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ORIGINAL ARTICLE

Temporal variations in phytoplankton biomass over the past 150 years in the western Seto Inland Sea, Japan

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Abstract We examined fossil pigments in a ^{210}Pb-dated sediment core to document the temporal variations in phytoplankton biomass over the past 150 years in a semi-enclosed bay, Beppu Bay, in the western Seto Inland Sea, Japan. The flux of fossil pigments was used as an index of phytoplankton biomass, which we reconstructed after removing the effect of post-burial degradation on the concentrations of fossil pigments. The flux doubled from the 1960s to the early 1970s, decreased or remained stable in the early 1980s, and increased again from the late 1980s to the early 1990s. The first increase in phytoplankton biomass during the 1960s was likely caused by eutrophication due to an increase in terrestrial nutrient fluxes from watersheds. The decreasing phytoplankton biomass in the early 1980s was likely related to the establishment of a sewage treatment system that reduced the terrestrial nutrient fluxes to the sea. However, the terrestrial nutrient fluxes could not explain the second increase from the late 1980s to the early 1990s. Intensification of the influx of nutrients from the shelf slope to the sea was likely the cause of the second increase in phytoplankton biomass. This is supported by the inverse relationship between phytoplankton biomass and sea level at the shelf slope, the latter being an index of the intensity of the influx of oceanic nutrients from the shelf slope to the sea. The supply of oceanic nutrients may be therefore a critical factor in the determination of primary production in the western Seto Inland Sea.

Keywords Primary production · Pigments · Eutrophication · Kuroshio current · Palaeoceanography · Seto Inland Sea

1 Introduction

In recent decades, inland sea ecosystems have been severely impacted by nutrient inputs associated with anthropogenic activities in their watersheds (Carpenter et al. 1998; Cloern 2001; Yamamoto 2003). Excessive nutrient inputs from watersheds have led to eutrophic conditions in coastal seas and induced the frequent occurrence of red tides, which have become a widespread social problem (Cloern 2001). For example, in the Seto Inland Sea, red tide blooms have been apparent since the 1960s, their occurrence being contemporaneous with increased inputs of terrestrial nutrients due to industrialization and urbanization in the watershed (Honjo 1993; Takeoka 2002; Yamamoto 2003).

The input of oceanic nutrients from shelf slope waters, however, can also be an important determinant of primary production in the coastal ocean (Prezelin et al. 2004; Nakano et al. 2005). Field observations have demonstrated that primary production in shelf waters along the Japanese Pacific coast is greatly stimulated by nutrients from the shelf slope (Ozaki et al. 2004; Nakano et al. 2005). It has also been demonstrated that the intrusion of oceanic water dramatically changes nutrient concentrations and the biomass of the dominant phytoplankton in shelf waters

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(Kaneda et al. 2002a; Hayami et al. 2004; Kuroda et al. 2010; Nakano et al. 2005). Nutrient concentrations within the inland sea, in addition to those in shelf waters, also show remarkable year-to-year variability that is concordant with variations in nutrient inputs from the shelf slope (Sugimatsu and Isobe 2010). Many studies have drawn attention to the importance of nutrients supplied to the Seto Inland Sea from the ocean (e.g., Fujiwara et al. 1997; Takeoka 2002; Hayami et al. 2004). Those studies have demonstrated that, along with the input of nutrients from land runoff, the supply of nutrients from the offshore ocean may intensify primary production in the inland sea. However, all of these studies have been limited to a timeframe shorter than several years. Therefore, we actually do not know how variations in oceanic and terrestrial nutrient supplies over a timeframe longer than a few decades affect primary production in a coastal or inland sea (Ozaki et al. 2004; Yamamoto 2003). Understanding how such long-term variations affect lower tropic level ecosystems is useful for developing good strategies for the management and rehabilitation of coastal and inland aquatic ecosystems (Boesch 2002).

Long-term monitoring data are necessary to elucidate how inland-sea ecosystems respond to nutrients supplied from the land and ocean. However, inland-sea monitoring programs were not started until the 1970s, when eutrophication was already recognized as a problem (Yamamoto 2003). Consequently, those monitoring programs cannot provide the data needed to assess environmental conditions before the occurrence of eutrophication.

A paleoceanographic approach using appropriate proxies in sediment cores can potentially enable the productivity in oceans to be reconstructed (Cooper 1995; Chmura et al. 2004; Rabalais et al. 2007). Fossil pigment of chlorophyll derivatives derived from phytoplankton and preserved in coastal sediments is used to reconstruct the phytoplankton biomass that existed in the past (Chmura et al. 2004; Rabalais et al. 2007). Some studies have successfully reconstructed past phytoplankton biomass in coastal ecosystems and identified the main factors that controlled historical changes in biomass, such as the effects of nutrient loads from urbanizing watersheds (Chmura et al. 2004; Rabalais et al. 2007).

Beppu Bay is a suitable site for investigating the effects of oceanic and terrestrial nutrient supplies on primary production in an inland sea because the bay enables us to obtain undisturbed sediment cores that reflect high sediment accumulation rates (Kuwae et al. 2013) and because it is very close to the Bungo Channel, a conduit to the Pacific Ocean that is frequently stimulated by nutrient inputs from the shelf slope (Fig. 1b; Kaneda et al. 2002a). In this study, we applied the paleoceanographic approach to the analysis of sediment collected from Beppu Bay in the western Seto Inland Sea to examine long-term changes in phytoplankton biomass.
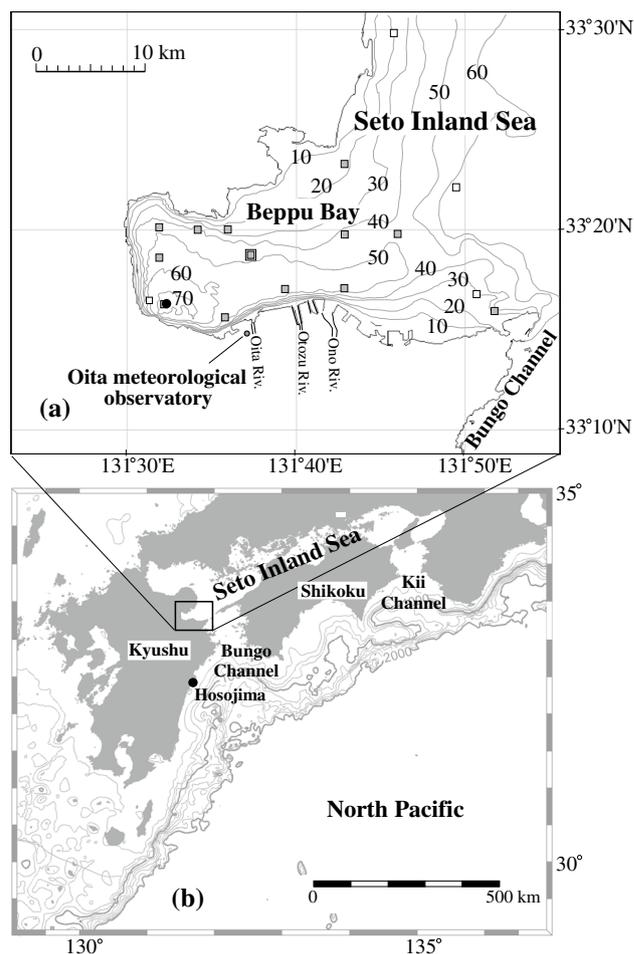
Fig. 1 Bathymetric maps (contour intervals in m) showing **a** the location of the coring site (*filled circle*) and **b** the location of Beppu Bay in the western Seto Inland Sea, Japan. *Open and closed squares* in **a** show monitoring sites where measurements of nutrients (*closed*) and chlorophyll *a* concentrations (*open*) were made in the bay (see Sect. 3.3)

