Biogeochemistry **23**: 35–51, 1994.

Pre-industrial particulate emissions and carbon sequestration from biomass burning in North America

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Received 13 September 1993; accepted 22 November 1993

Key words: biomass burning, carbon sequestration, charcoal, fire emissions

Abstract. Spatial trends in pre-industrial biomass burning emissions for eastern North America were reconstructed from sediment charcoal data. Petrographic thin sections were prepared from varved lake sediments along a transect of sites extending from NW Minnesota eastward to NE Maine. Results showed an exponential decline in charcoal abundance with distance east from the prairie/forest border. This result quantifies burning along the broad climate/vegetation gradient from xeric woodland to mesic eastern deciduous forest. Post-settlement charcoal accumulation showed no such geographic pattern, varying from site-to-site, likely reflecting local variability in land use and combustion sources. Results suggest the total emissions of large (> 10 μm diameter) charcoal particles decreased by a factor of three during the twentieth century.

Introduction

Seiler & Crutzen (1980) demonstrated that large quantities of C might be sequestered during biomass burning in relatively inert elemental form. Goldberg (1985) calculates that, failing oxidation losses, Seiler & Crutzen's (1980) estimated rates of C sequestration during biomass burning would tie up all of the globe's surface C in less than 100,000 yr. Such estimates are difficult, because they involve scaling small numbers with high uncertainty to large regions. Radke et al. (1978) point out that our best estimates of particulate emissions from US forest fires span a range that overlaps with the estimated global anthropogenic particulate emissions. These observations reveal two important facts about emissions from biomass burning, a) they are massive, and b) the estimated rates are highly imprecise.

Significant amounts of relatively stable charcoal and other combustion products are released during biomass burning. Seiler & Crutzen (1980) concluded that C sequestered in inert form might represent greater than 20% of the current total anthropogenic C in charcoal (e.g., Shneour 1966; Herring 1984) making this a potentially important sink (Seiler & Crutzen 1980; Robinson 1989; Crutzen & Andreae 1990). Particulates in smoke emissions affect visibility (Wolff et al. 1981), radiation budgets and, therefore, climate

(Rosen et al. 1984; Ch'ylek et al. 1984). Also of concern are aerosol-bound carcinogens, polycyclic aromatic hydrocarbons (PAH's), produced by combustion of wood and fossil fuel (Gschwend & Hites 1981). Finally, C sequestration in elemental form affects the cycling of other atmospheric constituents, notably oxygen (Budyko 1987; Robinson 1989). The long-term consequences of C sequestration on atmospheric O_2 with implications for fire behavior (Robinson 1989) and evolution of vertebrate body size and physiology (Budyko 1987) makes it necessary to begin exploration of the role of fire and the C cycle from time scales of 10^0 to 10^6 yr (Clark & Robinson 1993). Despite calls by atmospheric scientists for baseline (pre-land clearance, pre-industrial) rates of C sequestration during fire (Malingreau et al. 1993), Seiler & Crutzen's (1980) review remains the principle source for these estimates.

Fortunately, lakes continuously "sample" the atmosphere, integrate spatial variability in deposition patterns of many constituents, and deposit and preserve samples in orderly sequences (Wreight 1982). Many landscapes provide spatial arrays of lakes that can be sampled at a variety of spatial scales to produce continuous records of atmospheric deposition spanning years to millennia. The products of combustion that are preserved and can be quantified in lakes include fossil charcoal. Such records provide opportunity for determining how emissions have changed through the past.

We used charcoal analysis of varved (annually laminated) lake sediments to determine geographic patterns of pre-industrial charcoal accumulation in some different vegetation types of eastern North America and to determine how those patterns changed sine A.D. 1900, with recent land clearance, fire suppression, altered ignition patterns, and industrialization. Our sites span from near the prairie/forest border in NW Minnesota eastward through eastern Maine. Our network of sites includes a gradient in climate, vegetation, and modern fire importance. Results are a first step toward establishing baseline estimates of emissions from pre-industrial time as perspective for evaluating modern particulate measurements in the atmosphere.

Study areas

Sites were selected to provide a high temporal resolution of emissions across the climate and vegetation gradient of eastern North America from pre- vs post-industrial time (Fig. 1). High temporal resolution was achieved through use of varved sediments (Swain 1973). Presettlement time was identified using varve counts that were confirmed by pollen indicators of land-clearance (Brugam 1978). All lakes range from 3 to 15 ha in surface area and are from 9 to 34 m deep. We selected sites known to have varves, so more detailed site descriptions are contained in previous publications referenced under individual sites.

NW Minnesota lakes (47°10'N, 95°10'W, elevation 464 m). Charcoal and

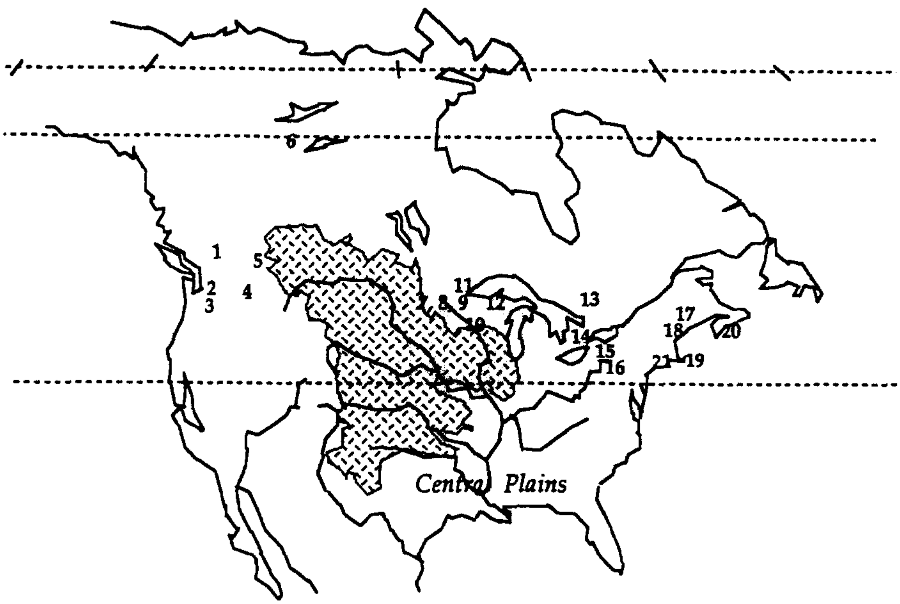


Fig. 1. Sites investigated in this study (7, 8, 9, 10, 12, 14, 15, 16, 17, 18) and others mentioned in the text. Site names are listed in Table 1.

pollen analyses of three kettle lakes in ground moraine were completed by Clark (1988b, 1990). The canopy is dominated by a mixture of pines (*Pinus resinosa, P. strobus*), early successional hardwoods (*Betula papyrifera, Populus* spp.), and *Abies balsamea*. The understory contains dense *Corylus* thickets in places. Logging in the region progressed from the east during the late 19th and early 20th century. Widespread cultivation in the nearby Red River Valley also began at the time. The catchments of these lakes were never cleared for farming. Selective logging of the large pines occurred early in the century, and fire suppression became effective by A.D. 1915.

