

Temporal Variation of Nitrate and Phosphate Transport in Headwater Catchments: the Hydrological Controls and Landuse Alteration

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Abstract

Oceania Rivers are hotspots of DIN (dissolved inorganic nitrogen) and DIP (dissolved inorganic phosphorus) transport due to humid/warm climate, typhoon-induced episodic rainfall and high tectonic activity that create an environment favorable for high/rapid runoff and soil erosion. In spite of its uniqueness, effects of hydrologic controls and land use on the transport behaviors of DIN and DIP are rarely documented. A 2-years monitoring study for DIN and DIP from three headwater catchments with different cultivation gradient (0 To 8.9%) was implemented during a ~3-day interval with additional monitoring campaign at 3-hour interval during typhoon periods. Results showed the DIN yields in the pristine, moderately cultivated (2.7%), and intensively cultivated (8.9%) watersheds were 8.3, 26, and 37 kg-N/ha/yr, respectively. For the DIP yields, they were 0.36, 0.35, and 0.56 kg-P/ha/yr, separately. Year-roundly higher DIN concentrations and five times increase in DIN yield in intensively cultivated watershed indicates DIN is more sensitive to landuse changes. The high background DIN yield from relatively pristine condition is attributed to high atmospheric nitrogen deposition and large subterranean N pool. The correlations of runoff-concentration reveals that typhoon flood purges out more DIN sourced from subterranean reservoir, by contrast, washes off surface soil resulting in higher suspended sediment accompanying with higher DIP. Collectively, typhoon runoff contributes 20-70% and 47-80%, respectively, to the annual DIN and DIP exports. The DIN yield to DIP yield ratio varied from 97 to 410, higher than the global mean of ~18, indicating a P-limiting condition in stream and downstream aquatic environment. Based on our field observation, we constructed a conceptual model illustrating different remobilization mechanisms for DIN and DIP from headwaters in a mountainous river, which is analogous to typical Oceania Rivers and the headwater of large rivers in similar climate zone. Our study advanced our understanding about the role of cyclone, which exerts hydrological control, and landuse on nutrient export in Oceania region benefiting watershed managements under the context of climate change.

Keywords: Oceania Rivers, headwater catchment, DIN, DIP, nutrient fluxes

50 1. Introduction

51 The global biogeochemical cycles of nitrogen (N) and phosphorus (P) have been significantly
52 altered due to the increasing demand of food and energy consumption caused by increasing
53 population and human activities (Galloway and Cowling, 2002; Seitzinger et al., 2010). In the past
54 five decades, the rate at which biologically available nitrogen enters the terrestrial biosphere has
55 doubled. The increase in the rate stems from human disturbances, such as fertilizer production, fossil
56 fuel combustion, cultivation, and livestock industry (Galloway et al., 2004). The phosphorus enters
57 into the environment has also been doubled due to mining and the use of rock phosphate as fertilizer,
58 detergent additives, animal feed supplements, and other technical uses (Bennett et al., 2001; USGS,
59 2008). The increasing discharges of dissolved inorganic nitrogen (DIN) and phosphorus (DIP) from
60 rivers may subsequently induce eutrophication in freshwater and coastal marine ecosystems (Turner
61 et al., 2003; Duan et al., 2007; Conley et al., 2009) resulting in seasonal hypoxia, harmful algal
62 blooms, and losses in fishery production in aquatic ecosystems (Lu et al., 2011; Billen and Garnier,
63 2007; Diaz and Rosenberg, 2008; Howarth et al., 1996; Rabalais, 1996). Moreover, the imbalanced
64 export of DIN and DIP from land/soil that cause a change in the riverine DIN:DIP ratio may alter
65 phytoplankton community structures, consequently, deteriorating the ecosystem (Justic et al., 1995;
66 Howarth et al., 1996; Rabalais et al., 1996; Elser et al., 2009).

67 Oceania is a region centered on the islands of the tropical Pacific Ocean. Oceania ranges from
68 the coral atolls and volcanic islands of South Pacific to the entire insular region between Asia and the
69 Americas, including Australasia and the Malay Archipelago. Oceania Rivers accounts for 4.5% of the
70 total land surface area on the Earth and exports up to 12% and 35% of the global water and sediment
71 discharge, respectively (Milliman et al., 1999). High precipitation, steep slopes, small basin areas,
72 and frequent flood events can induce high erosion rates on Oceania islands (Kao and Milliman,
73 2008). The mass flux from such small rivers might also have unique biogeochemical significance
74 since many of these rivers discharge material onto narrow shelves and canyons, facilitating material
75 bypass to the deep sea (Milliman, 1995; Nitttrouer et al. 1995; Carey et al. 2002; Lyons et al. 2002;

76 Kao et al. 2006; Leithold et al. 2006). Model studies have demonstrated that Oceania Rivers are
77 significant sources of global DIN and DIP export (Seitzinger et al., 2005; 2010) and projected that
78 the riverine N flux in Oceania will increase to over 10% by 2030 (Bouwman et al., 2005). However,
79 those models are mainly extrapolation from large rivers with limited data from Oceania Rivers
80 (Meybeck, 1982; Caraco and Cole, 1999; Smith et al., 2003). A comprehensive study for Oceania
81 Rivers is difficult due to the effect of frequent typhoon events, which plays the critical role in
82 material transport, on field sampling. By comparison, Taiwan has relatively well resources for
83 researchers working on these topics (Kao and Liu, 2000; Kao et al., 2005).

84 Taiwan is a subtropical mountainous island with the maximum elevation of ~4000ma.s.l. and
85 ~70% area is above 100ma.s.l. The island-wide annual rainfall is approximately ~2400 mm, which is
86 over 3 times of the global average (Legates, 1995). Due to steep slope and small watershed, about
87 70% annual rainfall turns into runoff. During May to October, the season of growing and fertilizer
88 application for agriculture, 3–5 typhoons invade Taiwan bringing up episodic runoff facilitating high
89 erosion and nutrient export to the aquatic system. In headwater catchments, agriculture-associated
90 landuse is limited by government rules yet nutrient export is extremely sensitive to the runoff and
91 landuse type coverage (Huang et al., 2012).

92 In this paper, we investigated the DIN and DIP fluxes for headwater catchments for two years
93 (2007–2008). The nitrate and phosphate concentrations were measured in three headwater
94 catchments representing different cultivation extents. These catchments were monitored during a 3
95 -day interval, and were supplemented by typhoon sampling at a 3-hour interval. The study is useful
96 to the watershed biogeochemistry field because our work captured these fluxes for (1) the Oceania, a
97 globally significant region where exports from Oceania Rivers are known to be significant and
98 increasing; (2) smaller catchments which represent lotic environments that are most vulnerable to
99 landuse activities while at the same time critical to our understanding of source inputs to larger river
100 systems; and (3) typhoon events of varying strength enabling some assessment of how fluxes behave
101 during these events which are likely to continue to change with climate variation.

102

103 **2. Study site**

104 The Chichiawan Watershed located in Central Taiwan has a drainage area of 105 km² and
105 elevations ranging from 1131 to 3882 m above sea level. The mean daily air temperature is 13.5 °C
106 with an average of 8.9 °C in January and 17.7°C in July (see more details in Fig. 2). The annual
107 runoff is ~3300 mm, and ~75% of the runoff occurs during the wet season (May to Oct), primarily
108 during the typhoon periods. The rest months (Nov. to April in the next year, dry season) occupies the
109 other 25%.

110 This creek consists of three major tributaries, namely, Gaoshan (area = 21 km²), Yikawan (area
111 = 53 km²), and Yusheng Creeks (area = 31 km²)(Fig. 1). The Gaoshan and Yikawan Creeks are the
112 only habitat for the Formosan Landlocked Salmon (*Oncorhynchus masou formosanus*) (Tung et al.,
113 2006; Lee et al., 2012). In this study, the high-frequency water sampling sites were installed. G
114 stands for pristine watershed, Gaoshan Creek (0% cultivated land). K locates at downstream of
115 intercept of Gaoshan Creek and Yikawan, which represents moderately cultivation (2.7% cultivation).
116 Y stands for Yusheng Creek, which represents intensively cultivated watershed (Table 1). Although
117 Yusheng Creek Watershed has only 8.9% agriculture land which is not intensive at all when
118 comparing with the cultivated extent in the Continents, it not only resulted in a low water quality
119 index but also a degradation in benthic algae biodiversity (Yu et al., 2005). Moreover, the
120 mountainous watersheds are more sensitive to farming activity (Huang et al., 2012) even a few
121 percent cultivation is influential particularly the water quality demand of upstream biome is very
122 high.

123 Three precipitation gauges maintained by the Taiwan Power Company are located in this area
124 (Fig.1). The most west gauge monitors air temperature as well. Two hydrologic gauges monitor
125 water levels, one for Yusheng Creek and one for the entire Chichiawan (all three creeks). The
126 consecutive water level will be transferred into hourly water discharge via a rating curve of water
127 discharge against water level, which is calibrated each year. The most downstream gauge (Chichi

128 awan) subtracts the discharge for Yusheng Creek will be the discharge for the Sta. K. The discharge
129 for G is derived from area proportion of Sta. K (Kao et al., 2004; Huang et al., 2012). The average
130 daily discharge for Chichiawan and Yusheng Creeks are 7.94 and 2.41 m³/sec, respectively. During
131 the wet season, the average daily discharges are 11.80 and 4.07 m³/sec, respectively.

132 The study watersheds represent the typical landscape in mountainous regions (i.e., small
133 proportion of agricultural activities located along the riparian zone) where some of the residents earn
134 their living by growing vegetables and fruits (Huang et al., 2012). The landuse pattern and the
135 landscape characteristics for each catchment are shown in Fig. 1 and Table 1. The natural forest,
136 mixed forest, and secondary forest are the main land use types, and are patched by grass, bare land,
137 orchard, active farm, and inactive farm. To rehabilitate the land locked salmon, the cultivated farms
138 along Yusheng Creek were expropriated by the government since 2005. The expropriated vegetable
139 farms were categorized as inactive farms in this study. On the other hand, the currently growing
140 farms, mainly cabbage, are designated as active farms (Huang et al., 2012). The three adjacent creeks
141 have similar environmental features, i.e. precipitation, atmospheric deposition, and geology, with
142 varying cultivation gradients providing us a good experimental ground to explore the nutrient
143 dynamics and differentiate anthropogenic alteration from natural background.

