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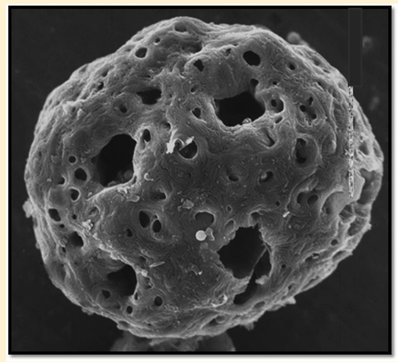
Spheroidal Carbonaceous Fly Ash Particles Provide a Globally Synchronous Stratigraphic Marker for the Anthropocene

Neil L. Rose*

Environmental Change Research Centre, Department of Geography, University College London, Gower Street, London WC1E 6BT, United Kingdom

Supporting Information

ABSTRACT: Human impacts on Earth are now so great that they have led to the concept of a new geological epoch defined by this global human influence: the Anthropocene. While not universally accepted, the term is increasingly popular and widely used. However, even among proponents, there is considerable debate regarding when the epoch may have started, from coeval with the Holocene, through the Industrial Revolution, to the mid-20th century when unprecedented human activities resulted in exponential increases in population, resource consumption, and pollutant emission. Recently, this latter period, known as the Great Acceleration, appears to be becoming the more widely accepted start date. To define any start point, a global stratigraphic marker or Global Boundary Stratotype Section and Point (GSSP) is typically required. Here, spheroidal carbonaceous fly ash particles (SCPs), byproducts of industrial fossil-fuel combustion, are proposed as a primary marker for a GSSP at the time of the Great Acceleration. Data from over 75 lake sediment records show a global, synchronous, and dramatic increase in particle accumulation starting in c. 1950 driven by the increased demand for electricity and the introduction of fuel-oil combustion, in addition to coal, as a means to produce it. SCPs are morphologically distinct and solely anthropogenic in origin, providing an unambiguous marker. This is a clear signal of great stratigraphic utility representing a primary driving force for global anthropogenic change.



INTRODUCTION

Recognition that human activities are having far-reaching impacts upon the Earth is not new,[1] but the scale and extent of that influence on biodiversity, biogeochemical cycles, and climate[2] is increasingly apparent. In 2000, it was proposed that anthropogenic environmental change had become so dominant that it justifies a new geological epoch, and the name "Anthropocene" was proposed for it.[3] Since then, although not universally accepted,[4] the term has become increasingly widely used in both the scientific and broader literature. However, there has been considerable controversy regarding the point at which the Anthropocene may be considered to have succeeded the current Holocene Epoch. Some arguments effectively eliminate any distinction with the Holocene[5] by proposing the megafaunal extinction c. 12 500 BP[6,7] and the start of domestication as the onset point, while Ruddiman's "early-anthropogenic era"[8] uses agriculturally-derived increases in atmospheric methane and CO_2 at 5000 and 8000 BP, respectively. Others argue that the start of the Industrial Revolution in the latter half of the 18th century, with the expansion of fossil-fuel exploitation on an industrial scale and the subsequent inflection in greenhouse gas concentrations above the Holocene background values, marks the logical start date.[1] Still others suggest an even later date in the mid-20th century marked by dramatic increases in human population, unprecedented exploitation and consumption of natural resources, and emissions of pollutants to the atmosphere and surface waters.[9]

It is apparent that the earlier suggested dates are neither global nor synchronous.[9] Fuller et al.[10] present archaeozoological data to show how the spread of livestock and domestication across Africa, India, and southeast Asia each cover c. 4000 years, while similar studies show, for example, how rice cultivation spread over periods of millennia both regionally[11,12] and within a single country.[13] Similarly, the Industrial Revolution can no longer be regarded as the abrupt discontinuity that its name suggests, having its origins in economic expansion in Britain in the 16th century[14] and with subsequent industrialization spreading across the world over a period of 200 years between the 18th and 20th centuries.[15] In contrast, the mid-20th century period, termed the "Great Acceleration" or the "Atomic Age",[2,16] is marked by global and synchronous rather than local or regional and time-transgressive changes. For example, Steffen et al.[1,17] demonstrate how dramatic and unprecedented changes in human population, damming of rivers, fertilizer consumption, vehicle numbers, increases in atmospheric N_2O and depletion of

