

Title	Seasonal and site-specific variability in terrigenous particulate organic carbon concentration in near-shore waters of Lake Biwa, Japan
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25

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.

44

45 **Keywords**

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

47

48

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

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

81           Here, we measured carbon ( $\delta^{13}\text{C}$ ) and nitrogen ( $\delta^{15}\text{N}$ ) stable isotope ratios of  
82 POM in near-shore water samples collected during four seasons at different locations set  
83 along the shore-line of Lake Biwa. In general,  $\text{C}_3$  terrestrial plants (T-POM) have  
84 markedly different  $\delta^{13}\text{C}$  (-27‰) from that of aquatic microalgae (P-POM and E-POM;

85 Marshall et al. 2007). Moreover, among aquatic primary producers, phytoplankton and  
86 epilithic algae have different  $\delta^{13}\text{C}$ , due to the  $\text{CO}_2$  diffusion limitation within the  
87 boundary layer surrounding algal cells (Phytoplankton:  $-32 \pm 2$  (SD) ‰, epilithic algae:  
88  $-17 \pm 2$  (SD) ‰; Fry and Sherr 1984; France 1995). In addition,  $\delta^{15}\text{N}$  of aquatic primary  
89 producers may be variable among aquatic environments depending on the extent of  
90 anthropogenic nitrogen inputs (McClelland and Valiela 1998; Kohzu et al. 2008; Karube  
91 et al. 2010). The above differences in isotope signatures of POM with different origins  
92 are useful for estimating the relative contribution of P-POM, E-POM and T-POM by  
93 means of stable isotope mixing models (Fry 2006).

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



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

109

## 110 Materials and Methods

### 111 Sample collections

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

121 HORIBA).

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

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

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

142           Second, to determine isotopic values of epilithon-derived POM (E-POM),  
143 epilithon samples were collected at all the sites except for some sites where boulders of  
144 appropriate size were not found (Fig. 1). At each site, epilithic organic matter scraped  
145 off, with a brush, from five boulders of moderate size (30-40 cm) which have a surface  
146 area of more than 36cm<sup>2</sup> on the upper side were mixed, filtered through precombusted  
147 GF/F filters, and served for the stable isotope analyses.

148           Third, we collected riverine particulate organic matter (R-POM) as a proxy of  
149 terrigenous POM (T-POM) at sites 0.5-4 km upstream from the mouth, in the same  
150 months as the near-shore sampling but in the previous year (i.e., July 2004, November  
151 2004, February 2005 and May 2005: see Kohzu et al. 2009 for details). In general,  
152 R-POM is composed of T-POM and fluvial E-POM (Kohzu et al. 2009). In our study  
153 rivers, however,  $\delta^{13}\text{C}$  of R-POM was significantly different from that of fluvial E-POM  
154 in all four months (Tukey's post-hoc test, R-POM vs. fluvial E-POM, Nov:  $p < 0.001$ ,  
155 Feb:  $p < 0.01$ , May:  $p < 0.01$ , Jul.:  $p < 0.001$ ) but not from C<sub>3</sub> terrestrial plant detritus, i.e.,  
156 rice straw collected from paddy fields in any months (Tukey's post-hoc test, R-POM vs.

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

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

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

175 composition of near-shore POM. All these POM samples collected on precombusted  
176 GF/F filters were exposed to 0.1 M HCl to eliminate carbonates, rinsed with distilled  
177 water, and then stored at  $-20^{\circ}\text{C}$  until the stable isotope analysis.

178

179 Stable isotope analysis

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

187 
$$\delta^{13}\text{C} \text{ or } \delta^{15}\text{N} = (R_{\text{sample}} / R_{\text{standard}} - 1) \times 1000 (\text{‰}),$$

