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Abstract

Spheroidal carbonaceous particles (SCPs) are carbonaceous fly ash particles produced solely from industrial fossil fuel combustion. SCPs in sediments can be an indicator of past industrialization. We examined the sedimentary SCP record in Beppu Bay, southern Japan, and compared this record to historical trends of industrial activity and monitoring data for atmospheric pollutions in the region. Beppu Bay has varved sediments in the absence of bioturbation, providing an ideal situation for SCP study. Our results show that the temporal variation of SCP deposition is consistent with the trends of industrial activity and the temporal variation of status of atmospheric pollution. We conclude that undisturbed sediments such as Beppu Bay sediments have SCP records that adequately represent historical trends of industrial activity and atmospheric pollution in detail. These findings support that SCPs are a potential marker for the Anthropocene, characterized by industrialization.

Keywords

Spheroidal carbonaceous particles (SCPs), fly ash, industrial activity, atmospheric pollutions, Anthropocene, Global boundary Stratotype Section and Point (GSSP)

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Introduction

Spheroidal carbonaceous particles (SCPs) are carbonaceous fly ash particles with a size approximating several to tens of microns; they originate from industrial high-temperature fossil fuel combustion (Rose, 2001). Importantly, these particles are produced solely from industrial activities and are well preserved in various sediments owing to their carbon-rich composition. Consequently, SCPs are found only in modern sediments, especially sediments from the late twentieth century, which generally yield a large quantity of SCPs (Rose, 2015; Swindles, 2015), which is taken to indicate global industrialization during this period. In that respect, SCPs are regarded as a primary defining marker for the Anthropocene period, resulting from the "Great Acceleration" of global industrialization and associated factors (Waters et al., 2016, 2018; Zalasiewicz et al., 2017). Sedimentary SCPs have been studied for several decades (e.g., Griffin and Goldberg, 1979, 1981; Wik et al., 1986; Wik and Renberg, 1991; Rose and Juggins, 1994; Rose, 1995; Rose et al., 2004;Nagafuchi et al., 2009; Engels et al., 2018; Inoue et al., 2020), and their temporal records represent past industrial activity in the respective regions.

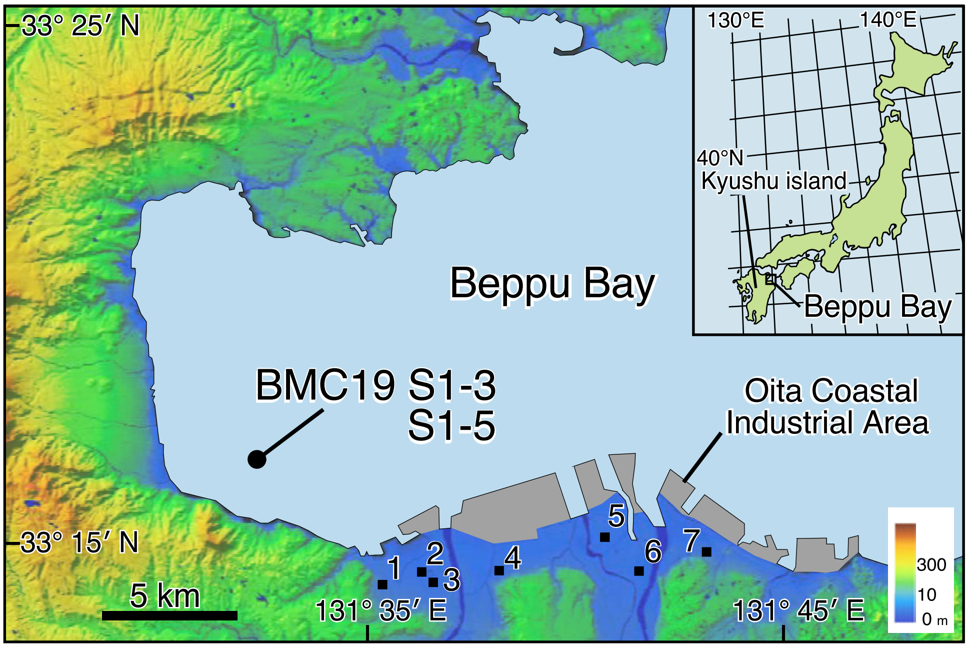

Figure 1. Map of Beppu Bay (and location within Japan) showing sites for BMC19 S1-3 and S1-5 cores. The gray region in the map shows the Oita Coastal Industrial Area. Black squares with numbers 1–7 indicate the sites of monitoring stations for atmospheric pollutants. The monitoring data are shown in Figure 5. The base is the topographic relief map of Oita Prefecture (modified from the Geospatial Information Authority of Japan, https://www.gsi.go.jp/kankyochiri/degitalelevationmap_kyushu.html).

Beppu Bay in southern Japan (Figure 1), the focus of this study, exhibits autumnal oxygen depletion in its bottom waters (Kameda and Fujiwara, 1995), resulting in the formation of sediment varves due to the absence of bioturbation (Kuwae et al., 2013). Radioactive dating of these sediments shows a high and constant sedimentation rate of ~1 cm years^{-1} in surface sediments, which provide undisturbed proxy records with high temporal precision (^{137}Cs, Kuwae et al., 2013; Takahashi et al., 2020; polychlorinated biphenyls, Takahashi et al., 2020, dichlorodiphenyltrichloroethane, Nishimuta et al., 2020; brominated fire retardants, Hoang et al., 2021; and microplastics, Masumoto et al., 2018). Such proxy records suggest that Beppu Bay sediments meet the requirements to act as potential candidate Global boundary Stratotype Section and Point (GSSP) for the basement of the Anthropocene or auxiliary stratotypes (Kuwae et al., 2022). Distinctive sediment characteristics are also ideal for SCP studies, enabling the assessment of annual SCP deposition and comparisons between the SCP record and monitored atmospheric pollutants and historical trends of industrial activity in the study region. Detailed information on past industrial activity in the region has been recorded, especially for thermal power plants, which are a primary source of SCP emissions. Comparing sedimentary SCP records to the industrial trends may show a relationship between SCP deposition and industrial activities. In addition, detailed monitoring data for atmospheric pollutants (e.g., sulfur oxides and deposited dust) cover the past over 50 years are available. Combining the sedimentary SCP record and monitoring data enables us to assess the contribution of the sources of SCP emissions to these pollutant emissions. As noted above, Beppu Bay sediments provide an ideal, well-contextualized situation for SCP study to assess how SCP records represent past industrial activities and atmospheric pollution conditions.