We analyzed fossil chlorophyll *a* pigments in the sediment from Beppu Bay to (1) reveal the temporal variations of phytoplankton biomass in the western Seto Inland Sea over the past 150 years, and (2) identify the causes of those changes.

2 Study site

Beppu Bay is a semi-enclosed bay in the western Seto Inland Sea, Japan (Fig. 1). The basin of the bay has a maximum depth of 72 m in its innermost region and a deep sill at its mouth, where the water depth is ~50 m. These bathymetric features result in highly stagnant bottom-water conditions (e.g., Kameda and Fujiwara 1995; Kuwae et al. 2007) and enable the bay to provide undisturbed sediment cores that reflect high sediment accumulation rates (Kuwae et al. 2013).

The main rivers that flow into the bay are the Oita and Ono rivers (Fig. 1a). The combined discharge from these rivers allowed us to examine the effects of terrestrial nutrient inputs on primary production. On the other hand, Beppu Bay is also a suitable site for investigating the effects of oceanic nutrient supplies on primary production in the inland sea because the bay is very close to the Bungo Channel, a conduit to the ocean that is frequently stimulated by nutrient inputs from the shelf slope (Fig. 1b; Kaneda et al. 2002a).

3 Materials and methods

3.1 Sampling and chronology

We used a gravity corer (model HRL, Rigo Co. Ltd., Saitama, Japan) to collect an undisturbed sediment core (BG091010-1) from the innermost region of Beppu Bay in September 2009 (water depth 71.5 m; 33°16.4958′N, 131°32.1845′E). The core sample was undisturbed during the coring process and carefully sliced at 1-cm intervals from the surface to the bottom. Aliquots of the sliced samples were sealed in shielding bags and stored at -20 °C for analysis of fossil pigments. Core chronology was determined by the constant initial concentration (CIC) method of ^{210}Pb dating (Robbins and Edgington 1975). The validity of the ^{210}Pb dating was checked by comparison with the peak in ^{137}Cs activity, which is associated with the fallout maximum of 1964 in Japan (Hirose et al. 2008a). The activities of ^{210}Pb, ^{214}Pb, and ^{137}Cs were determined by γ-spectrometric analysis of dried sediment. Dried samples were sealed in standard holders for two weeks to allow ^{222}Rn and its short-lived daughter (^{214}Pb) to equilibrate. Radioactivities were determined with a high-purity Ge detector (GXM25P, EG&G ORTEC, Tokyo, Japan) equipped with a multichannel analyzer (MCA7700, Seiko EG&G, Tokyo, Japan). The activity of ^{210}Pb$_{supported}$ was estimated by measuring the activity of ^{214}Pb, whereas the activity of ^{210}Pb$_{excess}$ was determined from the difference between the total and supported ^{210}Pb $\left(^{210}\text{Pb}_{excess} = {}^{210}\text{Pb}_{total} - {}^{214}\text{Pb}\right)$.

3.2 Fossil pigments

Concentrations of fossil pigments were quantified by high-performance liquid chromatography (HPLC). The frozen sediments were placed in 10-mL glass vials. HPLC-grade acetone was added (3 mL), and the pigments were extracted at 0 °C during agitation in an ultrasonic bath (Tani et al. 2009a, b). The extracts were collected after centrifugation at 2000 rpm for 5 min. These procedures were repeated at least three times for each sample until the supernatant was completely colorless. The combined extract was evaporated to dryness under N$_2$, dissolved in 3 mL diethyl ether, and washed with an aqueous solution of NaCl (1 M). After evaporating the ether phase to dryness under N2, the residue was dissolved in 200–500 μL of acetone together with an internal standard, mesoporphyrin IX dimethyl ester (Sigma–Aldrich, St. Louis, MO, USA), and analyzed by HPLC (LC-10AD, Shimadzu, Kyoto, Japan) using a photodiode array detector (SPD-M10AVP, Shimadzu). A Wakopack Navi C30-5 reverse-phase column (4.6 mm diameter and 250 mm length; Wako Pure Chemical Industries, Ltd., Osaka, Japan) was used for separation. Mobile-phase solvent A consisted of a mixture of acetonitrile and water (90:10 by volume); solvent B was 100% ethyl acetate. A linear gradient from 100% A to 100% B over 40 min was followed by an isocratic hold for 10 min at 100% B. The flow rate was 1.0 mL min^{-1}. Pigments analyzed in this study were chlorophyll a (a major photosynthetic pigment ubiquitous in all taxonomic algae) and its derivatives. Pheophytin a and pyropheophytin a were identified based on their retention times, absorption spectra, and quantified using commercially available reference compounds and absorption coefficients, as shown in Tani et al. (2002). More hydrophilic chlorophyll a derivatives with absorption spectra similar to pheophorbide a (pheophytin a) were eluted before intact chlorophyll a when analyzed by the C30-5 reversed-phase HPLC. We did not identify these derivatives because of a lack of commercially available reference compounds for them. In this study, we therefore grouped these derivatives together as "pheophorbide a" (Nara et al. 2005) and calculated the total concentration of them under the assumption that they have the same specific absorption coefficient as pheophorbide a. The sum of chlorophyll a and its derivatives (pheophytin a, pyropheophytin a, and "pheophorbide a") is denoted here "Chl-a."

3.3 Meteorological and in situ data

We used the meteorological, physical, chemical and biological monitoring data to examine the effects of nutrients from the land and shelf slope on the reconstructed fluxes of Chl-a into the sediments, and to test the reliability of the reconstructed Chl-a fluxes.

