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Seasonal and site-specific variability in terrigenous particulate organic carbon
concentration in near-shore waters of Lake Biwa, Japan

Authors

¹Sakai Y, ²Karube Z, ³Takeyama T, ²Kohzu A, ¹Yoshimizu C, ⁴Nagata T, ¹Tayasu I and
¹Okuda N

¹Center for Ecological Research, Kyoto University, 509-3, 2-chome, Hirano, Otsu,
Shiga 520-2113, Japan

²National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba-City, Ibaraki,
305-8506, Japan

³Department of Biology and Geosciences, Osaka City University, 3-3-138 Sugimoto,
Sumiyoshi, Osaka, 558-8585, Japan

⁴Atmosphere and Ocean Research Institute, The University of Tokyo, Kashiwa, Chiba,
277-8564, Japan

Correspondence: Yoichiro SAKAI

19 Center for Ecological Research, Kyoto University, 509-3, 2-chome, Hirano, Otsu, Shiga
20 520-2113, Japan.
21 Tel: 077-549-8020
22 Fax: 077-549-8201
23 E-mail: biwaensis.2002@gmail.com
24

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26 Abstract

27 Identifying sources of particulate organic matter (POM) is important for
28 clarifying fundamental mechanisms by which lake food webs are sustained. We
29 determined carbon and nitrogen stable isotope ratios of POM in near-shore waters of
30 Lake Biwa, a large, meso-eutrophic lake in Japan, to estimate relative contributions of
31 terrigenous particulate organic carbon (T-POC), plankton-derived POC (P-POC), and
32 epilithon-derived POC (E-POC) to POC in near-shore waters. Samples were collected
33 during different months (November, February, May and July) at 29 sites located near the
34 mouth of tributary rivers with different discharge and catchment land use. The data
35 revealed that POC mainly consisted of P-POC and T-POC, with relative contributions
36 varying widely over season and among locations. E-POC generally contributed little to
37 the near-shore POC. Path analyses revealed that concentration of riverine POC whose
38 isotopic signatures were similar to those of rice straws increased with a larger %paddy
39 field area in the catchment of tributary rivers, which subsequently enhanced T-POC
40 inputs to near-shore waters through riverine transportation. Furthermore, our results
41 suggested that T-POC contribution was influenced, with a time lag, by wave-driven
42 turbulence and shore topography, which appear to affect sedimentation and resuspension
43 of T-POC.

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45 **Keywords**

46 Allochthonous input, Isotope mixing model, Land use, Path analysis, Terrigenous POM

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49 Introduction

50 Particulate organic matter (POM) is a fundamental energy source for food webs
51 in lake ecosystems. POM in lake waters consists of diverse organic compounds with
52 different origins, including phytoplankton-derived POM (P-POM), epilithon-derived
53 POM (E-POM) and terrigenous POM (T-POM). Although P-POM is generally
54 considered to be a major energy source for planktonic food webs in pelagic
55 environments (Dodson 2005), E-POM and T-POM may also play an important role as
56 food resources for suspension feeders. In fact, in small to medium-sized lakes in North
57 America, some studies have revealed that terrigenous particulate organic carbon
58 (T-POC) derived from tributary rivers can subsidize pelagic food webs (Cole et al.
59 2006; Pace et al. 2007; Cole et al. 2011). Other studies, however, have suggested that
60 zooplankton growth and reproduction is slowed by the inputs of T-POM, which have
61 high C/N ratios and are low-quality food (Karlsson 2007; Brett et al. 2009). In other
62 systems, it has been reported that pelagic food webs were subsidized by benthic algal
63 products (Rautio and Vincent 2007; Karlsson and Sawstrom 2009). For aquatic
64 consumers, reliance on each of POM originated from different sources appears to vary
65 widely among lakes, depending on lake size, depth and trophic status (Chandra et al.
66 2005; Doi 2009; Vander Zanden et al. 2011). The relative contribution of POM with

different origins may also vary among different locations of a single lake, especially in near-shore waters. For example, allochthonous input of T-POM might be large in near-shore environments adjacent to river mouths, although the extent and nature of this “river effect” may depend on river discharge, T-POM concentrations in river waters, and physical settings of the locations (shore topography and turbulence that affect sedimentation and resuspension of T-POM). Spatial and temporal variability in contributions of POM with different origins within a single lake has important implications for understanding mechanisms by which lake food webs are differentially organized in the same species pool. Indeed, a previous work has reported that allochthonous organic matter inputs to Lake Biwa from tributary catchments led to spatial heterogeneity in trophic pathways within coastal macro-invertebrate communities (Karube et al. 2010). However, data are highly limited to elucidate mechanisms underlying spatio-temporal variability in relative contributions of P-POM, T-POM and E-POM to total POM in near-shore environments.

Here, we measured carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$) stable isotope ratios of POM in near-shore water samples collected during four seasons at different locations set along the shore-line of Lake Biwa. In general, C_3 terrestrial plants (T-POM) have markedly different $\delta^{13}\text{C}$ (-27‰) from that of aquatic microalgae (P-POM and E-POM;

Marshall et al. 2007). Moreover, among aquatic primary producers, phytoplankton and epilithic algae have different $\delta^{13}\text{C}$, due to the CO_2 diffusion limitation within the boundary layer surrounding algal cells (Phytoplankton: -32 ± 2 (SD) ‰, epilithic algae: -17 ± 2 (SD) ‰; Fry and Sherr 1984; France 1995). In addition, $\delta^{15}\text{N}$ of aquatic primary producers may be variable among aquatic environments depending on the extent of anthropogenic nitrogen inputs (McClelland and Valiela 1998; Kohzu et al. 2008; Karube et al. 2010). The above differences in isotope signatures of POM with different origins are useful for estimating the relative contribution of P-POM, E-POM and T-POM by means of stable isotope mixing models (Fry 2006).

Lake Biwa, the largest lake in Japan, consists of two basins: the large (616 km^2), deep (average depth 41 m), mesotrophic northern basin and the small (58 km^2), shallow (average depth 4 m), eutrophic southern basin (Rossiter 2000; Somiya 2000). The lake receives water from 121 major tributary rivers and hundreds of other small creeks. The catchments of the major rivers show a wide range of land use patterns, including those prevailed by agricultural field, forest, or residence area (Somiya 2000). The 29 sampling sites of the present study covered a range of situations with variable influences of rivers that have different land use patterns in their catchments. We hypothesize that the quantity and composition of the near-shore POM can vary, on a

within-lake scale, depending on land use patterns in tributary catchments as well as on coastal physical characteristics. Assuming that POM with different origins, i.e., P-POM, E-POM and T-POM, have different stable isotopic signatures, we aim to examine what factors determine within-lake variability in relative contributions of these potential organic sources to the near-shore POM, using an isotope mixing model and a path analysis.

Materials and Methods

Sample collections

We collected POM samples at 29 sites along the shoreline of Lake Biwa in November 2005 and February 2006, May 2006, and July 2006 (Fig. 1). These sites were located near the mouth of tributary rivers, which have widely variable land use patterns (Appendix A). In most cases, the sampling sites were set within 100 m of the river mouth. For some sites, however, our research boat could not get access to the target location because these sites have a gentle coastal slope and/or are luxuriated by submerged plants. In such cases, the sampling site was set on the position of 5 m-depth contour where the distance between the site and river mouth was shortest. We measured pH and electric conductivity using a multiprofiler at each sampling site (U-22,

HORIBA).

At each sampling site, we used a water pump to collect water samples at the depth of 2 m. The water samples were first filtered through a 150 μ m-mesh net to remove coarse particles and then filtered through precombusted (450°C for 2 h) glass fiber filters (GF/F, 0.7 μ m, Whatman). The particle size of 0.7-150 μ m covers the most size range of phytoplankton found in Lake Biwa (Tsuda et al. 1992). The POMs collected on the glass fiber filters were used for stable isotope analysis (see below). We extracted Chlorophyll *a* from these POM samples in 90% acetone solution and measured its concentration with a fluorometer (Turner Designs, 10-AU), according to Wetzel and Linkens (2000). The Chlorophyll *a* concentration was used as an indicator of local productivity in near-shore waters.

We assumed three end members (plankton, epilithon and terrigenous organic matter) as primary organic sources for the near-shore POM. First, to determine isotopic values of plankton-derived POM (P-POM), we collected water samples from the depth of 2 m at an offshore site (Station 30, see Fig. 1) in the same months as POM sampling in near-shore waters. The P-POM was filtered on precombusted GF/F filters after screening with a 150 μ m-mesh net. The P-POM collected at this site was assumed to have isotope signatures typical for phytoplankton-derived organic matter in the lake

because previous work has found that $\delta^{13}\text{C}$ values of POM collected there were identical to those of chlorophyll *a* extracted from offshore phytoplankton after the correction of isotope fractionation (Maki et al. 2010).

Second, to determine isotopic values of epilithon-derived POM (E-POM), epilithon samples were collected at all the sites except for some sites where boulders of appropriate size were not found (Fig. 1). At each site, epilithic organic matter scraped off, with a brush, from five boulders of moderate size (30-40 cm) which have a surface area of more than 36cm² on the upper side were mixed, filtered through precombusted GF/F filters, and served for the stable isotope analyses.

Third, we collected riverine particulate organic matter (R-POM) as a proxy of terrigenous POM (T-POM) at sites 0.5-4 km upstream from the mouth, in the same months as the near-shore sampling but in the previous year (i.e., July 2004, November 2004, February 2005 and May 2005: see Kohzu et al. 2009 for details). In general, R-POM is composed of T-POM and fluvial E-POM (Kohzu et al. 2009). In our study rivers, however, $\delta^{13}\text{C}$ of R-POM was significantly different from that of fluvial E-POM in all four months (Tukey's post-hoc test, R-POM vs. fluvial E-POM, Nov: $p < 0.001$, Feb: $p < 0.01$, May: $p < 0.01$, Jul.: $p < 0.001$) but not from C₃ terrestrial plant detritus, i.e., rice straw collected from paddy fields in any months (Tukey's post-hoc test, R-POM vs.

T-POM, Nov: $p=0.82$, Feb: $p=0.65$, May: $p=0.79$, Jul.: $p=0.91$), suggesting that R-POM is dominated by T-POM in tributary rivers of Lake Biwa.

One may expect that submerged macrophytes can be a dominant source for the near-shore POM. However, we did not incorporate them as an end member of our mixing model for the following reasons. First, live macrophytes cannot contribute to the POM pool in near-shore waters during summer growing season. It may be true that their detritus contribute to the near-shore POM during decomposition process in winter, especially at sampling sites in shallow south basin where macrophytes luxuriate vigorously (Haga et al. 2007). However, we have little knowledge on stable isotopic fractionation during their decomposition and have technical difficulty in separate their detritus from a mixture of POM with different origins in the near-shore waters for the stable isotope analysis. If macrophyte contribution is not much great for near-shore waters of whole lake basin, we have a disadvantage to incorporate additional end member into our mixing model, which will lead to low estimation accuracy for major organic sources. Therefore, we did not regard submerged macrophytes as primary organic sources in the near-shore water.