144

145 **3. Materials and Methods**

146 **3.1 Water sampling and chemistry**

147 Water samples from the three stations were collected twice per week and analyzed during 2007
148 and 2008. Four typhoons in 2007 and four typhoons in 2008 brought the study site relatively large
149 amounts of rainfall. Three among the four in 2007, Pabuk, Sepat, and Krosa during 6-8 Aug., 16-19
150 Aug., and 4-7 Oct., respectively, were sampled intensively at a 3-hour interval for 62 to 87 hours.
151 Depth integrated water samples were obtained using a vertically mounted 1 L bottle attached to a
152 weighted metal frame that was gradually lowered from the bridge. The U.S. Geological Survey
153 DH-48 sampler was not used because of its difficulty in sinking in turbulent flows >2.5 m/s

154 (Milliman et al., 2008).

155 Water samples were immediately filtered through GF/F filters (0.7 μm). The filtrates were
156 quick-frozen in liquid nitrogen for water chemistry analyses. Nitrate, nitrite and ammonium content
157 was determined by ion chromatography (IC) using a Dionex ICS-1500 instrument with a detection
158 limit of 0.2, 0.2, and 0.4 μM , respectively. For all samples, nitrite and ammonium concentrations
159 were lower than the detection limit. Hence, nitrates are the dominant species of dissolved inorganic
160 nitrogen (DIN). Phosphate, dissolved inorganic phosphorus (DIP), was determined with standard
161 Molybdenum blue method with the detection limit of 0.01 μM (Parson, 1984). During typhoon
162 period, total suspended matter (TSM) was determined gravimetrically using pre-combusted 0.7 μm
163 GF/F filters. The mean of the blank, calculated from ten replicates, was 0.05 ± 0.01 mg/l. This blank
164 value was well below the weight of sediment on the filter (generally $>10\text{g/l}$).

165

166 **3.2 Flux calculation**

167 The flux is the total amount of export elements from a watershed within a given period. The
168 elemental concentrations measured in the stream were transformed into flux by multiplying the
169 corresponding discharge to obtain the elemental mass load. We need flux estimator to derive the flux
170 within a given period when there are limited samples. These estimators are performed when
171 continuous measurement (e.g. daily) of the constituent's concentration (C) and discharge (Q) is not
172 possible. Two estimators, flow-weighted method and rating curve methods, are implemented in this
173 study for different circumstances. The flow-weighted method, which considers hydrological controls
174 on element concentration, was applied to calculate the flux on non-typhoon days because typhoon
175 discharge could change the C-Q relation of the non-typhoon period (Lee et al., 2009). However, the
176 close C-Q relationship observed during the typhoon period makes the rating curve method suitable,
177 particularly when the samples cover the higher-discharge spectrum (Kao and Liu, 2001; Kao et al.,
178 2004; 2005; Kao and Milliman, 2008).

179 The formula for the flow-weighted method is shown below (Dolan et al., 1981; Coats et al.,

180 2002).

$$181 \quad Load_{NT,m} = K_1 \frac{\sum_{i=1}^{N_m} C_{i,m} Q_{i,m}}{\sum_{i=1}^{N_m} Q_{i,m}} \times Q_{t,m}, m=1 \sim 12 \quad (1)$$

182 where K_1 is the conversion factor ($=10^{-3}$, converting [g] to [kg]) used to calculate $Load_{NT,m}$ [kg] for a
 183 month, m. Subscript NT denotes non-typhoon days. $C_{i,m}$ [mg/l] is the nutrient concentration of i^{th}
 184 sample in a month. $Q_{i,m}$ [m^3/sec] is the daily discharge on the day while the i^{th} sample is taken. $Q_{t,m}$
 185 [m^3] is the total monthly discharge excluding typhoon days, and N_m represents the total number of
 186 samples in a month.

187 The rating curve method presumes that a power function (i.e., $F=aQ^b$) exists between the
 188 observed elemental flux (F) and discharge (Q). The coefficients of power function, a and b , in Eqn.
 189 (2), can be derived from the observed elemental fluxes and the water discharge rates by the log-linear
 190 least-square method. In this study, 3-hour-interval samples are used to construct the power-function
 191 relation, which is further applied on the entire typhoon period to estimate the total typhoon load.

$$192 \quad Load_T = K_2 \sum_{j=1}^t F_j = K_2 \sum_{j=1}^t a Q_j^b \quad (2)$$

193 where K_2 is also a conversion factor ($=3600$ [sec]) used to calculate $Load_T$ [kg] for a specific period
 194 (in this case, typhoon hours). T denotes typhoon period. F_j [g/sec] is the hourly elemental flux, which
 195 can be estimated by the hourly discharge rate Q_j [m^3/sec] on the j^{th} hour. t [hours] is the total typhoon
 196 hour.

197

198 4. Results

199 The air temperature during observation varied from -4 °C to ~25 °C showing a significant
 200 annual cycle. In Figure 2 we plotted daily average thus the range is from 2 to 20°C (Fig. 2a). The
 201 water discharges are shown in Fig. 2 and Fig. 3. The discharge shows a spiky feature ranging over
 202 three orders of magnitude, which is common and mostly attributed to typhoons, characterizing small

203 mountainous rivers (Milliman and Kao, 2005; Kao and Milliman, 2008).

204

205 **4.1 Temporal variation of nitrate and concentration-runoff relation**

206 In the pristine catchment (Sta. G) the nitrate concentration varied between ~ 0.2 to ~ 40 μM (Fig.
207 2b). The average nitrate concentration of typhoon samples, 23 ± 8.4 μM , was higher than that in
208 non-typhoon period, 10 ± 5.7 μM (Table 2). The nitrate concentration began to rise in the beginning of
209 spring, i.e. onset of growing season in Taiwan, resembling the annual cycle of temperature. Generally,
210 the higher concentration was found in the wet season and the nitrate concentration seemed to
211 correspond to the discharge patterns resulting in a positive concentration-runoff (C-Q, Q in [mm/day])
212 relation during non-typhoon period in Fig. 4a-1 (hallow circles). However, during flood event period
213 (i.e. typhoon period) dilution had occurred that the nitrate concentrations decreased with the
214 increasing discharge (solid circles in Fig. 4a-1). The C-Q relation during typhoon event thus differed
215 from that of non-typhoon periods.

216 Compared with the pristine watershed, nitrate concentrations at Sta. K (Fig. 2c) were higher
217 ranging from ~ 10 to ~ 200 μM due to cultivation. Unlike in the pristine watershed (Fig. 2b), the
218 overall mean nitrate concentration of the typhoon samples, 39 ± 12 μM , was below the mean
219 concentration measured during non-typhoon days, 55 ± 21 μM (Fig. 2c and Table 2). The annual cycle
220 was not as apparent as in the pristine watershed. However, it could be still found the rising nitrate
221 concentration in the beginning of the growing season in 2007 but not in 2008. There were
222 corresponding changes between nitrate concentration and discharge during non-typhoon period in
223 2007, reflecting the positive C-Q relation in Fig. 4a-2. However, in 2008 the nitrate concentration
224 had only subtle fluctuation until the first typhoon raised the concentration, and the nitrate
225 concentration kept decreasing afterwards (Fig. 2c). Note that in this year we did not monitor the
226 water chemistry during flood peak. Nevertheless, nitrate concentrations were diluted by water
227 discharge during the typhoon period in 2007 (Fig. 4a-2) as seen in the pristine watershed and the
228 dilution effect seemed to be more obvious.

229 At Sta. Y, the intensively cultivated watershed (Fig. 2d), nitrate concentration varied between ~5
230 to 300 μ M. A significant higher baseline can be found in 2007 when comparing with that in 2008.
231 The average nitrate concentration were $108\pm20\mu\text{M}$ for typhoon samples and $92\pm54\mu\text{M}$ for
232 non-typhoon samples (Table 2). The observed concentrations at this station sometimes surpassed the
233 criterion for drinking water quality, ~160 μ M. The nitrate concentration started to decrease since the
234 midyear of 2007. Among the three watersheds, the decrement of this nitrate falling from wet to dry
235 season was the most significant. Again, dilution occurred during the typhoon period that can be seen
236 in the C-Q relation (Fig. 4a-3). For three observed stations, the behaviors of nitrate concentration
237 enhancement and dilution were rotating among non-typhoon and typhoon periods.

238

239 **4.2 Temporal variation of phosphate and concentration-runoff relation**

240 The temporal variations of phosphate for three stations were shown in Figure 3. Overall,
241 phosphate did not vary seasonally as significantly as nitrate. At Sta. G, the pristine watershed, the
242 phosphate concentration ranged from ~0.01 to ~0.8 μ M with a mean of $0.19\pm0.09\mu\text{M}$ (Table 2).
243 During typhoon flood phosphate was much higher ($0.42\pm0.10\mu\text{M}$, red triangles in Fig. 3b) compared
244 to non-typhoon periods. We could not find the tendency of rising phosphate concentration in the
245 beginning of the growing season in either 2007 or 2008. Opposite to nitrate, the phosphate
246 concentrations were more enhanced during typhoon flood. The running average of the phosphate
247 revealed a correspondence to the discharge pattern (Fig. 3b); however not as significantly as the
248 nitrate did (Fig. 2b). A positive C-Q relation was observed regardless of non-typhoon or typhoon
249 period (Fig. 4b-1).

250 At Sta. K, the phosphate concentration did not surpass 0.7 μ M (Fig. 3c). The mean of phosphate
251 concentration for non-typhoon samples were $0.18\pm0.1\mu\text{M}$ comparable to the pristine watershed
252 (Table 2). The average of typhoon samples was $0.36\pm0.08\mu\text{M}$. A significant correspondence between
253 discharge and phosphate concentration reflected again a positive C-Q relation (Fig. 4b-2).

254 In the intensively cultivated watershed (Sta. Y), the observed phosphate concentrations during

255 non-typhoon period were in similar range, $0.18 \pm 0.13 \mu\text{M}$ (Table 2), as the other two (Fig. 4b-3). By
256 contrast, phosphate concentration surged during typhoon period with a maximum of $>2.8 \mu\text{M}$, which
257 is the highest concentration been observed in this study, reflecting the influence of fertilization. For
258 all three stations, phosphate concentrations were enhanced as the increase of runoff depth (right
259 panels of Fig. 4) regardless of agriculture and flow conditions.