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

192

193 Isotope mixing model

194 Relative contributions of three potential organic sources (i.e., P-POM, E-POM  
195 and T-POM) to POM collected at each near-shore site were estimated using an isotope  
196 mixing model with  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  (IsoSource Program; Phillips and Gregg 2003). This  
197 model enabled us to estimate all possible combinations of each source contribution  
198 (0–100%) in 1% increments to create a set of predicted sources. The initial tolerance  
199 was set at 0.1‰. If the estimated values were outliers (i.e., < 0% or > 100%), the  
200 tolerance was increased in 0.1‰ steps to 2‰. The model outputs are expressed as the  
201 mean value and range (minimum–maximum) for each source. From the estimated mean  
202 contribution of each organic carbon source to POC concentration ( $\text{mgL}^{-1}$ ) at each  
203 near-shore site, we calculated the concentrations of P-POC, E-POC and T-POC  
204 according to the following equation.

$$205 \quad \text{P-POC (mgL}^{-1}\text{)} = \text{POC (mgL}^{-1}\text{)} \times f_p$$

$$206 \quad \text{E-POC (mgL}^{-1}\text{)} = \text{POC (mgL}^{-1}\text{)} \times f_e$$

$$207 \quad \text{T-POC (mgL}^{-1}\text{)} = \text{POC (mgL}^{-1}\text{)} \times f_t$$

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

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

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

216

217 Land use pattern and physical environments

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

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

228 River discharge (m<sup>3</sup> S<sup>-1</sup>) = 0.0284 × catchment area<sup>0.9502</sup> (R<sup>2</sup>=0.846, p<0.01)

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

235 As physical characteristics of coastal environments, we used wave height (m),  
236 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.  
237 2006), and coastal slope ( $1 \text{ d}^{-1}$ ;  $d$  = horizontal distance from the shoreline to the point  
238 with a depth of 2 m). These data are available at  
239 <http://www.biwakokasen.go.jp/others/kankyoujyouhou/index.html> (Kinki Regional  
240 Development Bureau, Ministry of Land, Infrastructure, Transport and Tourism).

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

242

243 Statistical analysis

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



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

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

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

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

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

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

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

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

312

## 313 Results

### 314 Spatiotemporal variation in POC concentration and composition

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

319 showed high estimation accuracy (Appendix D), except for some sites (e.g., site no. 3,  
320 10, 11, 13, 20 in November, 2005), in which P-POM and R-POM had similar isotopic  
321 signatures (Appendix C). Based on the relative contribution of these sources, we  
322 calculated the concentration of P-POC, E-POC, and T-POC (Table 1 & Fig. 4). The  
323 near-shore POC concentrations differed among sites (Fig. 4) and seasons ( $F_{3, 83} = 13.56$ ,  
324 Jul. > May = Feb. > Nov.,  $p < 0.05$ ). P-POC and T-POC were the dominant organic  
325 sources of the near-shore POC, whereas E-POC concentration was consistently low (Fig.  
326 5). In July, the concentration of P-POC was generally higher than that T-POC (Fig. 5).  
327 T-POC concentrations tended to be high at the sites located along southern and eastern  
328 shores, especially in February and May (Fig. 4).

329

330 Possible factors that influences the spatiotemporal dynamics of T-POC

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

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

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

348

#### 349 Discussion

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

355 near-shore T-POC concentration via R-POC inputs (Path 2 & 4). Large contribution of  
356 T-POC in May could be related to agricultural practices in this season. Previous work  
357 has reported that a large quantity of paddy-derived wastewater flows into the lake basin  
358 at the onset of the rice irrigation period between late April and early May (Fujii et al.  
359 2002; Haga and Ohtsuka 2003; Ohkubo and Azuma 2005; Hama et al. 2010). The  
360 results of the present study support the notion that agricultural activities, especially  
361 paddy irrigation, can be a significant factor that affects T-POC concentrations in  
362 near-shore environments of Lake Biwa. It is also interesting to note that %paddy field  
363 area had a positive effect on R-POC concentration even in February when paddy field  
364 activities are considered to be minimal. This suggests the possibility that organic-rich  
365 soils can be eroded from paddy fields even in winter, due to snowing and melting.  
366 Although the underlying mechanisms have not yet been poorly understood, they remain  
367 to be investigated in the future.