Meteorological data, including annual averages of air temperature and wind speed, and the monthly sum of precipitation and sunshine duration were obtained from the Oita Local Meteorological Observatory, which is located on the south coast of Beppu Bay (Fig. 1). At the Oita Observatory, air temperature, precipitation, and daylight hours have been recorded since the late nineteenth century, whereas wind speed has been recorded since 1951. Because the Japan Meteorological Agency changed the methods used to measure daylight hours and wind speed in 1985 and

1975, respectively, we used information on daylight hours and wind speed only after 1985 and 1975, respectively.

There are two major rivers that flow into Beppu Bay: the Oita and Ono rivers (Fig. 1a). The monthly flow volume in each river (Oita and Ono; the Funai-Ohashi and Shirataki-Bashi observation sites, respectively) was obtained from the database of the Water Information System (http://www1.river.go.jp/) operated by the Japanese Ministry of Land, Infrastructure, Transport and Tourism (Ministry of Land, Infrastructure, Transport and Tourism 2008).

Concentrations of dissolved inorganic nitrogen (DIN) and orthophosphate (PO_4-P) in Beppu Bay from 1973 to 2003 were obtained from a regular survey conducted by the National Research Institute of Fisheries and Environment of the Inland Sea. Monthly measurements of DIN and PO_4-P concentrations have been made at 13 sites (closed squares in Fig. 1a) in the bay since 1973. Concentrations of chlorophyll *a* in Beppu Bay were obtained from the Japanese Ministry of the Environment. Measurements of chlorophyll *a* concentrations have been made at four sites in Beppu Bay (open squares in Fig. 1a) as a part of an area-wide comprehensive water-quality survey since 1981. Terrestrial nutrient inputs of total nitrogen (TN) and phosphorus (TP) into the Seto Inland Sea were obtained from Sekine and Ukita (1997) and SECA (2001).

The sea level data at Station Hosojima on the Pacific Ocean side of Kyushu Island (Fig. 1b) since 1894 were obtained from the Japan Metrological Agency. The sea level at Hosojima Station is associated with the strength of intrusions of cold, nutrient-rich bottom water from the shelf slope onto the shelf and the thermal contraction of the water column along the coasts of western Kyushu and southern Shikoku (Fujiwara et al. 2004). Because the intrusion of cold slope water occurs primarily in the summer (Kaneda et al. 2002a), we used the average sea-level anomalies (i.e., the differences from the long-term monthly means) in early summer to fall (May–October) as proxies for nutrient supply from the slope region offshore the Bungo Channel. In addition, the bottom water temperatures in early summer to fall at an offshore site near the sampling station (double square in Fig. 1a) were used as a proxy for the historical variation in the strength of bottom intrusion from the shelf slope. The bottom-water temperatures since 1979 were obtained from the monthly survey conducted by the National Research Institute of Fisheries and Environment of the Inland Sea.

3.4 Data processing

Chl-*a* concentrations per unit of dry sediment mass were converted to fluxes (mg m^{-2} year^{-1}) using mass accumulation rates obtained from ^{210}Pb dating (see Sect. 4.1). To check the reliability of the reconstructed Chl-*a* fluxes, we

investigated the relationship between the fluxes and the monitoring chlorophyll *a* concentrations in Beppu Bay since 1981. The time resolution per sample of the sediment core after the 1980s was about 2–3 years depending on the sediment depth. Thus, we averaged concentrations of chlorophyll *a* for the years that correspond with the sediment samples that were used to estimate the fluxes. To elucidate the effects of nutrients supplied from the shelf slope on temporal variations in phytoplankton biomass in Beppu Bay, we compared sea level anomalies from early summer to fall at Station Hosojima and the reconstructed Chl-*a* anomalies (i.e., differences from the long-term mean) through the sediment core. Prior to analysis, the fluxes of reconstructed Chl-*a* were log(*x*)-transformed to stabilize the variance, where *x* is the flux of reconstructed Chl-*a*.

4 Results

4.1 Lithology, ^{137}Cs and ^{210}Pb$_{excess}$ radioactivity, and age–depth model

Our sediment core was almost constant in terms of grain size except for the turbidite layers (as described below), and the lithology was characterized by finely laminated black and olive-gray silty clay above a depth of about 20 cm, but below that it was characterized by massive olive-black or grayish olive silty clay (for a detailed account of the lithology in the Beppu Bay sediments, see Kuwae et al. 2013). Many turbidite layers were recognized in the Beppu Bay sediment and thought to be formed as a result of earthquakes, tsunamis, and flooding in the region surrounding the bay (Kuwae et al. 2013). These layers were characterized by high density associated with high magnetic susceptibility (Kuwae et al. 2013). In our sediment core, there were also high-density, high-magnetic-susceptibility layers at depths of 19–21, 37–39, 42–44, and 84–90 cm. We removed these layers for dating purposes due to the rapidity of the sedimentation event (Kuwae et al. 2013) and thereafter expressed the sediment depth as the corrected depth for subsequent analysis.

The fact that the vertical profile of ^{210}Pb$_{excess}$ in the sediment core decreased exponentially with mass depth (Fig. 2a) suggests that the sedimentation rate was constant and that there had been no vertical disturbance. Under such conditions, the CIC model (Robbins and Edgington 1975) is the most appropriate for determining sediment age (Andersen 2006). The ^{210}Pb$_{excess}$ activities plotted against the corrected depth of sediment decreased systematically from 0.24 Bq g^{-1} in the surface layer to 0.016 Bq g^{-1} at a core depth of 31 cm. Based on the CIC age model (Robbins and Edgington 1975), the average mass accumulation rate of the sediment was estimated to be 0.0713 g cm^{-2} year^{-1},