That is why we used the dominant three end members, P-POM, E-POM and R-POM representative of T-POM, for our isotopic mixing model to estimate

composition of near-shore POM. All these POM samples collected on precombusted GF/F filters were exposed to 0.1 M HCl to eliminate carbonates, rinsed with distilled water, and then stored at –20°C until the stable isotope analysis.

Stable isotope analysis

The GF/F filters were dried at 60°C for 24 h, and then the residues were scraped off from the surface of filters. Carbon and nitrogen stable isotope ratios of the residues were determined using continuous-flow isotope ratio mass spectrometers (CF/IRMS; Conflo II and Delta S, Finnigan MAT, Germany and Conflo III, delta plus XP, Thermo Fisher, Germany) and their carbon content was measured using elemental analyzers (EA1108, Fisons, Italy and EA1112, Thermo Fisher, Germany). The isotope ratios were expressed as the per mil deviation from standards as follows:

$$\delta^{13}\text{C} \text{ or } \delta^{15}\text{N} = (R_{\text{sample}} / R_{\text{standard}} - 1) \times 1000 (\text{‰}),$$

where $R = {}^{13}\text{C}/{}^{12}\text{C}$ or ${}^{15}\text{N}/{}^{14}\text{N}$. Vienna Pee Dee belemnite (VPDB) and atmospheric nitrogen were used as standards for carbon and nitrogen, respectively. The analytical precision based on working standards (Tayasu et al. 2011) was $\pm 0.3\text{‰}$ for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$.

Isotope mixing model

Relative contributions of three potential organic sources (i.e., P-POM, E-POM and T-POM) to POM collected at each near-shore site were estimated using an isotope mixing model with $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ (IsoSource Program; Phillips and Gregg 2003). This model enabled us to estimate all possible combinations of each source contribution (0–100%) in 1% increments to create a set of predicted sources. The initial tolerance was set at 0.1‰. If the estimated values were outliers (i.e., < 0% or > 100%), the tolerance was increased in 0.1‰ steps to 2‰. The model outputs are expressed as the mean value and range (minimum–maximum) for each source. From the estimated mean contribution of each organic carbon source to POC concentration (mgL^{-1}) at each near-shore site, we calculated the concentrations of P-POC, E-POC and T-POC according to the following equation.

$$\text{P-POC (mgL}^{-1}\text{)} = \text{POC (mgL}^{-1}\text{)} \times f_p$$

$$\text{E-POC (mgL}^{-1}\text{)} = \text{POC (mgL}^{-1}\text{)} \times f_e$$

$$\text{T-POC (mgL}^{-1}\text{)} = \text{POC (mgL}^{-1}\text{)} \times f_t$$

where f_p , f_e and f_t are the fraction of P-POC, E-POC and T-POC relative to the near-shore POC (i.e., $f_p + f_e + f_t = 1$).

At some sites, we could not estimate each source contribution for the lack of

E-POC data or because the estimated contribution remained outlier for either of the three sources under the above criteria for our isotopic mixing model. In the present study, we present results of simulation model only for carbon source estimation but analytical results on nitrogen sources were qualitatively same as the carbon source estimation model.

Land use pattern and physical environments

Human population density (HPD in populationkm⁻²) and proportions (%) of paddy field, forest, and residential areas of tributary catchment were calculated using a geographical information system (GIS: Appendix A). We assumed that these indices reflect the loading of terrigenous organic matter with different origins: e.g., the HPD or %residential area is an indicator of organic matter derived from sewage, the %paddy field area is an indicator of that from agricultural wastewater, and the %forest area an indicator of that from leaf litter transported through upper streams.

Because there is a strong positive correlation between river discharge and catchment area, we estimated the river discharge, based on the following regression for tributary rivers for which discharge data are available (Shiga Prefecture1992-2002):

$$\text{River discharge (m}^3 \text{ S}^{-1}\text{)} = 0.0284 \times \text{catchment area}^{0.9502} \text{ (R}^2\text{=0.846, p<0.01)}$$

where the river discharge is the average for February, May, July, and November during the recent 10 years (1992-2002). Although the river discharge shows daily variation depending on weather condition, its monitoring date never accorded with our sampling date. Considering this variation, the inter-annual average can be a better indicator for river discharge compared to single monitoring data corresponding to our sampling months.

As physical characteristics of coastal environments, we used wave height (m), wave energy flux ($\text{kg m day}^{-1}\text{m}^{-1}$), shore energy flux ($\text{kg m day}^{-1}\text{m}^{-1}$, see Nakatsuji et al. 2006), and coastal slope (1 d^{-1} ; d = horizontal distance from the shoreline to the point with a depth of 2 m). These data are available at <http://www.biwakokasen.go.jp/others/kankyoujyouhou/index.html> (Kinki Regional Development Bureau, Ministry of Land, Infrastructure, Transport and Tourism).

The GIS and coastal physical characteristics data are summarized in Appendix A and B.

Statistical analysis

We performed a nested analysis of variance (ANOVA) to test the temporal variation in POC concentrations, incorporating sampling sites as a random factor into the model (the nlme package of R Statistics; Pinheiro et al. 2010), which assumes that

error term distribution accords to gaussian distribution. If the data distribution did not show normality and homoscedasticity, the dependent variable (POC concentration) was $\log(x+0.5)$ -transformed satisfy the criteria for the ANOVA (Yamamura 1999). Then we performed a post-hoc test for multiple comparisons using the Tukey method (the multcomp package of R Statistics; Hothorn et al. 2008) to compare POC concentrations among seasons. Furthermore, we performed ANOVA to compare the carbon concentrations among the three organic sources (P-POC, E-POC, and T-POC) in each season. All analyses were conducted using the statistical package R ver. 2.13.1 (R Development Core Team 2011).

We performed a path analysis to examine how the land use patterns in the tributary catchments and the physico-chemical characteristics of the sampling sites may affect spatiotemporal variation in measured and estimated values of R-POC and T-POC concentrations, respectively, incorporating GIS and environmental variables into the model (see Appendix A and B). First, we screened the explanatory variables because models with too many variables generate redundant information. We checked the correlations among variables to remove multicollinearity and then selected the variables that best accounted for the variation in R-POC and T-POC concentrations. For instance, we selected wave height from among the three wave-associated variables and HPD but

not %residential area. Because forest and paddy field are the most dominant land use types in the Lake Biwa Watershed, it is a matter of course that there is a strong negative correlation between their proportional data ($R^2=0.93$, $p<0.001$). We performed two preliminary models to test effects on the R-POC for each of these two land use types. First, when we used %forest area solely as a GIS variable, instead of %paddy field area, we found that the former showed significantly negative but not positive effects (Path coefficients, Nov.: -0.44, Feb.: -0.53, May: -0.83, Jul.: -0.64, $p<0.01$). Second, when we incorporated residuals of the %forest area regressed against the %paddy field area into the preliminary model, we did not find any significantly positive effect of %forest area on R-POC concentration, suggesting that forests cannot be a primary source of R-POC loadings in catchment areas. Based on results of these preliminary analyses, therefore, we excluded %forest area from a final version of our *a priori* model to reduce the redundancy and the multicollinearity between potentially correlated GIS variables.

After screening the variables, we constructed the basic framework of an *a priori* model consisting of two components (Fig. 3). The model was separated because the data distributions were needed to be $\log(x+0.5)$ -transformed for some of explanatory variables in component 2 to ensure normality and homoscedasticity for statistical criteria. Component 1 was a sub-model that accounts for the variation in measured

values of R-POC (upper panel in Fig. 3). Assuming that human sewage and agricultural wastewater have strong effects on R-POC, we incorporated HPD (Path 1 in Fig. 2) and the %paddy field area (Path 2) into the model. We also assumed that the HPD and %paddy field areas have a positive interactive effect because agricultural activities are usually high in areas with a high population density and vice versa (Path 3).

Component 2 was constructed to examine which physical characteristics of coastal environments explain the site-specific variation in estimated values of T-POC concentration in near-shore waters, assuming that T-POC concentration can be affected by a dilution effect due to river discharge (Path 5), resuspension due to wave-driven turbulence (Path 6), and the potential for resuspension, which is intensified in sites with a gentle coastal slope (Path 7). These paths were assumed to have an additive effect on T-POC concentration.

The model analysis consisted of two steps. First, we conducted a simultaneous analysis of several groups, considering the four seasons as different groups. This allowed us to test for significant differences in the causal mechanisms of the seasonal dynamics of R-POC and T-POC concentrations and provided more accurate model estimation than what would have been obtained from separate analyses of each of the four groups (Arbuckle 2007). Then we selected the best-fit model, setting equality

constraints as a null hypothesis that the coefficient of each path is not significantly different among the four seasons (Tabei 2001; Arbuckle 2007). In the initial condition, the level of significance was set at 0.05 for the seasonal difference in each path coefficient and was then increased up to 0.6 in intervals of 0.05. In this way, we constructed a total of 12 models and compared them using the Akaike information criterion (AIC). We selected the best-fit model based on the minimum AIC. The best-fit model was tested using the chi-square goodness-of-fit. We also evaluated its goodness-of-fit based on the root mean square error of approximation (RMSEA; criterion $RMSEA < 0.05$: good, $RMSEA > 0.08$: mediocre, $RMSEA > 0.1$: not accepted, Browne and Cudeck 1993). The path analysis was performed using the statistical package AMOS (ver. 16, IBM-SPSS, Tokyo).

Results

Spatiotemporal variation in POC concentration and composition

Using carbon and nitrogen isotope mixing model with three end members, P-POM, E-POM and R-POM, we estimated the relative contributions of three organic sources (P-POC, E-POC, and T-POC) to POC at each near-shore site (Appendices C&D). As indicated by a narrow range of estimated values, our isotope mixing model

showed high estimation accuracy (Appendix D), except for some sites (e.g., site no. 3, 10, 11, 13, 20 in November, 2005), in which P-POM and R-POM had similar isotopic signatures (Appendix C). Based on the relative contribution of these sources, we calculated the concentration of P-POC, E-POC, and T-POC (Table 1 & Fig. 4). The near-shore POC concentrations differed among sites (Fig. 4) and seasons ($F_{3, 83} = 13.56$, Jul. > May = Feb. > Nov., $p < 0.05$). P-POC and T-POC were the dominant organic sources of the near-shore POC, whereas E-POC concentration was consistently low (Fig. 5). In July, the concentration of P-POC was generally higher than that T-POC (Fig. 5). T-POC concentrations tended to be high at the sites located along southern and eastern shores, especially in February and May (Fig. 4).

Possible factors that influences the spatiotemporal dynamics of T-POC

T-POC concentrations in near-shore waters showed marked spatiotemporal variation at sampling sites (Fig. 4). In our path analysis, a sub-model of component 1 gave the best fit when the significance level for the equality constraints was set at 0.6 ($\chi^2=0.721$, $df=7$, $RMSEA=0.000$, $AIC=58.721$). The total effect of human land use on R-POC concentration showed great seasonal variation (Fig. 6). Tributary rivers with a larger %paddy field area had a consistently high R-POC concentration year-round (Path

2). In contrast, HPD did not have significantly positive effects on R-POC concentration (Path 1).