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

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

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



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

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

409 the near-shore POM for the following two reasons. First, in Lake Biwa, especially in  
410 deep north basin, the area of shallow habitats available to macrophytes is  
411 disproportionately small relative to that of the whole basin, so that their total primary  
412 production is much lower than that of phytoplankton and of even epilithon whose  
413 euphotic zone is deeper than the macrophytes (Sakamoto 2000). Second, we have  
414 technical difficulty in separating macrophyte detrital fraction from a mixture of POM  
415 with different origins in the near-shore water for the stable isotope analysis and thus in  
416 characterizing isotopic alteration during their decomposition in nature. To evaluate their  
417 contribution, one of promising approaches is polyunsaturated fatty acid (PUFA) analysis,  
418 which can quantify each of organic sources by characterizing the composition of  
419 primary producer-specific polyunsaturated fatty acids (Kelly and Scheibling2012). In  
420 floodplain lakes of the central Brazilian Amazon basin, for instance, the technique  
421 revealed that macrophyte detritus is a primary component of suspended POM during  
422 low water season (Mortillaro et al. 2011). The application of PUFA analysis, even if it is  
423 beyond our scope in the present study, will be a future challenge for better  
424 understanding of seasonal changes inorganic matter flows in Lake Biwa, especially in  
425 shallow south basin where submerged plants have recently luxuriated.

426           Using stable isotope approaches, we demonstrated that allochthonous inputs of

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

433

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445

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574

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**

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

599

600 **Fig. 5**

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

603

604 **Fig. 6**

605 The path model based on the simultaneous analysis of multiple populations with  
606 structured means. The basic framework is the same as that in Figure 2. Dark and light  
607 arrows are significant ( $p < 0.05$ ) and not significant ( $p > 0.05$ ) paths, respectively.

608 Values with arrows are path coefficients. The subscripts of R-POC and log (T-POC)  
609 indicate the total effect on these variables.