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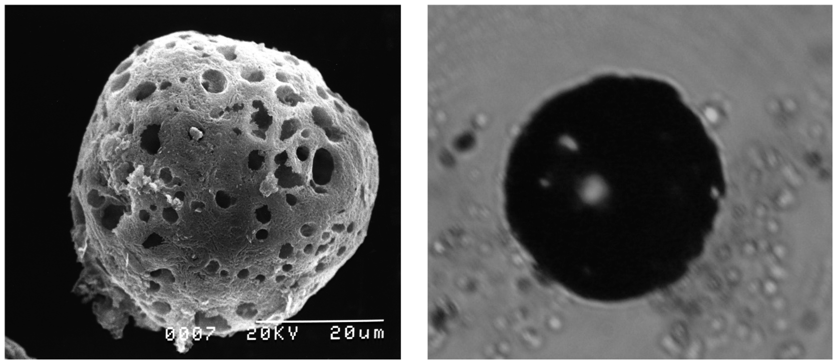

Figure 1. Scanning electron microscopy (SEM) and light microscopy photographs of SCPs extracted from lake sediments. The SEM image shows 20 μm scale bar, and the SCP in light microscope image is 18 μm in diameter.

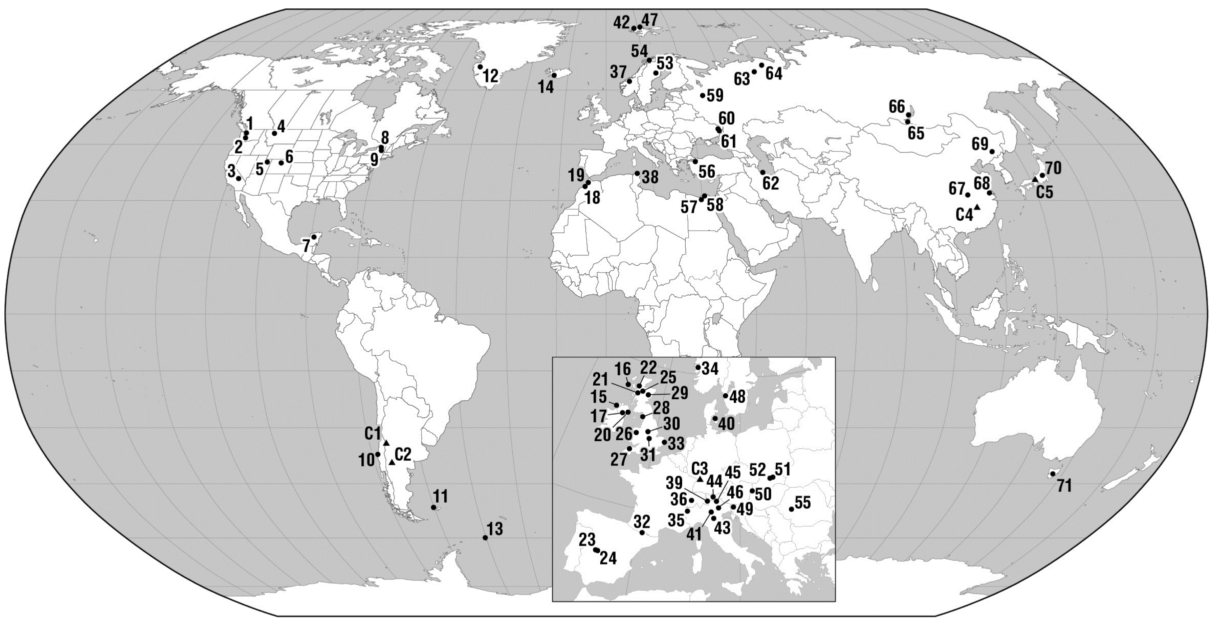

Figure 2. Site location map. A Robinson projection with sites used in this study. Labels 1−71 are lakes with radiometrically dated sediment cores and SCP accumulation rate data. (Inset) Enlarged map of European sites. Presented data for Europe, especially U.K. and Ireland, are not exhaustive but representative of these regions. C1−C5 are sites, taken from the literature, for areas where less SCP accumulation rate data exist. Cores from these lakes have radiometric dates and SCP sediment concentration data (see SI Figure 1 of the Supporting Information) but not SCP accumulation rate data. All site names, latitudes, longitudes, and data sources are provided in the Supporting Information.