Fig. 2 Vertical profiles of **a** $^{210}Pb_{excess}$ and **b** ^{137}Cs activity (Bq dry g^{-1}) for the sediment core in Beppu Bay. **c** Age–depth model based on the constant initial concentration model of Robbins and Edgington (1975)
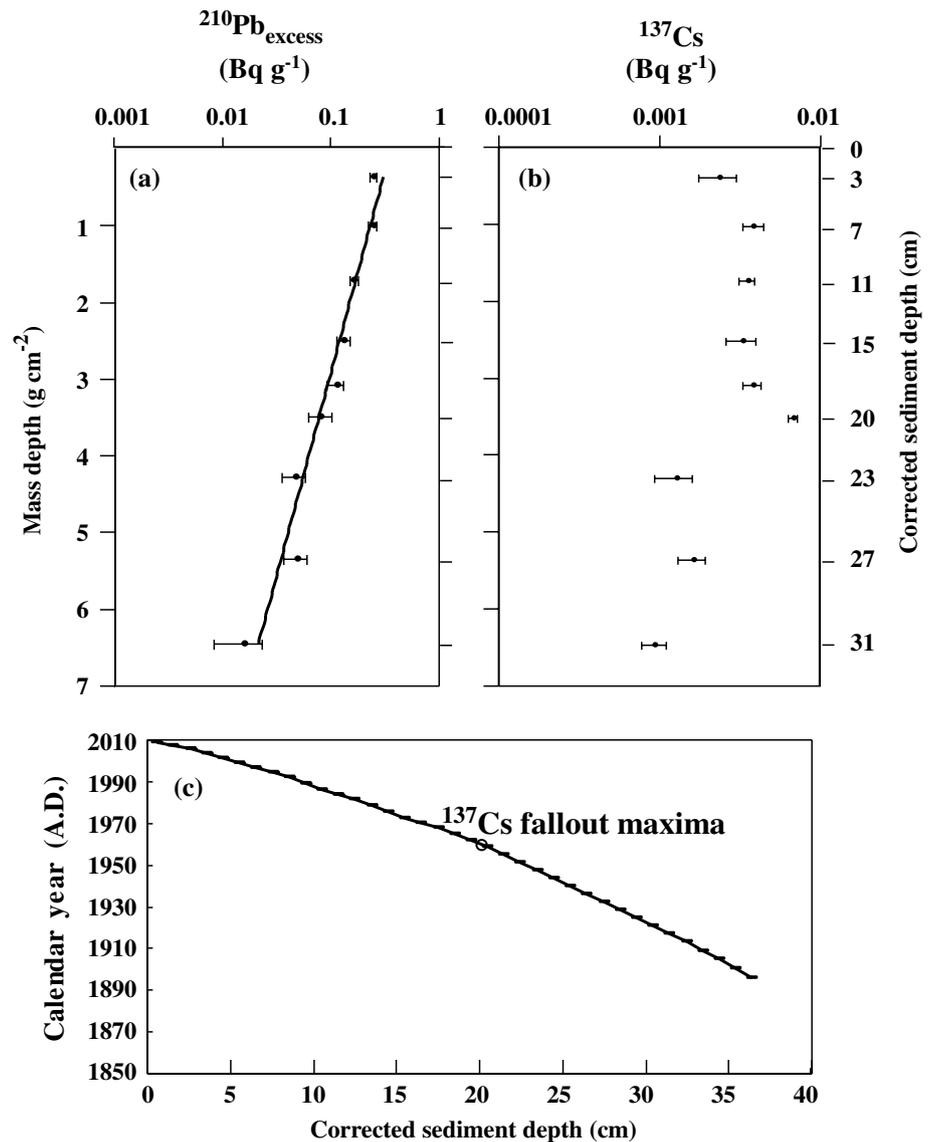
and the calendar year ranged from 1896 at a core depth of 36.5 cm to 2009 at the surface. Because the ^{137}Cs fallout maximum in Japan occurred in 1963 (Hirose et al. 2008a), the peak in ^{137}Cs activity at a depth of about 20 cm (Fig. 2b) is considered to have occurred in the early 1960s. This timing agrees well with the age estimated by ^{210}Pb dating (1963 AD, Fig. 2c). As described before, lamination was visible in the uppermost 20 cm above the horizon at the peak of ^{137}Cs activity. In the corresponding layers of the cores collected near the site of the core employed in this study, the thicknesses of laminae were almost constant (Kuwae et al. 2013), probably due to constant formation of annual varves. The lithological evidence implies that the sedimentation rate and dilution effect were almost unchanged after the 1960s, which is consistent with the linear trend in $^{210}Pb_{excess}$.

4.2 Fossil pigments and temporal variations in phytoplankton biomass

In the Beppu Bay sediment, pheophytin *a*, pyropheophytin *a*, and "pheophorbide *a*" were detected as chlorophyll *a* derivatives (Fig. 3a). Pheophytin *a* and pyropheophytin *a* comprised around 60% or more of the total Chl-*a* (chlorophyll *a* and its derivatives) throughout the core, and "pheophorbide *a*" comprised the remainder (9–44%). The concentration of Chl-*a* in our core was low prior to the 1950s (Fig. 3b), but there was a small amount of temporal variation prior to the 1950s, although the magnitude of the variation was small compared to the variations after the 1960s. The Chl-*a* concentration in the 1960s reached levels 2.3-fold higher than those prior to the 1950s. In the 1970s, the Chl-*a* concentrations decreased but remained higher than