Similarly, component 2 of our path model was the best fit when the significance level was set at 0.3 ($\chi^2=0.488$, $df=42$, $RMSEA=0.057$, $AIC=140.288$). The total effect of proximate physical factors on T-POC showed great seasonal variation (Fig. 6). The allochthonous input of R-POC had a significantly positive effect on the near-shore T-POC concentration only in May (Path 4), whereas river discharge had a negative effect on the near-shore T-POC concentration in July (Path 5). Wave height always had a significantly positive effect on the near-shore T-POC concentration (Path 6), and the near-shore T-POC concentration was higher for sites with a gentle coastal slope in February and May (Path 7).

Discussion

Our data demonstrated that near-shore T-POC concentrations varied widely among sites and over season. The results of path analysis suggested that agricultural wastewater from the paddy fields was the primary driver affecting the near-shore T-POC concentration via increased inputs of R-POC. Especially in May, we detected a significant, robust path in that the %paddy field area had a positive effect on the

near-shore T-POC concentration via R-POC inputs (Path 2 & 4). Large contribution of T-POC in May could be related to agricultural practices in this season. Previous work has reported that a large quantity of paddy-derived wastewater flows into the lake basin at the onset of the rice irrigation period between late April and early May (Fujii et al. 2002; Haga and Ohtsuka 2003; Ohkubo and Azuma 2005; Hama et al. 2010). The results of the present study support the notion that agricultural activities, especially paddy irrigation, can be a significant factor that affects T-POC concentrations in near-shore environments of Lake Biwa. It is also interesting to note that %paddy field area had a positive effect on R-POC concentration even in February when paddy field activities are considered to be minimal. This suggests the possibility that organic-rich soils can be eroded from paddy fields even in winter, due to snowing and melting. Although the underlying mechanisms have not yet been poorly understood, they remain to be investigated in the future.

The results of path analysis also suggested that physical characteristics of coastal environment, including coastal slope and wave height, have significant effects on T-POC concentration in near-shore waters. We speculate that waves and gentle coastal slopes might synergistically enhance the resuspension of sedimentary T-POC, resulting in increased terrestrial contribution to the near-shore POC. This notion is

partly supported by findings of Murase and Sakamoto (2000) who reported that the near-shore sediments of Lake Biwa contained a large quantity of terrigenous organic matter. Intriguingly, in the present study, although R-POC had a positive effect on amount of near-shore T-POC only in May (Path 4), the wave effects were strongest in November, after rice field irrigation season (Fig. 6). These results suggest that agricultural wastewater loading had a time lag effect on the T-POC concentration via sedimentation and wave-driven resuspension in near-shore waters.

One might expect allochthonous inputs of R-POC to be magnified by river discharge, as well as by its concentration. However, our analysis did not support this prediction and revealed that river discharge had strongly negative effects on the near-shore T-POC concentrations in July with high precipitation. In general, flooding has the potential to increase concentration of suspended organic matter in river water and consequently organic matter loadings on near-shore waters. However, we avoided field samplings during flooding events because our interest was in land use effects on allochthonous inputs but not in physical transportation mechanisms under ordinary flow conditions. During the rainy season, discharge of larger rivers which receive larger quantity of rain waters in their larger catchment areas might have a greater dilution effect on near-shore POC.

In near-shore waters of Lake Biwa, POC was composed mainly of P-POC and T-POC, whereas E-POC contributed little to POC regardless of the season and site. In some shallow and oligotrophic lakes, it has been reported that epilithic-derived trophic energy flows dominate in coastal food webs (Rautio and Vincent 2007; Karlsson and Sawstrom 2009). There are two possible mechanisms in which relative contribution of E-POC was low in Lake Biwa basins. One is associated with lake morphological features: Lake Biwa has a deep structure with a steep slope, especially for large north basin, in which the proportional area of shallow waters to the whole basin was considerably small (Sakamoto 2000), suggesting that habitats are limited for epilithic growth. Another mechanism is that an increase in P-POM and T-POM decreases light penetration to the lake bottom, which has negative effects on epilithic growth (Vadeboncoeur et al. 2001, 2003). Consistent with our observation, previous studies have reported that filter-feeding macrozoobenthos and zooplankton in near-shore environments of Lake Biwa relied mainly on P-POC and T-POC rather than on E-POC (Karube et al. 2010). Their production reliance is also trophically transferred to that of predatory fishes, characterizing food web configuration of whole communities (Okuda et al. 2012).

In the present study, we did not consider contribution of detrital macrophytes to

the near-shore POM for the following two reasons. First, in Lake Biwa, especially in deep north basin, the area of shallow habitats available to macrophytes is disproportionately small relative to that of the whole basin, so that their total primary production is much lower than that of phytoplankton and of even epilithon whose euphotic zone is deeper than the macrophytes (Sakamoto 2000). Second, we have technical difficulty in separating macrophyte detrital fraction from a mixture of POM with different origins in the near-shore water for the stable isotope analysis and thus in characterizing isotopic alteration during their decomposition in nature. To evaluate their contribution, one of promising approaches is polyunsaturated fatty acid (PUFA) analysis, which can quantify each of organic sources by characterizing the composition of primary producer-specific polyunsaturated fatty acids (Kelly and Scheibling 2012). In floodplain lakes of the central Brazilian Amazon basin, for instance, the technique revealed that macrophyte detritus is a primary component of suspended POM during low water season (Mortillaro et al. 2011). The application of PUFA analysis, even if it is beyond our scope in the present study, will be a future challenge for better understanding of seasonal changes inorganic matter flows in Lake Biwa, especially in shallow south basin where submerged plants have recently luxuriated.

Using stable isotope approaches, we demonstrated that allochthonous inputs of

T-POC to near-shore waters were affected not only by the land use pattern in tributary catchment areas, but also by the physical characteristics of local coastal environments. This suggests that the spatiotemporal heterogeneity of food quality and quantity for aquatic consumers will create a dynamic pattern of coastal food webs, altering the relative importance of autochthonous grazing food chain and allochthonous microbial loop in lake ecosystems.

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575

576 **Figure and Table Legends**

577 **Fig. 1**

578 A map of POC sampling sites (no. 1-30) in the near-shore waters of Lake Biwa.

579 No. 30 is the reference site for P-POM. See Appendix A and B for details of site
580 information.

581

582 **Fig. 2**

583 Carbon and nitrogen isotopic signatures of near-shore POM and their potential organic
584 sources (P-POM, E-POM and R-POM). Each plot represents mean from all sampling
585 sites, and vertical and horizontal bars indicate range (also see Appendix C). Note that
586 P-POM data are derived from only a pelagic site.

587

588 **Fig. 3**

589 An *a priori* path model to explain the variation in T-POC concentrations in the
590 near-shore waters of Lake Biwa. The model consists of two components. Unidirectional
591 arrows indicate a causal effect and double headed arrows interactive effect.

592

593 **Fig. 4**

Spatiotemporal variations in the concentrations of three components, P-POC, E-POC and T-POC, of POC in the near-shore waters of Lake Biwa. The size of each circle is proportional to the POC concentration. Transparent circles represent sites where the relative contributions of three potential organic sources could not be estimated (see Materials and Methods).

Fig. 5

Seasonal changes in the composition of three potential organic sources of POC in the near-shore waters of Lake Biwa. Vertical bars are the standard deviation.

Fig. 6

The path model based on the simultaneous analysis of multiple populations with structured means. The basic framework is the same as that in Figure 2. Dark and light arrows are significant ($p < 0.05$) and not significant ($p > 0.05$) paths, respectively. Values with arrows are path coefficients. The subscripts of R-POC and $\log(T\text{-POC})$ indicate the total effect on these variables.

611

612 **Table 1**

613 Measured POC concentration and estimated P-POC, E-POC and T-POC concentrations

614 (mgL^{-1}) at each coastal site in Lake Biwa.

615

616

617

618 **Appendix A**

619 Geographic information system (GIS) data and riverine organic matter for each tributary
620 catchment area. Land use pattern is expressed as the proportional area.

621

622 **Appendix B**

623 The major environmental data and the physical characteristics of the corresponding
624 coastal site.

625

626 **Appendix C**

627 Carbon and nitrogen stable isotope ratios (‰) of POM and its three potential sources
628 (P-POM, E-POM, and R-POM) at each coastal site.

629

630 **Appendix D**

631 The relative contribution (%) of the three potential sources (P-POM, E-POM and
632 T-POM) to POM at each coastal site.

