakes, adjusted by ^{210}Pb flux for sediment focusing, varied from 9 to 318 g m^{-2} y^{-1} (Fig. 3A). Regional estimates of cumulative dry mass for northern and southern lakes, surface ^{210}Pb activities (decay-corrected to 10 y) and OC concentrations were used to calculate modern OC burial rates according to Eq. 5 (Fig. 3A). These single-sample estimates do a very reliable job of describing whole-core OC burial measured for each lake. This validation step relies on common parameters between the predicted and observed values, however the derivation of both $^{210}Pb_{atm}$ and $^{210}Pb_{site}$ is sufficiently different so as not to consider the comparison of OC burial rates a circular relationship. The observed whole-core values of modern OC burial rely on (1) a measured whole-core ^{210}Pb inventory to derive ^{210}Pb flux, which is used to explicitly correct for sediment focusing; (2) a site-specific sediment accumulation derived from the c.r.s model; (3) a $^{210}Pb_{site}$ which is decay-corrected using site-specific accumulation. The predicted single-sample estimates rely on (1) a regional $^{210}Pb_{atm}$ and make no assumptions about site-specific ^{210}Pb flux to the core site; (2) a regional estimate of modern (~10 y) cumulative dry mass accumulation; (3) a regional decay correction $^{210}Pb_{site}$. We therefore feel that there are sufficient differences between how the OC burial values are derived that this validation step demonstrates single-samples can reliably estimate whole-core OC burial. Based on the residuals of the estimated values from the 1:1 line as a percentage of the measured OC burial, we estimated a % error for OC burial from the surface-sample approach. The probability density distribution of the residuals suggested that 6.5% is a suitable inter-lake predictive error, where the 75th percentile was 24% error (Fig. 3c).

As an additional validation of the surface-sample approach, we re-cored and independently analyzed nine of the original study lakes (Fig. 3). The sediments from the additional lakes were collected in the vicinity of the original core site, somewhere near the center of the depositional basin, as is common practice. The OC burial rates for the repeat cores were calculated using our Eq. 5 (Fig. 3). This cross-validation of the estimated OC burial values yielded a root mean square error of prediction of 1.9 g m^{-2} y^{-1}. We therefore conclude that single surface-sediment samples can be used to provide reliable estimates of whole-core OC burial.

Method assumptions and applicability

Our surface-sample approach, originally proposed by Binford and Brenner (1986) to assess trophic state, requires the acceptance of a number of assumptions (Benoit and Hemond 1988). Whereas some concerns have been addressed (Binford and Brenner 1988), a lack of data at the time prevented explicit validation for the main assumptions. Here, we revisit some of these original concerns and defend the assumptions made in using a single sediment sample.