260 During typhoon period, those samples revealed positive yet significantly different correlations
261 (see equations in Fig. 5) of phosphate concentration against TSM, The positive relations
262 demonstrated that phosphate is highly particulate-associated. At a given TSM concentration the
263 phosphate concentration is significantly higher but more scattering at Sta. Y where cultivated land
264 proportion is the largest among the three. Interestingly, the pristine watershed may provide slightly
265 higher phosphate concentration at a given TSM when compared with the moderately cultivated
266 watershed.

267

268 **4.3 DIN to DIP ratio**

269 The monthly variations in ratio of DIN concentration (DIN_C) to DIP concentration (DIP_C) were
270 illustrated in Fig. 6. The $\text{DIN}_C:\text{DIP}_C$ fluctuated up to two orders of magnitude revealing the
271 seasonality. At Sta. G, the DIN_C to DIP_C ratios varied from ~ 2 to ~ 200 (Fig. 6b) mimicking the
272 temporal trend of air temperature (Fig. 6a) with lowest ratios in winter (dry period). The lowest DIN_C
273 to DIP_C ratios were mainly due to low DIN concentrations. The ratios during the typhoon periods
274 were slightly lower comparing with that in the same month but the overall trend still followed the
275 annual temperature cycle.

276 At Sta. K, $\text{DIN}_C:\text{DIP}_C$ ranged from ~ 50 up to $\sim 5,000$ (Fig. 6c) revealing remarkable difference
277 to the pristine watershed reflecting agricultural effect. In early summer, the a significant increase in
278 $\text{DIN}_C:\text{DIP}_C$ was observed due to fertilizer application. After the peak, a gradually descending trend in
279 monthly $\text{DIN}_C:\text{DIP}_C$ was seen as runoff increases. Note that during our typhoon observation,
280 $\text{DIN}_C:\text{DIP}_C$ values were the lowest throughout the year. These observed values dropped instantly to a

level similar to the pristine watershed in summer (dashed line in Fig. 6c), and an instant rebound to higher level was also observed afterward in following water samples. There was no evident correlations either between $\text{DIN}_C:\text{DIP}_C$ and air temperature or between $\text{DIN}_C:\text{DIP}_C$ and discharge at Sta. K.

At Sta. Y, except the typhoon observation the $\text{DIN}_C:\text{DIP}_C$ ranged from ~ 200 to $\sim 8,000$, which is even higher than that at Sta. K. A gradually increase pattern was detected from ~ 500 in January to ~ 8000 in August. Similar to Sta. K, a one-step jump in July and rapid drops of $\text{DIN}_C:\text{DIP}_C$ in October can be seen (Fig. 6d). A significant feature is the DIN_C to DIP_C ratio restarted to build up gradually. The instant drops to the level of ~ 100 during typhoon sampling and rebounds back after typhoon sampling were observed indicating such phenomenon is common in agriculture watersheds.

The $\text{DIN}_C:\text{DIP}_C$ versus DIP_C (Fig. 7a) and $\text{DIN}_C:\text{DIP}_C$ versus runoff depth (Fig. 7b) were plotted to describe the hydrological influence on $\text{DIN}_C:\text{DIP}_C$ under various cultivation gradients. In intensively cultivated condition, $\text{DIN}_C:\text{DIP}_C$ and DIP_C correlated negatively and tightly in a much wider range, in which $\text{DIN}_C:\text{DIP}_C$ is also roughly negatively correlated over a full range of runoff (Fig. 7b). At a given DIP_C , the intensively cultivated watershed held the highest $\text{DIN}_C:\text{DIP}_C$ when compared with moderately cultivated and pristine watersheds (Fig. 7a). Pristine watershed held a negative yet much loose relation between $\text{DIN}_C:\text{DIP}_C$ and DIP_C . A dome shape in $\text{DIN}_C:\text{DIP}_C$ forms over the full runoff range (Fig. 7d). Moderately cultivated watershed possessed in the middle of aforementioned two extreme cases. For comparison, we put dry period samples together in color fields (Fig. 7a). Interestingly, $\text{DIN}_C:\text{DIP}_C$ tends to be higher at middle runoff condition regardless cultivation degree (Fig. 7b, 7c and 7d) though less significant in pristine watershed.

4.4 DIN and DIP production

The log-log relations of observed N and P fluxes against discharges were shown in Fig. 8. The strong positive correlations illustrates that hydrology exerts strong control on both DIN and DIP exports, particularly, for the typhoon samples. The relations could be well depicted by power

function, i.e. rating curve method (Table 3). The coefficients of determination, i.e. R^2 , were high (>0.85), indicating the applicability of rating curve method on estimating nutrient fluxes during typhoon flood. Thus, the hourly discharge reported for the eight typhoon periods were substituted into the power functions to estimate typhoon-triggered nutrient fluxes. For the non-typhoon period (hollow circles in Fig. 8) the relations were relatively scattering, and the degree of scatterness was much higher at the low-flow end. Accordingly, during non-typhoon period the monthly flow-weighted method was used to estimate monthly nutrient fluxes.

Since hydrology plays an important role in nutrient export, we would like to examine the fractional contribution from typhoon events in term of nutrient export. In 2007, four typhoons collectively brought 30-50% of annual total runoff into three watersheds resulting in 20-48% and 47-84% of annual DIN and DIP export, respectively. Four typhoons in 2008 contributed 48-55% of annual runoff carrying slightly higher (40-70% and 60-80% of annual DIN and DIP export) amount of nutrient transported off the watersheds.

The nutrient loads were converted into yield per area to illustrate the production rate of DIN and DIP (represented by DIN_y and DIP_y , respectively), eliminating the watershed size effect in convenience for comparison (Table 4). Cultivation is more influential on DIN_y than DIP_y . DIN_y ranged from 7.9-8.6 kg-N/ha/yr in the pristine watershed to 30-43 kg-N/ha/yr in the intensively cultivated watershed. This result reflected the cultivation gradient captured by these catchments. For DIP_y , the yields for the three watersheds were ~ 0.36 , ~ 0.35 , and ~ 0.56 kg-P/ha/yr, respectively. As mentioned above, hydrology regulates relative yield of DIN to DIP; therefore, very different $DIN_y:DIP_y$ can be seen during non-typhoon periods (103 to 1036) and typhoon periods (93 to 225), also typhoon's higher fractional contribution resulted in annual means of $DIN_y:DIP_y$ varying from 97 to 410 for the three watersheds (Table 4).

5. Discussion

Following the results we observed, this discussion section will start with the landuse alteration

on nutrient export (5.1) and then hydrological controls the nutrient export (5.2). Afterwards, nutrient export will be depicted by a conceptual model (5.3). The significance of the Oceania will be revealed in the end of this section (5.4).

5.1 Importance of human activity on nutrient export

Human activities have been demonstrated the most significant factors influencing nutrient export (Galloway and Cowling, 2002; Smith et al., 2003; Seitzinger et al., 2010), of which fertilization is highlighted. Our study showed that nitrate and phosphate concentration increase with the increasing proportion of agricultural activities in a watershed. However, the increase in nutrient concentration is so responsive to agricultural activities that even a 2.7% occupation of agricultural landuse, the average nitrate concentration increased ~500% (Table 2) throughout ~95% time period in a year (typhoon excluded) revealing how vulnerable the lotic environments are to the landuse changes in subtropical mountainous small rivers. There is ~1000% increase of nitrate concentration in a watershed with 8.9% agricultural land.

In Taiwan, the frequent summer typhoon rain forces the farmers to apply much higher amount of ammonium sulfate and urea (~3,750 kg-N/ha/yr) to speed up cabbage growth (N demand ~600-900 kg-N/ha/yr) and harvest earlier before typhoon invasion. The over-fertilization is also due to farmers' anticipation of a substantial flush away in wet season. Basing on watershed monitoring network in our study area and a sophisticated deconvolution method, Huang et al. (2012) obtained a very high nitrate yield number for the active farms, ~3,000 kg-N/ha/yr, which apparently the major contributor to enhance nitrate in stream water. 1% increment of active farm will increase DIN_y from background value (i.e. ~8.3 kg-N/ha/yr) to ~38 kg-N/ha/yr. Even if the yield for the inactive farm, ~770 kg-N/ha/yr, was obtained revealing the lingering effect of agricultural activities.

Besides the fertilization, widespread anthropogenic N deposition (ANN) prevails in the Oceania. Pristine is just pristine in terms of landuse pattern. Atmospheric deposition (long-range transport mainly from China, ~21 to ~34 kg-N/ha/yr) supplements the N input elevating the background value

(King et al., 1994; Chen et al., 1998; Lin et al., 2000; Fang et al., 2008). The DIN_y for Sta. G, ~ 8.3 kg-N/ha/yr, is higher than most of the rivers draining land to ocean (Smith et al., 2003) revealing the latent effects of ANN. In the pristine watershed (Sta. G), net increase of DIN inventory is likely occurring in the system. Besides, the DIN leaks out of the forest in growing season (DIN concentration follows air temperature) showing a syndrome of nitrogen saturation. However, this speculation is difficult to evaluate due to the large N pool in the soil (see below); meanwhile, the rates of N mineralization, nitrification (Owen et al., 2010), retention (Fang et al., 2008), and gaseous N losses (Koba et al., 2012) are influential to nitrogen dynamics and cycling. Whether N is saturated needs further investigations on N:P in plant foliage (Tessier and Raynal, 2003) and monitoring extra N budget terms such as gaseous nitrogen in the watersheds in Taiwan.

Landuse effects on phosphate concentration was also found during typhoon flood; however, there seems to have a threshold of cultivation level thus only in the intensively cultivated watershed we can observe $\sim 2.5\times$ higher concentration compared to the other two watersheds (Table 2). At a given TSM in Fig. 5, phosphate concentration is the highest at Sta. Y implying cultivated soil contains more dissolvable phosphate. The DIP_y in the intensively cultivated watershed, ~ 0.56 kg-P/ha/yr, increased 50% compared to that in pristine watershed (~ 0.36 kg-P/ha/yr, Table 4). There is also DIP from the atmospheric deposition (~ 0.1 kg-P/ha/yr, Mahowald et al., 2008) in this area. Beside the unjustified N saturation, more DIP output than input puzzles us. These findings and speculations excite our future studies to unpuzzle the fate of P and N in the subtropical watersheds.