stratospheric ozone, exploitation of global fisheries, and perturbation of the nitrogen cycle all started from c. 1950.

In 2009, the Anthropocene Working Group (AWG) of the Subcommission on Quaternary Stratigraphy was established to consider the evidence for environmental signatures that could be uniquely attributed to a new epoch.[15] The AWG is responsible for producing a recommendation to be considered by the International Commission on Stratigraphy in 2016, including a requirement for defining its lower boundary.[18] Therefore, the recognition of an unequivocal human-induced stratigraphic marker[19] that may underpin a Global Boundary Stratotype Section and Point (GSSP) (or "golden spike") or alternatively a Global Standard Stratigraphic Age (GSSA), with which to define the lower boundary of the new epoch, is essential to this process and, hence, the definition of the Anthropocene itself. Recent indications are that the Great Acceleration currently has the most support within the AWG,[9,20] who are "working on a stratigraphical boundary for the mid-20th century".[21] While not all mid-century markers show the same precise temporal patterns,[22] the beginning of the atmospheric ^{14}C peak[23] or that of ^{239}Pu and ^{240}Pu[24] resulting from the fallout of nuclear weapons testing may be considered as markers for a global stratigraphically synchronous boundary for the mid-20th century. However, as described above, the mid-1960s is a decade later than many of the observed emissions and impacts that define the Great Acceleration[1] and almost two decades later than the proposed boundary level at the time of the world's first nuclear explosion at Almagordo, NM, at 11:29:21 ± 2 s [UTC or Greenwich Mean Time (GMT)] on July 16, 1945.[9]

Here, it is proposed that the fly ash particle record in natural archives could provide a more suitable globally synchronous indicator of anthropogenic impact for c. 1950. Fly-ash particles are the particulate byproducts of high-temperature fossil-fuel (coal-series; fuel-oil) combustion and are emitted to the atmosphere along with flue gases. There are two main particle types: inorganic ash spheres, which are derived from non-combustible minerals present within the fuel, and spheroidal carbonaceous particles (SCPs) (Figure 1), which result from the incomplete combustion of the pulverized coal particles or