610

611

612 **Table 1**

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

614 ( $\text{mgL}^{-1}$ ) 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

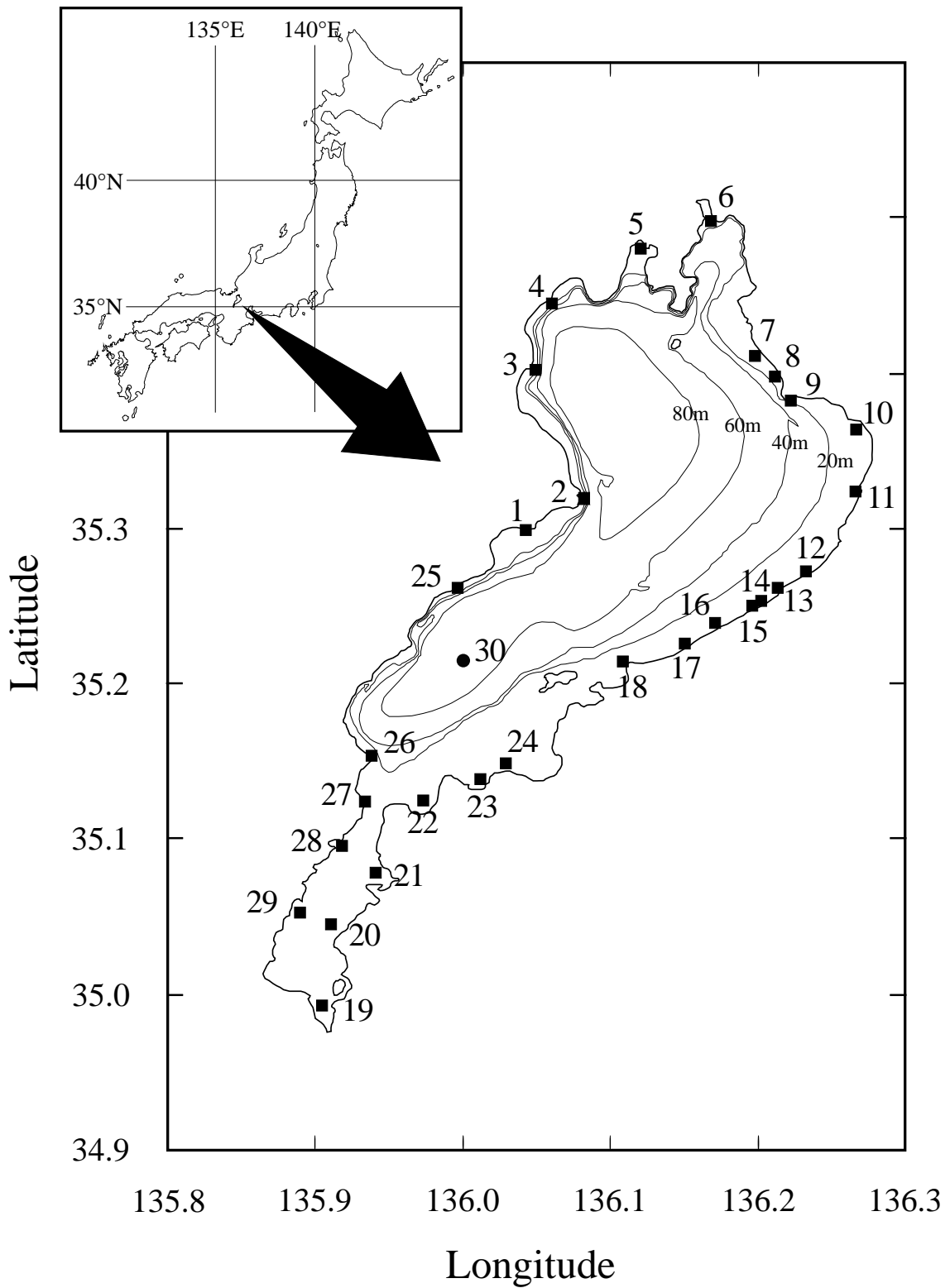


Fig. 1



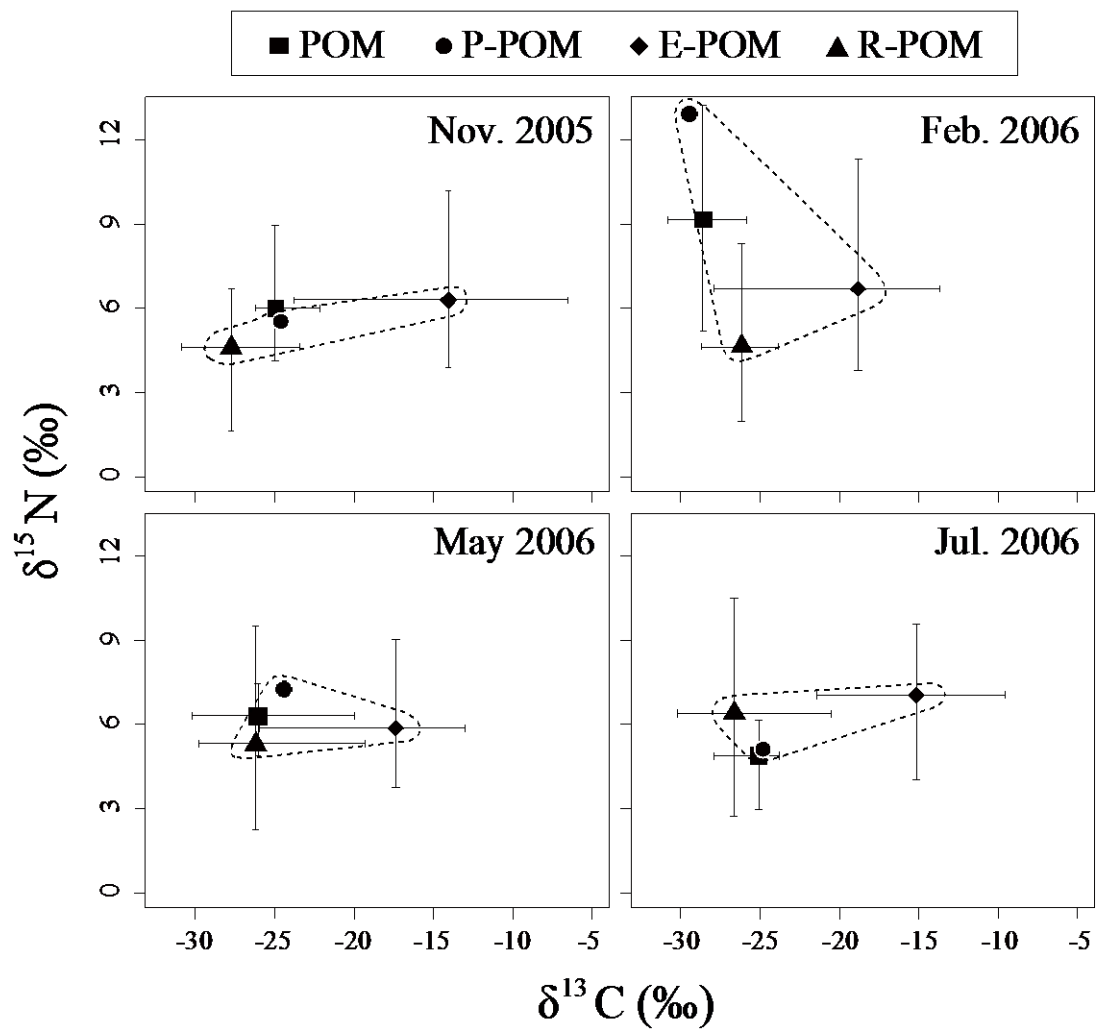