633

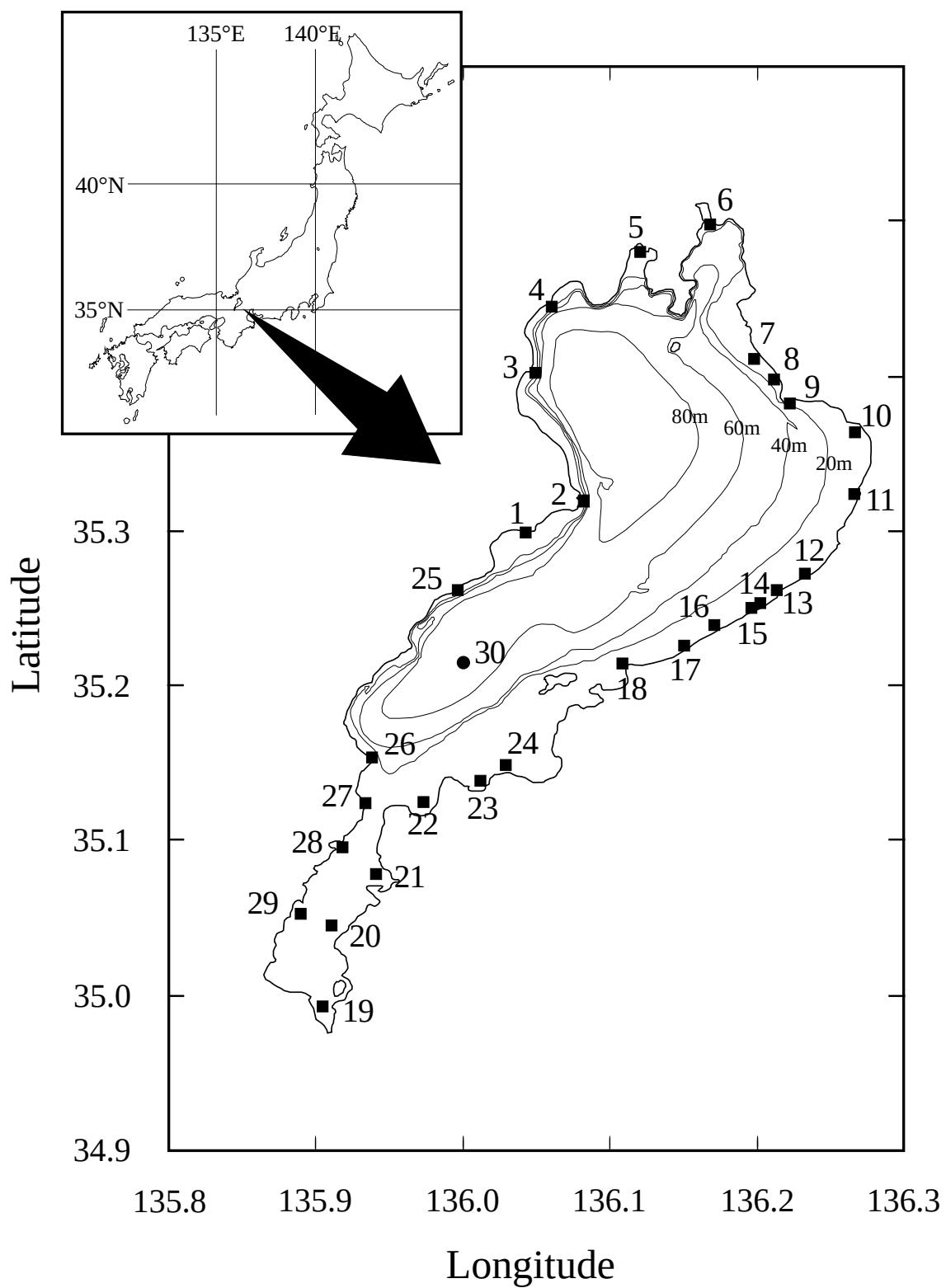


Fig. 1

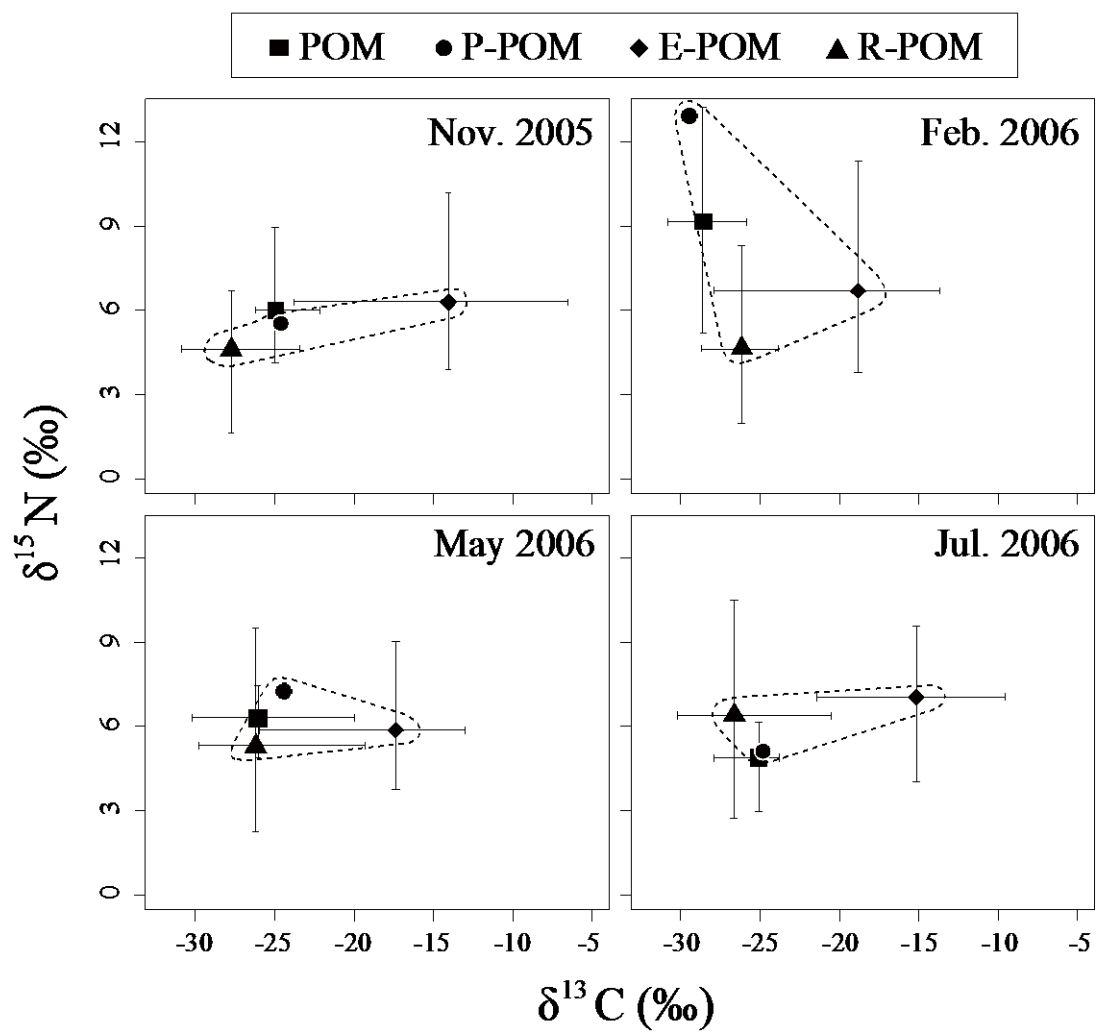


Fig. 2

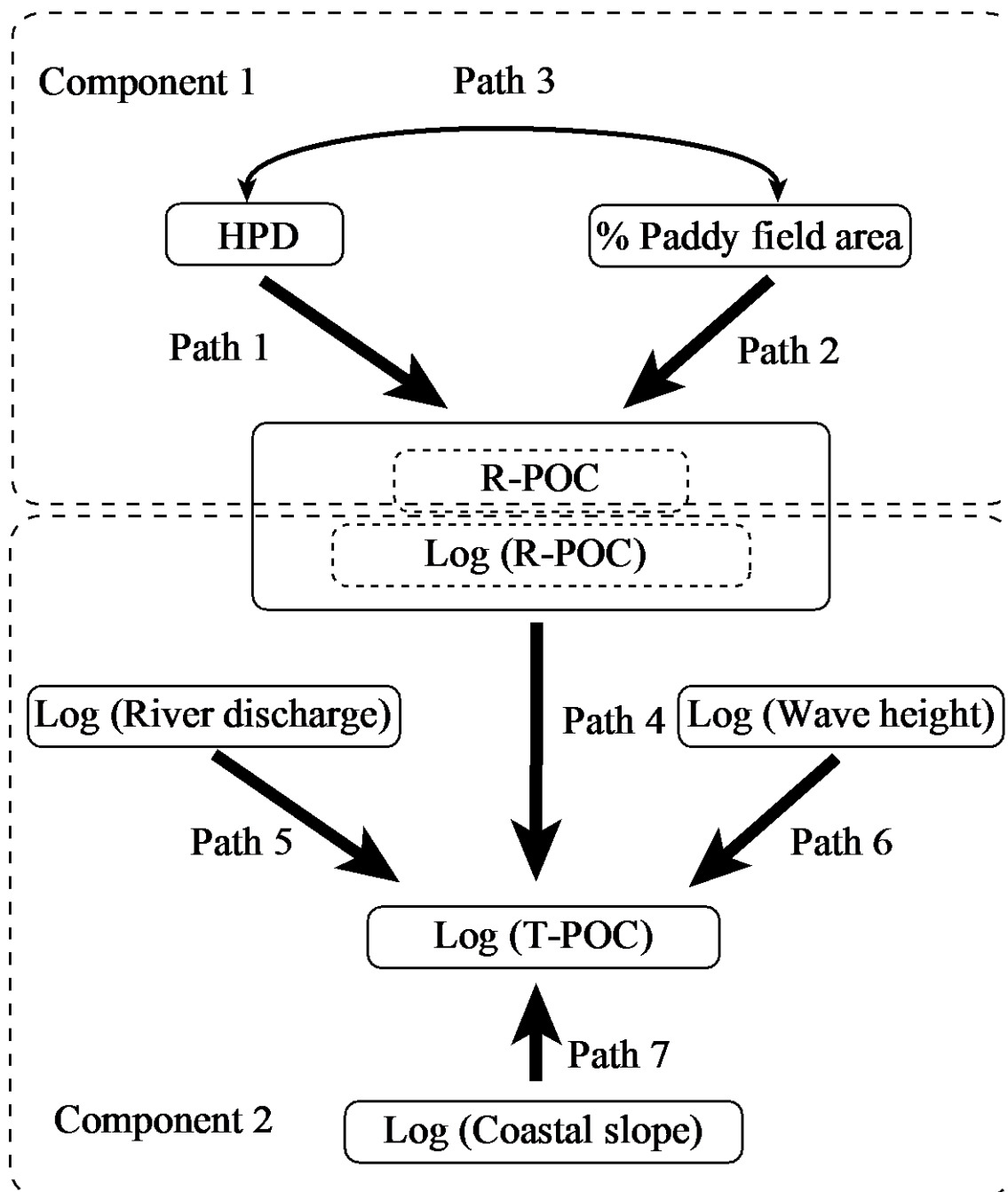


Fig. 3

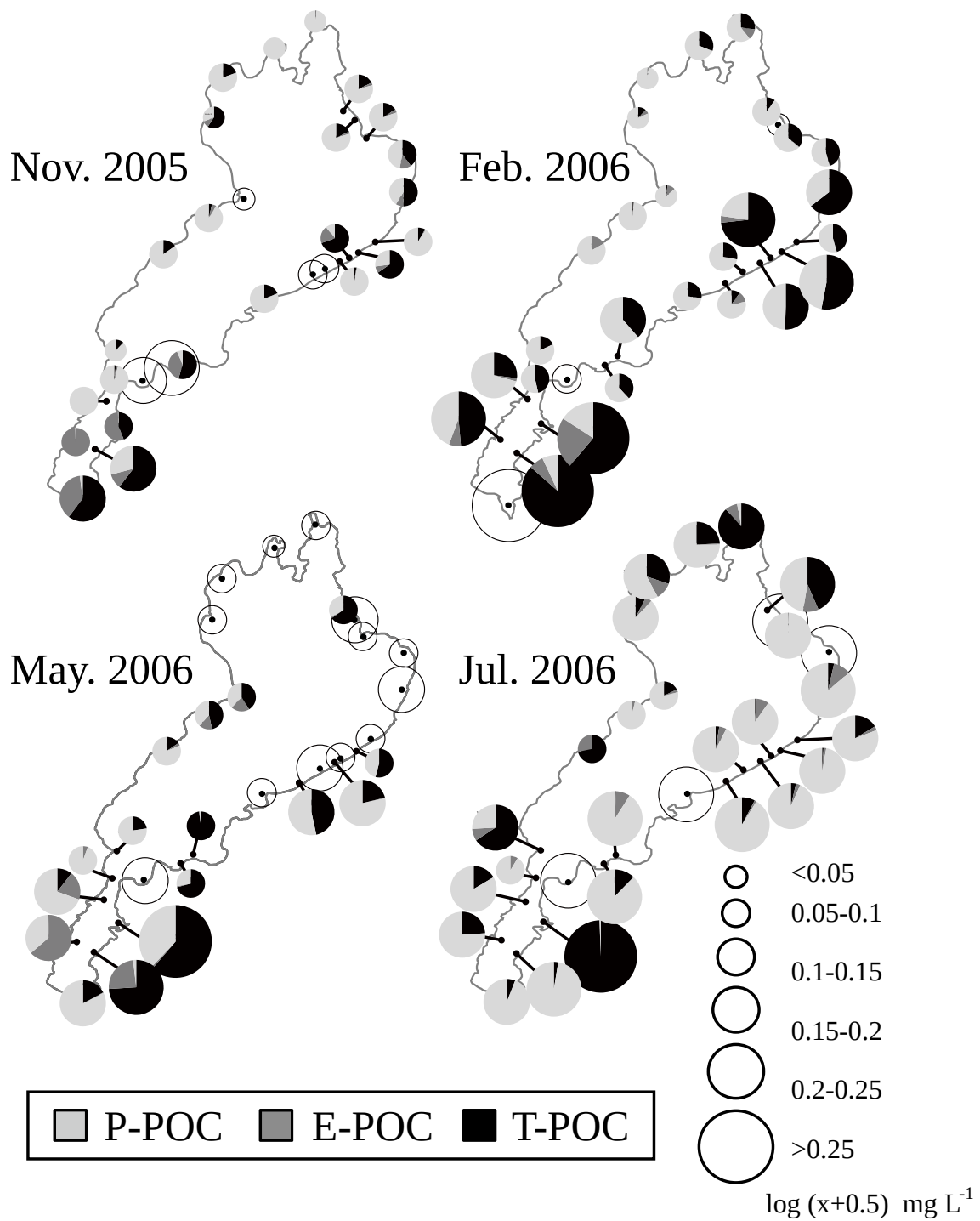


Fig. 4

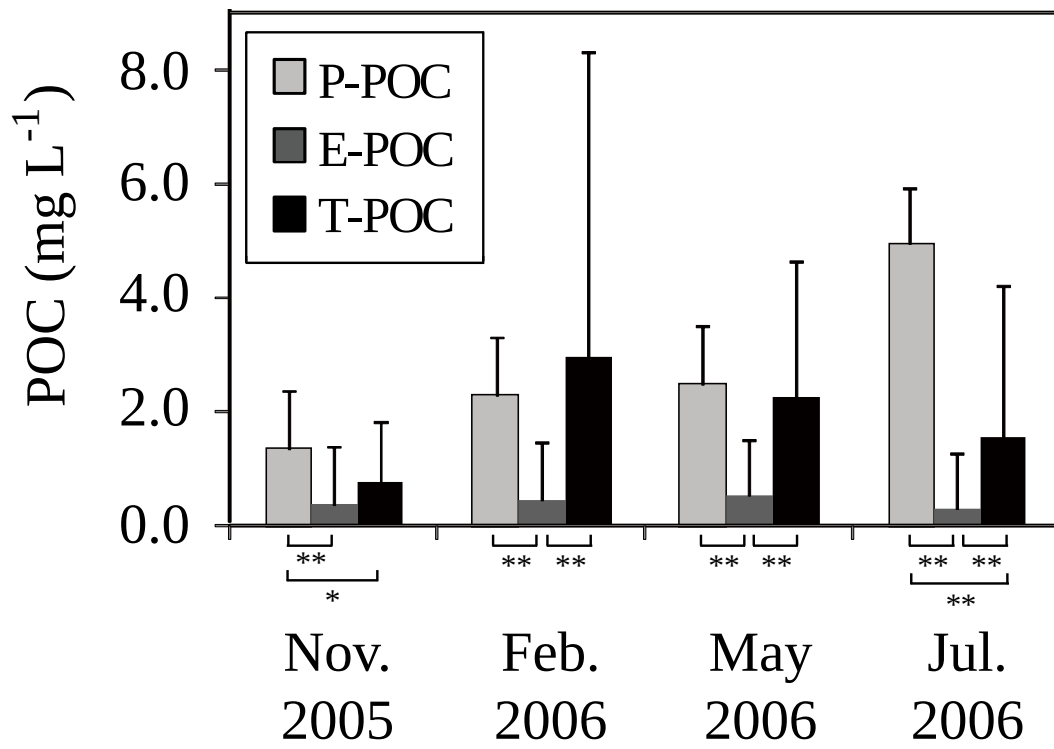
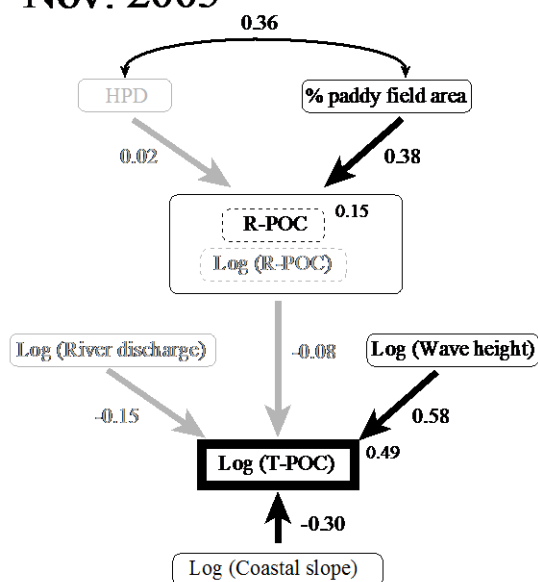
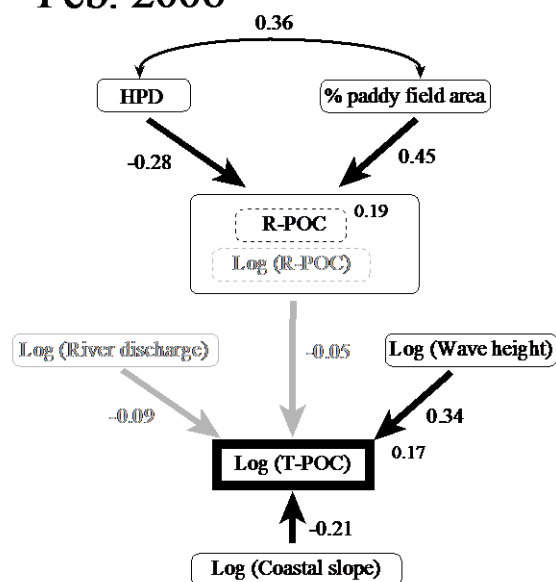