Hell's Kitchen, WI (46°11′N, 89°42′W, elevation 500 m). Pollen and charcoal analyses were completed by Swain (1978). Because previous charcoal data are reported as pollen: charcoal they could not be used in this analysis. The tree canopy is dominated by early successional hardwoods (*Betula papyrifera, Populus* spp., northern hardwoods (*Acer saccharum, Betula lutea*), and conifers (*Abies balsamifera, Tsuga canadensis, Thuja occidentalis, Pinus strobus, P. resinosa, P. banksiana*). Logging of *Pinus strobus* and *Tsuga canadensis* occurred from 1890 to 1920. Today the area is forested with scattered houses.

Crawford Lake, Ontario (44°N, 80°W, elevation 279 m). Pollen analysis was completed by McAndrews (McAndrews & Boyko-Diakonow 1989). They interpret pollen and archaeological evidence from the lake catchment to suggest forest clearance by cutting and burning about A.D. 1360 followed by Indian agriculture until A.D. 1660, when forests were invaded by *Populus, Quercus* and *Pinus strobus,* dominant species today. Eurocanadian cultivation became widespread after A.D. 1800. The catchment now supports a forest, a reconstructed Iroquois village, and cleared fields.

Devil's Bathtub, NY (43°N, 78°W, elevation 550 m). A detailed analysis of the full Holocene record of this kettle lake is presented elsewhere (Clark et al., in prep.). Devil's Bathtub lies at a vegetation transition depicted by reconstruction of pre-settlement forests (Seischab 1990). On till plains to the north were northern hardwoods, dominated by *Fagus grandifolia, Acer saccharum,* and *Tilia americana.* On the Allegheny Plateau to the south were mixed hardwoods, containing a more diverse mixture of these northern hardwoods with *Quercus* spp, *Tsuga canadensis,* and *Pinus strobus.* The catchment is today forested and dominated by *Quercus* spp., which probably followed cutting, and an understory of *Acer saccharum* and *Fagus grandifolia.*

Ely Lake, PA (41°46′N, 75°50′W, elevation 384 m). Pollen analysis of this basin in ground moraine was completed by Gajewski et al. (1987). The canopy includes a mixture of *Tsuga canadensis, Quercus* spp., and northern hardwoods (*Fagus grandifolia, Betula lutea, Acer saccharum*), which likely dominated in presettlement time. The region witnessed several waves of cutting following settlement (Whitney 1990). Clearing for agriculture became important after A.D. 1800. *Tsuga* is among the most abundant canopy species in the lake catchment, which also contains much cleared pasture.

Basin Pond, ME (44°28′N, 70°3′W, elevation 125 m). Pollen analysis was completed by Gajewski et al. (1987). The lake is surrounded by mixed hardwood/conifer forest typical of the region, including *Fagus grandifolia, Betula papyrifera, B. lutea, Acer saccharum, Tsuga canadensis, and Pinus strobus.* Catchment forests have been logged in the past.

Conroy Lake, ME (46°17′N, 67°53′W, elevation 140 m). Pollen analysis was completed by Gajewski et al. (1987). The lake catchment contains *Betula papyrifera, Picea glauca, P. rubens, Abies balsamea,* and *Pinus strobus.* Northern white cedar (*Thuja occidentalis*) fringes the lake itself. Regional forests are spruce-fir (*Picea glauca, P. rubens, Abies balsamea*), with northern hardwoods (*Fagus grandifolia, Betula lutea, Acer saccharum*) on mesic sites. The catchment is mostly forested, but includes several houses. Cleared fields are common in the region.

Methods

Charcoal analysis (to determine past emissions) and varve counts (to establish accumulation rates) were completed on petrographic thin sections of lake sediments (Clark 1988a). Sediment profiles containing the last 750 to 2000 yr were obtained by two methods. A 1.5 m long frozen slab of the flocculant surface sediments was extracted using an aluminum box filled with dry ice and kept frozen until preparation of thin sections. A piston corer was used to obtain longer cores from each of the same lakes (Wright et al. 1984).

Petrographic thin sections (Merkt et al. 1971) of lake sediments were prepared on contiguous 5-cm long by 2-cm wide sections of sediment. Samples were dehydrated in acetone, embedded in epoxy resin, and sectioned petrographically (Clark 1988a). Laminations were identified and marked directly on thin sections. The annual nature of laminations was confirmed in all cases using combinations of pollen indicators of agricultural activity, other sediment indicators of fossil fuel combustion, and/or ^{14}C chronologies (Clark et al., in prep). Pollen analysis, to confirm varve chronologies, was completed by us (Clark 1988a) or in separate studies of these sites (see **Study areas**).

Charcoal was quantified at 63× on a binocular microscope. Charcoal particles are taken as those that are angular and entirely opaque (e.g., Harris 1958; Cope & Chaloner 1985; Goldberg 1985; Clark 1988a). Charred wood and leaf tissue are readily discernible from the combustion products of fossil fuels, such as fly ash (Clark 1988a). Counts were completed along each varve. Particle geometric mean areas were determined using a calibrated reticule, values were scaled to correct for magnification, summed for each varve, and the total divided by the width of the section counted. These calculations yield mm^2 charcoal per mm^2 sediment per year ($mm^2\ mm^{-2}\ yr^{-1}$). Scaling by 100 gives the charcoal index C (percent) reported by Clark (1988a, 1990).