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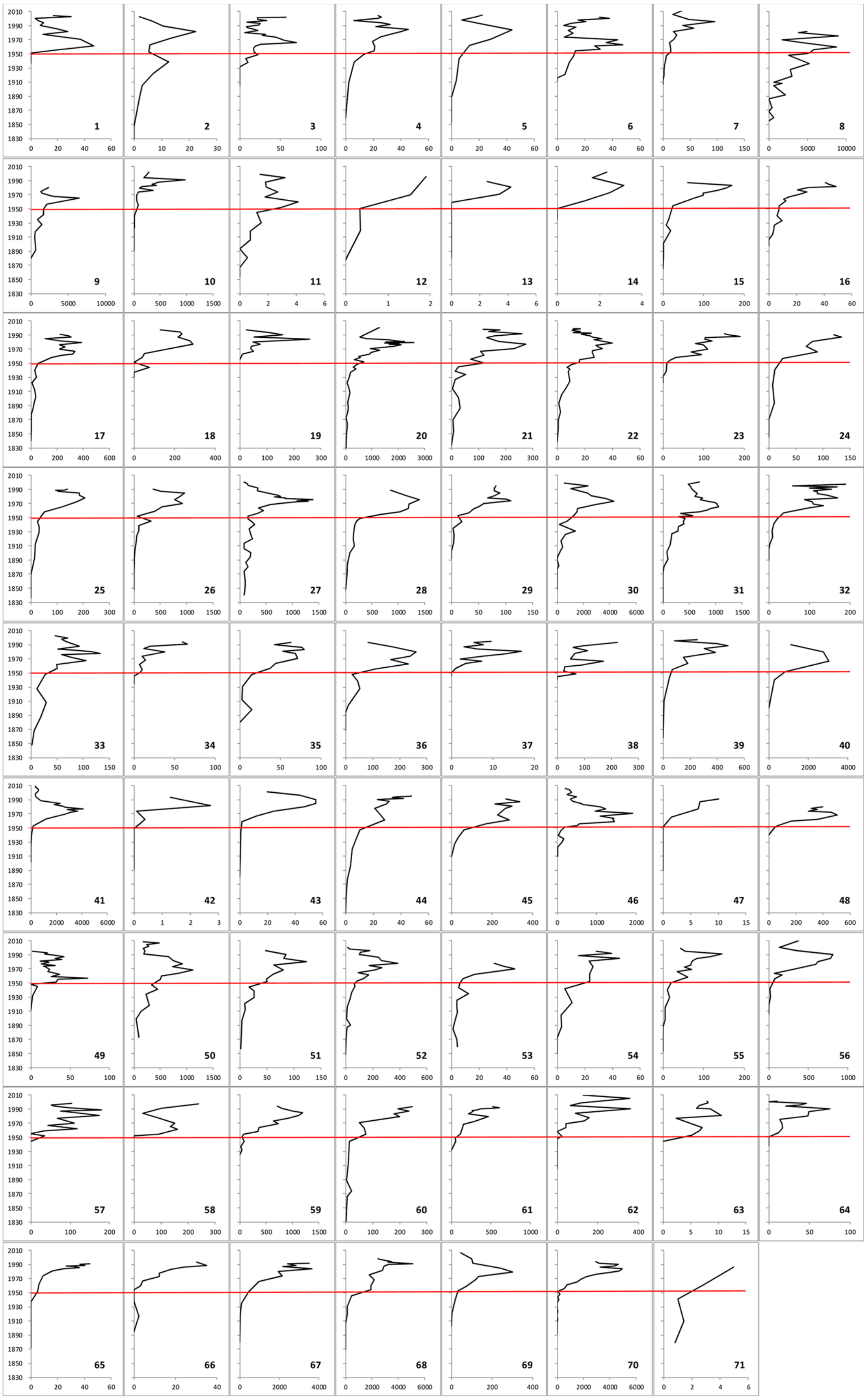

Figure 3. SCP accumulation rate data (number of particles cm^{-2} year^{-1}) for the 71 lake sediment cores (1−71 in Figure 1) plotted on a radiometrically derived chronological y axis (years AD). The red line indicates 1950. Additional site information and data sources are provided in SI Table 1 of the Supporting Information.

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oil droplets themselves[25] and are a component of the "black carbon continuum".[26] They have no natural sources, and their characteristic shape, color, and morphology (Figure 1) make them unambiguous indicators of contamination from this industrial source.[25,27] Further, they preserve in a range of accumulating media, including freshwater[28,29] and marine[30] sediments, peats,[31] and ice.[32] However, most data are available from lake sediments because their continuous accumulation, global spread, clarity of record, and comparative ease with which reliable, highly resolved chronologies can be produced make them ideal archives.

■ MATERIALS AND METHODS

Sites and Sampling. The data presented here represent an amalgamation of over 20 years of research undertaken at the Environmental Change Research Centre (ECRC), University College London, and additionally through collaboration with many institutions around the world. These studies have included assessments of acidification and recovery, eutrophication, and the scale and extent of atmospherically deposited contamination using the lake sediment archive as an historical record. Because of the varying nature of these studies, there are no specific selection criteria that cover all sites. Criteria for inclusion in this current study include historical SCP flux data and a reliable sediment record with reasonable radionuclide-derived chronologies. However, within some regions, for example, across the U.K.,[33] a large number of sites fulfilling these criteria were available and only a typical few have been selected for inclusion. Consequently, SCP flux data from 71 lakes are presented, and while the majority of these are within Europe (Figure 2 and SI Table 1 of the Supporting Information), every continent is represented. Where data are sparse, additional sites with only SCP concentration data (sites C1−C5; Figure 2) and radiometric chronologies have also been included (see the Discussion).