Herein, in order to clarify the pattern of SCP deposition in Beppu Bay for approximately 100 years and to identify its sources, we examined the concentrations and chemical compositions of SCPs in cores collected from the bay in 2019 (Figure 1). The SCP record in Beppu Bay was compared to those in other regions across the globe, showing similarities and the regional differences between records. SCPs, as well as other proxies in Beppu Bay sediments, show that the sediments record significant environmental changes related to modern human activities from the middle of the 20th century.

Materials and Methods

Sampling locations and sediment cores

We collected sediment cores BMC19 S1-3 and S1-5 over a depth of 69.1 m in Beppu Bay (33°16′23.5″N, 131°32′21.5″E and 33°16′23.8″N, 131°32′22.2″E, respectively; Figure 1) on September 10, 2019, using a 120 cm gravity corer (Ashura, RIGO Co. Ltd., Saitama, Japan). These cores consist primarily of hemipelagic silty clay-rich sediments with high-density varve interbedding event layers, high-magnetic susceptibility, and coarse grains. Since these event layers may be found anywhere in the deepest areas of Beppu Bay, cores collected in the area can be correlated based on lithological and stratigraphic features identified *via* visual inspection, CT imaging, and magnetic susceptibility (Kuwae et al., 2013). In the current study, we used the BMC19 S1-5 core for SCP analysis (Figure 2). We established the chronology of the core (i.e., an age-depth model) based on the ages of the event layers (i.e., layers with high-density, high-magnetic susceptibility, and coarser grains, which were probably formed by floods or earthquakes) of Ev 0a (AD 1924), −1a (1967), −1b (1994), −1cU (2005), and −1dU (2013) (Supplementary 1), assuming that mass deposition rates (g cm^{-2} year^{-1}) are constant between the layers. The ages of event layers were obtained from the chronology of core BMC19 S1-3 according to a constant rate of supply model obtained *via* ^{210}Pb dating (Takahashi et al., 2020). The age–depth model of core BMC19 S1-5 shows that the core covers the last 100 years (Figure 2).