Comparison with emission estimates from measurements on experimental and wildland fires requires an estimate of charcoal mass. Conversion of area estimates (from paleo studies) to mass (from emissions studies) requires estimates of charcoal volume and charcoal density. The volume estimate was obtained by use of a correction factor to account for the preferential orientation of particles with thin axes parallel to the sediment bedding plane. A simple stereology argument would determine volume as the cube of particle diameter (Clark 1988a). In fact, the nonrandom shape and therefore arrangement of particles with respect to the angle of observation must upwardly bias the estimate of actual charcoal amount. Charcoal particles span a great range. Average length-to-breadth ratios are 1.9 (Harris 1958) to 2.0 (our measurements, Clark, in prep). Our calculation of particle volume assumed length-to-breadth ratio of 2.0. The density of charcoal particles ranges from about 0.4 to 0.7 g cm^{-3}, depending mostly on porosity (Sander & Gee 1990) and less on the relative fractions of organic and elemental C (Goldberg 1985). We assumed a density of 0.5 g cm^{-3} in our calculations to transform area estimates to mass fluxes. Errors associated with these transformations are mentioned in the **Discussion** section and considered in detail by Clark (in prep).

Conversion to common units of mass flux allowed us to compare ours and previously published charcoal data analyzed by other methods. We surveyed published literature for charcoal studies from temperate and boreal North America. Only those studies that included accumulation rates of charcoal could be used for these comparisons. Studies reporting only sediment concentrations of charcoal could not be used, because accumulation rates of other sediment constituents vary within and among lakes. Nor could we use published ratios of charcoal: pollen in sediments. Those values depend on pollen production, which can vary greatly with vegetation composition (pines produce much more pollen than do many hardwoods, for instance). Values from these studies were converted to g $m^{-2} yr^{-1}$ in the manner described above.

Results

Time control of charcoal sequences

Annual laminations were confirmed in all but Devil's Bathtub by pollen analysis of agricultural indicators (Fig. 2). The time scale in Figure 2 is from varve counts. *Ambrosia* pollen, an invader of cleared fields, is so common in post-settlement lake sediments of the eastern United States that it alone is frequently used as a datable horizon. It typically increases after A.D. 1800 and remains high at least until A.D. 1900 (e.g., Brugam 1978). We used *Ambrosia* pollen to confirm that laminations in cores are annual and so could be used as basis for sediment age estimates. Increases in *Ambrosia* with clearance activities in recent sediments provide strong support for the chronology, because only near the sediment surfaces were sediments sometimes disrupted. Below the *Ambrosia* increases, laminations are distinct in all cores. Disruption of surface sediments of Devil's Bathtub made lamination counts tentative for the 20th century. That chronology is well dated with ten ^{14}C dates all of which agree well with over 10,000 lamination counts in the rest of the core (Clark et al., in prep).

Geographic patterns in presettlement charcoal

Presettlement charcoal accumulation varied systematically across the study transect. An example of two contrasting patterns of charcoal accumulation since A.D. 1500 is included in Figure 3. The relatively high values of northwest Minnesota (Fig. 3a) show extreme variability resulting from individual fires and from changes in fire regime that attended climate fluctuations of the last several centuries (Clark 1990). Charcoal accumulation declines abruptly with 20th century fire suppression. Crawford Lake, southern Ontario (Fig. 3b), shows low values throughout, but especially before 1900. An exception is a significant increases between A.D. 1360 and 1660, when Iroquois settlements occupied the lake catchment (McAndrews & Boyko-Diakonow 1989; Clark & Royall, in prep.). The increases after 1900 might be related to regional combustion activities. An optimal (Weiner) filter using