Fig. 2

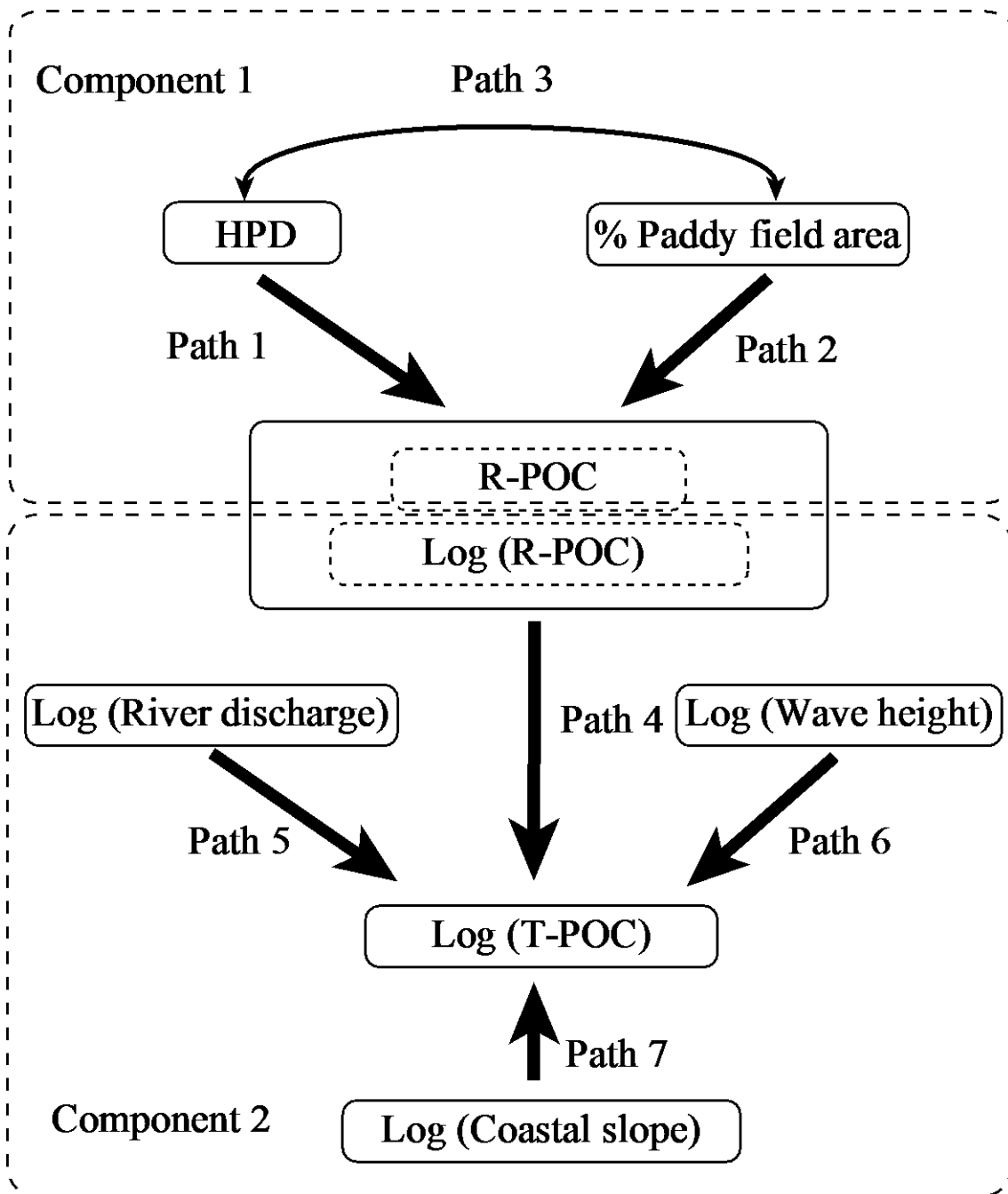


Fig. 3

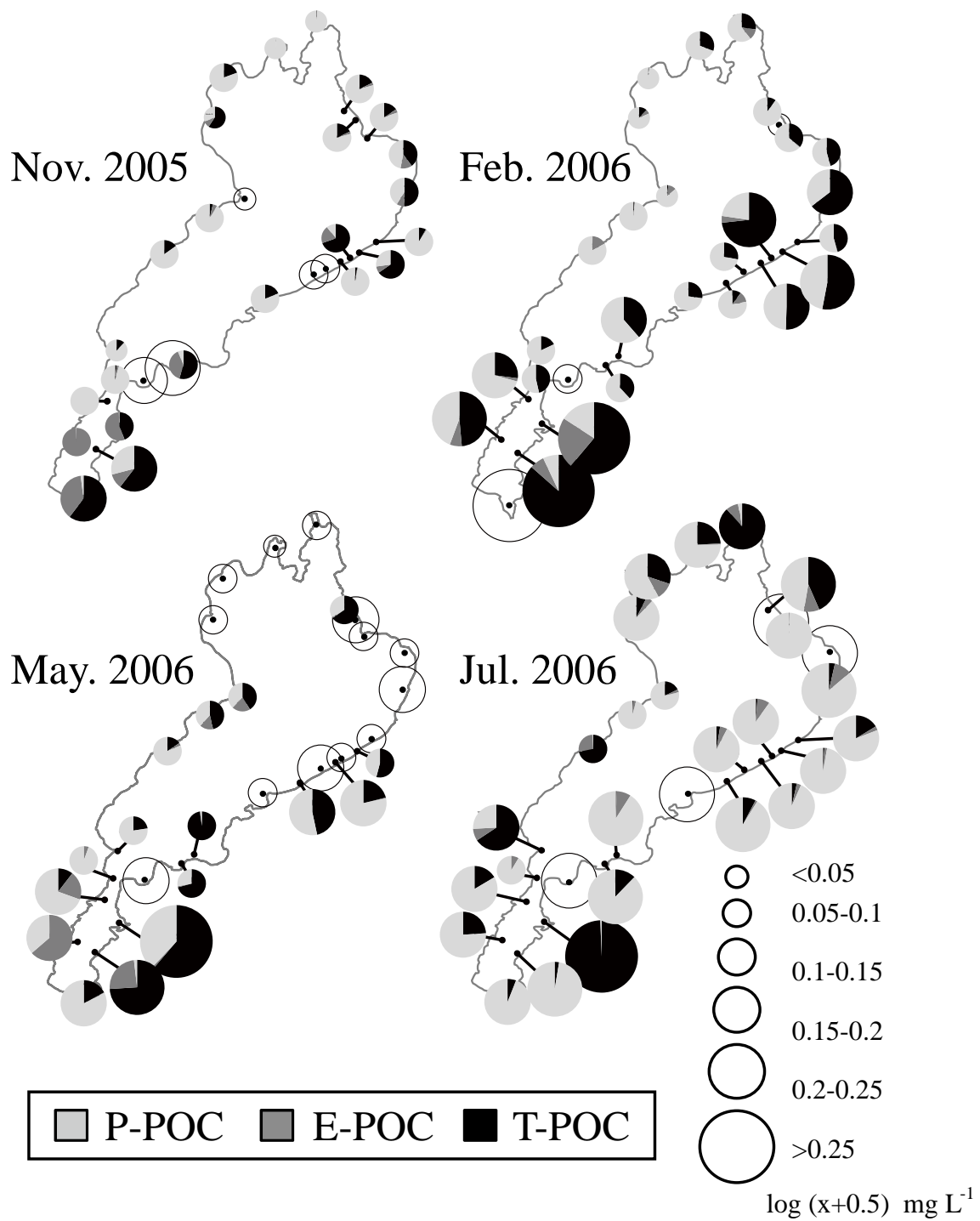


Fig. 4