Sediment cores were taken using a variety of different techniques appropriate to the requirements of these studies. These included short gravity cores,[34,35] mini-Mackereth cores,[36] and modified Livingstone cores.[37] Further, they have been sub-sampled at a variety of different resolutions from 0.2 cm intervals for remote, slowly accumulating upland and mountain lakes to 1 cm intervals for rapidly accumulating lowland ponds and pools. All sediment samples were air- or freeze-dried prior to SCP analysis and radiometric dating.

SCP Analysis. SCP analysis involved sequential treatments of nitric, hydrofluoric, and hydrochloric acids to remove organic, siliceous, and carbonate fractions, respectively, from the sediment, resulting in a suspension in water.[38] A known fraction of this suspension was then evaporated onto a coverslip and mounted onto a glass slide, and the number of SCPs was counted using a light microscope at 400 times magnification. Standard criteria for SCP identification were followed.[27] SCP concentrations were calculated as the number of particles per gram of dry mass of sediment (g^{-1} of DM), and SCP fluxes were calculated as the product of the SCP concentration and bulk dry sediment accumulation rate (number of particles per centimeter squared per year; cm^{-2} $year^{-1}$). SCP accumulation rate data are used preferentially to SCP concentrations because they take into account the effect that variations in the bulk sediment accumulation rate may have on contaminant concentration data (e.g., a dilution when sediment accumulation increases). Analytical blanks and SCP reference material[27] were included with all sample digestions. The detection limit for the technique is typically less than 100 g^{-1} of DM, and calculated concentrations generally have an accuracy of c. ±45 g^{-1} of DM.

Radiometric Dating of Sediment Cores. Dried sediment samples were analyzed for ^{210}Pb, ^{226}Ra, ^{137}Cs, and ^{241}Am by direct gamma assay using ORTEC HPGe GWL series well-type coaxial low background intrinsic germanium detectors. ^{210}Pb was determined via its gamma emissions at 46.5 keV, and ^{226}Ra was determined by the 295 and 352 keV gamma rays emitted by its daughter isotope ^{214}Pb, following 3 weeks of storage in sealed containers to allow for radioactive equilibration. ^{137}Cs and ^{241}Am are artificial radionuclides measured by their emissions at 662 and 59.5 keV, respectively.[39] They were introduced into the environment by atmospheric fallout from nuclear weapons testing and by nuclear accidents. Sediment records of these radionuclides generally begin in the 1950s and peak in 1963, the year prior to the global atmospheric test ban treaty. Absolute efficiencies of the detectors were determined using calibrated sources and sediment samples of known activity. Corrections were made for the effect of self-absorption of low-energy gamma rays within the sample. There are two principal methods for determining the initial ^{210}Pb activity of a sediment layer necessary for the calculation of its ^{210}Pb date. These are the CRS (constant rate of unsupported ^{210}Pb supply) model and the CIC (constant initial ^{210}Pb concentration) model.[40] The CRS model is perhaps the most widely accepted and is based on the hypothesis that the ^{210}Pb supply to the sediments is dominated by constant atmospheric fallout. This model is not valid where there are interruptions or changes to the ^{210}Pb supply, for example, with a sediment hiatus. The independently derived 1963 fallout date is used to help constrain the ^{210}Pb dates.[40] Final chronologies for all cores were derived using all of these available data. ^{210}Pb has a half-life of 22.26 years, allowing for reliable chronologies of 150−170 years to be produced. Sediment core chronological errors for most recent decades are typically ±2 years, with sediments in the mid-20th century typically ±4−7 years and the mid-late 19th century typically ±18−25 years.