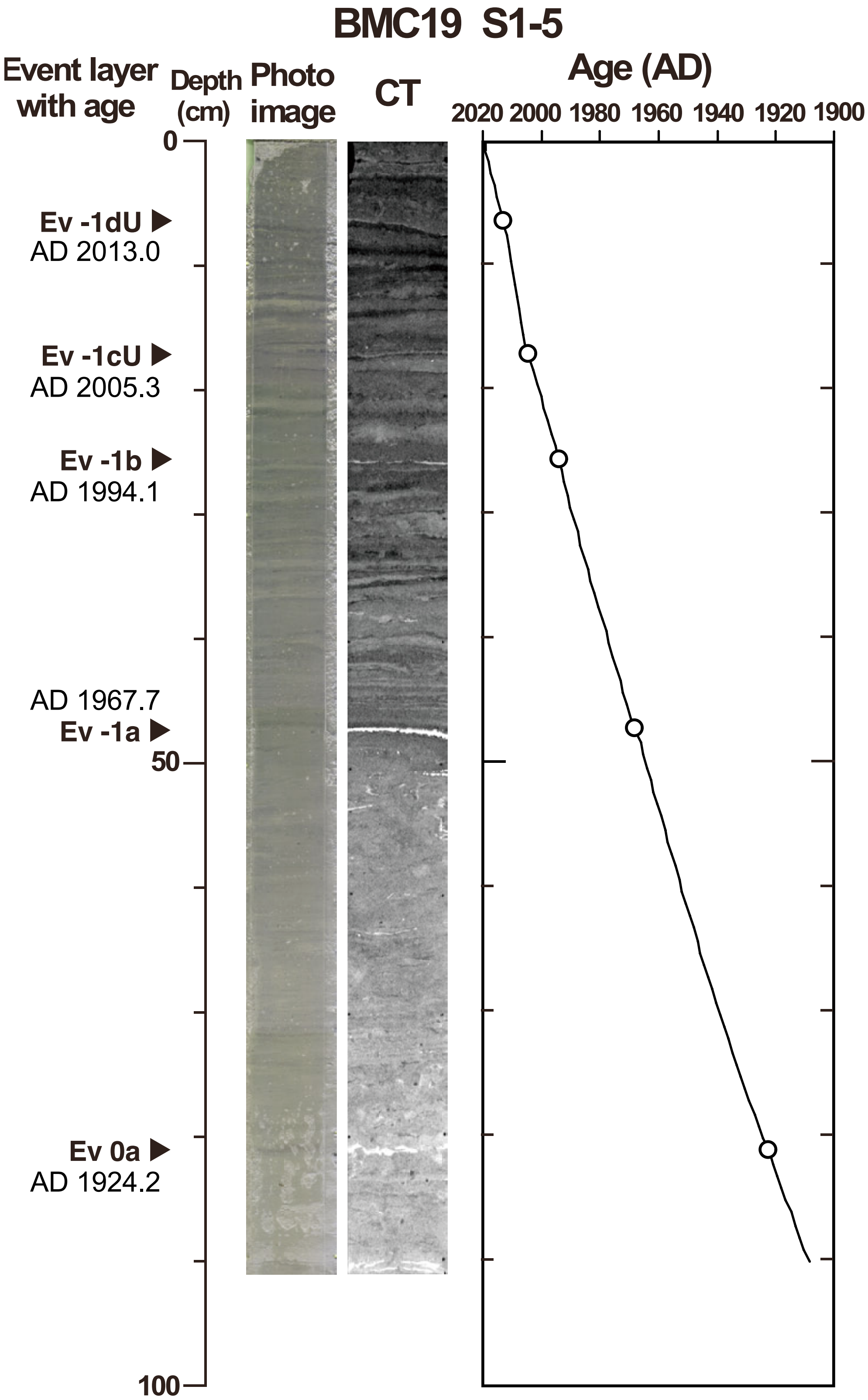

Figure 2. Photograph and computerized tomography (CT) image of BMC19 S1-5 collected in Beppu Bay and its age–depth model constructed on the basis of the ages of event layers Ev 0a, −1a, −1b,−1cU, and −1dU.

SCP analysis

For SCP analysis, we used 0.1–0.2 g of dry sediments from subsamples of 1 cm thickness taken continuously throughout the core, with the exception of a subsample from a depth of 47–48 cm, which corresponds to event layer −1cU, which is composed of sand. For SCP extraction, dried sediment subsamples were chemically digested using HNO_3, H_2O_2, HF, and HCl following the methods of Momose et al. (2012) and Inoue et al. (2014), which represent as modified versions of the method proposed by Rose (1994). Approximately 10% of the residue after the treatments was dropped onto a membrane filter with a diameter of 47 mm and a porosity of 0.45 μm and filtered using a suction filtration device. Areas covered by the residue were mounted onto a glass slide for microscopic observation. This preparation enables us to observe objects under both transmitted and incident light using microscopy. This method was originally applied to the observation of charcoal fragments in sediments (e.g., Inoue et al., 2018; 2021).

We counted the number of SCPs in all areas covered by the residue using a microscope under both transmitted and incident light at 400× magnification. Criteria for SCP identification followed Rose (2008); SCPs were identified as particles with black color, rounded appearance, three-dimensional shape, and porous features, as well as using more detailed criteria, i.e., particles with the aspect ratio between 0.7 and 1.0, and greater than one half of a particle if the particle is broken. Although SCPs are characterized by porous features, pores are not necessarily observed under transmitted light (Figure 3a), as noted by Rose (2008). In order to identify the porous features, we observed SCPs using both transmitted and incident light for the particle identification. Observations under incident light aid in recognizing pores and other features of particles (Figure 3b).

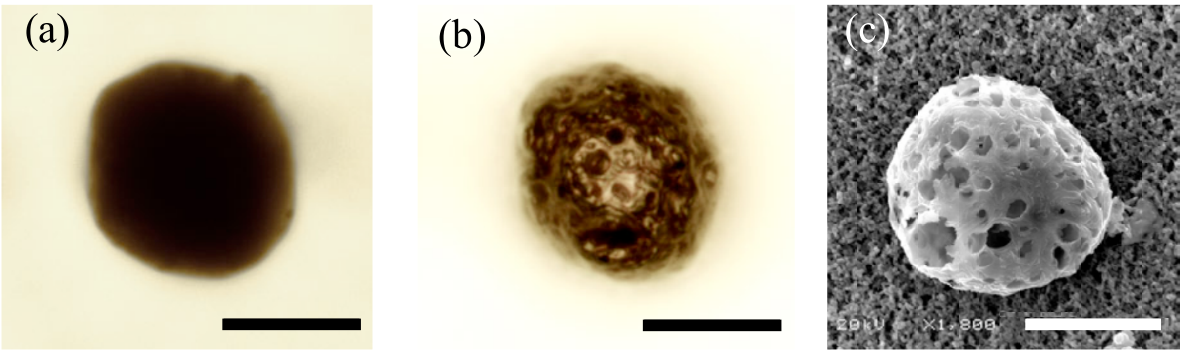

Figure 3. Light microscopy images under transmitted light (a) and incident light (b) and SEM image (c) of SCPs. The image under incident light is an all-in-focus image generated from multiple images. Scale bars in the images show 20 μm.

We counted the number of particles on the slides of the subsamples from the surface (0–1 cm in depth) to the depth at which no SCPs were found in four consecutive samples. Consequently, we counted the particles on the slides of subsamples from 0 to 78 cm depth. SCP concentration was calculated based on the weight of dry sediments used for analysis, the proportion of the residue dropped onto the filter, and the number of SCPs counted on the filter. SCP flux was calculated according to the concentration and the mass sedimentation rate, based on the following equation:

SCP flux (number of particles cm^{-2} year^{-1}) = SCP concentration (number of particles g^{-1}) × mass sedimentation rate (g cm^{-2} year^{-1})

The mass sedimentation rate was estimated using the ages of event layers and the volumetric density of subsamples with a 1 cm thickness.

SCP elemental analysis was conducted for seven subsamples at depths of 1–2, 9–10, 21–22, 32–33, 44–45, 49–50, and 55–56 cm. For SCP elemental analysis, a portion of the residue after chemical treatment was dropped onto the membrane filter, and a part of each membrane was used after coating a part of each filter with a thin carbon layer. Subsequently, both observational and elemental SCP analysis were performed using a JEOL JSM-5500 scanning electron microscope (SEM) equipped with an EDAX–energy-dispersive spectroscopy (EDS) system (APOLLOX detector; Genesis Software) at the Department of Geosciences, Osaka City University, Osaka, Japan. The SEM was operated at an accelerating voltage of 20 kV and a beam current of 500 pA. We used EDS to quantitatively analyze Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, and Zn concentrations. The ZAF-correction method was used for quantitative analysis. We restricted SCP identification to rounded particles with many pores (Figure 3c), and 20–30 particles were analyzed in each sample. We also measured the sizes of particles examined by elemental analysis using the corresponding SEM images.