Fig. 5

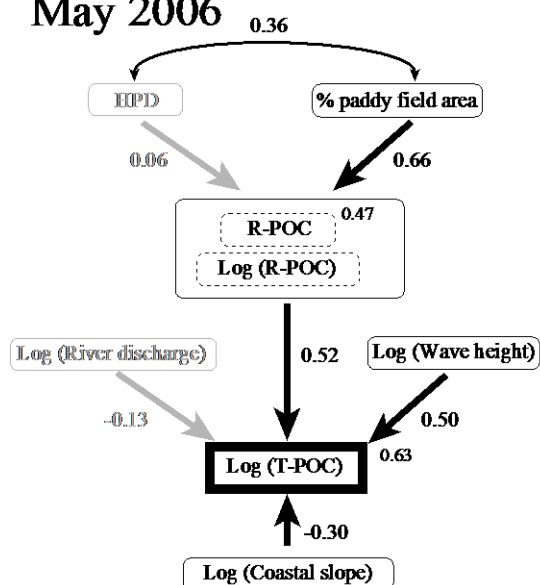
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Feb. 2006



May 2006



Jul. 2006

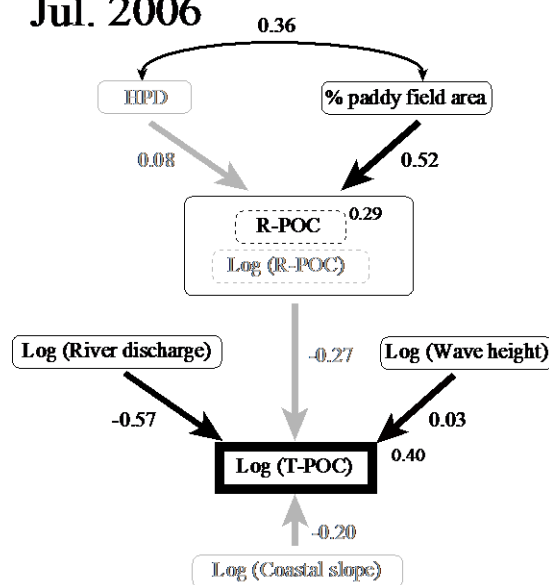


Fig. 6

Table 1

Measured POC concentration and estimated P-POC, E-POC and T-POC concentrations (mg L⁻¹) at each coastal site in Lake Biwa.