■ RESULTS AND DISCUSSION

The SCP flux data for the 71 individual lakes are presented on chronological axes in Figure 3. Across Europe and North America, the SCP record typically begins in the mid-19th century.[28,29] The observable start of the SCP record varies depending upon regional developments in the industrial combustion of coal-series fuels but also the accumulation rate of the depositing archive. For example, a rapid accumulation rate may dilute the concentration of SCPs (or any contaminant) to a level below the analytical limit of detection, thereby delaying the observable start of the record until such time that deposited contamination increases to a level that exceeds that detection limit. SCP concentrations and fluxes typically increase gradually from the start of the record (Figure 3) until the mid-20th century, when there is a "rapid increase" starting from c. 1950. This dramatic increase in SCP deposition is concomitant with the many other indicators of the Great Acceleration,[1,2] including, of course, primary energy use,[17] and is linked with the post-World War II (WWII) increase in the demand for electricity and the introduction of cheap fuel oil, which led to the development of the first large-scale oil-fired power plants.[25] The duration of this rapid increase varies between regions because the SCP concentration and flux peaks are often followed by a decline in inputs (Figure 3). This is due

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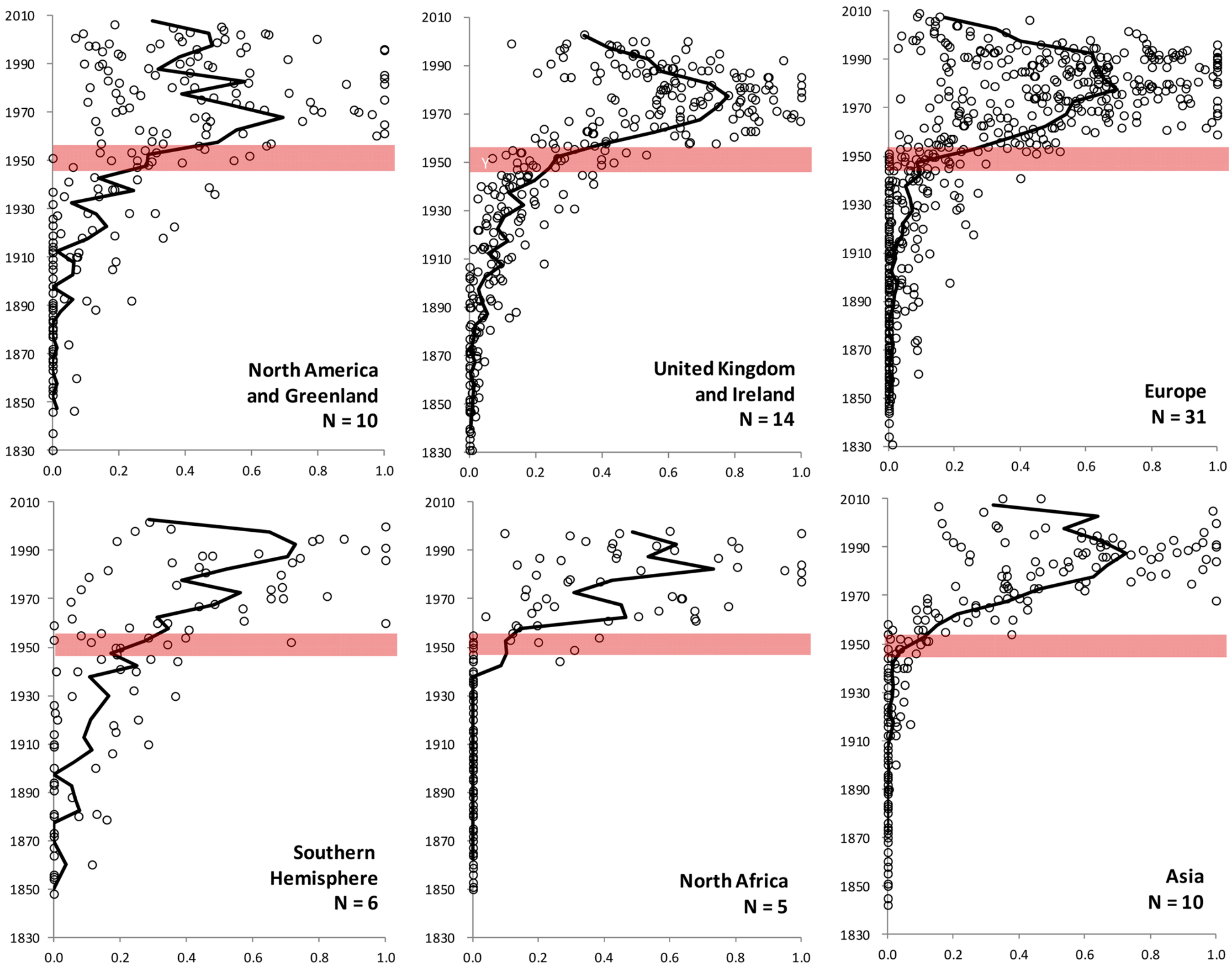