Fig. 3 Age profiles of **a** the relative abundance of the concentration of the biological proxy common to all algal taxa (chlorophyll *a* and its derivatives: "pheophorbide *a*", pyropheophytin *a*, and pheophytin *a*), **b** the concentration of Chl-*a* (chlorophyll *a* and its derivatives), and **c** the fluxes of reconstructed Chl-*a* [Chl-*a* was corrected by removing the effect of post-burial degradation; *dashed and solid lines* represent the fluxes calculated using the first and second methods, respectively; see Sect. 4.2], and **d** the ratio of chlorophyll *a* to pheophytin *a*. The *thick solid line with closed squares* in **c** shows the average of the chlorophyll *a* concentrations during four seasons in the surface water of Beppu Bay based on the regular monitoring data
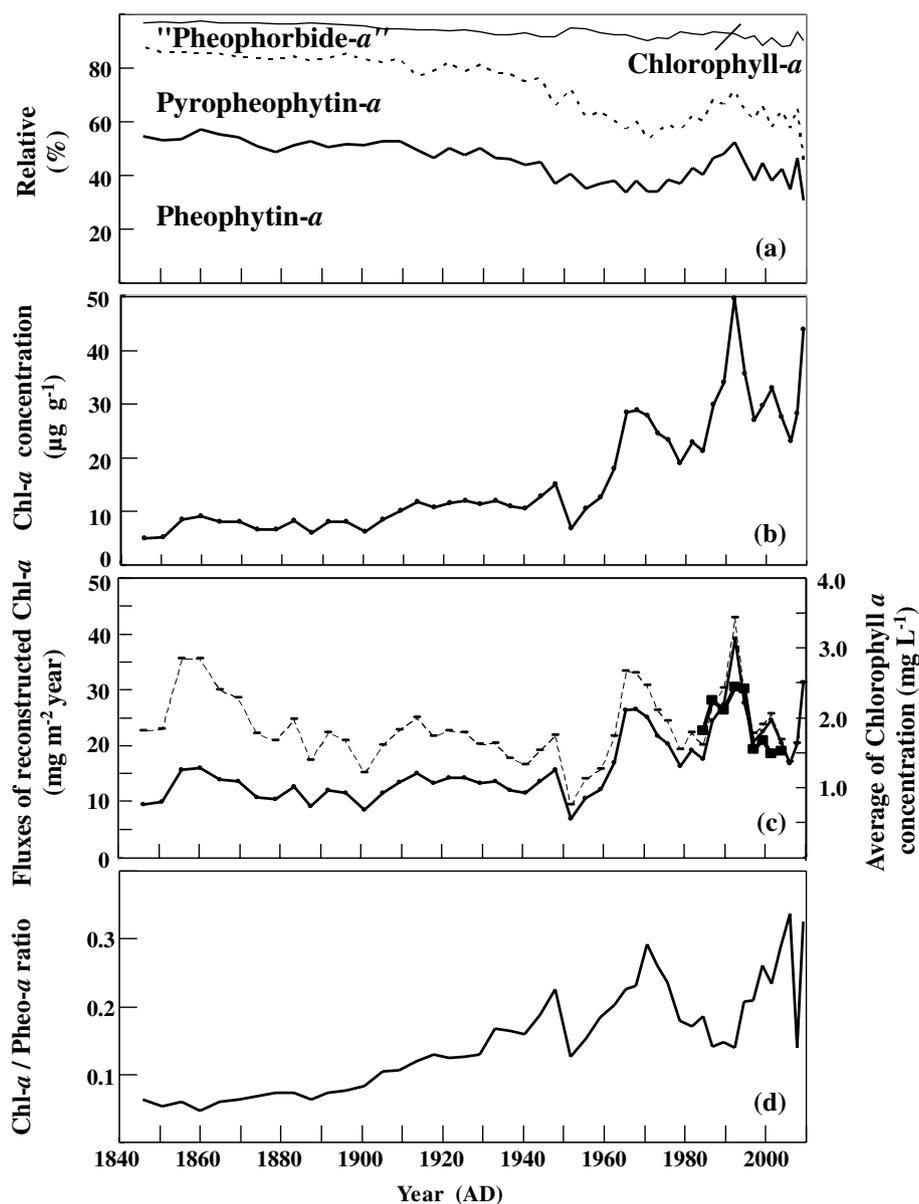
the concentrations during the 1950s. In the early 1990s, the Chl-*a* concentrations again increased greatly.

The temporal variations in the concentrations of Chl-*a* indicate that phytoplankton production rates in Beppu Bay were considerably lower before the 1960s. However, to reach a robust conclusion, we needed to consider biotic and abiotic degradation of Chl-*a* in sediments that may have caused the concentrations to decrease over time (Leavitt and Carpenter 1990; Ming-Yi et al. 1993) and consequently may have led to the apparently low Chl-*a* concentrations before the 1950s. In order to eliminate the effects of degradation on the temporal variations in the Chl-*a* concentration, we calculated a degradation rate based on the assumption that pigment degradation follows first-order kinetics (e.g., Ishiwatari et al. 2009):

$$(Chl - a)_t = (Chl - a)_0 \times \exp[-kt],$$

where $(Chl-a)_t$ is the normalized concentration of Chl-a at time t; t is the number of years after sedimentation of the Chl-*a* (=0 at the sediment surface); and k is a first-order rate constant (year^{-1}). We estimated the first-order rate constant using two calculation methods. First, we used the all data from the late 1840s to 2009 (the surface), as shown in Fig. 3b, yielding the following least squares equation:

$$(Chl - a)_t = 32.0 \times \exp[-0.011t] \ (r^2 = 0.81). \tag{1}$$

Second, we used the data before the 1950s because the rapid increase in Chl-*a* concentration that occurred after the 1960s was probably caused by an intensification of phytoplankton production associated with anthropogenic nutrient

loading—an intensification also seen in other bays of the Seto Inland Sea (Hirose et al. 2008b; Yoshioka et al. 2012). Therefore, including data obtained after the 1960s may bias the estimated effects of degradation. Consequently, we obtained the following least squares equation:

$$(Chl - a)_t = 16.9 \times \exp[-0.0062t] \ (r^2 = 0.53). \quad (2)$$

We then reconstructed the Chl-*a* concentration prior to post-burial degradation (Fig. 3c) using the Chl-*a* concentration at the sediment surface obtained from Eqs. 1 and 2 ([Chl-*a*]$_0$) and the ratio of the measured Chl-*a* concentration in a sample to the normalized Chl-*a* concentration at time *t*, respectively:

$$(\text{Reconstructed Chl} - a)_{\text{sample}}$$
$$= (Chl - a)_0 \times (Chl - a)_{\text{sample}}/(Chl - a)_t \quad (3)$$

Flux of reconstructed Chl − *a*
$$= \text{Reconstructed Chl} - a \times AR, \quad (4)$$

where AR is the mass accumulation rate (0.0713 g cm^{-2} year^{-1}) obtained from the ^{210}Pb-based CIC model (Robbins and Edgington 1975). The temporal variations in the fluxes of reconstructed Chl-*a* calculated using the two methods described above showed obvious increases during the 1960s and around 1990, though only small-amplitude changes occurred before the 1950s (Figs. 3c, 8a). The ratio of chlorophyll *a* to pheophytin *a*, which is indicative of the degree of preservation of labile pigments (Leavitt and Hodgson 2001; Reuss et al. 2010), gradually increased until the late 1940s and then decreased, but again increased towards the 1960s, although the value was lower than 0.23, which was the maximum value before the late 1940s (Fig. 3d). Afterwards, it varied among years by less than 0.23 except during the early 1970s and after the late 1990s. It should be noted that the ratio greatly decreased around 1990. These results suggest that the marked increase in the fluxes of reconstructed Chl-*a* during the 1960s and around 1990 compared to those before the 1940s cannot be fully explained by the degradation effect, but rather can be explained by increased productivity.