The distinct transport behaviors between nitrate and phosphate results in high $\text{DIN}_C:\text{DIP}_C$, particularly evident in cultivated watersheds. $\text{DIN}_C:\text{DIP}_C$ at a lowest given Q (Fig. 8a), i.e. groundwater, reflects the cultivation gradients as well, implying the long-term agricultural activities may have influenced the groundwater. Such high $\text{DIN}_C:\text{DIP}_C$ indicates the aquatic ecosystem is mainly limited by DIP (Tseng et al., 2010). Without the DIP export to the stream during typhoon periods, the leakage of DIP from terrestrial system is likely limited. Overall speaking, the increase in agricultural activity causes a higher $\text{DIN}_C:\text{DIP}_C$ with larger variation.

385

386 **5.2 Hydrological control on nutrient exports**

387 The nitrate and phosphate concentrations showed a positively correlation with discharge
388 implying the dominance of rainfall-runoff process on nutrient export in small mountainous rivers
389 (Fig.2, 3, and 4). The dominance of discharge on nutrient export is also illustrated by Fig. 8, although
390 in the low-flow condition there seem some additional factors scattering the discharge-driven export.
391 The scattering at low-flow, particularly for the phosphate in two cultivate watersheds, may result
392 from local agricultural practice, in-stream biota uptake or some unknown in-stream processes.

393 Basically, nitrate concentrations increase with increasing water discharge illustrating typical
394 diffuse source that nitrate is carried along the flow pathways (Salmon et al., 2001; Kao et al., 2004).
395 In general, the hydro-biogeochemical system showed dilution and concentration phenomenon due to
396 alternation of hydrological access for chemicals with large concentration differences between deep
397 and shallow soil components. In our study the nitrate concentrations increase as the increasing runoff
398 may resemble the case of enhanced hydrological access on shallow soil source (Salmon et al., 2001).
399 Dissimilarly, nitrate concentration in surface soil is low in forest of mountainous Taiwan (Lin et al.,
400 2004). The major reason is that soils on steep slope in Taiwan are mainly porous old landsliding
401 material being easy to infiltrate and the uptake by the shallow-root vegetation is fast.

402 While rainfall amount is large exceeding some certain threshold during typhoon or rainstorm
403 periods (e.g., 200mm/day for nitrate; see Fig. 4a), surface runoff with depleted nitrate dilutes the
404 nitrate concentration in the river. Although the nitrate concentration is diluted during flood periods,
405 the increase in discharge by three orders of magnitude compensated the dilution effect (~one-order
406 magnitude decrease in concentration) and leads to greater transports (Table 3 and Table 4). The
407 unceasing DIN export is very likely the case in Taiwan (Kao et al., 2004) regardless cultivation
408 degree.

409 This may imply either the nitrogen storage in the watershed is sufficient to afford frequent
410 flooding or the supplement of nitrogen to the watershed system is fast. The estimation of the soil

total N pool in the neighboring watershed (Owen et al., 2010) was 6,909 kg-N/ha, which is 1000x the export from pristine watersheds, being supportive to our speculation of sufficient nitrogen storage.

The hydrological control on phosphate is more remarkable than on nitrate. The consistent positive correlation between phosphate concentration and discharge indicates there is always more phosphate input to the stream while discharge increases. Increasing phosphate concentration accompanies the increase in discharge, congruent with the hydrologic controls on the enhancement of phosphate concentration in other watersheds (Sharpley and Menzel, 1987; Correll et al., 1999, Fig. 4b). Increasing TSM triggered by increasing surface runoff is the most possible cause (Fig. 5). In a year, the phosphate concentration reached its highest value during typhoon events due to greater soil erosion. The attached phosphate is carried into the river with soil particles. During the typhoon periods, the increase of the discharge and phosphate concentration by three and two orders of magnitude, respectively (Fig. 8), substantially enhance the phosphorus export (Correll et al., 1999; Green and Finlay, 2010).

Beside the total export, hydrology also modulates the $DIN_C:DIP_C$ through different hydrological pathways. The imbalanced DIN and DIP productions responding to different hydrological conditions lead to the transition of $DIN_C:DIP_C$ from dry to wet seasons (Fig. 8b, 8c, 8d). In dry season, i.e. low-flow condition, while groundwater dominates hydrograph, the $DIN_C:DIP_C$ is categorized as background condition. While approaching high-flow condition, increase of water level induces more flushable nitrate, mainly from soil, to the stream leading higher $DIN_C:DIP_C$. When discharge surges during typhoon or rainstorm periods, surface runoff dominates hydrograph (>50% total discharge, Lee (2012)) washes out enhanced phosphate and diluted nitrate reducing the $DIN_C:DIP_C$ significantly (Saunders et al., 2006; Green et al., 2007). From the previous section, we have known that $DIN_C:DIP_C$ is agriculture-dependent. Concentration of DIN and DIP sources vary with the cultivation gradient, i.e. higher cultivation levels have higher DIN and DIP concentrations.

Although hydrological forcing (i.e. typhoon and rainstorm flood) caused a sharp drop in the $DIN_C:DIP_C$ (not vivid in the pristine watershed), the $DIN_C:DIP_C$ in the sequential months (Sep. after

437 Aug. and Nov. after Oct.) immediately rebounded (red arrows in Fig. 6), which reveals the quick
438 resilience of lotic environment to the episodic natural disturbances, i.e. typhoons. It is hardly to see
439 the lingering influences of typhoons, e.g. brash and fallen foliage, since most of them are carried off
440 the watershed by dominant surface runoff (West et al., 2009). If we went into the detailed
441 chemograph, we could see that $DIN_C:DIP_C$ returned to pre-event level gradually while hydrograph
442 recession (not shown). The dramatic change of fluvial nutrient status may be one of the unique
443 characteristics for small mountainous rivers.

444

445 **5.3 Differential transport for DIN and DIP**

446 In this study, a conceptual model is proposed to illustrate the transport of nitrate and phosphate
447 in different hydrological conditions (Fig. 9). During the dry period (from Nov. to Mar. in the next
448 year is also off-growing season, Fig. 9a), water level is low and dominated by groundwater which
449 represents the background nitrate and phosphate concentration in a watershed. Foliage from
450 deciduous forest is fallen on the ground and begins decomposed. For the cultivated land, in the
451 off-growing season farmers usually overturn the residuals from the harvest crop into the surface soil
452 producing nutrient for the next growing period. In addition, the atmospheric deposition prevails in
453 Taiwan. Small rain event in the dry season leaches nutrient, mostly nitrate, into the shallow soil (Fig.
454 9a) as the N-draining mechanism addressed by Creed et al. (1996). Most of phosphate remains in the
455 surface attached to the soil particles. In the beginning of wet season (from Apr. to Oct. excluding
456 typhoon and rainstorm events, Fig. 9b), relatively large rain events increase water level flushing out
457 the nitrate stored in the soil to the stream. In this condition, most of the additional stream discharge
458 comes from the subsurface that contains more nitrate yet less phosphate; therefore, $DIN_C:DIP_C$
459 elevates significantly (Fig. 8). Meanwhile, soil erosion occurs in the hill foot where is most likely
460 saturated and generates surface runoff increasing phosphate concentration in the river (Hung et al.,
461 2009). During the typhoon or rainstorm flood (Fig. 9c), stream discharge, dominated by surface
462 runoff in which nitrate is depleted, decreases nitrate concentration in the river. But the surface runoff

463 containing abundant particle-associated phosphate increases phosphate concentration, resulting in
464 significantly lower $\text{DIN}_\text{C}:\text{DIP}_\text{C}$ (Fig. 8). The transport mechanism is applicable for different
465 cultivation levels which only influence the nutrient concentrations of sources, i.e. surface, subsurface
466 and groundwater. Various proportions of different flow pathways determine the behaviors of stream
467 nutrient concentration (Mulholland and Hill, 1997; Katsuyama et al., 2001; Poor and McDonnell,
468 2007). Both hydrological controls and land use alternation are important to regulate DIN and DIP
469 transport in Oceania Rivers.

470

471 **5.4 Significance of Oceania streams**

472 Smith et al. (2003) investigated DIN and DIP loads from 165 sites occupying 35% of total land
473 area on the Earth. About 60% of the catchments they used have areas between 10^3 and 10^5 km^2 (10^1
474 to 10^7 km^2 of full range). For both DIN_y and DIP_y , our three watersheds overwhelm most of these
475 sites. Among 111 large rivers (8011 km^2 of the minimum watershed area) in the world that Harrison
476 et al. (2005) analyzed, only 14 of them had higher DIP_y than our pristine watershed, ~ 0.36 kg-P/ha/yr .
477 Unlike these large rivers having only ~ 0.33 m/yr in runoff depth, the Oceania have ~ 2 m/yr . Runoff
478 depth, i.e. hydrological control, leads to the high DIN and DIP yield for Oceania Rivers. Of which,
479 frequent cyclone invasions are responsible for the high fluvial DIP_y (Green and Finlay, 2010). If the
480 contributions from the typhoons were removed (Table 4), DIP_y would be comparable to the large
481 rivers. However, even if the contribution of typhoons is removed, DIN_y still remains a high level
482 compared to the world rivers, attributed to unceasing leakage from terrestrial N storage. Facing the
483 increasing typhoon invasion in terms of frequency and intensity due to climate change (Tu et al.,
484 2009); the alteration of nutrient export and the consequent effects on coastal environment deserve
485 more attentions.

486 Smith et al. (2003) developed a statistic model using $\log(\text{population density [people/km}^2\text{]})$ and
487 $\log(\text{runoff depth [m/yr]})$ to estimate DIN_y and DIP_y . However, the parameterized function only
488 estimates less 50% of DIN_y in our pristine watershed (Table 4). In their study, only six catchments,

489 catchment size smaller than 10^2 km^2 , were used in the analysis and basin characteristics were poorly
490 resolved. The underestimation is understandable since the function is basically determined by the
491 large rivers. The lack of a landuse-associated factor in the function will aggravate the
492 underestimation. It is particularly crucial for the Oceania where human population usually
493 concentrates in certain districts. This human population actually consumes the agricultural products
494 from other areas, e.g. headwater catchments. For the concept of ecological footprint, the population
495 who fed on the agricultural products should transfer into the landuse alteration effects. However, the
496 function overestimated the DIP_y in the pristine and moderately cultivated watershed but
497 underestimated in the intensively cultivated watershed, which need more detailed investigation in the
498 future.