Results

Concentrations, fluxes, and chemical compositions for SCPs in the BMC19 1-5 core are shown in Figures 4 and 5, respectively. Patterns of variation in SCP fluxes are similar to those of SCP concentrations (Figure 4) since the mass sedimentation rates of the core are almost constant (~0.2 g cm^{-2} year^{-1}).

The SCP records of the core exhibit the first appearances of SCPs around 1940, after which gradual increases are observed until around 1960 (concentrations of 200–2000 particles g^{-1} and fluxes of <400 particles cm^{-2} year^{-1}). The maximum concentration reaches over 30,000 particles g^{-1} (the fluxes of 6000 particles cm^{-2} year^{-1}) in the terminal 1960s, during a period of rapid increase. Between the 1970s and the 1980s, the values fluctuate with time (10,000–20,000 particles g^{-1} and 2000–4000 particles cm^{-2} year^{-1}) and are generally higher than those in subsequent periods. SCP concentrations and fluxes (values) generally decline from the early 1990s to the late 2010s. SCP values in these decades are relatively low (<~2000 particles cm^{-2} year^{-1}) compared to those between 1970 and 1990.

The chemical compositions of SCPs over seven periods (subsamples) are characterized as follows. Chemical characteristics of SCPs in the early 1960s, early 1970s, early 1980s, late 1990s, early 2000s, early 2010s, and late 2010s are similar; i.e., most of the particles are characterized by high percentages of sulfur (20–90 wt%) and chlorine (10–40 wt%), except for some particles with a higher percentage of silicon, aluminum, magnesium, and sodium. In contrast, the chemical compositions of particles in the late 1950s differ. Approximately half of the particles are characterized by high percentages of sulfur and chlorine, similar to those in other periods. However, several particles contained more aluminum and silicon (10–20 wt%) than other particles did, and some particles are characterized as titanium-rich (up to 80 wt%). The sizes of SCPs over these seven periods are largely <20 μm, and the particle size averages in the respective ages ranged from 11.3 to 14.9 μm.

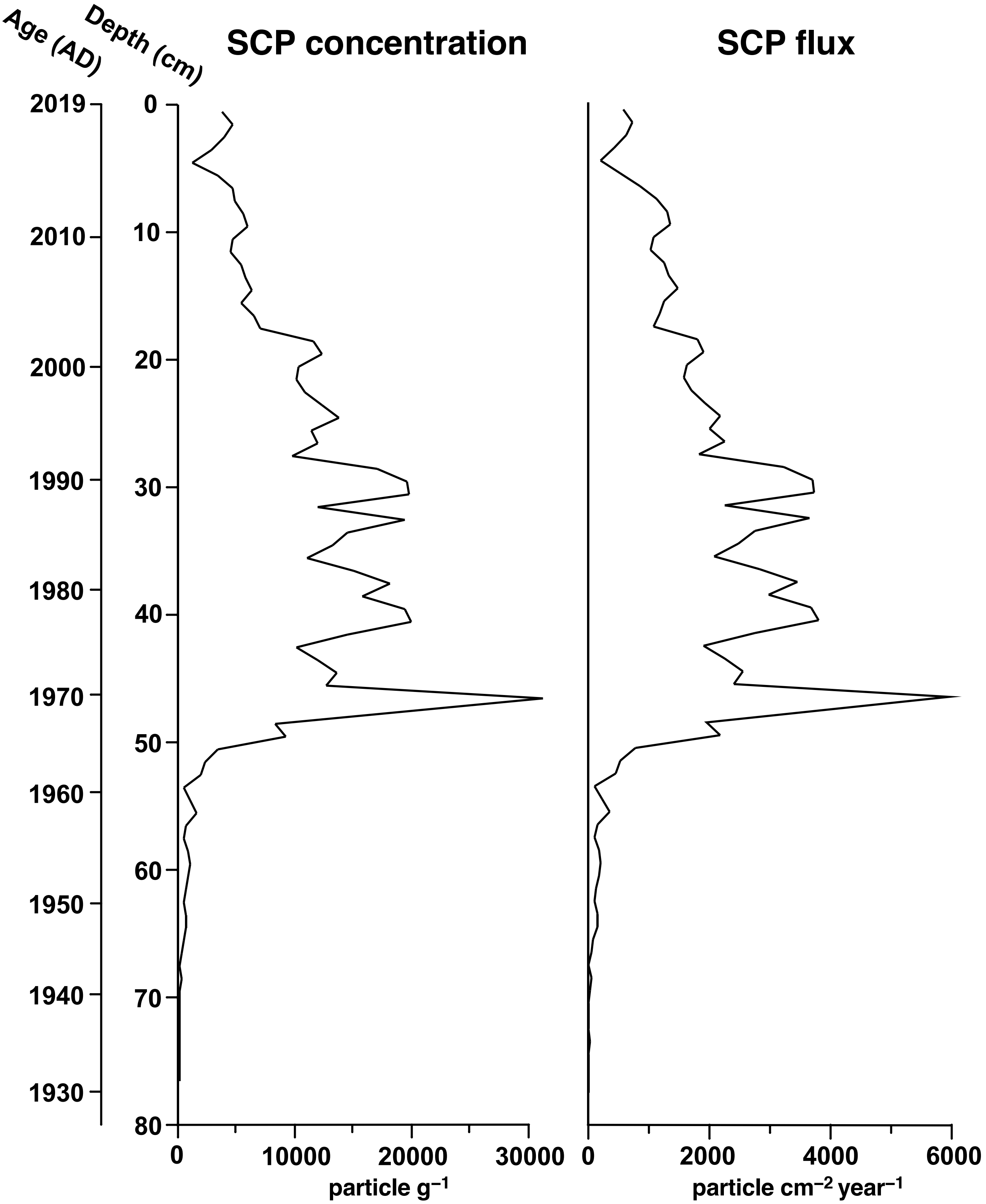

Figure 4. Concentrations and fluxes of SCPs in BMC19 S1-5 core collected from Beppu Bay.