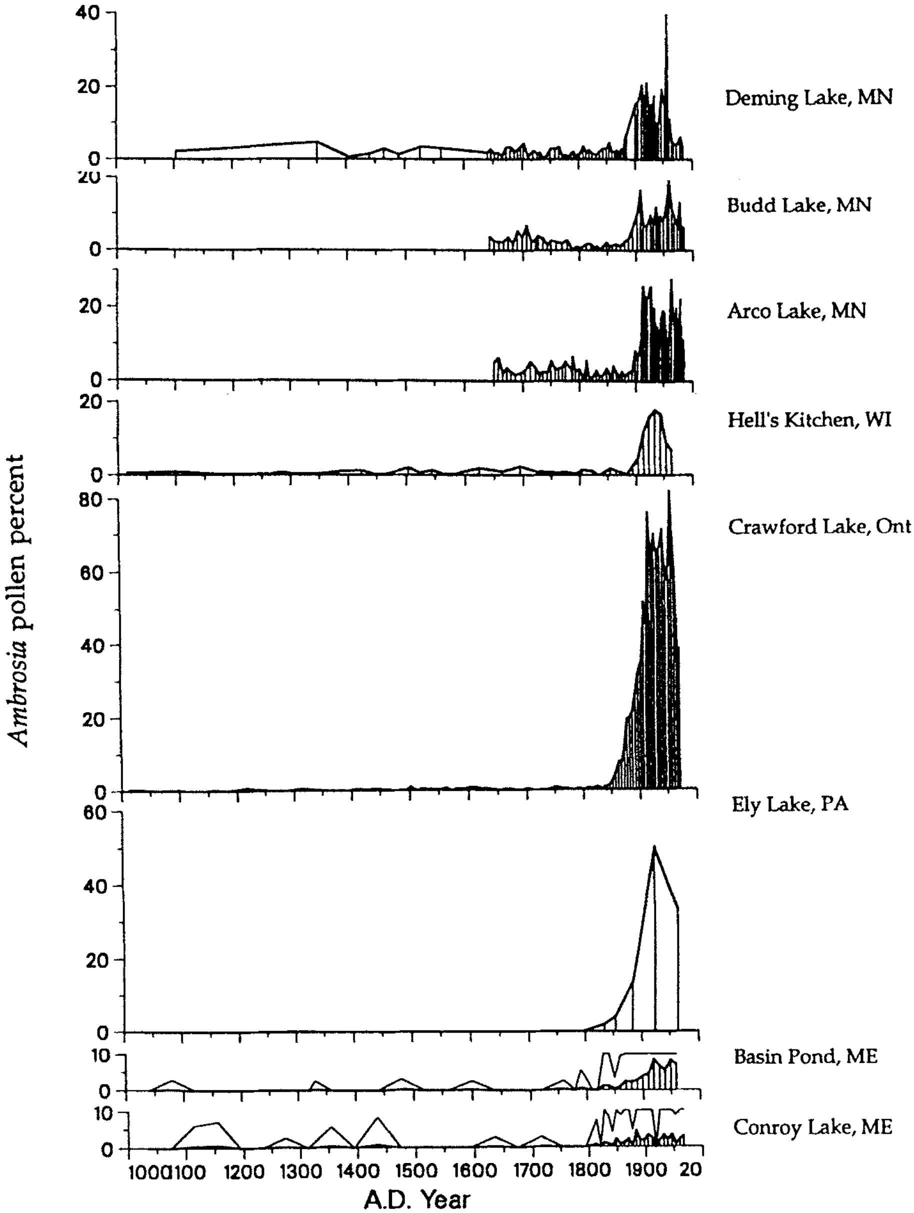


Fig. 2. Percentages of *Ambrosia* pollen, an indicator of land clearance, in cores. Chronologies come from varve counts. Pollen is expressed as percent of the total arboreal pollen.

a fourier transform was used to smooth series (Fig. 2c, 2d) to more clearly illustrate contrasting patterns between the two sites.

The full suite of sites show that differences in charcoal amount between Minnesota and southern Ontario are consistent with a geographic trend in

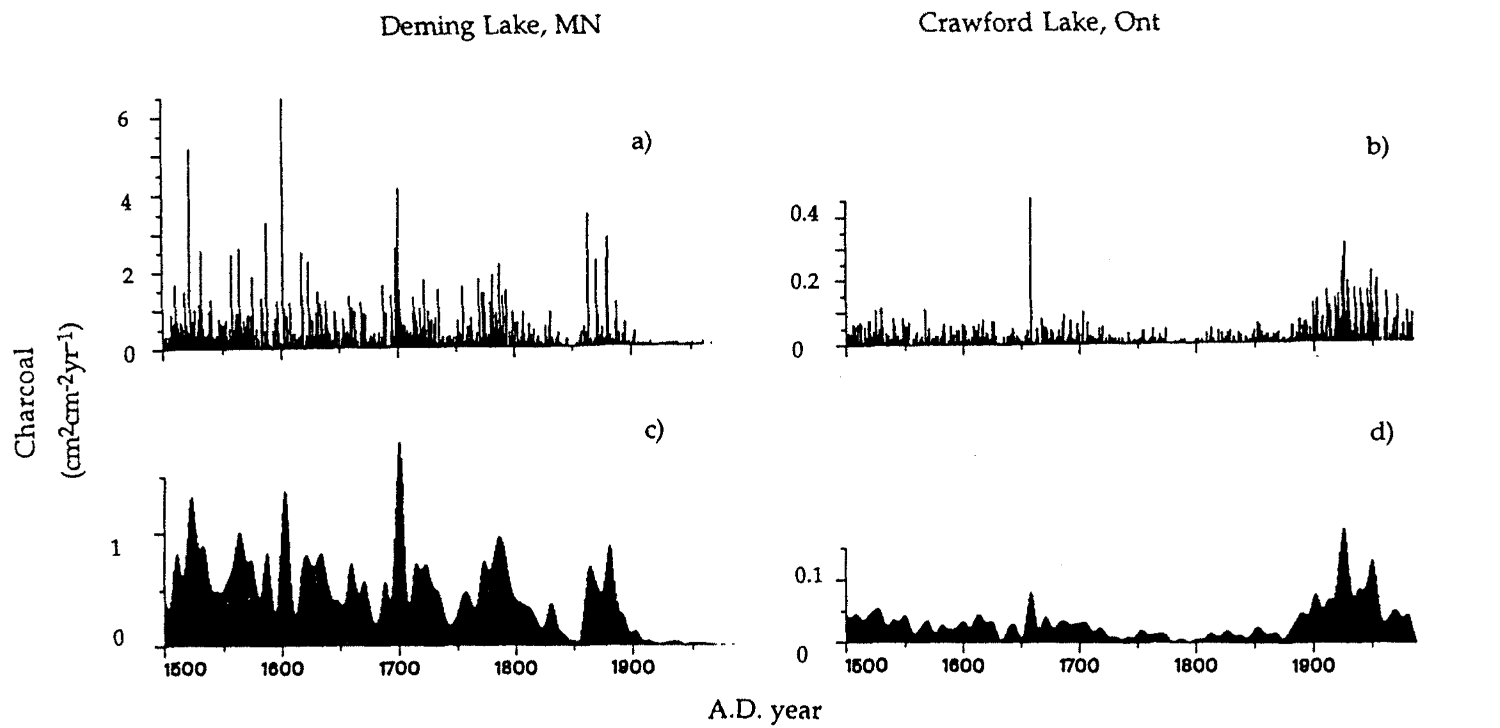


Fig. 3. Charcoal series showing changes in charcoal accumulation since A.D. 1500. In Minnesota (a, c), charcoal accumulation decreases abruptly with fire suppression after 1900. A southern Ontario site (b, d) shows the opposite trend. Panels (c) and (d) are filtered series of (a) and (b), respectively.

charcoal abundance. Accumulation rates were averaged for the two portions of each charcoal series, before and after A.D. 1900. Before 1900, charcoal accumulation rates display a more-or-less exponential decline with distance eastward from the prairie/forest border (Fig. 4a, b). Distance "zero" is placed at the 96th meridian. High values of > 10^{-1} cm^2 cm^{-2} yr^{-1} near (about 60 km from) the prairie/forest border in Minnesota decline by an order of magnitude to the eastern end of the gradient in Maine. Despite relatively consistent patterns from these sites, the sample is small in view of the large area spanned by the transect.

After A.D. 1900, there is no clear geographic trend in charcoal accumulation (Fig. 4c). All values fall below 10^{-1} cm^{-2} yr^{-1}. A t test for unequal variances between pre- vs post-settlement log-transformed charcoal data (a conservative test, because log-transformed data were near-normal) gave site-specific results. Some sites show decreases (the three Minnesota lakes, Hell's Kitchen), and others show no significant change (Ely Lake, Basin Pond).