499 Besides population density and runoff depth in Smith et al., (2003) equation, air temperature
500 also plays a role though minor. Temperature negatively correlated to DIN_y in their model suggests
501 the dependence of denitrification on temperature (Knowles, 1981). However, it seems not the case
502 for small mountainous rivers. Positive correlation between nitrate concentration and air temperature
503 was found in our pristine watershed (Fig. 2b). Warmer temperature seems to enhance the rates of
504 organic matter decomposition and nitrification within a watershed. On the other hand, rapid
505 infiltration in Oceania Rivers diminishes denitrification potentials; therefore, even in the cultivated
506 watershed denitrification signal cannot be detected in stream water (Peng et al., 2012). The
507 temperature dependence disappeared due to the significant influence on nitrate in stream water
508 sourced from limited cultivated land in our study. As for phosphate, both from Smith compilation
509 and our study phosphate did not show any temperature-dependent relations.

510 Seitzinger et al. (2005) developed a complicated model, NEWS, to estimate nutrient export
511 from 5761 watersheds. Their DIN_y and DIP_y estimations for the Oceania, as a function of land use,
512 nutrient inputs, hydrology, and other factors, were $\sim 7.2 \text{ kg-N/ha/yr}$ and $\sim 0.28 \text{ kg-P/ha/yr}$,
513 respectively, the highest among eight Continents highlighting the significance of nutrient yields from
514 the Oceania. This number for nitrogen is almost identical to our yield from pristine watershed

515 (7.9-8.6 kg-N/ha/yr). If our case in Taiwan is representative of the background of entire Oceania, the
516 N yields from Seitzinger et al. (2005) highly likely underestimate the Oceania export, although their
517 number is already the highest among world continents. Similar situation existed in DIP_y estimation
518 that their model-estimated DIP_y was even smaller than our pristine background (~ 0.36 kg-P/ha/yr).
519 Once we superimposed the influence of population, such as sewage, the largest anthropogenic source
520 of DIP to the costal zones (Harrison et al., 2005), the DIP_y yield from entire Oceania should be much
521 higher than their model expected. It is well known that the Oceania characterizes the world highest
522 sediment yields (Milliman and Syvitski, 1992) and Taiwan Rivers features more extraordinary values
523 (Mulder and Syvitski, 1995; Milliman and Kao, 2005), implying the significance of particulate
524 nitrogen and phosphorus export. However, this entire study focuses on DIN and DIP export which
525 are more directly accessible by biota and influence the aquatic ecosystem.

526 N-limited ocean is demanding nitrogen input. N-access fluvial water from the Oceania is hence
527 very important supplement for the ocean. It is particularly important because the Oceania is
528 surrounded by oligotrophic ocean where additional N input will significantly enhance its productivity
529 and trigger biogeochemical cycles. Our study showed that the ratio of annual DIN_y to DIP_y ranged
530 from 97-410 (Table 4) higher than the global average (18:1, Smith et al., 2003), featuring evident
531 N-access. Massive loadings of nutrients and organics to rivers have caused dramatic changes of
532 nutrient status in adjacent estuarine and coastal ecosystems (Jickells, 1998; Galloway et al. 2004)
533 stemming from rapid urbanization. Oceania is facing rapid development leading to abundant
534 enriched nutrient fluvial water ($\sim 12\%$ annual global discharge) to the ocean, which might further
535 influence global biogeochemical cycles.

536

537 **6. Conclusion**

538 The rainfall-runoff process plays critical role in releasing DIN and DIP via different
539 mechanisms, respectively, from subsurface and surface reservoirs into stream, subsequently,
540 modulate the ratio of DIN_C to DIP_C that may influence the production and phytoplankton/microbial

community structures in downstream aquatic system. Cultivation gradient and hydrological conditions interplays resulting in wide range of DIN_C : DIP_C varying from ~ 20 to ~ 8000 . Such high DIN_C : DIP_C indicates the aquatic ecosystem is mainly limited by DIP. Without heavy rain brought by typhoons or rainstorms to generate surface erosion, the leakage of DIP from terrestrial system is likely limited. On the other hand, the intensive typhoon rain engenders deeper irrigation to flush out DIN stored in deeper soil, of which plants are not easy to access. Obviously, typhoon is crucial in regulating the absolute and relative amount of DIN and DIP discharges. Much higher increase in DIN relative to DIP caused by cultivation implies Oceania Rivers are sensitive to DIN addition. The rapid flushing and irrigation caused by typhoon rain in summer may push farmer to overdose in order to enhance vegetable production, similarly, the DIN leaks out of the forest in growing season showing a syndrome of nitrogen saturation. However, in comparison with the atmospheric deposition (~ 21 - 34 kg-N/ha/y) the pristine watershed retains $\sim 70\%$ of atmospheric DIN input showing a strong uptake capability. Such result points out the next task to properly define the degree of nitrogen saturation for Oceania watersheds. Besides, more DIP output than input deserves our attention as well. Our study also shed lights on how to project the future trend of DIN and DIP export from Oceania Rivers under the scenario of increasing frequency and intensity of tropical cyclones under global warming condition.

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565

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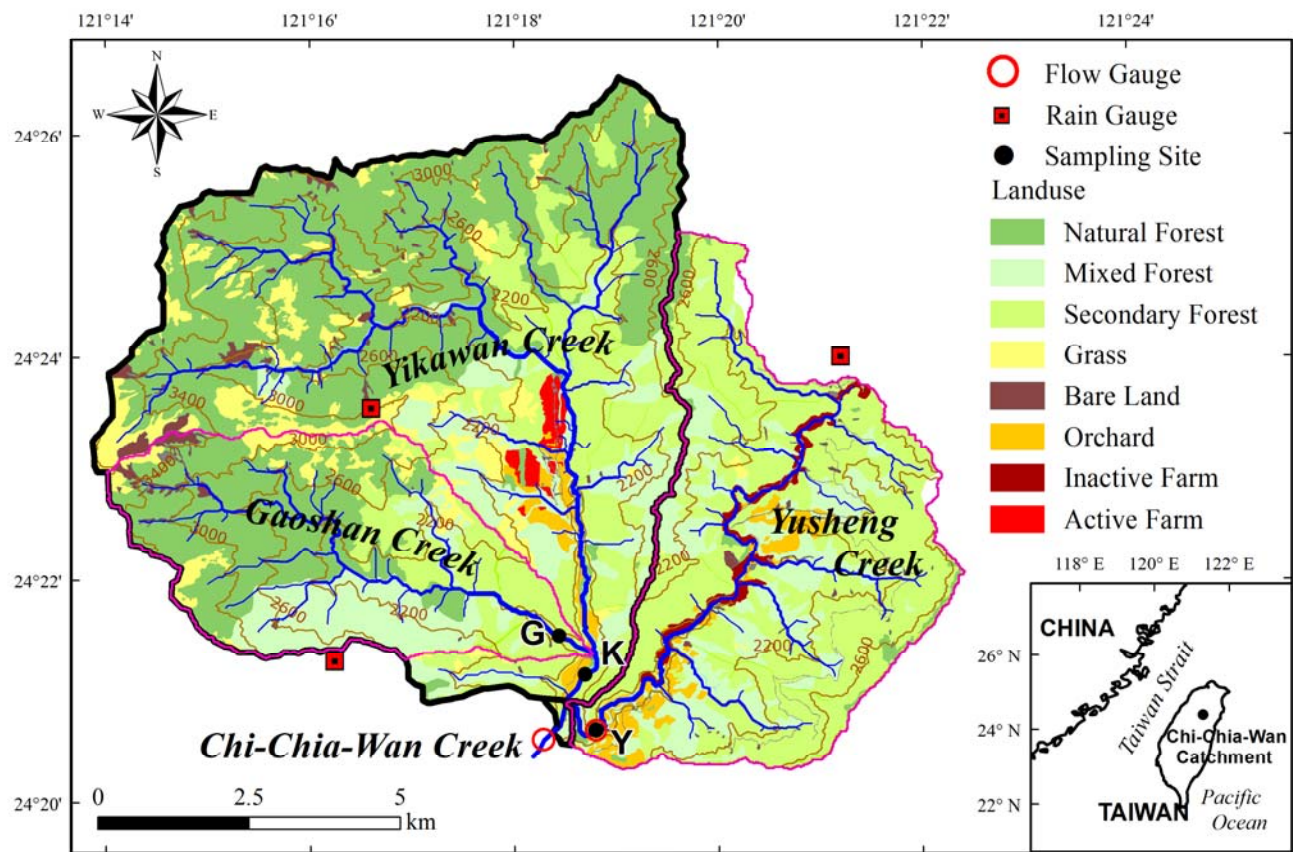
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769 **Figure 1.** The landscape of the study watershed, including sampling sites, rainfall gauges, discharge
770 gauges and landuse patterns. The study watershed was divided into three sub-catchments
771 representing pristine (water samples taken at Sta. G), moderately-cultivated (Sta. K), and
772 intensively-cultivated watersheds (Sta. Y).