4.3 Meteorological, environmental, and oceanographic conditions

Air temperatures in Beppu Bay area have been gradually increasing, especially since (roughly) 1980 (Fig. 4a). The monthly sum of precipitation has varied from year to year but does not exhibit a temporal trend (Fig. 4b). Daylight hours has varied among years, but they decreased slightly from the 1980s to the early 1990s (Fig. 4c). The wind speed decreased and then increased around 1980 but has changed little since that time (Fig. 4d).
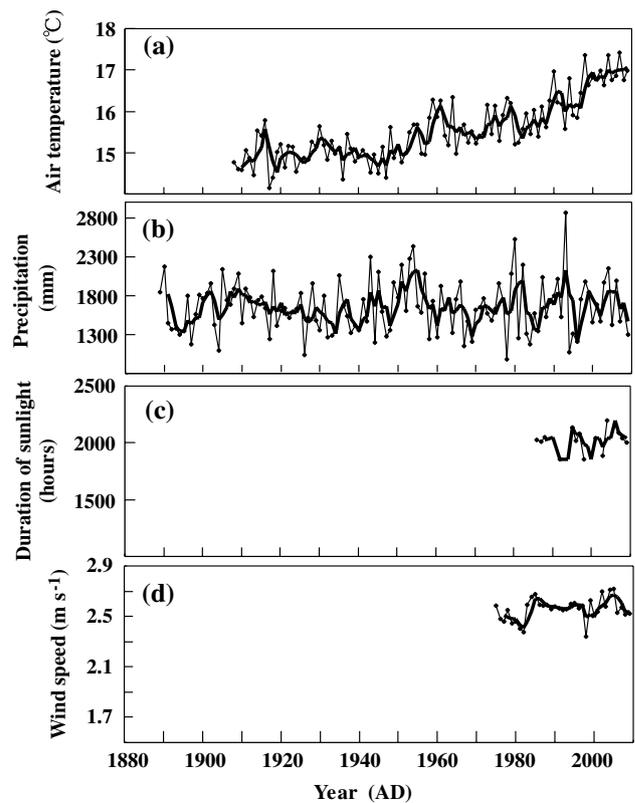
Fig. 4 Long-term changes in **a** the annual average air temperature, **b** the monthly sum of precipitation, **c** the duration of sunlight, and **d** the annual average wind speed at Oita, the site of the meteorological observatory nearest to Beppu Bay, are shown by *thin lines*. In each panel, the 3-year moving average is indicated by a *bold line*

Monthly flow volumes in the Oita and Ono rivers around Beppu Bay are shown in Fig. 5. The volumes of both rivers increased in the early 1980s and around 1993, though they fluctuated slightly in the 1970s, but were about the same before and after the early 1990s.

The DIN concentrations in Beppu Bay were relatively high until the late 1970s, but subsequently decreased. They increased temporarily in the mid-1980s and again increased greatly in the early 1990s (Fig. 6a). The concentrations of PO$_4$-P (Fig. 6b) varied in a similar manner. According to the monitoring data, the annual mean chlorophyll *a* concentration in Beppu Bay was high from the late 1980s to the early 1990s and then decreased abruptly (thick solid line with closed squares in Fig. 3c).

The riverine nutrient inputs to the Seto Inland Sea, such as TN and TP, dramatically increased from the late 1950s to the early 1970s, and then decreased towards the early 1980s (Fig. 7). Since then, the terrestrial nutrient input of TN has barely changed, although it has gradually decreased since the 1990s (Fig. 7a), whereas that of TP has also gradual decreased (Fig. 7b).
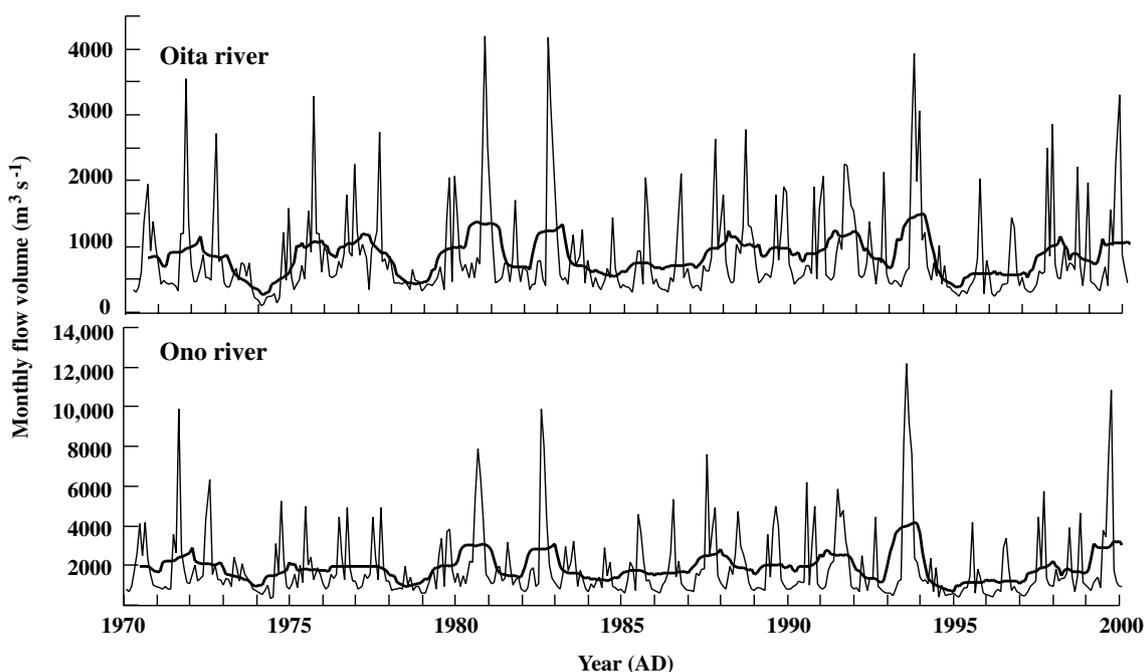
Fig. 5 Monthly flow volumes in the Oita and Ono rivers (*upper and lower panels*, the Funai-Ohashi and Shirataki-Bashi sites, respectively) around Beppu Bay from 1970 to 2000. The *bold line in each panel* shows the 12-month moving average
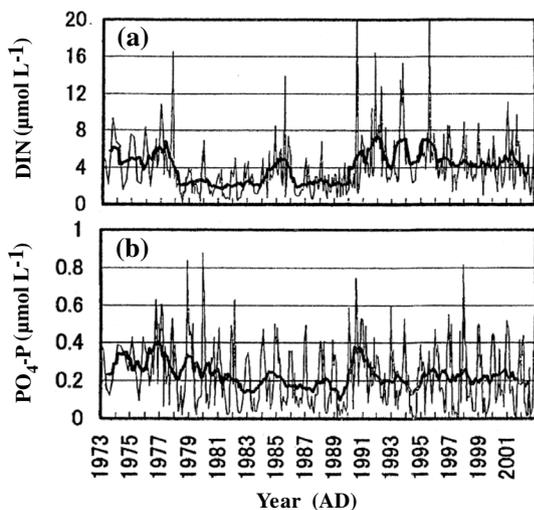
Fig. 6 Long-term changes in the monthly average **a** DIN and **b** PO$_4$-P concentrations in the surface layer (*thin lines*) measured at 13 sites in Beppu Bay from 1973 to 2003. The *thick line in each panel* shows the 12-month moving average