A rough estimate of change in annual charcoal emissions from pre- to post-settlement time at this latitude for the eastern US can be made by integration across the transect. Using Fig. 4b as a crude estimate of spatial trends in the presettlement,

$$c(s) = \alpha \exp(-\beta s),$$

this integral is

$$C = \int_0^D c(s)ds$$

for transect length D (km) and distance s (km). Parameter estimates are given on Figure 3. Provided $D \gg 1/\beta$, as in this case, this result is roughly

$$c \approx \frac{\alpha}{\beta} \quad = 510 \text{ cm}^2\text{cm}^{-2} \text{ yr}^{-1}$$

The best we can do given lack of trend in post-settlement time is a simple scaling of the average value (i.e., the approach used in almost every calculation of this sort)

$$0.06 \times D = 180 \text{ cm}^2\text{cm}^{-2} \text{ yr}^{-1}$$

The result of this simple analysis is an estimated three-fold decrease in emissions of particles in diameter range > 10^1 µm for the region since A.D. 1900. We view these estimates as highly tentative, serving as a first effort at understanding charcoal fluxes beyond the scale of individual sites. Better geographic coverage should provide means for evaluating these patterns in the future.

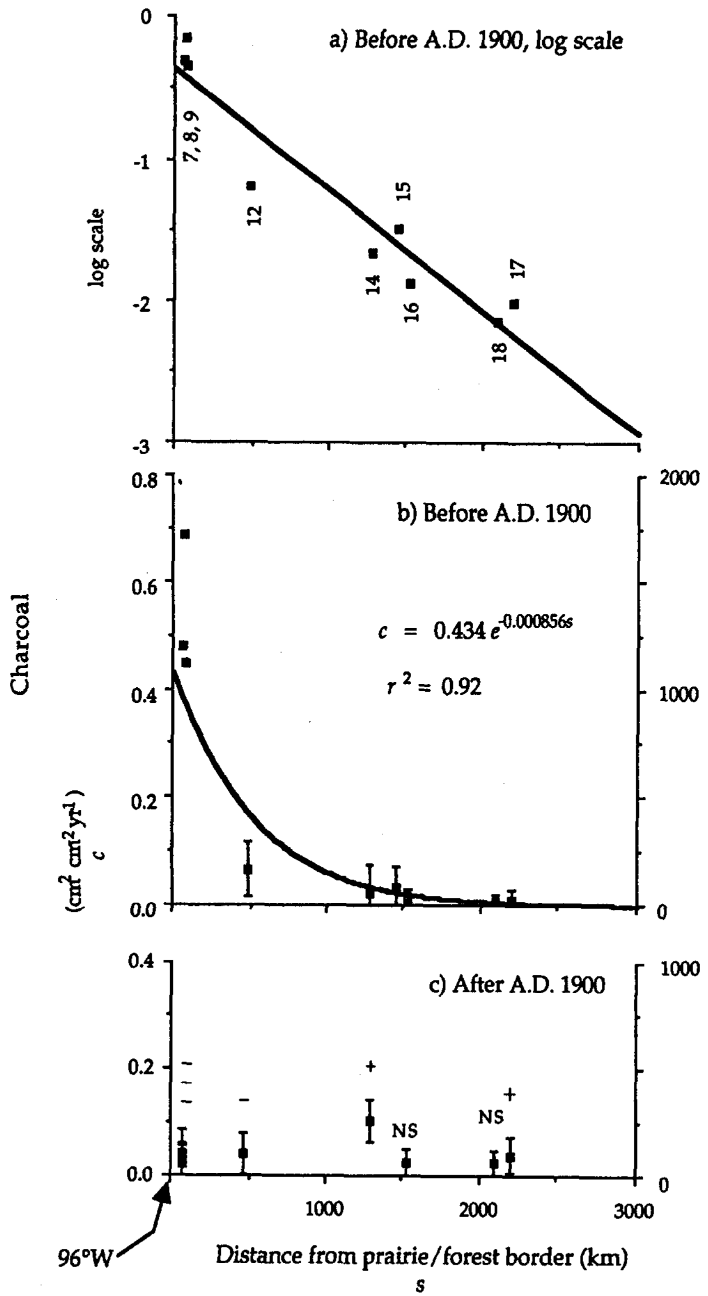


Fig. 4. Trends in average charcoal accumulation before (a and b) and after (c) A.D. 1900 with distance from the prairie/forest border (~96°W at this latitude). Site numbers on Figure 1 and Table 1 are given in (a). The exponential model provides a good fit to presettlement data. There is no trend after 1900. Symbolism for (c) includes the mean, one s.d. (error bar), "+" indicates an increase over presettlement rates, "–" indicates a decrease, and "NS" means no significant change.

Synthesis of patterns from mid-latitude North America

Results from this study were integrated with transformed charcoal data from previously published analyses (Fig. 5a). Together they span mid-latitude forest sites from the Pacific Northwest to Newfoundland (Table 1). The methods used in these different analyses include analysis of charcoal using optical microscroscopy (pollen slides, thin sections (this study)), nitric acid digestions, and infrared absorption (Table 1). Although other studies include charcoal data, we are unaware of other studies that report accumulation rates. The average presettlement charcoal accumulation rates from these studies were mapped, and contour lines drawn (Fig. 5b).