Site no.	Name of tributary rivers	Nov. 2005				Feb. 2006				May 2006				Jul. 2006			
		POC	P-POC	E-POC	T-POC	POC	P-POC	E-POC	T-POC	POC	P-POC	E-POC	T-POC	POC	P-POC	E-POC	T-POC
1	Kamo River	1.614	1.473	0.077	0.061	3.346	3.286	0.013	0.047	3.897	1.489	0.596	1.812	3.421	3.281	0.109	0.031
2	Ado R.	1.493				1.226	1.060	0.147	0.020	1.724	0.655	0.360	0.708	4.256	3.349	0.145	0.757
3	Ishida R.	1.446	0.450	0.139	0.857	0.981	0.801	0.061	0.120	1.796				6.145	5.408	0.277	0.461
4	Momose R.	2.300	1.851	0.007	0.442	1.242	1.226	0.009	0.009	2.531				5.720	3.318	0.686	1.722
5	Oura R.	1.222	1.213	0.004	0.004	2.274	1.569	0.000	0.705	0.921				7.029	5.286	0.056	1.687
6	Shiotsu-o R.	1.249	1.225	0.014	0.009	2.310	1.400	0.284	0.626	4.259				5.567	0.167	0.484	4.916
7	Chonoki R.	1.709	1.355	0.048	0.306	1.771	1.593	0.000	0.177	4.395	1.481	0.040	2.875	8.603	4.069	0.792	3.742
8	Ta R.	1.713	1.358	0.048	0.307	1.391				4.586	0.000	0.000	0.000	7.001			
9	Ane R.	1.963	1.580	0.075	0.308	1.862	1.178	0.013	0.670	2.595				6.974	6.939	0.035	0.000
10	Yone R.	1.607	0.754	0.212	0.640	2.953	1.603	0.009	1.343	3.291	0.000	0.000	0.000	10.145			
11	Amano R.	1.711	0.707	0.147	0.857	6.035	2.124	0.024	3.886	4.758				8.501	7.337	0.867	0.298
12	Seri R.	2.166	1.967	0.022	0.178	4.101	2.215	0.021	1.866	3.021	0.000	0.000	0.000	6.820	5.524	0.184	1.112
13	Inukami R.	1.866	0.507	0.140	1.216	8.151	3.831	0.000	4.320	4.348	1.991	0.009	2.348	6.617	6.432	0.119	0.066
14	Ajiki R.	2.116	0.224	0.421	1.473	9.595	2.178	0.384	7.024	3.378	0.000	0.000	0.000	6.102	5.504	0.531	0.073
15	Uso R.	4.483	4.339	0.031	0.112	6.672	3.303	0.000	3.369	4.984	3.908	0.030	1.052	6.047	5.654	0.181	0.212
16	Bunroku R.	2.601				2.533	1.824	0.000	0.709	4.699	0.000	0.000	0.000	5.470	5.054	0.284	0.137
17	Nomazu R.	2.794				3.336	2.605	0.400	0.327	6.483	3.468	0.000	3.015	9.929	9.055	0.109	0.764
18	Echi R.	3.920	3.175	0.008	0.737	2.327	1.682	0.016	0.628	3.701				12.012			
19	Nagaso R.	5.833	0.117	2.187	3.529	12.609				7.392	6.099	0.022	1.271	6.220	5.847	0.000	0.373
20	Hayama R.	6.262	1.828	0.645	3.782	21.669	1.517	1.408	18.744	8.838	0.168	2.121	6.549	9.259	9.027	0.000	0.231
21	Shin-moriyama R.	3.095	0.025	1.711	1.362	34.982	5.527	8.046	21.409	14.337	5.448	0.115	8.774	12.896	0.064	0.000	12.831
22	Yasu R.	6.179				3.684				6.571				11.995			
23	Yanomune R.	11.544				3.388	2.101	0.000	1.287	3.883	1.091	0.043	2.753	11.108	9.742	0.022	1.344
24	Hino R.	3.567	0.235	1.384	1.948	5.953	3.661	0.000	2.292	3.339	0.053	0.013	3.273	9.522	8.665	0.809	0.048
25	U R.	2.136	1.816	0.000	0.320	2.263	1.878	0.373	0.011	3.161	2.580	0.095	0.487	3.545	0.025	0.982	2.542
26	Wani R.	1.117	0.978	0.003	0.136	3.539	2.902	0.000	0.637	2.741	2.119	0.005	0.617	4.516	1.165	0.379	2.972
27	Mano R.	2.322	2.220	0.063	0.039	2.585	1.383	0.000	1.202	4.185	3.955	0.230	0.000	3.694	3.380	0.314	0.000
28	Tenjin R.	3.431	3.431	0.000	0.000	5.342	3.776	0.118	1.448	4.860	3.377	0.967	0.510	5.831	4.869	0.000	0.962
29	Fujinoki R.	2.081	0.010	2.071	0.000	8.922	3.953	0.625	4.345	6.095	2.206	3.870	0.018	6.197	4.709	0.000	1.487
Offshore reference site																	
30	Ie-1	2.38				2.22				3.01				4.73			

Appendix A

Geographic information system (GIS) data and riverine organic matter for each tributary catchment area. Land use pattern is expressed as the proportional area.

Station No.	Station Name	Human population (ind)	Human population density (ind. km ⁻²)	Catchment area (km ²)	% Paddy field	% Other crop land area	% Forest area	% Waste land area	% Residential area	% Traffic area	% Liver and Lake area	% Other area	Riverin Particulate Organic Nutrient (PON)				Riverin Particulate Organic Carbon (POC)			
													(μ mol L ⁻¹)				(μ mol L ⁻¹)			
													Nov. 2005	Feb. 2006	May 2006	Jul. 2006	Nov. 2005	Feb. 2006	May 2006	Jul. 2006
1	Kamo River	4149	89.16	46.531	18.13	0.48	74.80	1.67	1.20	0.09	2.05	1.58	1.014	2.779	2.722	2.331	9.872	29.989	22.935	20.056
2	Ado R.	8185	26.74	306.123	3.11	0.54	91.45	1.42	0.71	0.01	1.76	0.80	0.979	0.331	1.842	1.280	7.518	2.459	11.818	10.687
3	Ishida R.	5023	83.83	59.926	9.58	0.93	81.11	4.13	1.96	0.02	0.86	1.42	0.666	0.696	2.450	1.190	5.820	7.297	16.998	11.342
4	Momose R.	851	64.74	13.142	4.02	0.34	87.69	1.23	0.56	0.00	3.07	3.10	0.731	0.818	3.508	2.757	5.797	7.669	31.119	27.666
5	Oura R.	3828	97.62	39.209	11.28	1.61	78.52	2.19	2.67	0.16	0.94	2.63	7.627	9.810	4.809	4.032	96.905	136.730	45.918	46.646
6	Shiotsu-o R.	1111	55.03	20.242	6.66	0.38	86.85	1.29	1.57	1.34	1.38	0.51	11.435	9.279	2.485	1.822	143.682	148.315	21.167	19.257
7	Chonoki R.	4219	412.28	10.234	76.29	3.61	0.09	0.08	13.52	0.01	5.13	1.27	5.815	31.818	9.420	2.305	51.361	264.691	69.923	21.837
8	Ta R.	10853	301.33	36.016	46.68	2.08	37.10	0.25	9.59	1.39	1.93	0.99	11.907	12.720	5.298	9.858	118.184	133.328	44.291	120.706
9	Ane R.	22516	60.57	372.261	5.91	1.08	88.16	0.81	1.70	0.06	1.63	0.65	2.926	6.996	1.618	1.064	28.161	90.280	12.117	10.066
10	Yone R.	31047	2047.40	15.164	54.84	1.27	0.76	0.00	34.54	1.82	1.50	5.27	2.825	2.312	1.858	3.281	23.010	25.849	17.330	34.578
11	Amano R.	24994	225.70	110.935	18.48	2.48	66.31	1.49	5.82	1.44	1.97	2.01	21.215	1.770	2.883	1.538	160.075	18.646	24.513	14.668
12	Seri R.	34039	462.06	73.859	6.96	0.91	75.20	2.20	8.09	1.32	2.36	2.60	5.529	1.807	1.467	2.036	37.959	20.657	11.392	17.104
13	Inukami R.	11066	109.47	101.626	6.74	0.86	82.16	3.30	2.79	0.41	3.14	0.59	7.760	2.186	0.820	0.802	45.100	24.647	7.002	7.234
14	Ajiki R.	14870	1001.57	14.847	70.24	0.68	1.06	0.14	20.11	1.25	1.80	4.72	24.544	4.425	5.132	4.172	185.768	40.110	36.507	47.858
15	Uso R.	34452	411.42	83.740	51.72	0.97	28.60	1.22	11.05	1.58	2.41	2.45	39.132	7.403	3.870	3.420	312.742	81.385	30.546	27.136
16	Bunroku R.	8323	595.25	13.981	66.79	1.22	15.68	0.90	11.69	0.00	2.35	1.35	21.247	4.018	6.447	5.407	170.537	39.113	55.116	60.821
17	Nomazu R.	5465	757.58	7.214	65.99	0.71	3.58	0.00	20.17	1.13	2.21	6.22	14.473	2.336	4.710	4.045	119.538	24.225	38.565	36.029
18	Echi R.	22957	109.59	211.139	10.41	0.72	77.97	2.85	2.54	0.10	4.79	0.61	8.799	0.936	2.579	0.997	64.881	7.945	22.102	9.721
19	Nagaso R.	11728	3174.29	3.695	18.35	2.40	20.63	1.09	31.40	7.95	5.86	12.32	17.375	4.968	4.116	1.917	138.208	53.009	34.308	46.767
20	Hayama R.	68910	2047.95	33.648	43.70	0.59	7.52	1.19	29.70	3.18	3.53	8.75	14.215	2.556	9.411	2.274	120.184	27.370	69.935	22.470
21	Shin-moriyama R.	14426	2539.60	5.680	54.53	0.19	1.16	0.37	33.73	0.10	0.59	9.34	16.716	4.585	7.420	3.444	134.247	36.162	62.000	29.964
22	Yasu R.	126122	324.12	391.183	19.96	2.51	57.67	2.16	6.50	0.40	5.58	3.11	12.572	2.348	3.311	2.595	92.455	19.354	23.234	20.113
23	Yanomune R.	35935	858.81	41.842	52.27	1.38	19.47	2.18	15.72	0.99	3.91	4.07	22.867	3.154	7.158	4.218	179.294	27.223	61.422	43.261
24	Hino R.	76360	338.11	225.846	31.26	2.14	44.32	2.02	8.42	0.43	4.89	4.41	13.271	3.116	6.309	3.253	82.902	25.800	45.762	29.626
25	U R.	465	66.15	7.028	5.96	0.00	89.06	0.45	0.81	0.32	0.78	2.62	12.367	0.278	1.063	0.917	158.484	2.551	10.894	12.313
26	Wani R.	3203	186.48	17.174	15.96	0.12	65.86	4.66	5.42	1.89	0.93	2.67	8.895	3.876	2.124	1.292	75.623	29.538	16.125	11.239
27	Mano R.	24305	1048.33	23.184	28.45	0.33	42.77	6.63	15.60	1.96	1.13	2.99	9.137	2.074	3.458	2.101	69.424	22.013	24.245	16.789
28	Tenjin R.	5268	539.33	9.768	27.48	0.95	54.70	4.15	7.31	2.79	0.86	1.76	11.925	1.573	5.300	2.673	87.948	14.400	34.817	28.289
29	Fujinoki R.	7116	1804.68	3.943	16.19	0.53	49.08	0.28	20.76	3.49	0.27	9.39	16.805	1.161	0.896	1.596	169.476	10.000	6.440	21.301

Appendix B

The major environmental data and the physical characteristics of the corresponding coastal site.