Fig. 7 Historical variations in the terrestrial nutrient inputs of total nitrogen (TN) and phosphorus (TP) to the Seto Inland Sea

The bottom-water temperature from early summer to fall in Beppu Bay showed a decreasing trend from the 1980s to the early 1990s (Fig. 8b). Since then, it has greatly increased, although it has varied among years. The sea level at Hosojima Station changed greatly from 1894 to 2005 (Fig. 8c), but was (similar to the bottom-water

temperature) relatively low during the period from the 1980s to the early 1990s. In addition to that period, the sea level was low around the late 1910s, in the 1930s, and in the 1960s, whereas values were high around 1900, in the

Fig. 8 Anomalies in the **a** fluxes of reconstructed Chl-*a* (corrected by removing the effect of post-burial degradation; *dashed* and *solid lines* indicate the flux anomaly calculated using the first and second methods, respectively; see Sect. 4.2), **b** bottom-water temperatures in early summer to fall at an offshore site near the sampling station, and **c** sea level at Hosojima Station, averaged from early summer to fall (May to October)
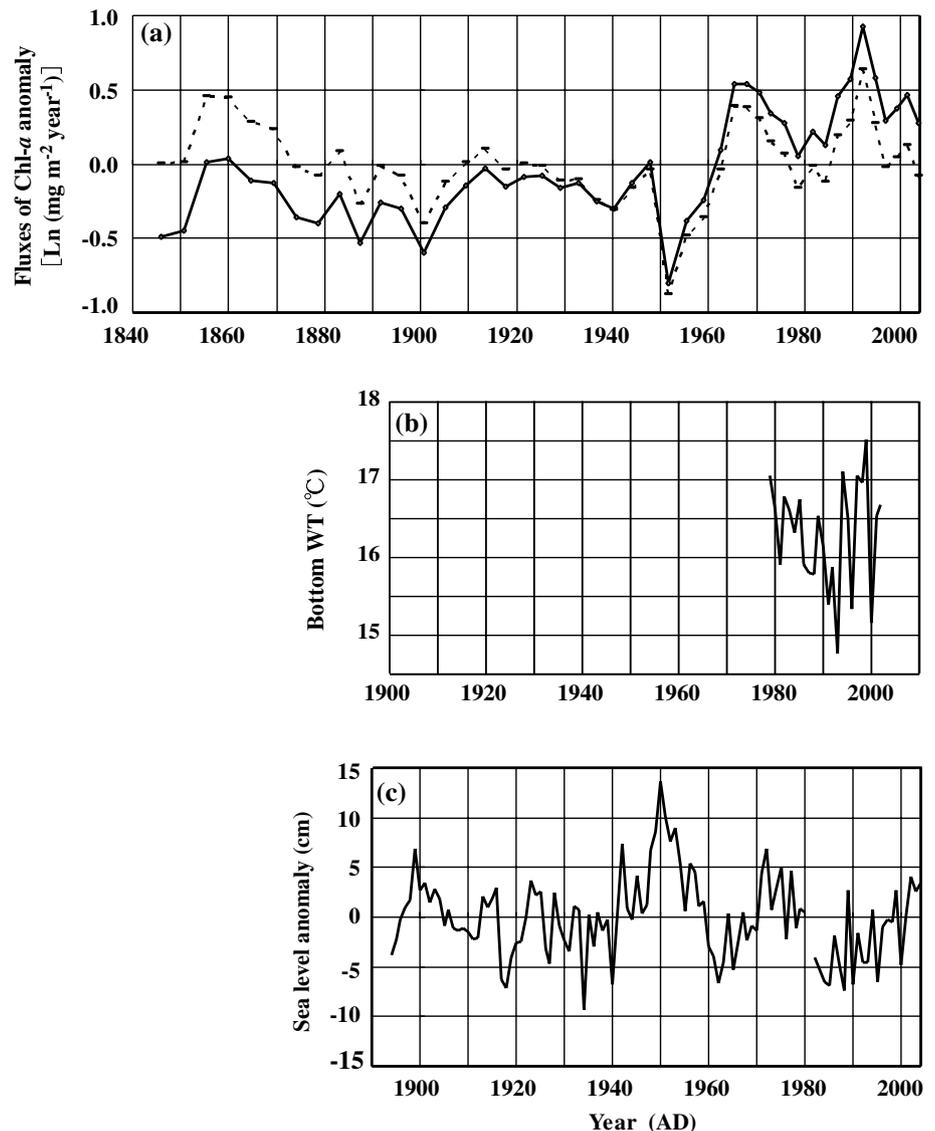
period from the 1940s to the 1950s, and in the early 1970s. The recent period of low sea level is known to be consistent with thermal effects given that the water temperature in the Bungo Channel has also been low recently due to the intensified intrusion of bottom water from the shelf slope into the channel (Fujiwara et al. 2004).

5 Discussion

5.1 Reconstructed primary productivities and the factors that caused changes

The Seto Inland Sea is a major fishing ground in Japan (Imai et al. 2006), and its watershed is one of the most industrialized areas. Many industrial facilities were built in the coastal zone of the sea after 1963, when the New

Industrial City Law was enacted (Takeoka 2002). Industrialization and urbanization since the 1960s have greatly increased terrestrial nutrient inputs to the Seto Inland Sea (Fig. 7; Sekine and Ukita 1997; SECA 2001). This increase in nutrient loading was responsible for a sevenfold increase in the number of red tide blooms between 1965 and 1976 (Okaichi 1997). In accordance with previous reports (Honjo 1993; Okaichi 1997; Yamamoto 2003), the primary productivity in Beppu Bay, as reconstructed from our sedimentary records, was relatively low until roughly 1960 and then increased greatly until the early 1970s. In the Seto Inland Sea, an environmental conservation law was enacted in the late 1970s to reduce nutrient loading from the watershed. Since then, terrestrial nutrient input has reduced (Fig. 7; see also Yamamoto 2003). Indeed, our sedimentary records showed that primary productivity in the bay decreased from the 1970s to the early 1980s, a

time during which nutrient concentrations in the bay also declined (Fig. 6). Honjo (1993) and Yamamoto (2003) have also reported that the frequency of red tide blooms in the Seto Inland Sea decreased after the 1970s, and attributed this decrease to the reduction in nutrient loading from the watershed. The density of flagellate cysts in the sediments of the eastern Seto Inland Sea (Harima-Nada) peaked around 1970 and decreased thereafter (Ichimi et al. 2005). All these results suggest that the degree of eutrophication in the Seto Inland Sea has decreased since the late 1970s.