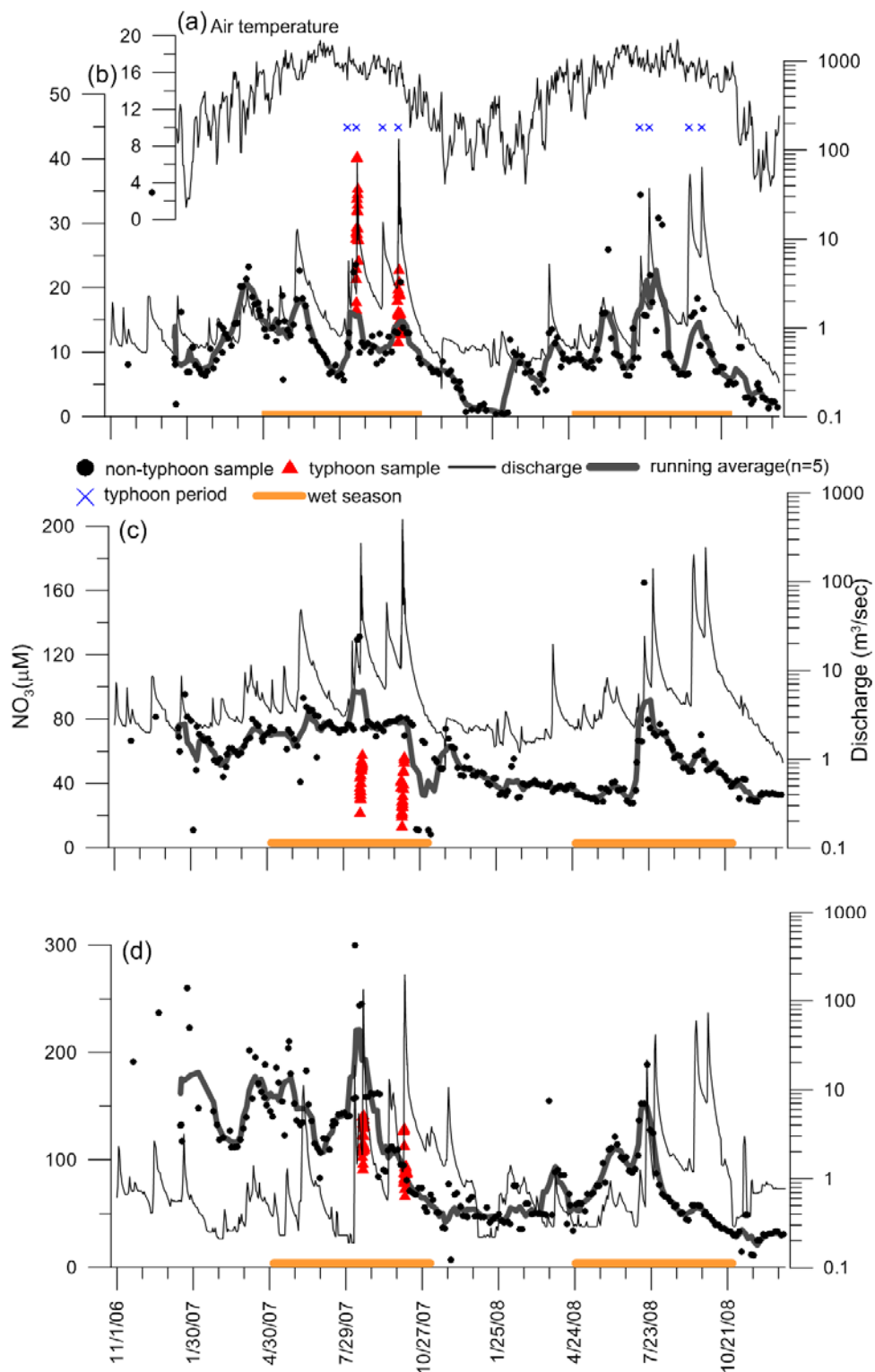


Figure 2. The monitored (a) air temperature; discharge, and nitrate concentration in the (b) pristine, (c) moderately-cultivated and (d) intensively-cultivated watersheds. Water samples include typhoon (red triangle) and non-typhoon (black circle) samples. Eight typhoons were marked by cross symbol. The running average of 5 adjacent nitrate concentrations was illustrated in thick grey line. Wet seasons were highlighted in orange line on the x-axis. Note that the data for the first typhoon we sampled was missing due to our negligence on sample storage

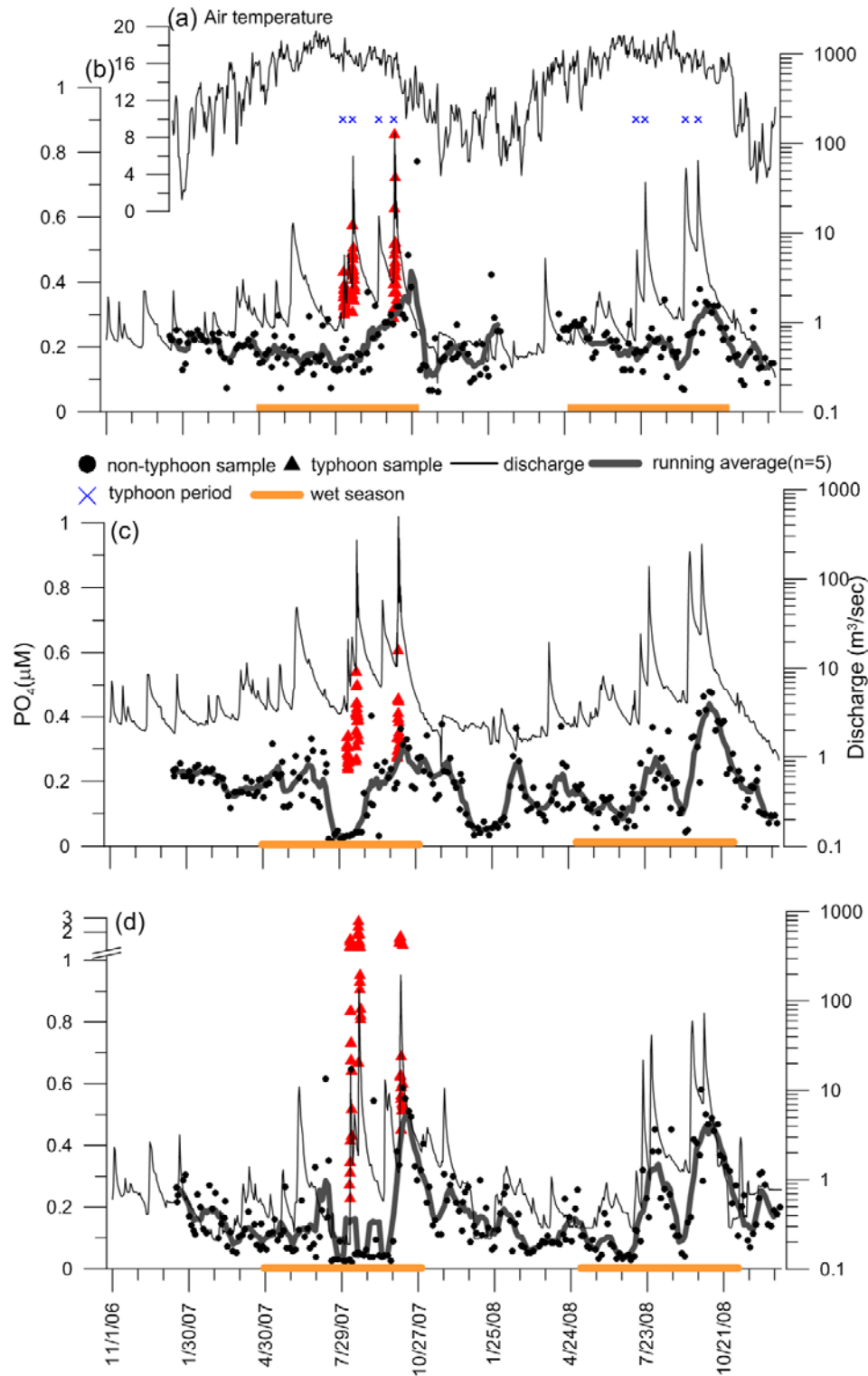
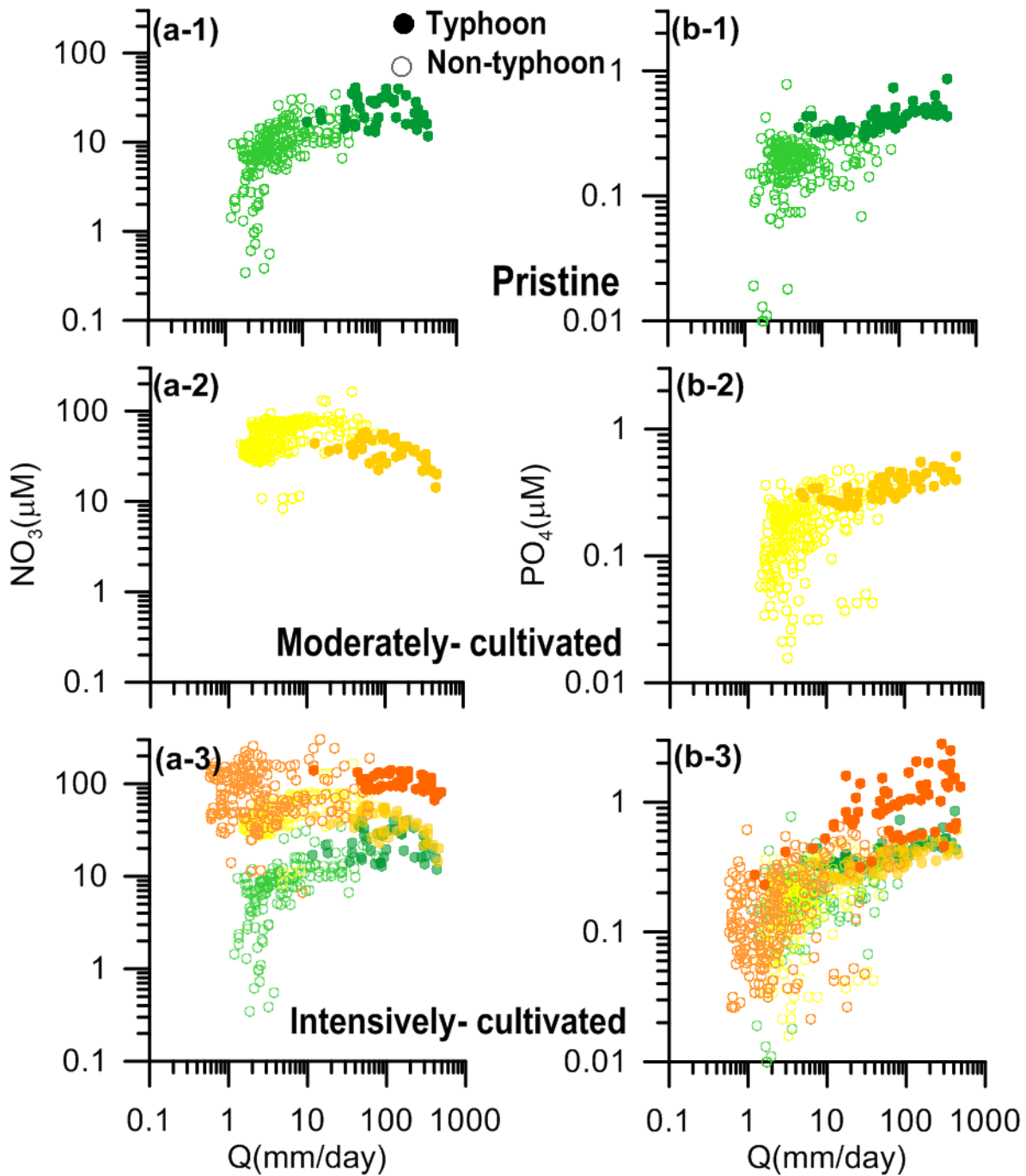


Figure 3. The monitored (a) air temperature; discharge, and phosphate concentration in the (b) pristine, (c) moderately-cultivated and (d) intensively-cultivated watersheds. The same symbols were used as in Figure 2. Note that, the y-axis for (d) plot is rescaled above 1 μM . The missing data in the beginning 2008 was due to our negligence on sample storage.