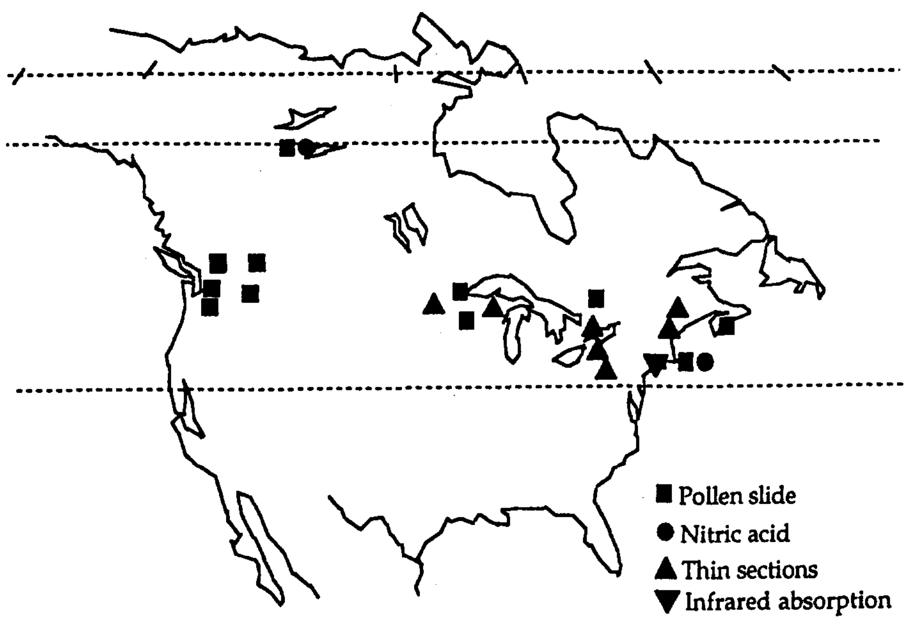
Despite differences in method and objective, the mapped accumulation rates show consistent geographic trends that appear related to broad climate and vegetation gradients across temperate North America (Fig. 5b). Highest values border the Central Plains, coming from xeric forests in Minnesota and Alberta. Because prairies sites are lacking from this data set, it is not possible to determine whether values would continue to increase from forest to prairie or if values indeed reach a maximum in these ecotonal forests. The few sites from the Northwest and from the eastern deciduous forest are lowest. The increasing trends from temperate forest to boreal in Ontario are suggestive of the expected increase in fire importance with rising fire frequencies.

Discussion

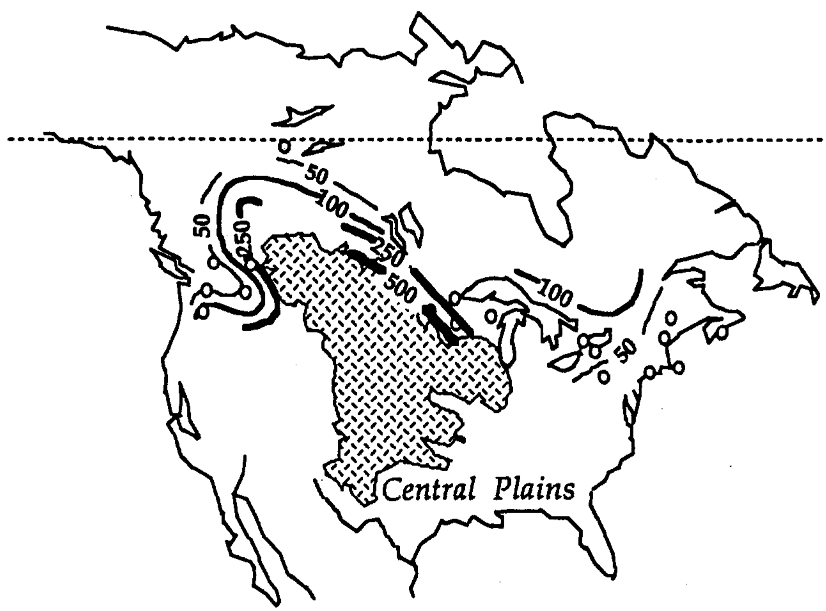
Charcoal analyses of varved lake sediments show geographic patterns in the emissions of combustion-derived particulates of diameters > 10^1 μm. A clear eastwardly declining trend in presettlement time is consistent with broad climate and vegetation gradients. Frequent fire is expected on prairie sites near the west end of the gradient. Low charcoal accumulation rates near the eastern end likely reflect low flammability of broad-leaved deciduous forests, inefficient burns, and low emission rates (Clark & Stocks, in prep).

Twentieth century land-use patterns (including effective fire suppression) and industrialization saw the geographic pattern in emissions replaced by high site-to-site variability. Whereas past emissions appear to reflect subcontinental scale climate controls, recent patterns apparently reflect variability in more local sources. Combustion-derived particulates in smaller size ranges, especially aerosols, are currently concentrated near urban centers, although they may be transported long distances (e.g., Andreae 1983; Rosen et al. 1984). The 20th-century patterns we observed are expected to reflect even more local emission patterns, because the large particles we measure have short residence times in the atmosphere. It thus appears that modern practices have profoundly affected both the total amount and the scale of spatial variability in atmospheric particulate loads.

Simple integration of charcoal estimates across our transect suggests a



a) Presettlement sites reporting accumulation rates



b) g/m²/yr

Fig. 5. Charcoal accumulation before A.D. 1900 from ours and previous work in North America. a) Sites used to estimate charcoal accumulation indicated by the method used to quantify sediment charcoal. b) Contours of charcoal accumulation using the values listed in Table 1.

Table 1. Sites used to estimate presettlement charcoal accumulation rates in lakes.

Site # on map	Site name	Lat (°N)	Long (°W)	Yr start/ Yr end (yr B.P.)	Vegetation	Method	Charcoal accumulation ($g\ m^{-2}\ yr^{-1}$)
1.	Kirk Cwynar (1978)	48	122	6800 6000	mixed conifer	pollen slides	38
2.	Hall Tsukada et al. (1981)	48	122	5000 90	Hemlock/ Doug fir	pollen slides	38
3.	Mineral Tsukada and Sugita (1982)	46	122	5000 90	Hemlock/ Doug fir	pollen slides	100
4.	Lost Trial Mehringer et al. (1977)	46	114	1600 20	Pine	pollen slides	9
5.	Tobaggan MacDonald (1989)	50	115	5500 0	Pine/ Spruce	pollen slides	380
6.	Rainbow MacDonald et al. (1991)	60	112	90 0	Spruce/ Pine	pollen slides	25
7.	Deming this study	47	95	750 90	Pine/ Hardwood	thin sections	1200
8.	Arco this study	47	95	350 90	Pine/ Hardwood	thin sections	1700
9.	Budd this study	47	95	350 90	Pine/ Hardwood	thin sections	1200
10.	Dark Gajewski et al. (1985)	45	92	1000 90	Hardwood/ Pine	pollen slides	130
11.	Clouds Swain (1973)	48	92	240 40	Pine/ Hardwood	pollen slides	100
12.	Hell's Kit this study	46	90	3000 1000	Pine/ Hardwood	this slides	150
13.	Green Cwyner (1978)	46	78	1220 720	Hardwood/ Pine	pollen slides	100
14.	Crawford this study	44	77	2000 90	Hardwood/ Pine	thin sections	55
15.	Devil's this study	43	77	1900 500	Hardwood	thin sections	80
16.	Ely this study	42	76	1800 90	Hemlock/ Hardwood	this sections	33
17.	Conroy this study	46	68	2000 90	Hardwood/ Conifer	thin sections	25
18.	Basin this study	44	69	1900 90	Hardwood/ Pine	thin sections	18
19.	Duck Winkeler (1985)	42	70	7000 190	Pine/ Oak	pollen slides	2.5
20.	Nova Scotia Green (1980)	45	65	3100 10	Northern hardwoods	pollen slides	5.0
21.	Whitney Goldberg (1985)	41	73	pre industrial	Hardwood	infrared absorption	1.0