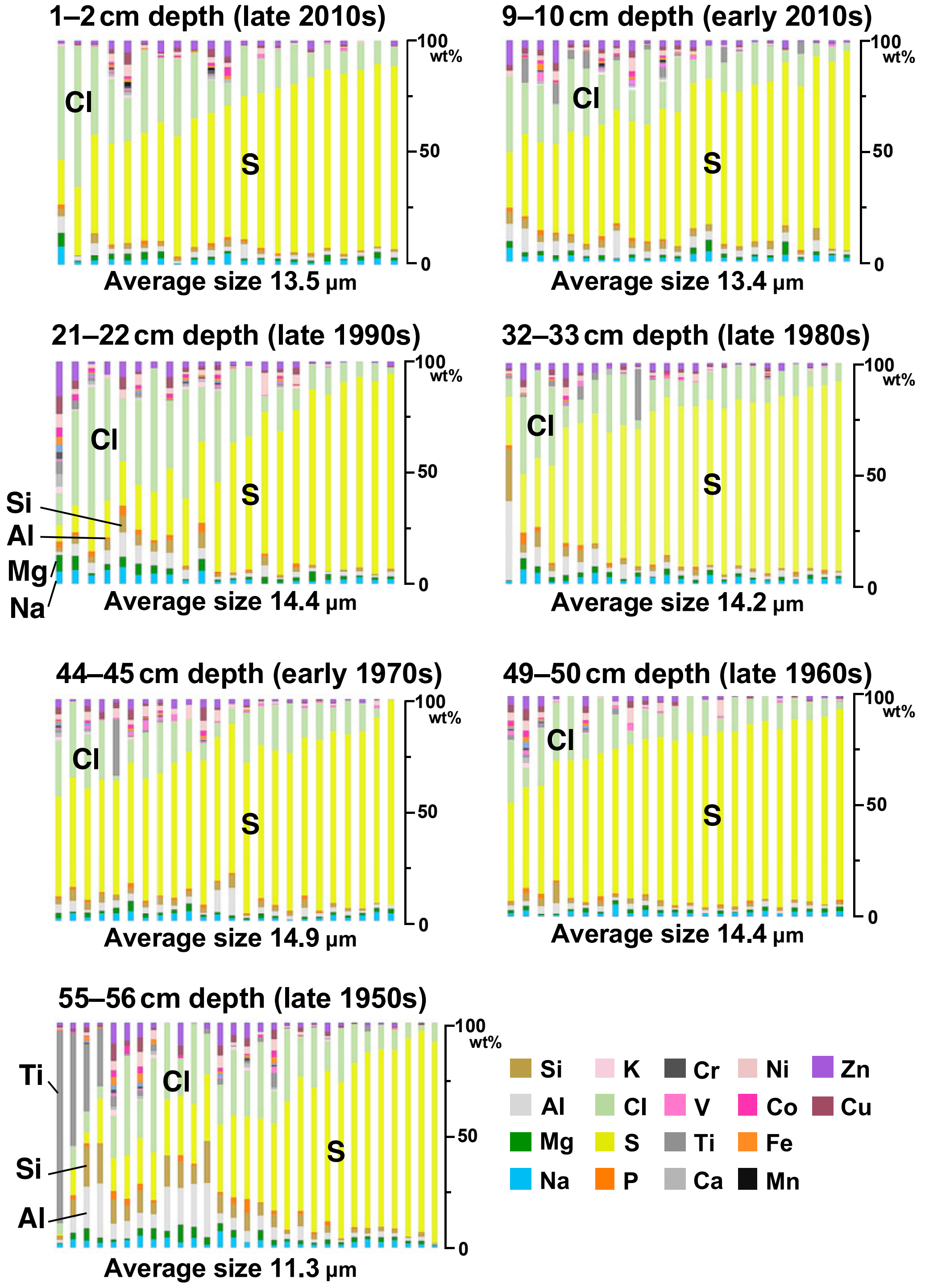

Figure 5. Elemental compositions of representative SCPs in seven subsamples of the BMC19 S1-5 core and their average size in each subsample.

Discussion

SCP records indicating factors behind temporal variations in particle emissions

To assess the relationship between SCP records and industrial trends in the study region, we compared the SCP record in Beppu Bay sediment (Figure 6B) to historical trends of industrial activities in the Oita Coastal Industrial Area nearby (Figure 6A), and annual power generated from oil thermal plants on Kyushu Island (Figure 6C).

The onset of SCP deposition in Beppu Bay dates to the 1940s, with SCP fluxes showing a slight increase until 1960 (Figure 6B). The chemical compositions of particles in the late 1950s show that half were enriched in sulfur, one-third were enriched in aluminum and silicon, and others were enriched in titanium (Figure 5). Variations in particle chemical composition suggest that the particles were likely derived from different fuel combustion processes. Previous studies have shown that oil-fired power plants are prone to emitting S-rich SCPs (Alliksaar et al., 1998; Rose et al., 1996, 1999; Murakami-Kitase et al., 2010). In contrast, Inoue et al. (2014) showed that in China, SCPs in surface sediments around 2010 were enriched in silicon and titanium, which are considered to have been emitted from coal-fired sources. Therefore, in the current study, S-rich particles in the late 1950s are presumably derived from oil-fired sources with high-temperature combustion, whereas Si-rich and Ti-rich particles throughout this period possibly reflect emission from coal-fired sources.