Site no.	Name of tributary rivers	Sampling location		Wave height (m)	Wave energy flux (kg m day ⁻¹ m ⁻¹)	Lake shore energy flux (kg m day ⁻¹ m ⁻¹)	Coastal slope (1 distance ⁻¹)	Sand beach width (m)	Reed grassfield width (m)	Chl.a of near-shore POM				Conductivity				pH			
		Latitude	Longitude							(μg l L ⁻¹)				(mS m ⁻¹)							
										Nov. 2005	Feb. 2006	May 2006	Jul. 2006	Nov. 2005	Feb. 2006	May 2006	Jul. 2006	Nov. 2005	Feb. 2006	May 2006	Jul. 2006
1	Kamo River	35°17.921'	136°02.645'	0.08	-93.08	15.82	10.72	19	0	2.145	1.168	1.565	1.872	14	14	13	13	8.040	7.340	7.900	7.170
2	Ado R.	35°19.186'	136°04.947'	0.09	15.07	14.42	8.12	8	0	1.333	0.752	1.275	1.872	13	13	13	13	7.440	7.360	6.800	7.620
3	Ishida R.	35°24.185'	136°03.058'	0.08	183.11	33.68	9.37	20	0	1.971	0.752	1.275	3.188	13	14	13	13	7.310	7.660	7.500	7.830
4	Momose R.	35°26.660'	136°03.776'	0.08	109.58	11.76	7.00	18	0	1.739	0.869	2.027	3.237	13	21	12	13	8.050	7.650	6.500	7.600
5	Oura R.	35°28.851'	136°07.307'	-	-	-	8.61	-	-	1.913	0.788	1.722	4.943	13	14	13	12	8.090	7.560	8.800	8.010
6	Shiotsu-o R.	35°30.046'	136°10.137'	-	-	-	5.33	-	-	1.101	1.217	3.980	3.237	13	18	10	13	7.260	7.360	7.700	6.810
7	Chonoki R.	35°24.611'	136°11.902'	0.14	351.31	9.86	25.82	0	13	4.931	1.333	7.948	4.895	15	15	12	14	7.520	7.180	7.100	6.790
8	Ta R.	35°24.025'	136°12.716'	0.15	430.07	11.11	32.61	0	25	5.569	1.043	5.105	5.090	15	15	13	14	7.820	7.290	6.400	6.670
9	Ane R.	35°22.969'	136°13.387'	0.08	11.51	22.08	7.69	28	0	3.016	1.558	3.480	5.821	13	13	13	14	8.160	7.310	7.100	6.650
10	Yone R.	35°21.802'	136°16.010'	0.10	23.17	4.90	25.99	0	35	3.828	1.071	3.538	7.235	25	18	18	17	8.430	8.410	7.400	6.650
11	Amano R.	35°19.405'	136°16.057'	0.23	1214.29	58.72	7.44	23	0	4.292	2.951	2.900	5.382	15	20	15	16	8.210	8.720	8.400	7.250
12	Seri R.	35°16.322'	136°14.039'	0.22	1076.3	76.30	9.07	0	0	4.640	2.900	3.248	5.723	13	17	16	15	7.990		8.200	7.330
13	Inukami R.	35°15.663'	136°12.860'	0.22	950.15	69.40	6.50	10	0	4.350	3.341	4.329	4.212	19	15	15	14	8.050	7.770	8.400	7.430
14	Ajiki R.	35°15.094'	136°12.230'	0.21	830.06	72.24	7.47	11	0	3.654	3.944	3.527	4.309	28	16	17	15	7.360	7.590	7.500	6.920
15	Uso R.	35°14.939'	136°11.881'	0.21	722.72	43.03	32.79	22	0	5.743	3.654	2.935	4.992	15	15	15	14	7.880	7.690	8.000	6.640
16	Bunroku R.	35°14.275'	136°10.378'	0.20	597.17	23.28	6.99	7	0	4.002	2.241	5.105	4.846	16	18	16	14	7.760	7.440	7.500	6.890
17	Nomazu R.	35°13.496'	136°09.118'	0.20	494.56	9.07	23.71	9	0	4.118	3.132	3.538	8.308	16	23	18	18	7.630	7.250	7.000	6.120
18	Echi R.	35°12.803'	136°06.509'	0.18	598.94	40.55	2.06	0	0	2.551	2.319	4.582	5.675	15	13	14	14	7.900	7.790	8.000	5.520
19	Nagaso R.	34°59.772'	135°54.043'	-	-	-	0.82	-	-	1.913	7.716	7.600	3.237	24	17	14	16	7.340	7.650	6.500	6.520
20	Hayama R.	35°02.765'	135°54.453'	0.14	289.63	2.68	6.79	0	21	3.248	14.390	4.060	2.457	22	19	14	14	7.200	7.670	6.800	6.510
21	Shin-moriyama R.	35°04.737'	135°56.542'	0.15	-72.41	6.06	5.20	0	0	1.797	18.887	11.720	6.016	26	26	15	17	6.990	7.810	6.700	6.670
22	Yasu R.	35°07.495'	135°58.443'	0.16	185.82	26.01	15.14	0	0	3.596	3.016	4.234	3.627	29	25	14	18	6.760	6.580	7.200	6.370
23	Yanomune R.	35°08.261'	136°00.800'	0.19	527.06	11.72	7.01	5	0	5.458	3.422	2.881	6.796	21	20	14	17	6.850	6.650	7.000	6.500
24	Hino R.	35°08.908'	136°01.725'	0.20	-881.45	9.74	12.39	105	0	6.572	2.842	4.646	6.601	16	19	14	15	8.300	6.990	7.100	6.680
25	U R.	35°15.685'	135°59.845'	0.08	-68.06	14.17	14.91	14	0	1.449	0.869		2.067	13	13	13	13	8.320	7.720	7.300	7.150
26	Wani R.	35°09.254'	135°56.317'	0.10	-104.75	1.64	20.17	38	0	2.319	2.721	1.913	2.749	14	13	15	13	8.570	7.820	7.100	7.000
27	Mano R.	35°07.434'	135°56.109'	0.07	-65.63	2.89	22.29	11	0	4.060	2.839	5.105	2.310	14	13	13	13	8.560	7.600	7.700	6.170
28	Tenjin R.	35°05.971'	135°55.531'	0.05	55.79	2.57	10.83	0	16	3.016	1.333	3.712	2.993	16	15	13	14	8.290	7.910	7.000	5.580
29	Fujinoki R.	35°03.353'	135°53.729'	0.06	-25.88	14.80	4.56	0	31	1.449	4.733	7.403	2.944	17	16	15	14	7.920	8.050	6.300	7.300
Offshore reference site																					
30	Ie-1	35°12.970'	135°59.959'							3.654	0.869	1.449	4.114								

Appendix C

Carbon and nitrogen stable isotope ratios (‰) of POM and its three potential sources (P-POM, E-POM, and R-POM) at each coastal site.

Site no.	Name of tributary rivers	Nov. 2005						Feb. 2006						May 2006						Jul. 2006					
		POM		E-POM		R-POM		POM		E-POM		R-POM		POM		E-POM		R-POM		POM		E-POM		R-POM	
		$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$
1	Kamo River	5.47	-24.13	5.23	-11.35	3.52	-28.06	12.26	-29.92	4.90	-21.97	4.15	-25.56	5.47	-25.35	6.25	-17.36	3.76	-28.78	4.42	-24.05	6.73	-9.60	6.63	-30.19
2	Ado R.	5.14	-24.73			6.30	-23.37	11.67	-28.54	3.78	-22.93	3.58	-25.18	6.90	-24.69	4.51	-25.90	7.68	-24.19	4.96	-24.57	5.96	-12.44	4.56	-24.91
3	Ishida R.	5.23	-24.96	7.99	-15.99	4.64	-26.58	11.14	-28.27	5.86	-18.68	1.96	-25.18	6.26	-29.69	4.14	-13.02	3.93	-23.23	4.83	-24.39	4.56	-10.35	2.72	-25.58
4	Momose R.	5.48	-25.66	5.00	-9.40	6.69	-29.05	13.21	-28.99	5.15	-14.19	4.27	-27.59	6.39	-30.18	3.76	-15.23	4.97	-27.62	5.36	-24.60	4.79	-12.44	6.26	-28.66
5	Oura R.	6.11	-25.10	4.18	-10.05	3.01	-28.60	9.37	-29.10	4.44	-14.00	2.21	-26.93	5.89	-28.22	4.84	-13.81	2.24	-26.85	4.75	-25.38	5.23	-12.16	3.86	-26.98
6	Shiotsu-o R.	5.99	-24.91	6.49	-12.82	2.27	-28.55	9.21	-28.05	5.40	-21.71	2.73	-27.81	4.87	-28.70	4.10	-20.21	3.25	-26.48	4.67	-26.01	5.22	-11.37	4.71	-27.51
7	Chonoki R.	5.20	-24.84	5.95	-18.71	3.56	-26.62	11.90	-29.34	5.07	-20.79	3.93	-26.84	5.01	-27.43	3.89	-22.50	3.68	-28.91	4.55	-25.07	4.00	-16.19	4.16	-27.01
8	Ta R.	5.21	-24.69	5.63	-15.90	3.64	-27.18	9.32	-27.61			3.24	-27.18	5.05	-26.68			2.80	-27.18	4.99	-24.54			3.84	-27.88
9	Ane R.	5.26	-24.31	5.33	-6.76	3.79	-26.88	9.32	-28.42	6.19	-13.93	3.08	-26.90	6.27	-27.41	6.18	-15.28	4.27	-23.34	4.75	-24.70	7.26	-10.83	5.95	-25.73
10	Yone R.	5.47	-24.00	4.96	-14.46	5.57	-26.40	9.61	-27.54	4.78	-15.07	5.76	-25.26	5.72	-27.23			6.67	-26.91	4.86	-26.35			6.71	-26.40
11	Amano R.	5.09	-24.14	4.50	-12.72	5.55	-25.57	6.77	-27.78	6.60	-17.69	5.96	-24.39	5.76	-26.87	5.72	-15.40	5.33	-22.77	4.79	-23.77	7.96	-18.08	6.18	-24.77
12	Seri R.	5.60	-24.66	7.71	-16.22	6.25	-25.79	7.53	-28.44	8.29	-20.71	4.61	-23.89	7.20	-25.43			7.26	-21.16	5.98	-24.55	7.36	-16.70	10.48	-23.63
13	Inukami R.	5.30	-24.42	6.76	-11.27	5.28	-25.87	7.49	-28.59	7.88	-18.01	5.29	-25.23	7.05	-25.71	5.92	-13.49	5.45	-25.34	5.03	-24.84	6.56	-14.89	7.90	-27.51
14	Ajiki R.	5.64	-25.59	7.44	-16.52	4.87	-28.10	6.90	-28.04	9.65	-27.87	4.90	-27.53	6.71	-25.74			6.13	-28.57	5.34	-24.27	9.13	-16.42	6.61	-27.88
15	Uso R.	6.51	-25.40	7.19	-6.52	4.63	-26.69	7.75	-28.76	8.51	-13.68	5.10	-25.56	7.47	-25.33	7.07	-15.29	6.92	-28.27	5.19	-24.60	7.01	-11.62	8.50	-25.30
16	Bunroku R.	6.27	-25.26			5.78	-27.44	9.46	-29.53	8.52	-16.31	3.89	-26.28	4.99	-26.31			5.63	-28.54	5.25	-24.52	9.27	-15.09	6.79	-26.56
17	Nomazu R.	6.07	-25.25			5.12	-29.29	9.57	-30.68	7.96	-26.82	4.39	-25.42	6.35	-26.04	9.03	-15.71	4.10	-26.67	4.74	-25.05	9.07	-21.16	4.10	-28.22
18	Echi R.	5.73	-25.72	5.18	-13.37	4.01	-28.08	9.41	-29.67	4.75	-21.91	4.75	-25.78	6.38	-26.72	4.33	-21.25	4.61	-19.26	2.95	-26.50	7.36	-16.36	7.22	-20.52
19	Nagaso R.	7.50	-25.64	8.78	-17.14	6.35	-30.83	5.16	-26.91	11.30	-19.23	8.28	-28.66	6.95	-25.13	7.88	-15.45	4.61	-28.14	4.83	-25.42	9.57	-17.95	6.42	-27.90
20	Hayama R.	4.16	-25.30	9.09	-15.93	3.94	-28.46	7.12	-26.29	8.44	-15.76	6.55	-26.84	5.12	-23.91	6.68	-14.32	5.18	-27.21	4.90	-25.17	8.64	-19.10	6.49	-26.02
21	Shin-moriyama R.	8.95	-26.15	10.20	-22.62	4.98	-28.20	7.82	-25.86	8.43	-18.18	6.29	-27.83	7.13	-26.69	7.94	-20.44	6.24	-27.43	5.91	-27.89	9.04	-21.21	5.52	-26.11
22	Yasu R.	5.88	-25.31			4.47	-28.36	8.70	-29.72			5.21	-25.12	5.49	-25.56			9.52	-24.68	3.86	-26.97			7.69	-29.62
23	Yanomune R.	7.27	-24.99			4.26	-27.63	8.38	-29.81	7.17	-17.88	4.48	-26.62	6.32	-26.19	7.04	-21.39	5.30	-26.34	4.81	-25.54	7.28	-21.01	6.25	-26.59
24	Hino R.	7.13	-24.27	8.42	-17.74	6.41	-28.86	8.75	-29.48	8.87	-19.08	6.44	-25.20	6.73	-26.95	7.47	-21.06	7.23	-26.24	5.22	-24.51	9.11	-21.45	10.12	-23.40
25	U R.	5.64	-26.07	3.90	-11.95	1.64	-29.62	11.65	-27.67	4.77	-19.70	3.84	-27.47	7.21	-24.45	5.07	-14.35	7.26	-26.81	4.02	-24.07	4.22	-13.70	4.06	-28.06
26	Wani R.	5.56	-25.32	4.58	-9.58	3.75	-29.33	9.85	-30.83	4.63	-16.30	6.54	-26.38	6.45	-24.98	5.31	-15.74	4.15	-24.28	4.51	-24.26	5.93	-11.09	4.12	-25.65
27	Mano R.	6.66	-24.64	4.63	-8.73	4.09	-29.87	7.74	-28.66	5.00	-14.21	3.94	-25.52	7.22	-23.82	5.55	-14.86	2.69	-28.75	4.95	-24.12	6.36	-15.59	8.82	-28.99
28	Tenjin R.	6.24	-24.74	4.48	-17.52	4.41	-25.91	9.48	-28.64	6.33	-23.14	3.75	-23.91	7.13	-23.38	6.58	-18.69	6.93	-25.70	5.14	-25.69	6.46	-13.85	8.83	-25.63
29	Fujinoki R.	8.29	-22.09	7.10	-23.78	5.02	-28.48	8.77	-26.95	7.76	-18.85	5.16	-25.83	6.82	-19.98	5.52	-18.34	5.87	-29.71	6.14	-25.90	8.60	-13.86	10.12	-28.07
Offshore reference site																									
30	Ie-1 (offshore reference)	5.54	-24.65					12.90	-29.44					7.25	-24.38					5.02	-25.00				