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790 **Figure 4.** The relation of observed (a) nitrate concentration and (b) phosphate concentration against
 791 runoff depth in the the pristine (-1, green), moderately-cultivated (-2, yellow) and
 792 intensively-cultivated watersheds (-3, orange), respectively. Solid and hollow circles stand for
 793 typhoon and non-typhoon samples, separately. For the plots of intensively-cultivated watershed, data
 794 in the other two watersheds were overlaid as references.

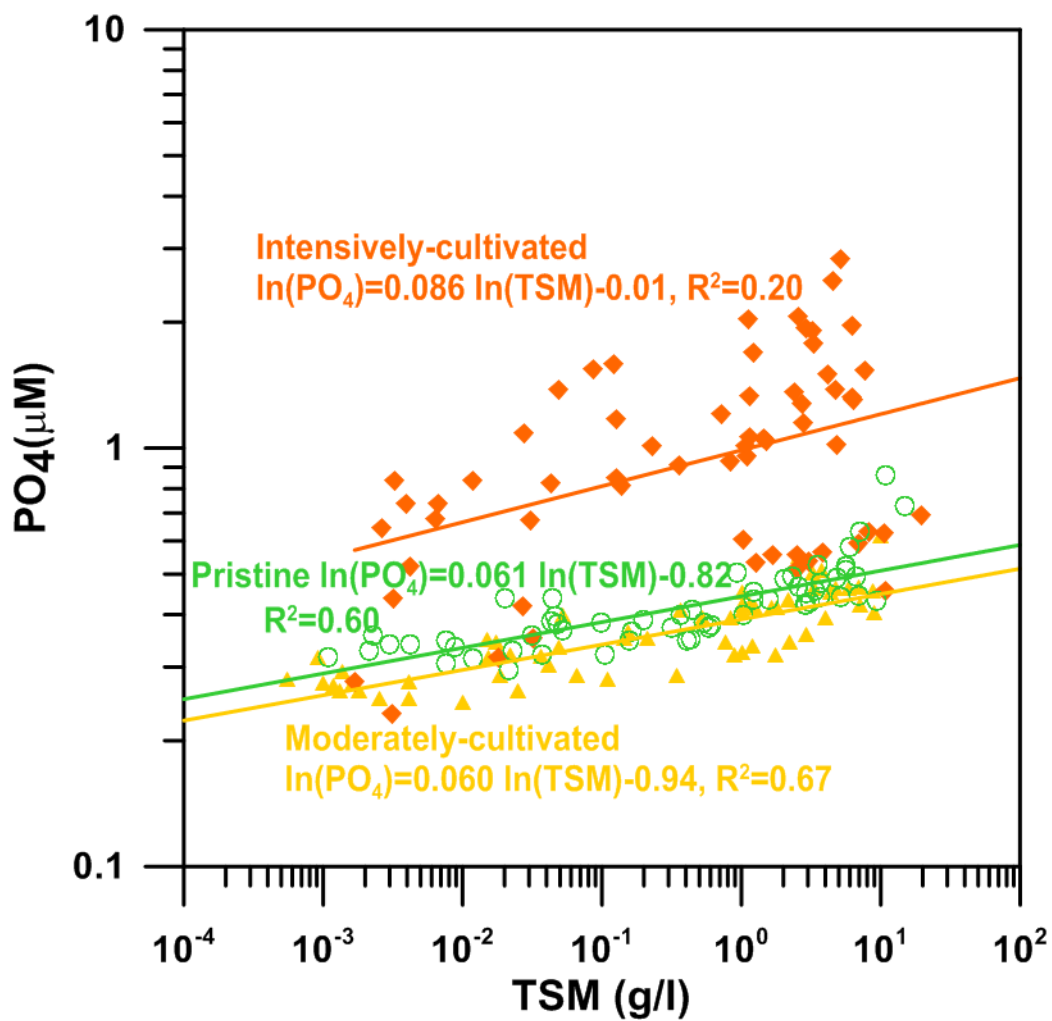


Figure 5. The relations between observed phosphate concentrations against total suspended matter (TSM) for typhoon samples in the pristine (green), moderately-cultivated (yellow) and intensively-cultivated watersheds (orange), respectively.

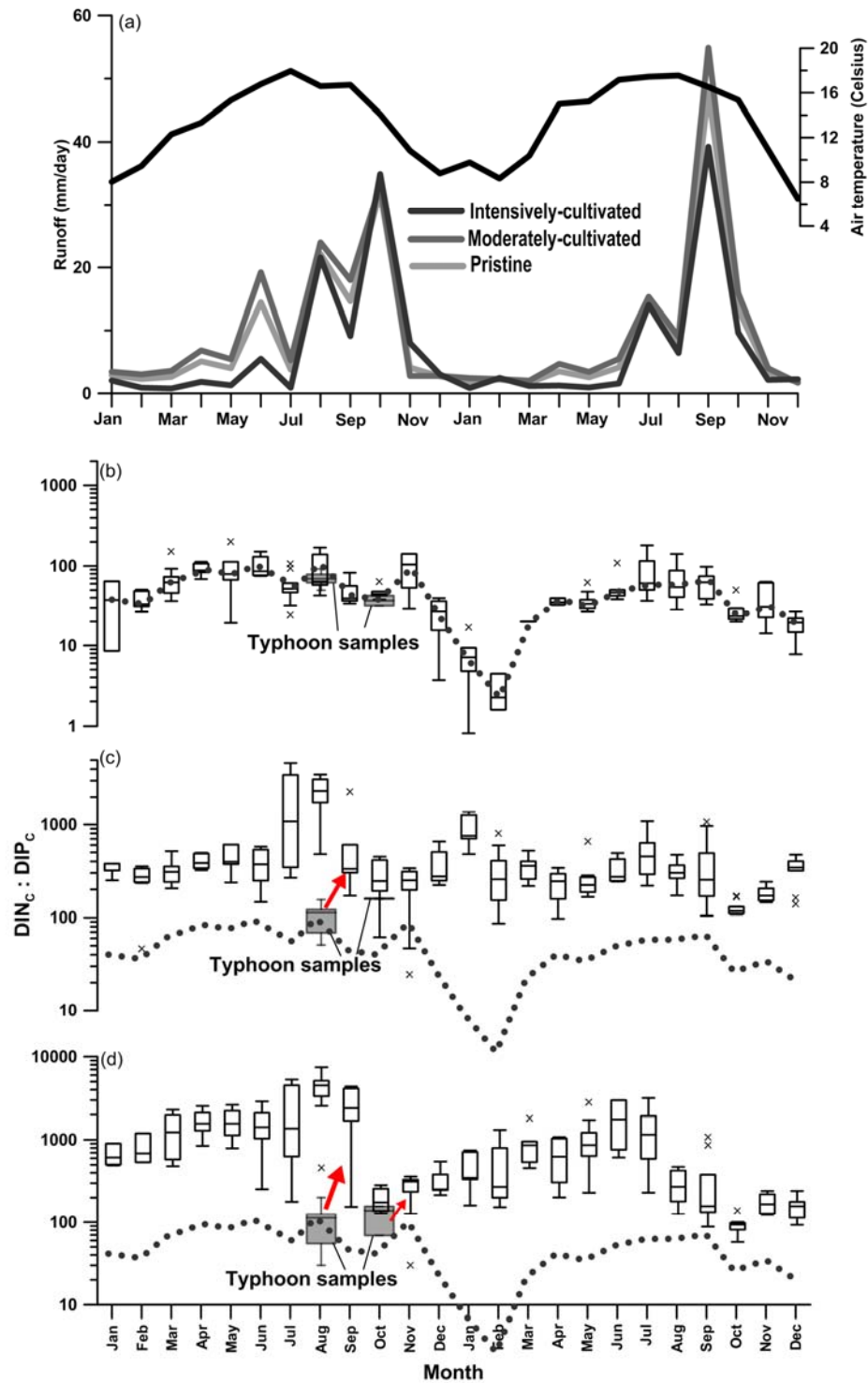


Figure 6. (a) The monthly distribution of daily air temperature, runoff depth; and box-and-whisker plot of sampled $DIN_c : DIP_c$ ratio in the (b) pristine, (c) moderately-cultivated, and (d) intensively-cultivated watershed. Typhoon samples are highlighted separately in grey box. Grey dashed line is the reference redrawn from the pristine watershed. Red arrows stand the resilience of $DIN_c : DIP_c$ ratio after typhoon invasion.