SCP records show a rapid increase in SCP fluxes from 1960 until around 1970 (Figure 6B). Comparing the SCP flux to industrial events in the study region (Figure 6A and B) shows that during this period, oil refineries with boilers, oil thermal plants, and petrochemical complexes with boilers started operating in 1964, 1969, and 1969, respectively, in the Oita Coastal Industrial Area within Beppu Bay (see Figure 1). Particle characteristics during this period are consistent with those assumed to be emitted from these facilities. In addition, rapid increases in SCP fluxes are consistent with increased power generation from oil thermal plants on Kyushu Island during the late 1960s (Figure 6C). Thus, the rapid increase in SCP fluxes is presumably due to the onset of the operation of these facilities. Decreases in SCP fluxes during the early 1970s corresponds closely to the installation of electrostatic precipitators in oil thermal plants in 1973, which were designed to control atmospheric particulate matter (Figure 6A). The decrease is therefore likely caused by the installation of this equipment, causing SCP fluxes to decline by half. Rose (2001) noted that dust arresters significantly reduced SCP emissions from thermal power plants.

The SCP record shows fluctuations between the middle 1970s and the earliest 1990s (Figure 6B). The fluctuation pattern roughly corresponds to that of power generation from oil thermal power plants on Kyushu Island (Figure 6 C); high SCP fluxes in the late 1970s and late 1980s correspond to relatively high power generation during those periods, whereas low SCP fluxes in the early 1980s correspond to relatively low power generation. Thus, SCP fluxes throughout these periods might largely depend on the operation status (i.e., power generations) of oil thermal power plants. Fluctuation in SCP fluxes and variations of power generation from oil during this period are possibly related to the fact that, on Kyushu Island, the primary energy source changed from thermal to nuclear power, and the primary fuel for thermal power generations changed from oil to LNG and coal since the early 1980s (Kyushu Electric Power Co., Inc., 2021).