three-fold decrease since the beginning of the twentieth century. Seiler & Crutzen (1980) argued that the fraction of total emissions contributed by biomass burning may have been more important prior to widespread fossil-fuel combustion. We predict that the average *absolute* flux of large particles may have been three times as large. It is important to bear in mind that the bulk of particulates fall in the aerosol (sub-micron) diameter range that are not considered by our method. We therefore do not argue that total emissions were higher in the past, just the fraction of large particles. Sediment records show that combustion-derived, aerosol-bound polycyclic aromatic hydrocarbons (PAH's) have increased in the 20th century near areas of concentrated fossil-fuel combustion in the eastern US (Laflamme & Hites 1987; Gschwend & Hites 1981) and Europe (Jones et al. 1989). There are no good records of PAH fluxes from regions of previous frequent fire (e.g., near the Central Plains), so this is an area for further analysis.

The charcoal accumulation rates determined by us (Fig. 4) and in previous analyses (Table 1, Fig 5b) are higher than expected considering current fuel loads, burning efficiencies, emission factors, and fire frequencies. There are several reasons why charcoal might be overrepresented in lakes that are beyond the scope of this analysis. Elsewhere we consider the implications of sediment charcoal for the C cycle on the basis of a global data set of sediment charcoal (Clark & Stocks, in prep.). Here we simply point out that the rates were observe are large relative to the amounts we expect to settle out of the atmosphere. And these high rates are consistently obtained across the different methods of analysis used in Figure 4b. This observation points to a need to calibrate sediment series if they are to be used to estimate absolute pre-settlement emission rates.

Although the absolute fluxes we observe are high, geographic patterns in carbon sequestration suggested by the data sets appear to agree with predictions one would make on the basis of fuel loads, burn frequencies and efficiencies, and emission factors (Clark & Stocks, in prep.). Despite the different methods used to quantify sediment charcoal and variability in flux estimates one would expect within and among lakes, conversion to common units shows clear geographic trends in charcoal accumulation. The paucity of sites makes mapping of this sort rather tentative. Our map does demonstrate geographic coverage of charcoal analyses, and it shows patterns that may provide insight into the importance of biomass burning through the past. Several aspects of the theses maps are noteworthy and suggest foci for future work. The highest rates appear near prairie/forest border areas. Sites are needed within the Central Plains to determine whether the maximum occurs within the Plains or at ecotones such as those included in this data set. Grasslands have relatively frequent fire with high burn efficiencies, but fuel loads and particulate emission factors can be low. Ecotonal woodland areas can not only have relatively frequent fire, but also larger fuel loads and emission factors can be expected.

Next highest values appear to come from conifer, largely boreal, forests.

Boreal fires occur at intervals of 10^2 yr, fuel loads are relatively high (crown fires are typical), and emissions can be large. Here again, few truly boreal-type sites are contained in the data set. The patterns in Figure 5b are suggestive and point to the need for more analyses at higher latitudes. Our lab is currently analyzing prairie, woodland, and boreal sites that will fill out some of the existing holes in the data set.

Lowest values are observed in eastern deciduous forest and at a high elevation site in the Rocky Mountains. Presettlement fire regimes are not well characterized in broad-leaved deciduous forests. Fires were not as common as in coniferous stands and the behavior of such fires is more difficult to quantify, lacking information on fuel loads and structure in presettlement stands. We cannot identify the source of low background levels we observe in our cores. It is likely that they do not represent transport from distant (e.g., prairie or boreal) regions of frequent burning, because atmospheric residence times of these large particles are low.

Acknowledgments

We thank K. Gajewski and J. McAndrews for providing pollen data for several sites reported here. For assistance coring lakes, we thank T. Abbott, E. Almgren, R. Davis, G. Jacobson, H. Jacobson, J. McAndrews, D. Porter, J. Porter and H. E. Wright, Jr. For their discussions we thank W. Cofer, W. Schlesinger, and B. Stocks. Two reviewers provided useful suggestions. Financial support was provided by NSF grant BSR-9107272.

References

Andreae MO (1983) Soot carbon and excess fine potassium: long range transport of combustion derived aerosol. Science 220: 1148–1151

Brugam RB (1978) Pollen indicators of land-use change in southern Connecticut. Quaternary Research 9: 349–362

Budyko KI (1987). History of the Earth's Atmosphere. Springer-Verlag, Berlin, Germany

Ch'ylek P, Ramaswamy V, & Srivastava V (1984) Graphitic carbon content of aerosols, clouds and snow, and its climatic implications. The Science of the Total Environment 36: 117–120

Clark JS (1988a) Charcoal-stratigraphic analysis on petrographic thin sections: recent fire history in northwest Minnesota. Quaternary Research 30: 67–80

Clark JS (1988b). Effects of climate change on fire regime in northwestern Minnesota. Nature 334: 233–235

Clark JS (1990) Fire and climate change during the last 750 years in northwestern Minnesota. Ecological Monographs 60: 135–159

Clark JS & Robinson J (1993) Paleoecology of fire. In: 193–214. in Crutzen PJ & Goldammer JG (Eds) Fire in the Environment: Its Ecological, Climatic, and atmospheric Chemical Importance. Wiley, New York, NY

Cope MJ & Chaloner WG (1985) Wildfire: an interaction of biological and physical processes. In: Tiffney BH (Ed) Geological Factors and the Evolution of Plants (pp 257–277). Yale University Press, New Haven, CT