Figure 4. SCP sediment profile data plotted for different regions. Data include 71 lake cores with SCP accumulation rate data and an additional 5 with SCP concentration data. All are normalized to the SCP accumulation or concentration peak (1.0) for that site, to take account of within and between region variations in the scale of contamination. Open circles are data points for the individual cores, and solid lines represent mean data (5 year time step) for the region. Horizontal bars indicate 1950 ± 5 years, i.e., "mid-20th century". See the Materials and Methods for radiometric dating errors across the time period represented by these data.

to a number of reasons, including the introduction of particle-arrestor technology to combustion sources, changes of fuel source (e.g., from coal to natural gas), and the regional decline of heavy industry. Therefore, while some features of SCP profiles (e.g., start of the record; the sub-surface peak) vary regionally,[25,33] the "rapid increase" in the mid-20th century appears to be a global signal.

The data presented in Figure 3 demonstrate this global signature and show that it is recorded in the sediments of a wide range of lake types from mountain lakes (e.g., sites 3, 12, and 65) to shallow lowland lakes (sites 31, 33, and 40) and coastal waterbodies (sites 58) and from small lakes a few hectares in area (sites 13 and 27) to some of the largest freshwater bodies in the world (sites 62, 66, and 68). The mid-20th century rapid increase in SCP accumulation is clearly observable in all locations. At some sites (e.g., sites 4, 15, and 51), this is recorded as an inflection in a gradually increasing SCP accumulation rate profile, marking the transition from lower levels of contamination to significantly enhanced contaminant deposition. At others, most noticeably in less contaminated areas (e.g., sites 13 and 14), regions of more recent industrial development (e.g., sites 58 and 62), or lakes with more rapid sediment accumulation rates (e.g., sites 38 and 67), this rapid increase can be observed as the start of the record, where the increase in SCP deposition leads to exceedance of the analytical detection limit for the first time. In areas where SCP accumulation rate data are sparse, additional concentration records from the literature have been used to provide supporting evidence for these temporal patterns. Locations are shown in Figure 2 (C1−C5) (details in SI Table 2 of the Supporting Information), and the concentration profiles are shown in SI Figure 1 of the Supporting Information, again on the same chronological axes. These SCP concentration profiles also show a clear and rapid increase from c. 1950. However, while these data are not as robust as SCP accumulation rate data for the reasons described above, the mid-20th century increases in concentration are so marked that significant and temporally coincidental changes in bulk sediment accumulation rates would be required for them to fail to agree with the observed global picture.

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Figure 4 shows these lake data (all 76 sites) divided into six geographical regions and normalized to peak SCP input (maximum SCP accumulation rate or concentration = 1.0) to take account of the variations in the scale of contamination between and within regions. While there are only limited data for the southern hemisphere and north Africa, the marked increase in SCP contamination at c. 1950 is clear in every region. Across Europe and North America, this is indicated by a rapid increase in SCPs over and above a gradual but already increasing level of contamination. In north Africa and Asia, where industrial development occurred later, this is more often observed as the start of the record. This geographical shift in global black carbon emissions (i.e., from North America and Europe to Asia) from the mid-late 20th century onward has also been reported from modeling studies.[41]