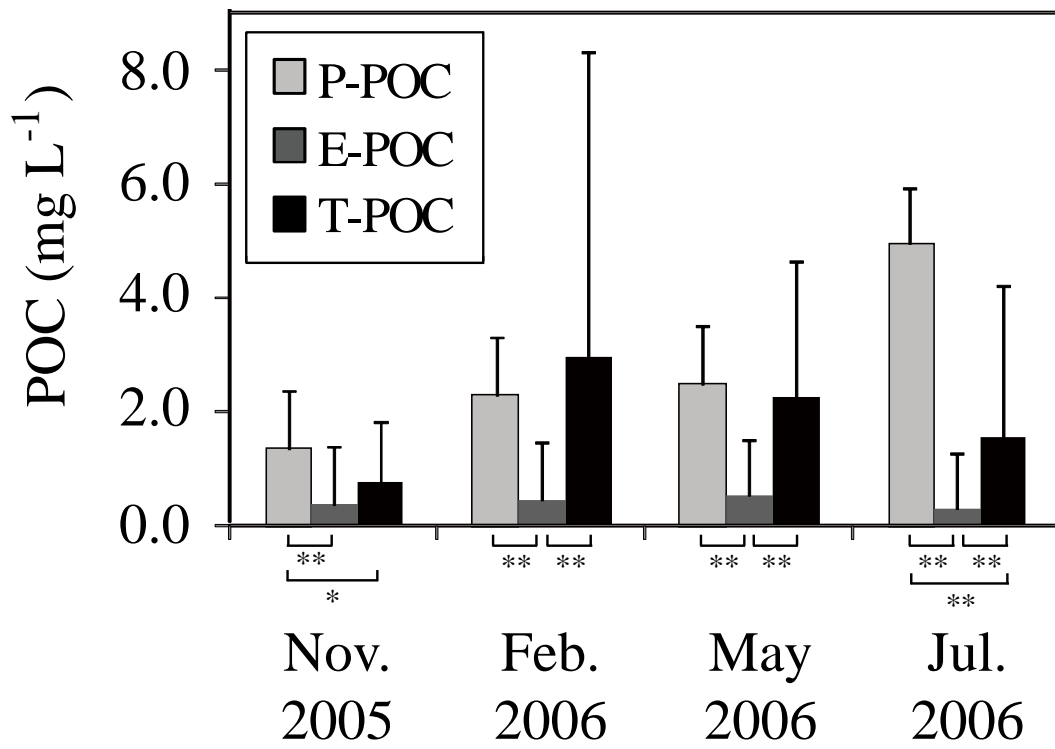
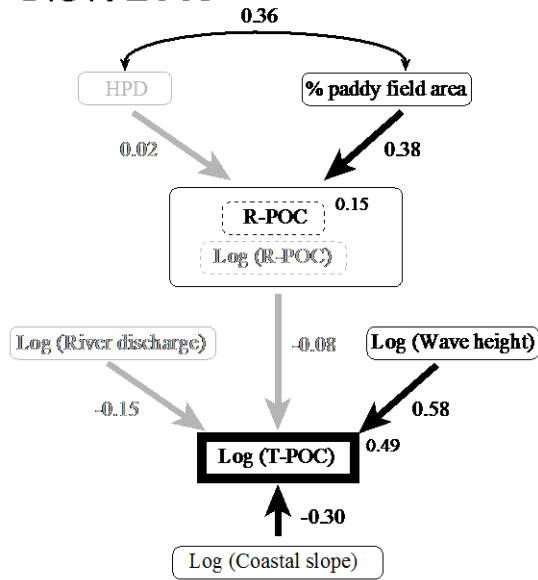
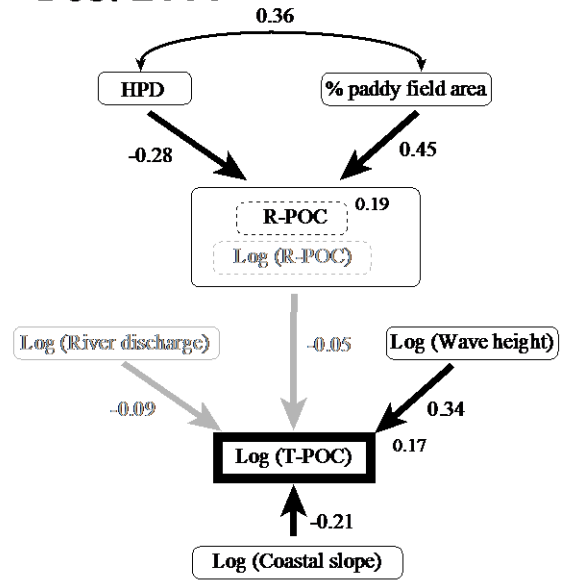