Crutzen PJ & Andreae MO (1990) Biomass burning in the tropics: impact on atmospheric chemistry and biogeochemical cycles. Science 250: 1669–1678

Cwynar LC (1978) Recent history of fire and vegetation from laminated sediment of Greenleaf Lake, Algonquin Park, Ontario. Canadian Journal of Botany 56: 10–21

Cwynar LC (1987) Fire and the forest history of the North Cascade Range. Ecology 68: 791–802

Gajewski K, Winkler MG, & Swain AM (1985) Vegetation and fire history from three lakes with varved sediments in northwestern Wisconsin. Review of Palaeobotany and Palynology 44: 277–292

Goldberg ED (1985) Black Carbon in the Environment. Wiley, New York, New York, USA

Green DG (1981) Time series and postglacial forest ecology. Quaternary Research 15: 265–277

Gschwend PM & Hites RA (1981) Fluxed of polycyclic aromatic hydrocarbons to marine and lacustrine sediments in the northeastern United States. Geobhimica et Cosmochimica Acta 45: 2359–2367

Harris TM (1958) Forest fire in the Mesozoic. Journal of Ecology 46: 447–453

Herring JR (1985) Charcoal fluxes into sediments of the North Pacific Ocean: The Cenozoic record of burning. In: Sundquist ET & Broecker WS (Eds) The Carbon Cycle and Atmospheric CO2: Natural Variations, Archaen to Present (pp 419–442). Geophyscal Monograph 32, American Geophysical Union, Washington, D.C.

Jones KC, Stratford JA, Waterhouse KS, Furlong ET, Giger W, Hites RA, Schaffner C, & Johnston, AE (1989) Increases in the polynuclear aromatic hydrocarbon content of an agricultural soil over the last century. Environmental Science and Technology 23: 95–101

Laflamme RE, & Hites RA (1978) The global distribution of polycyclic aromatic hydrocarbons in recent sediments. Geochimica et cosmochimica Acta 42: 289–303

Malingreau J-P, Albini FA, Andreae MO, Brown S, Levine JS, Lobert J, Kulbusch TA, Radke L, Setzer A, Vitousek PM, Ward DE, & Warnatz J (1993) Quantification of fire characteristics from local to global scales. In: Crutzen PJ & Goldammer JG (Eds) Fire in the Environment: Its Ecological, Climatic, and Atmospheric Chemical Importance (in press). Wiley, New York, NY

MacDonald GM (1989) Postglacial palaeoecology of the subalpine forest-grassland ecotone of southwestern Alberta: new insights on vegetation and climate change in the Canadian rocky Mountains and adjacent foothills. Palaeogeography, Palaeoclimatology, and Palaeoecology 73: 155–173

MacDonald GM, Larsen CPS, Szeicz JM, & Moser KA. (1991) The reconstruction of boreal forest fire history from lake sediments: a comparison of charcoal, pollen, sedimentological, and geochemical indices. Quaternary Science Reviews 10: 53–71

McAndrews JH & Boyko-Diakonow M (1989) Pollen analysis of varved sediment at Crawford Lake Ontario: evidence of Indian and European farming. In: Fulton RJ. (Ed) Quaternary Geology of Canada and Greenland (pp 528–530). Geological Survey of Canada

Mehringer PJ, Arno SF, & Petersen KL. (1977) Postglacial history of Lost Trail Pass Bog, Bitterroot Mountains, Montana. Actic and Alpine Research 9: 345–368

Merkt J (1971) Zuverlassige Auszahlungen von Jahresschichten in Seesedimenten mit Hilfe von Gross-Dunnschliffen. Archiv fur Hydrobiologie 69: 145–154

Radke LF, Wtith JL, Hegg DA, & Hobbs PV (1978) Airborne studies of particles and gases from forest fires. Journal of Air Pollution Control Association 28: 30–34

Robinson JM (1989) Phanerozoic O2 variation, fire, and terrestrial ecology. Palaeogeography, Palaeoclimatoloy, and Palaeoecology 75: 223–240

Rosen H, Hansen ADA, & Novakov T (1984) Role of graphitic carbon particles in radiative transfer in the Arctic haze. The Science of the Total Environment 36: 103–110

Sander PM & Gee CT (1990) Fossil charcoal: techniques and applications. Review of Palaeobotany and Palynology 63: 269–279

Seiler W & Crutzen PJ 91980) Estimates of gross and net fluxes of carbon between the biosphere and the atmosphere from biomass burning. Climatic Change 2: 207–247

Seischab FK (1990) Presettlement forests of the Phelps and Gorham Purchase in western New York. Bulletin of the Torrey Botanical Club 117: 27–38

Shneour EA (1966) Oxidation of graphitic carbon in certain soils. Science 151: 991–992

Swain AM (1973) A history of fire and vegetation in northeastern Minnesota as recorded in lake sediment. Quaternary Research 3: 383–396

Swain AM (1978) Environmental changes during the past 2000 years in north-central Wisconsin: analysis of pollen, charcoal and seeds from varved lake sediments. Quaternary Research 10: 55–68

Tsukada M & Sugita S (1982) Late Quaternary dynamics of pollen influx at Mineral Lake, Washington. Botanical Magazine (Tokyo) 95: 401–418

Tsukada M, Sugita S & Hibbert DM (1981) Paleoecology in the Pacific Northwest. I. Late Quaternary vegetation and climate. Verhandlungen des Internationalen Vereinigung für Limnologie 21: 730–737

Whitney GG (1990) The history and status of the hemlock-hardwood forests of the Allegheny Plateau. Journal of Ecology 78: 443–458

Winkler MG (1985) A 12,000-year history of vegetation and climate for Cape Cod, Massachusetts. Quaternary Research 23: 301–312

Wolff GT, Countess RJ, Groblicki PJ, Ferman MA, Cadle SH, & Muhlbaier JL (1981) Visibility-reducing species in the Denver "brown cloud" – II. Sources and temporal patterns. Atmospheric Environment 15: 2485–2502

Wright HE Jr (1982) Lake and wetland sediments as records of past atmospheric composition. In: Goldberg ED (Ed) Atmospheric Chemistry (pp 135–157). Springer-Verlag, Berlin

Wright HE Jr, Mann DH, & Glaser PH (1984) Piston corers for peat and lake sediments. Ecology 65: 657–659.