Appendix D

The relative contribution (%) of the three potential sources (P-POM, E-POM and T-POM) to POM at each coastal site.

Site no.	Name of tributary rivers	Nov. 2005						Feb. 2006						May 2006						Jul. 2006					
		P-POM		E-POM		T-POM		P-POM		E-POM		T-POM		P-POM		E-POM		T-POM		P-POM		E-POM		T-POM	
		Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
1	Kamo River	91.3	87 - 96	4.8	4 - 6	3.8	0 - 7	98.2	97 - 99	0.4	0 - 1	1.4	0 - 3	38.2	35 - 42	15.3	13 - 17	46.5	44 - 49	95.9	94 - 98	3.2	2 - 5	0.9	0 - 3
2	Ado R.							86.4	86 - 87	12.0	10 - 14	1.6	0 - 4	38.0	0 - 86	20.9	14 - 27	41.1	0 - 76	78.7	57 - 97	3.4	3 - 4	17.8	0 - 40
3	Ishida R.	31.1	0 - 69	9.6	2 - 16	59.3	29 - 85	81.6	81 - 82	6.2	5 - 7	12.2	11 - 13	-		-		-		88.0	84 - 92	4.5	4 - 5	7.5	4 - 11
4	Momose R.	80.5	79 - 83	0.3	0 - 1	19.2	17 - 21	98.7	98 - 100	0.7	0 - 2	0.7	0 - 2	-		-		-		58.0	47 - 69	12.0	9 - 15	30.1	22 - 38
5	Oura R.	99.3	99 - 100	0.3	0 - 1	0.3	0 - 1	69.0	67 - 71	0.0	0 - 0	31.0	29 - 33	-		-		-		75.2	66 - 85	0.8	0 - 2	24.0	15 - 32
6	Shiotsu-o R.	98.1	96 - 100	1.1	0 - 2	0.7	0 - 2	60.6	60 - 61	12.3	11 - 14	27.1	25 - 29	-		-		-		3.0	0 - 6	8.7	8 - 9	88.3	86 - 91
7	Chonoki R.	79.3	71 - 81	2.8	0 - 6	17.9	13 - 23	90.0	89 - 91	0.0	0 - 0	10.0	9 - 11	33.7	32 - 37	0.9	0 - 3	65.4	63 - 68	47.3	36 - 58	9.2	7 - 12	43.5	34 - 53
8	Ta R.	79.3	71 - 87	2.8	0 - 6	17.9	13 - 23																		
9	Ane R.	80.5	74 - 87	3.8	3 - 5	15.7	10 - 21	63.3	63 - 64	0.7	0 - 1	36.0	35 - 37	-		-		-		99.5	99 - 100	0.5	0 - 1	0.0	0 - 0
10	Yone R.	46.9	0 - 94	13.2	6 - 20	39.8	0 - 80	54.3	53 - 55	0.3	0 - 1	45.5	44 - 47												
11	Amano R.	41.3	0 - 95	8.6	5 - 14	50.1	0 - 91	35.2	34 - 36	0.4	0 - 1	64.4	63 - 66	-		-		-		86.3	82 - 90	10.2	9 - 12	3.5	0 - 9
12	Seri R.	90.8	82 - 100	1.0	0 - 2	8.2	0 - 17	54.0	54 - 54	0.5	0 - 1	45.5	45 - 46							81.0	79 - 83	2.7	2 - 4	16.3	15 - 18
13	Inukami R.	27.2	0 - 67	7.5	3 - 11	65.2	30 - 91	47.0	47 - 47	0.0	0 - 0	53.0	53 - 53	45.8	39 - 55	0.2	0 - 1	54.0	45 - 61	97.2	95 - 99	1.8	1 - 3	1.0	0 - 2
14	Ajiki R.	10.6	0 - 28	19.9	15 - 23	69.6	57 - 77	22.7	20 - 26	4.0	0 - 10	73.2	70 - 76							90.2	88 - 92	8.7	8 - 9	1.2	0 - 3
15	Uso R.	96.8	92 - 100	0.7	0 - 2	2.5	0 - 6	49.5	49 - 50	0.0	0 - 0	50.5	50 - 51	78.4	75 - 83	0.6	0 - 2	21.1	17 - 24	93.5	91 - 96	3.0	3 - 3	3.5	1 - 6
16	Bunroku R.							72.0	72 - 72	0.0	0 - 0	28.0	28 - 28							92.4	89 - 96	5.2	4 - 6	2.5	0 - 6
17	Nomazu R.							78.1	71 - 84	12.0	0 - 29	9.8	0 - 19	53.5	53 - 54	0.0	0 - 0	46.5	46 - 47	91.2	83 - 100	1.1	0 - 3	7.7	0 - 14
18	Echi R.	81.0	79 - 83	0.2	0 - 1	18.8	17 - 20	72.3	71 - 73	0.7	0 - 2	27.0	25 - 29	-		-		-		-		-	-	-	
19	Nagaso R.	2.0	0 - 6	37.5	36 - 40	60.5	57 - 64	-		-		-		82.5	80 - 85	0.3	0 - 1	17.2	15 - 19	94.0	93 - 95	0.0	0 - 0	6.0	5 - 7
20	Hayama R.	29.2	0 - 63	10.3	0 - 19	60.4	37 - 81	7.0	6 - 8	6.5	6 - 7	86.5	85 - 88	1.9	0 - 6	24.0	21 - 29	74.1	71 - 78	97.5	95 - 100	0.0	0 - 0	2.5	0 - 5
21	Shin-moriyama R	0.8	0 - 2	55.3	55 - 56	44.0	43 - 45	15.8	15 - 17	23.0	22 - 24	61.2	60 - 63	38.0	36 - 40	0.8	0 - 2	61.2	60 - 62	0.5	0 - 1	0.0	0 - 0	99.5	99 - 100
22	Yasu R.																								
23	Yanomune R.							62.0	60 - 64	0.0	0 - 0	38.0	36 - 40	28.1	24 - 33	1.1	0 - 3	70.9	67 - 73	87.7	85 - 91	0.2	0 - 1	12.1	9 - 15
24	Hino R.	6.6	1 - 13	38.8	37 - 41	54.6	50 - 59	61.5	61 - 62	0.0	0 - 0	38.5	38 - 39	1.6	0 - 4	0.4	0 - 1	98.0	96 - 100	91.0	91 - 91	8.5	8 - 9	0.5	0 - 1
25	U R.	85.0	85 - 85	0.0	0 - 0	15.0	15 - 15	83.0	83 - 83	16.5	16 - 17	0.5	0 - 1	81.6	63 - 100	3.0	0 - 6	15.4	0 - 31	0.7	0 - 1	27.7	27 - 28	71.7	71 - 72
26	Wani R.	87.5	84 - 92	0.3	0 - 1	12.2	8 - 15	82.0	81 - 83	0.0	0 - 0	18.0	17 - 19	77.3	52 - 96	0.2	0 - 1	22.5	3 - 48	25.8	15 - 38	8.4	8 - 9	65.8	54 - 76
27	Mano R.	95.6	92 - 100	2.7	0 - 7	1.7	0 - 5	53.5	53 - 54	0.0	0 - 0	46.5	46 - 47	94.5	94 - 95	5.5	5 - 6	0.0	0 - 0	91.5	91 - 92	8.5	8 - 9	0.0	0 - 0
28	Tenjin R.	100.0	100 - 100	0.0	0 - 0	0.0	0 - 0	70.7	70 - 72	2.2	0 - 5	27.1	24 - 30	69.5	56 - 84	19.9	16 - 23	10.5	0 - 22	83.5	82 - 85	0.0	0 - 0	16.5	15 - 18
29	Fujinoki R.	0.5	0 - 1	99.5	99 - 100	0.0	0 - 0	44.3	43 - 45	7.0	6 - 8	48.7	47 - 50	36.2	35 - 38	63.5	62 - 65	0.3	0 - 1	76.0	75 - 77	0.0	0 - 0	24.0	23 - 25