Fig. 5

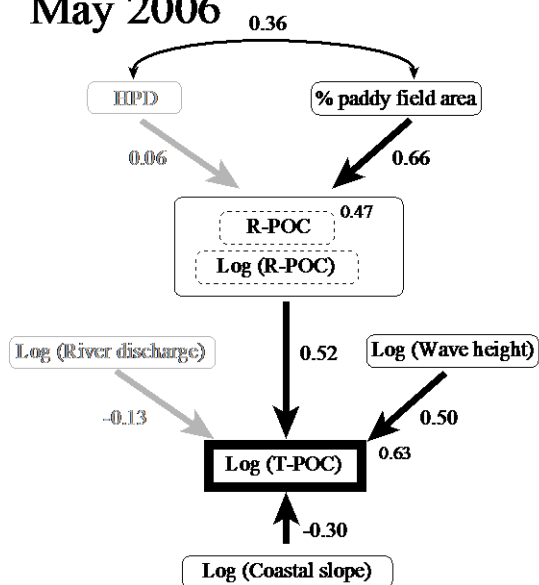
Nov. 2005



Feb. 2006



May 2006



Jul. 2006

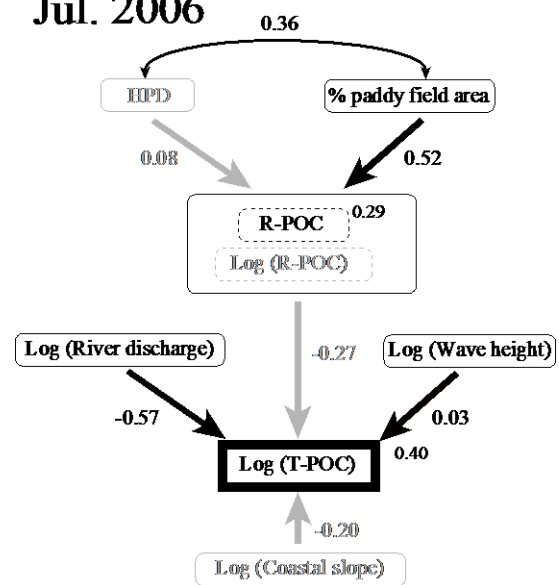


Fig. 6

Table 1

Measured POC concentration and estimated P-POC, E-POC and T-POC concentrations (mg L<sup>-1</sup>) 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 <sup>-2</sup> )	Catchment area (km <sup>2</sup> )	% 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) (μ mol L <sup>-1</sup> )				Riverin Particulate Organic Carbon (POC) (μ mol L <sup>-1</sup> )			
													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
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 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	5.75	-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	