Interestingly, our results also demonstrated a large increase in Chl-*a* fluxes from the late 1980s to the early 1990s. This temporal pattern of phytoplankton biomass after the late 1980s in Beppu Bay is consistent with monitoring data for chlorophyll *a* obtained by the Japanese Ministry of the Environment (Fig. 3c; $r = 0.774$ and 0.699, $p = 0.014$ and 0.036; dashed and solid lines showing the fluxes calculated using the first and second methods, respectively). Because the limiting factor for phytoplankton growth in this bay is probably nutrients (Horiguchi and Terasawa 2000), and because there was an increase in the nutrient concentrations around the early 1990s in the bay (Fig. 6), it is reasonable that the temporal increase in nutrient concentrations during this period was responsible for the increase in phytoplankton biomass at the same time.

The next question is why the nutrient concentrations increased in the early 1990s. One might suspect that nutrient supply from the watershed increased during this time. However, as mentioned above, an efficient nutrient control program for the Seto Inland Sea had been in place since the late 1970s (Yamamoto 2003), and accordingly, the terrestrial nutrient inputs such as TP and TN clearly started to decrease in the early 1970s, and they were almost constant or tended to decrease before and after the early 1990s (Fig. 7; Sekine and Ukita 1997; SECA 2001). The amount of precipitation and the volume of discharge from rivers into Beppu Bay did not almost change around the early 1990s (Figs. 4, 5). It is thus difficult to attribute the increase in nutrient concentration after the early 1990s to increased nutrient loading from the watershed.

Alternatively, an enhanced supply of nutrients from the shelf slope may have led to the increase in nutrient concentrations around the early 1990s in Beppu Bay. Cold and nutrient-rich water from the shelf slope intermittently intrudes into the bottom waters of the Bungo Channel (Kaneda et al. 2002a), which connects Beppu Bay to the Pacific Ocean. This phenomenon is called bottom intrusion (Kaneda et al. 2002a). When bottom intrusion occurs, nutrient concentrations and phytoplankton biomass change dramatically in the Bungo Channel (Koizumi 1999; Kaneda et al. 2002a; Nakano et al. 2005; Kuroda et al. 2010). For instance, Nakano et al. (2005) demonstrated that phytoplankton biomass in the Bungo Channel increased abruptly

in response to nutrients supplied from the ocean via bottom intrusion, and phytoplankton biomass was higher in a year of frequent occurrence of bottom intrusions than in a year of infrequent occurrence of such intrusions. In Beppu Bay, bottom-water temperatures decreased towards the 1980s and were very low during the early 1990s (Fig. 8b), which was likely due to intensified bottom intrusion from the shelf slope into the bay. Thus, it is likely that the temporary enhancement of nutrient supplies from the shelf slope in the early 1990s raised the nutrient concentrations in Beppu Bay, and consequently stimulated the primary productivity in the bay. Supporting this inference, it is recognized that diatom productivity in not only Beppu Bay (Kuwae, in preparation) but also the Bungo Channel (Kuwae et al. 2006), as estimated from diatom valves in the sediments, increased at around that time. Also, some studies have shown that nutrient concentrations in the eastern parts of the Seto Inland Sea (Osaka Bay; Sugimatsu and Isobe, 2010) and those in the Kii Channel (Ozaki et al. 2004), which connects the Seto Inland Sea and the Pacific Ocean, increased around the same period because of the intensified bottom intrusion from the shelf slope.

5.2 Effects of variability of the Kuroshio axis on primary productivity

Kaneda et al. (2002b) and Kuroda et al. (2010) have proposed that the intensity of bottom intrusions into the Bungo Channel is related to the position of the Kuroshio axis. Bottom intrusion waters originate from the upwelled water on the slope (Kaneda et al. 2002a), and the upwelling is controlled by Ekman dynamics near the bottom beneath the Kuroshio Current (Ichikawa and Kaneda 2000). When the Kuroshio flows near Kyushu, bottom intrusion into the Bungo Channel frequently occurs (Kaneda et al. 2002a). In contrast, when the Kuroshio moves further offshore from Kyushu, bottom intrusions occur infrequently. Therefore, the extent of nutrient stimulation from shelf slope water via bottom intrusions depends on the path of the Kuroshio (Sugimatsu and Isobe 2010).

The coastal sea level reflects the intensity of bottom intrusion because the temperature change associated with cold upwelled water can cause the sea level to vary (Fujiwara et al. 2004; Kuroda et al. 2010). When our data were compared with the sea-level data at Hosojima Station (Fig. 1b), significant negative correlations between the sea level and the fluxes of reconstructed Chl-*a* were detected (Fig. 8a, c; $r = -0.588$ and -0.507, $p = 0.010$ and 0.004; the r values are the correlation coefficients for the flux of reconstructed Chl-*a* calculated using the first and second methods, respectively). The inverse relationship between phytoplankton biomass and sea level implies that the intensity of nutrient inputs from the shelf slope caused by

variations in the position of the Kuroshio axis is an important influence on primary productivity in Beppu Bay. In concordance with our results, Kuwae et al. (2006) has previously reported similar patterns between the sea level at Hosojima and sedimentary records of diatom productivity in Bungo Channel.

6 Conclusions

Sedimentary records showed that phytoplankton biomass in Beppu Bay within the Seto Inland Sea increased greatly from the 1960s to the 1970s and decreased or remained stable in the early 1980s as a result of the establishment of a sewage treatment system that reduced the influx of terrestrial nutrients to the sea. However, phytoplankton biomass increased again from the late 1980s to the early 1990s. This increase was probably caused by an influx of nutrients from the shelf slope as a result of intensified frequency of bottom intrusion. This inference is supported by the inverse relationship between the reconstructed phytoplankton biomass and the sea level at Hosojima, which is a proxy for the intensification of bottom intrusion (Fujiwara et al. 2004). Because the sea level on the southeast coast of Japan varies on a decadal timescale that is related to oceanic currents such as the Kuroshio (Senjyu et al. 1999), the results presented here imply that primary productivity in the Seto Inland Sea has been greatly affected by variations in oceanic nutrient inputs on a decadal timescale.

In the Seto Inland Sea, the reduction in phytoplankton productivity induced by the regulation of terrestrial nutrient inputs is thought to have been the primary reason for the reduction in fishery production after the 1980s (Yamamoto 2003). Our results, however, imply that the remarkable increase in phytoplankton biomass in Beppu Bay after the 1980s was due to an influx of nutrients from the ocean. This conclusion raises another question, namely whether enhanced phytoplankton biomass affects fishery production in the bay. Further studies are therefore necessary to clarify whether and how the bottom-up effects of variations in primary productivity due to changes in the supply of nutrients associated with variations of the Kuroshio axis have impacted fishery production in the Seto Inland Sea.

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