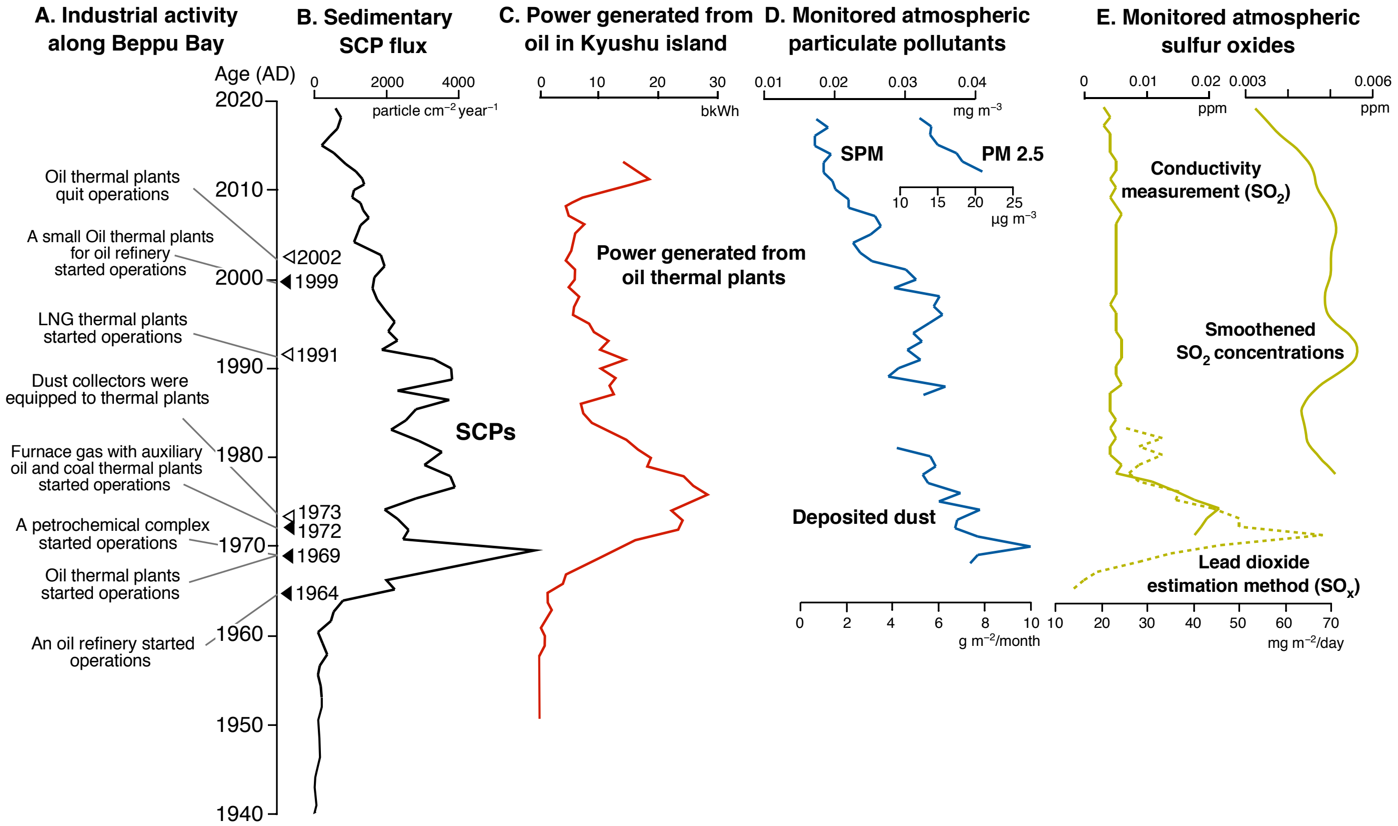

Figure 6. Comparison between SCP fluxes in the BMC19 S1-5 core and other data. (A) Historical trends of industrial activities in the Oita Coastal Industrial Area. Ages with black triangles and white triangles on the age axis show events with potential positive and negative effects on SCP emissions, respectively. (B) SCP fluxes in Beppu Bay sediment core (BMC19 S1-5 core). (C) Annual power generated from oil thermal plants on Kyushu Island (Kyushu Electric Power Co., Inc., 2021). (D) Temporal trends in atmospheric particulate pollutant concentrations or depositions monitored close to the industrial area, i.e., averages of the annual concentrations of suspended particulate matter (SPM) monitored at sites 1, 4, 5, and 7 (the locations of the sites are shown in Figure 1), PM2.5 monitored at sites 1, 4, and 7, and deposited dust monitored at sites 2, 3, and 6. (E) Annual average atmospheric sulfur oxides concentrations monitored at site 7 for over 50 years using the lead dioxide estimation method (SO_x: 1965–1983) and the conductivity measurement (SO_2: 1971–2019) with smoothened values at the far right. Values were smoothened using the LOESS smoothing function of the PAST software (Hammer et al., 2001). Data for SPM, deposited dust, and sulfur oxides are available at the website of the Oita Prefectural Government(https://www.pref.oita.jp/site/toukei/census.html). The data of PM2.5 are available at the website of the National Institute for Environmental Studies (https://tenbou.nies.go.jp/gis/monitor).

Since the 1990s, SCP fluxes have generally decreased, probably representing the reduction in power generations from oil and coal thermal power stations in the study area. Especially from the early 1990s to the early 2000s, SCP fluxes have been almost constant and lower than during the previous decades (Figure 6B). The trends of these fluxes are similar to those of power generation from oil (Figure 6C). High-powered LNG thermal power stations started operating in the study region in 1991 (Figure 6A), resulting in the replacement of primary fuel from oil to

LNG for power generations. Consequently, the operation of oil thermal power stations had been limited and, as a result, their SCP emissions remained low and stable (note that an LNG thermal power station never emits SCPs). Ceasing the operation of oil thermal plants coincided with the decrease of SCP fluxes in the early 2000s or 2002 (Figure 6A and B). Thus, limited SCP fluxes since the early 2000s were likely emitted from other facilities. Some particles with certain percentages of aluminum, silicon, and other elements from the early 2000s (Figure 5) were possibly derived from coal consumption in furnace gas with auxiliary oil and coal in another thermal plant. Throughout the 2000s, coal became the primary fuel for thermal plants on Kyushu Island, possibly resulting in the relative increase in coal-derived particles observed at this time.

The temporal variations in SCP deposition in the study region can be summarized as follows. SCP deposition started in the 1940s. Until 1960, SCP deposition was limited, particles being derived from various sources with coal or oil consumption. During the 1960s, SCP deposition increased rapidly due to the industrialization of the region, i.e., the construction of coastal industrial areas with oil thermal plants and an oil refinery. Rapid decreases in SCP deposition in the early 1970s presumably occurred due to the installation of dust collectors in thermal plants. Between the early 1970s and the earliest 1990s, the SCP record shows fluctuations in SCP deposition, mirroring the power generations dominating the region. Low SCP deposition since the early 1990s and especially since the early 2000s was likely caused by the replacement of the primary energy for thermal plants and the cessation of operation of thermal plants, respectively.

We conclude that the following suite of events caused the increases and decreases of SCP deposition in the study region. Events increasing deposition are 1) oil thermal operation, 2) oil refinery operation, and 3) increasing power generation in thermal power plants. Events decreasing deposition are 1) use of dust arresters in the thermal plants, 2) decrease of power generation in thermal power plants, 3) cessation of thermal power operations, and 4) change of the primary fuel for power generation. As noted above, SCPs are emitted solely from industrial high-temperature fossil fuel combustion (i.e., oil, coal, and oil shale). The primary facility meeting the condition for SCP emission is a thermal power plant, and it has been abundantly demonstrated that such facilities emit particles (e.g., Griffin and Goldberg, 1979; Rose, 1990; Rose et al., 1999). Indeed, in the current study, the operation period (1969–2002) of the oil thermal power plants corresponds to the period of relatively high SCP fluxes or concentrations. SCP fluxes during this period are generally more than double those of other periods, indicating that more than half of SCP emissions can be attributed to thermal power plants. SCP fluxes during this period significantly depend on the equipment of the thermal power plants (electrostatic precipitators) and changes in power generation within the context of changes in the energy policy.

Although SCPs are emitted from high-temperature industrial sources, SCP sources other than thermal stations have been poorly identified. In the Czech Republic, Vukic et al. (2006) identified boiler houses at uranium mining plants as SCP sources. Inoue et al. (2013) considered that SCPs are also emitted from oil refineries. In the current study, the onset of the rapid increase in the SCP values dates to the early 1960s, corresponding to the period prior to the operation of the oil thermal power plant. In the early 1960s, only the oil refinery with boilers operated in the study region as an industrial facility. Thus, the initial rapid increase in SCP values can presumably be attributed to the initial operation of the oil refinery. In an oil refinery, oil is generally heated at most 400℃ through the refinery process, resulting in no SCP emissions; however, these particles are presumably emitted from boilers installed in the facility.

SCP records compared to temporal variations in other atmospheric pollutants

To assess the relationship between SCPs and other atmospheric particulate pollutants in the study region, we compared the SCP record against monitoring records of deposited dust, suspended particulate matter (SPM: particulate matters <10 μm in size), and PM 2.5 close to the coastal industrial area (Figure 6D). In addition, we compared the SCP record to that of atmospheric sulfur oxides monitored over 50 years (Figure 6E), because in the northern UK, SCP sediment trap fluxes are correlated with fluxes of non-marine sulfate deposition (Rose and Monteith, 2005).