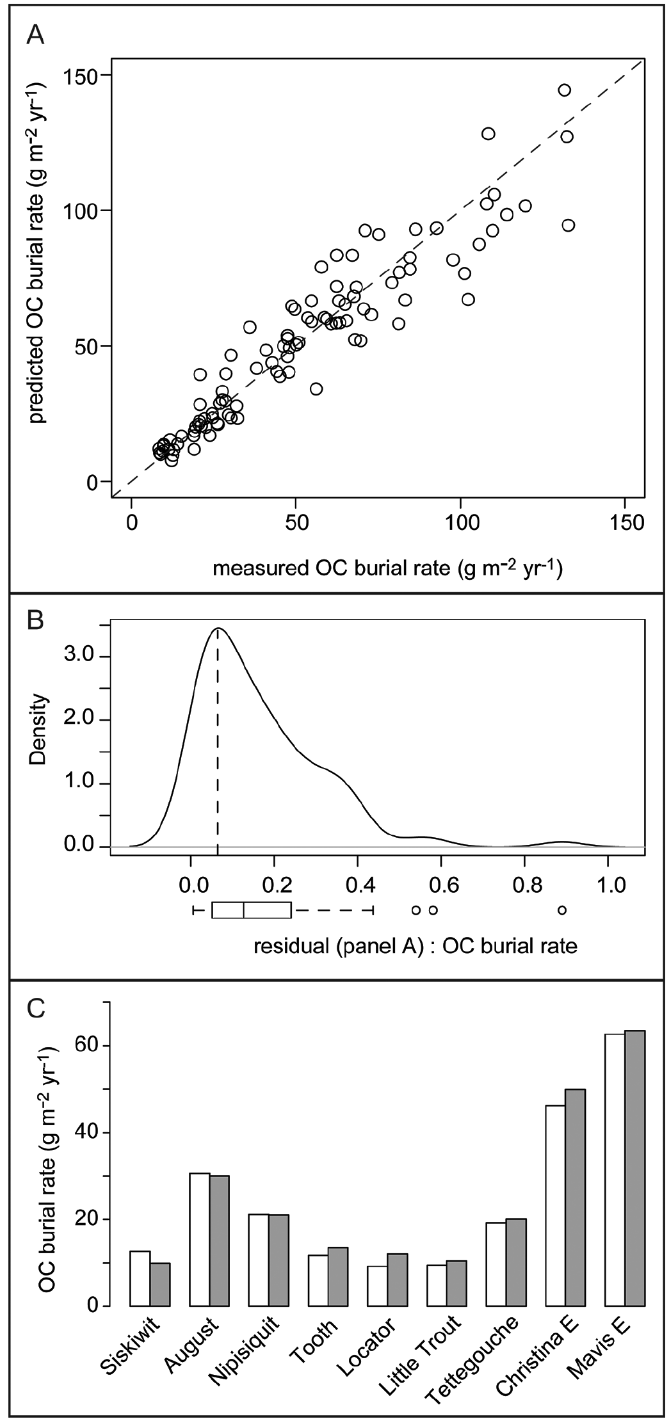


Fig. 3. (A) A scatterplot of the estimated OC burial rates using the surface-sample approach against the measured values based on fully dated cores over a 10-y period, adjusted for sediment focusing. (B) The density probability of the residuals from the 1:1 line of plot (A), expressed as a percentage of the measured value with the highest probable error shown as a dashed line. A boxplot of the data are shown below the probability plot, where the 75th percentile is 24% error (C). Repeat sediment cores were collected and independently analyzed in 9 of the lakes (4 to 10 years apart); OC burial rates of the repeat samples (gray bars) are calculated using the surface-sample approach.

A key condition of Binford and Brenner's (1986) formulation was the use of an estimated global average for the atmospheric ^{210}Pb flux, a value of 185 Bq m^{-2} y^{-1} (Turekian et al. 1977; Appleby and Oldfield 1983; Binford and Brenner 1988; Preiss et al. 1996). The range of published atmospheric ^{210}Pb fluxes spans almost an order of magnitude (30–200 Bq m^{-2} y^{-1}) and is a function of available landmass, climatic factors, and orographic influence. Whereas we now have a better understanding of the spatial variation of atmospheric ^{210}Pb flux in the Northern Hemisphere (Graustein and Turekian 1986; Binford and Brenner 1988; Preiss et al. 1996; Appleby 2008; Baskaran 2011; Lamborg et al. 2012), these data remain incomplete for many other parts of the world. Specific to our study, we have atmospheric ^{210}Pb flux measured from two sites in northeastern and southwestern Minnesota (Lamborg et al. 2012). In the absence of site-specific deposition data, an educated estimate based on published values is defendable in the Northern Hemisphere, acknowledging that there appears to be a west to east increase in fallout over the mid-latitudes of the major continents (Appleby 2008). Studies in polar and Southern Hemisphere regions should employ published values for these areas; because the smaller available land mass and high-latitude aridity will limit ^{222}Rn emanation and ^{210}Pb fallout (Appleby et al. 1995; Appleby 2008; Ribeiro Guevara et al. 2003). In the absence of published deposition data, ^{210}Pb-dated cores on a lake (or multiple lakes) in the region of interest can provide a robust estimate of the regional atmospheric ^{210}Pb flux (Fitzgerald et al. 2005). We caution against establishing ^{210}Pb flux from multiple soil profiles (Nozaki et al. 1978; Benoit and Hemond 1988), as it introduces uncertainty associated with local ^{210}Pb scavenging by terrestrial vegetation (Stankwitz et al. 2012) as well as down-slope redistribution of ^{210}Pb within the catchment.

A second assumption is that ^{210}Pb sedimentation should be linearly proportional to atmospheric deposition in order for the method to accurately describe sedimentation. This concern is based primarily on the understanding that sediment is focused within a lake basin, which affects the total ^{210}Pb inventory at the core site. We discussed earlier our contention that no additional correction is needed for focusing because it is explicitly included in the calculation (Eq. 5). Furthermore, results from our two multiple-core lakes demonstrated that the OC : ^{210}Pb ratio across the depositional basin remains relatively uniform because OC and ^{210}Pb are focused in a similar manner. Similar focusing likely results from the association of ^{210}Pb with fine-grained sediment particles, including organic seston. However, OC from littoral production and particulate carbon from terrestrial sources might focus differently than ^{210}Pb because of spatially non-uniform loading, especially in complex lake basins with multiple embayments (Bindler et al. 2001; Engstrom and Rose 2013). Nonetheless, the relative uniformity of OC:^{210}Pb ratios noted above suggests that both constituents are redistributed to the depositional region of a lake in roughly the same proportions.