Although the presented data are derived solely from lake sediment records, atmospherically deposited contaminants also accumulate in ice cores, peat sequences, and marine sediments. However, SCP data in the literature for these other natural archives are very sparse. While a number of papers consider historical profiles of various components of the black carbon continuum in ice cores,[42,43] the various definitions of black carbon cover more than just high-temperature fossil-fuel-derived particulates, such as SCPs, and to my knowledge, only a single ice core study to date specifically refers to SCP accumulation rate data.[32] This ice record only covers the period 1900−1995 but does show the rapid increase in SCP accumulation rate in c. 1950. SCP data for marine sites are restricted to coastal sediment concentrations. In all available locations, marine sediment SCP temporal trends reflect regional lake sediment profiles, indicating that SCP accumulation rates would likely also record the increase in SCP input in the mid-20th century. Similarly, SCP concentration profiles from accumulating peats are also reported to reflect adjacent lake sediment profiles, and the mid-20th century rapid increase in U.K. peat sequences has been used to provide a chronological control as a result.[31]

In proposing a stratigraphic marker for a new geological epoch, it is of benefit to demonstrate long-term preservation.[19] Because SCPs are only produced from high-temperature industrial sources, they are first recorded in the mid-19th century, and although they currently show no signs of decomposition or deterioration, the earliest particles are only a maximum of 170 years old. However, carbon-rich particles with a "bubble texture" thought to have been formed by bolide impact into organic-rich geology c. 65 Myr BP have been observed at the Cretaceous−Paleogene (K−P) boundary. These are thought to have been produced by low-temperature (300−800 °C) expansion of volatiles but not ignition[44] and, therefore, effectively represent a pre-combustion SCP-type particle. While easily distinguishable from fly ash, these "precursor SCPs" look structurally similar and have also been identified in Bronze Age peat sequences dated to c. AD 550.[45] The source of these latter particles remains to be determined but may be due to smelting processes. If SCPs preserve similarly well (and there is no reason to suggest that they will not), then they would easily meet the criterion for a long-term stratigraphic marker.

SCPs provide a more robust indicator for the mid-20th century Great Acceleration than many other potential contaminant markers. They provide a less ambiguous signal than trace metals, where changes to the weathering of natural sources[46] or local and regional activities can provide dramatic changes to concentrations across a variety of time periods (e.g., Bronze Age[47] and late 18th century[48]). As particulates formed from black carbon, they are not susceptible to photochemical and microbial degradation as are persistent organic pollutants[49,50] and other organic chemicals (e.g., pharmaceuticals[51,52]) nor are they affected by selective dissolution, as can be the case for magnetic minerals,[53] which may also be derived from a range of industrial and non-industrial sources.[54] The SCP rapid increase feature is the same and unidirectional across a wide range of site types and archives, unlike the signals that may be recorded by nitrogen isotopes, whereby upland systems experiencing enhanced nitrogen inputs from atmospheric sources show a decline in δ^{15}N values,[55] while those in lowland areas receiving increased nitrogen from agricultural inputs often show an increase in δ^{15}N.[56] Furthermore, unlike some radionuclides, they have no half-life precluding them from being long-term indicators (e.g., ^{137}Cs half-life = 30.17 years), while the extent and chronologies for other potential anthropogenic markers, such as plastiglomerates (the combination of melted plastic and lavas[57]) and technofossils,[58] remain to be fully evaluated.

As a marker for industrial fossil-fuel combustion, SCPs directly represent a globally distributed driving force of the Great Acceleration. They therefore provide not only a highly effective stratigraphic marker but are also a proxy for the rate and scale of anthropogenic global change. In conclusion, while further research is required to confirm temporal patterns in some regions and in the marine environment, the increase in SCP contamination shows an unambiguous, synchronous, and global stratigraphic indicator for the mid-20th century across a range of natural archives, making them a robust, near-ideal marker for the Anthropocene.

■ ASSOCIATED CONTENT

Supporting Information

Names, locations (latitudes and longitudes), and data sources for all lakes referred to in the paper (SI Tables 1 and 2) and SCP concentration profiles for lakes C1−C5 on radiometric chronological axes (SI Figure 1). This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*Telehone: +44-0-207-679-0543. Fax: +44-0-207-679. E-mail: n.rose@ucl.ac.uk.

Notes

The authors declare no competing financial interest.

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