The record of deposited dust covering 1968–1981 shows that the peak deposition in 1970 almost coincides with that of SCPs. The SCPs observed in this study range from several to tens of microns; since the size of deposited dust is over ten microns, some large SCPs are presumably classified as deposited dust. The coincidence of peaks is presumably due to that around 1970, SCPs emissions directly contributed to the increase of dust deposition, and/or deposited dust was mainly emitted from the same sources as SCPs, i.e., the oil thermal plants and oil refinery. Decreased dust deposition in the early 1970s is presumably attributed to dust precipitator equipment in the industrial facilities.

The SPM record covers the last ~30 years. Trends in the temporal variation in SPM concentrations correspond to those of SCP fluxes, especially in the case of the decreasing trends since the 1990s. The correspondence of the temporal trends in SPM concentrations and SCP fluxes is possibly due to the fact that the variation in SPM concentrations is partially attributed to the changes in fluxes of SCPs and other particles emitted from the same sources as SCPs.

Although the record of PM 2.5 is limited to the last seven years, there is a clear difference between the trends of temporal variations in SCPs (or SPM) and PM 2.5 during the period. This difference suggests that the primary sources of two particle types are different. The decreasing trend of PM 2.5 during the last decade has been observed throughout Japan, especially Kyushu Island. Many of PM 2.5 found in Kyushu Island in the early 2010s are believed to be due to the long transportation of particles from the Asian continent (e.g., Kaneyasu et al., 2014; Ikeda et al., 2014, 2015), and the decrease in the 2010s presumably results from declining particle emissions in the eastern Asian continent (e.g., Uno et al., 2017; Yin, 2021). The size of SCPs observed using a SEM in this study differs from that of PM 2.5. Inoue et al (2014, 2020) suggested that some SCPs deposited on Okinawa Island and Japanese remote islands during the early 2010s were potentially transported from the Asian continent. However, in the current study, since the chemical compositions of particles in the early 2010s are similar to those in previous periods, most particles were presumably emitted from local sources. Low SCP fluxes over recent decades suggest that particles transported over long distances have no or very little influence on SCP deposition in the study region.

Atmospheric sulfur oxides has been monitored for over 50 years at site 7 (see Figure 1). This record shows that sulfur oxides concentrations increased rapidly in the late 1960s and declined sharply in the 1970s, resulting in a peak around 1970 (Figure 6E). Since 1980, its concentrations have remained low and stable. The coincidence between increases of SCPs and sulfur oxides concentrations in the late 1960s is presumably due to the onset of large facilities conducting fossil fuel combustion, i.e., the oil thermal plants and oil refinery in the region. This is because the primary sources of sulfur oxide emission are assumably large facilities with fossil fuel combustion similar to those of SCPs. The decreases of sulfur oxides and SCP concentrations in the 1970s is probably due to the installation of flue-gas desulfurization equipment in these facilities. In Japan, desulfurization equipment was installed in most facilities with fossil fuel combustion during the 1970s due to the restriction of sulfur oxides emissions by the Air Pollution Control Law, which was revised in 1974. The smoothened values of SO_2 obtained *via* conductivity measurements are shown on the far right in Figure 6E. The variation of these values is similar to that of SCP fluxes, probably indicating that the little amount of sulfur oxides has been emitted from the same sources as SCPs even after installing desulfurization equipment and dust precipitators in the facilities.

SCP record in Beppu Bay compared to the records in other regions

Rose (2015) and Swindles et al. (2015) suggested that global sedimentary SCP values increased markedly around 1950, this increase coinciding with the Great Acceleration, i.e., the onset of the proposed Anthropocene Epoch. As noted above, SCP records vary between regions, i.e., their first appearance, rapid increases, and concentration peaks do not always indicate exact timings. However, in general, the first rapid increases occur in the mid-20th century, and the peaks rise in and after the third quarter of the century (Rose, 2015). In the Asian regions, the rapid increase largely occurred in the 1960s, as represented by the second peak in the density plot of rapid increases in global SCPs (Swindles et al., 2015). The rapid increase of SCPs in Beppu Bay in the late 1960s slightly lags behind the other industrial regions and the onset of Great Acceleration (assumed to be 1950), reflecting a lag in the acceleration of industrialization in the region behind the other industrial regions.

SCP records in developed and other countries show that few SCPs are found in sediments deposited in the 21st century, as also shown by the current study, probably due to reduced emissions following changes in the source of power generation (e.g., nuclear power, natural energy including solar energy, wind power generation, and natural gas for thermal power generation) related to policies for the reduction of CO_2 emissions. Although coal-fired power plants are still being built in some regions, since such policies will be further pursued, SCP emissions will continue to decline and may fall to zero within this century, even on a global scale. In that case, abundant SCPs and the concentration peak will be found solely in the late 20th and early 21st centuries in various types of sediments across the globe. In this respect, SCPs can play a role as an index fossil for the last and this century.

Conclusions

We examined the sedimentary SCP record in Beppu Bay, southern Japan, and compared this record to historical trends of industrial activity and monitoring data for atmospheric pollutions in the region. We obtained the following SCP record. SCP deposition started around 1940 and increased slightly until 1960. Subsequently, deposition rapidly increased in the 1960s and fluctuated between the early 1970s and early 1990s. Since the 1990s, deposition has been low relative to previous decades and has further decreased since the early 2000s. The chemical compositions of SCPs indicate that prior to 1960, particles were derived from both oil and coal combustion sources; by contrast, since 1960, particles have been emitted mainly from oil combustion sources. Comparing this record to the industrial trends and monitoring data shows that the temporal trend of SCP deposition and its source fuels represents the regional industrial trends and events in the study region. Additionally, this trend is correlated with those of atmospheric particulate pollutants (SPM and deposited dust) except PM2.5 and sulfur oxide in the region. We conclude that the undisturbed sediments possibly have SCP records adequately representing detailed historical trends of industrial activity and atmospheric pollution conditions. In this regard, SCPs are a potential marker for the Anthropocene, characterized by industrialization.

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Supplemental material

Supplemental material for this article is available online.

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