A final assumption is that nonatmospheric inputs of ^{210}Pb to the lake are negligible. This condition is generally true except in cases where catchment erosion rates are very high and the soil residence time of ^{210}Pb is short relative to its half-life (22 y). It is evident to us from studies within Minnesota and elsewhere that the overwhelming input of ^{210}Pb to lakes is atmospheric (Binford and Brenner 1988; Engstrom et al. 1994; Schottler et al. 2010). We do acknowledge that in lake systems with significant fluvial inputs the delivery of ^{210}Pb from the catchment may be significant and the retention of ^{210}Pb inputs may be incomplete (Cornett et al. 1984; Benoit and Hemond 1987). However, none of the lakes in our dataset represent systems that we would consider to have significant fluvial inputs, nor do they have very short hydraulic residence times (e.g., Dunnigan and Kjostad lakes have a residence time of 5 years; Swain et al. 1992). As the dataset of lakes used in this study covers a broad range of morphometric and trophic conditions over several biogeoclimatic zones (Table 1), we conclude that the surface-sample approach is broadly applicable to many temperate lake types and sizes.

Comments and recommendations

The obvious appeal of estimating modern OC burial using a single sediment sample is the increased efficiency and decreased cost of sample collection and analysis with greater potential spatial coverage. Applying this technique to large datasets provides a clearer picture of the spatial trends of OC burial in inland waters, whereas single-lake studies are more susceptible to the spatial variability that exists from lake to lake. This approach also allows a more accurate definition of spatial variability and the error associated with measuring OC burial in lakes. The within-lake variability (standard deviation) from our multiple core studies was ~4 g m^{-2} y^{-1}, while regionally an estimate of error for the method was 6.5%.

This method is broadly applicable to different lake types and offers a more accurate assessment of the role inland waters play in the global carbon cycle (*sensu* Cole et al. 2007; Tranvik et al 2009). As an example, there is a growing global dataset on OC burial as a function of lake area, which suggests smaller lakes bury more OC (Fig. 4). We find that this relationship holds when this dataset is populated with our results, providing confidence on the applicability of this method to large regional studies. The broader application of this method requires (1) measured or published data of atmospheric ^{210}Pb flux, (2) analysis of supported ^{210} Pb by γ spectrometry or a regional estimate of supported ^{210}Pb from cores previously dated by α spectrometry, and (3) an understanding of typical sedimentation rates in lakes from the study region to establish a realistic cumulative dry mass over which to integrate the sample (representing ~ 10 y). For many regions, these ancillary data already exist in the literature. In addition to clarifying the role of inland waters in the global C-cycle, this technique applied over large spatial gradients of land-use and water quality will allow us to address questions on the role that terrestrial–aquatic linkages play in the sequestration of OC by inland waters.

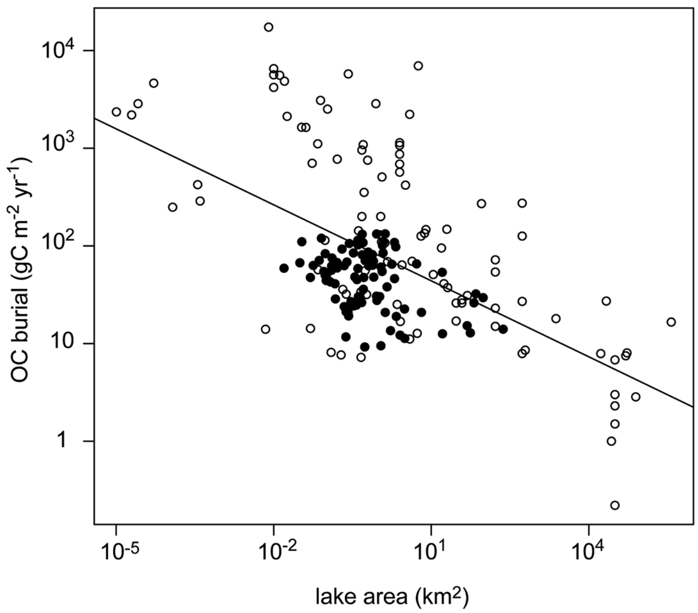


Fig. 4. A scatterplot of OC burial as a function of lake area for $n = 193$ lakes and reservoirs throughout the globe, including those from this study (solid circles) (Mulholland and Elwood 1982; Sobek et al. 2009; Downing et al. 2008; Heathcote and Downing 2012). A statistically significant linear model fit is shown with an adjusted r^2 of 0.33 and df = 189.

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