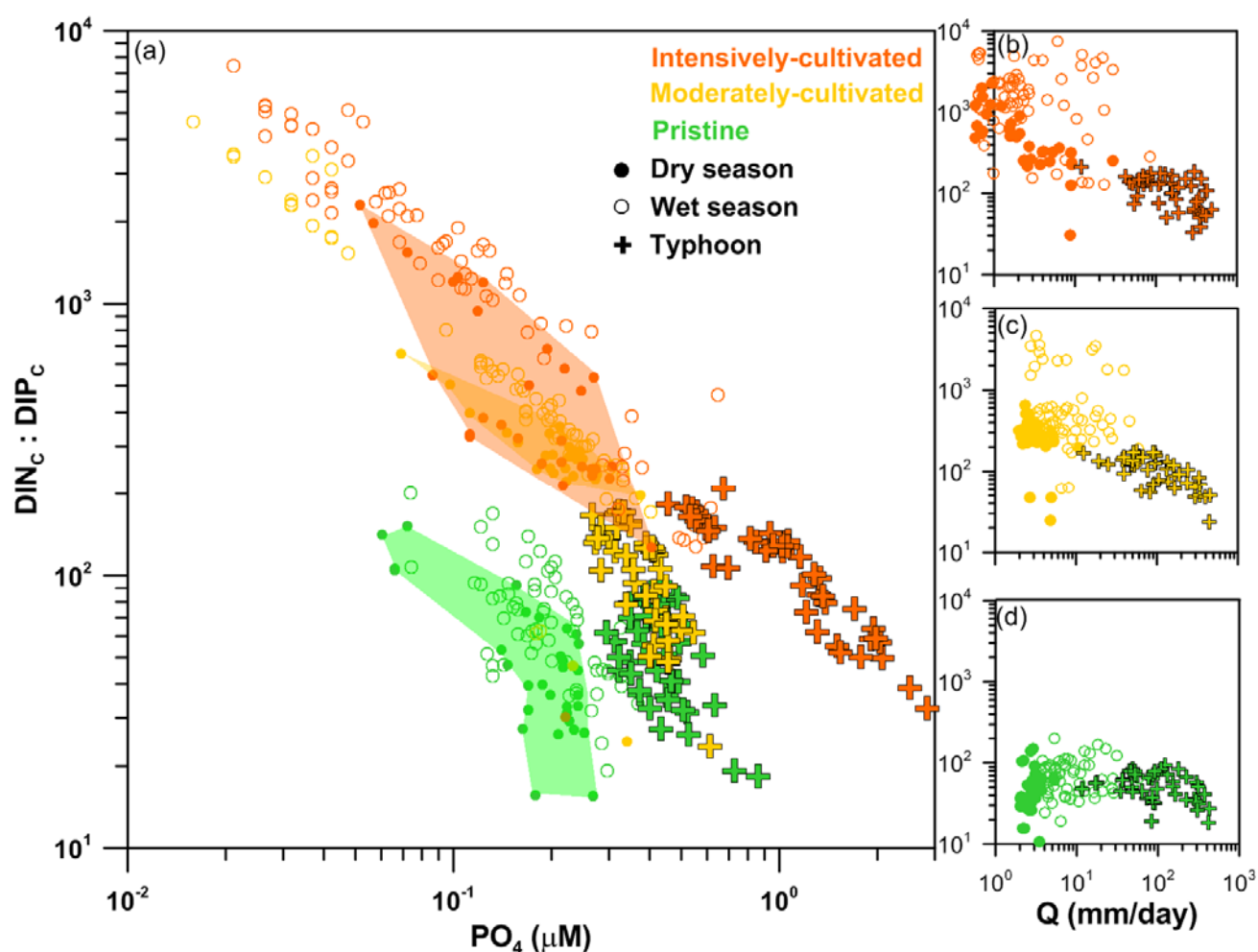


Figure 7. (a) The distribution of observed $\text{DIN}_C : \text{DIP}_C$ ratio against phosphate concentrations under different hydrologic controls and cultivation levels. The relationship between $\text{DIN}_C : \text{DIP}_C$ ratio versus discharge for the (b) pristine (in green), (c) moderately-cultivated (in yellow), and (d) intensively-cultivated watersheds (in orange). The solid circle, hollow circle, and cross symbol represent the $\text{DIN}_C : \text{DIP}_C$ sampled in dry season, wet season, and typhoon period, respectively. Except outliers, dry-season samples were grouped together in colored filed. Data collected in 2007 were used only.

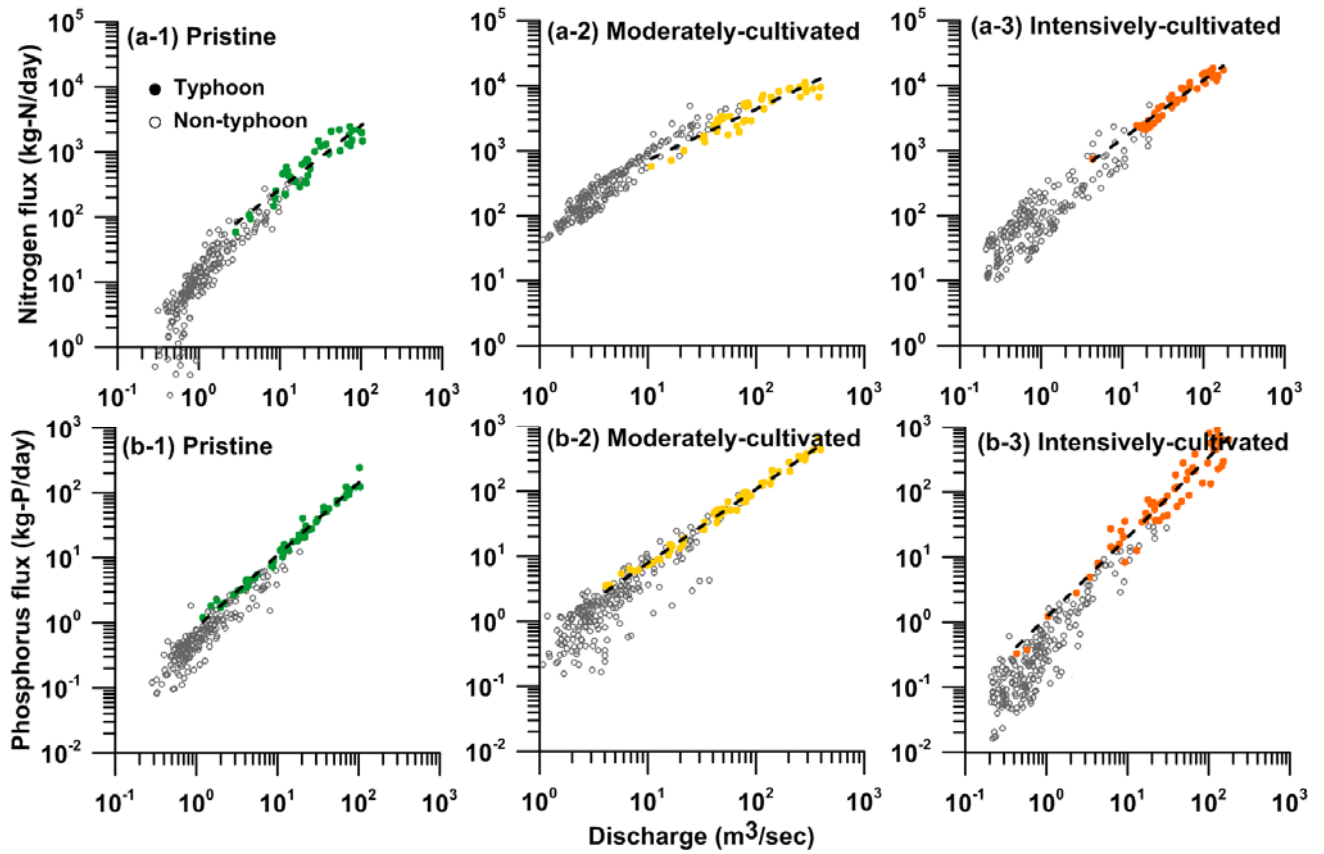
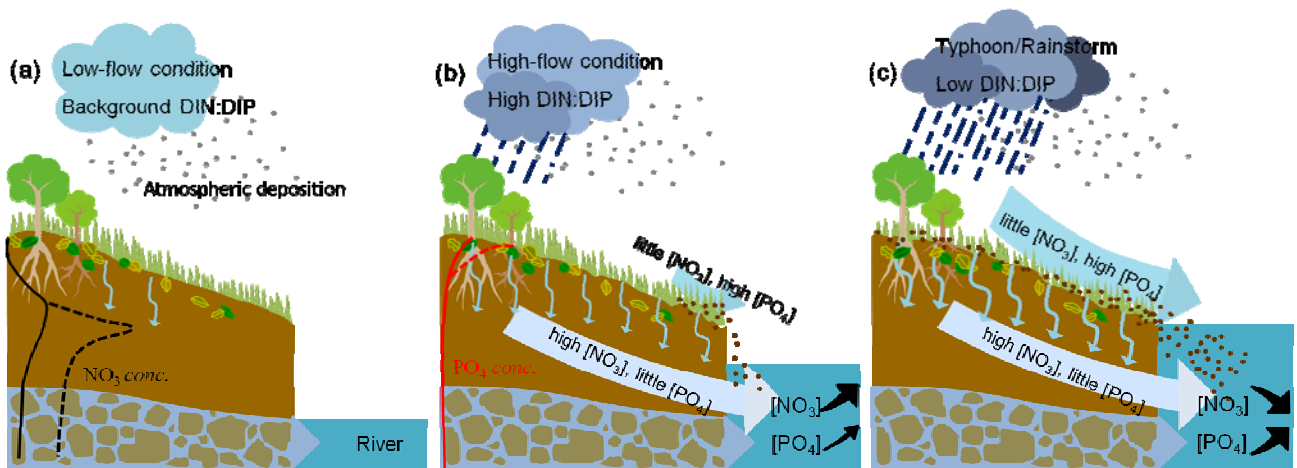


Figure 8. The log-log graphs of observed (a) nitrogen and (b) phosphorus fluxes against discharge in the pristine (-1), moderately-cultivated (-2), and intensively-cultivated watersheds (-3). Solid and hollow circles represent typhoon and non-typhoon samples, respectively. The dashed lines stand for the power regression equation of typhoon samples.



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Figure 9. The schematic diagram of the conceptual model illustrating nutrient export in (a) low-flow condition and dry season; (b) high-flow condition; (c) typhoon and rainstorm flood condition. The arrow size represents the runoff amount. [] denotes concentration. The increase or decrease of concentration and its significance was marked. NO_3 (black line) and PO_4 (red line) concentrations in the soil profile of pristine (solid line) and cultivated (dashed line) land are shown.

836 **Table 1.** Basic landscape characteristics and landuse patterns in the pristine, moderately-cultivated,
837 and intensively-cultivated watershed.

Landscape characteristics						
	Creek name	Sta. ID	Drainage area (km ²)	Slope (deg.)	Elevation (m)	Max. flow length (km)
Pristine	Gaoshan	G	21.05	74.4	2577	10.8
Moderately-cultivated	Yikawan	K	74.03	71.4	2581	17.09
Intensively-cultivated	Yusheng	Y	30.92	56.8	2182	15.48

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Landuses	Natural forest	Mixed forest	Secondary forest	Grass	Bare land	Orchard	Active farm	Inactive farm
unit	%	%	%	%	%	%	%	%
Pristine	45.8	22.5	19.7	9.7	2.2	0	0	0
Moderately-cultivated	47.4	15.9	21.8	9.7	2	1.7	1	0
Intensively-cultivated	2.6	23.5	62.9	0.8	1.9	5.2	0	3.7

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841 **Table 2.** The means and coefficients of variation for nitrate, phosphate, and TSM concentration of
842 water samples at three sites, including typhoon and non-typhoon samples. No TSM concentration
843 was measured for non-typhoon samples. Non-typhoon samples were taken twice per week from 2007
844 to 2008 and typhoon samples were taken during three typhoon periods in 2007. See more detail in
845 the text.

Non-typhoon data	Pristine(G)	Moderately-cultivated(K)	Intensively-cultivated(Y)
Nitrate [μM]	10(0.56)*	55(0.38)	92(0.58)
Phosphate [μM]	0.19(0.46)	0.18(0.53)	0.18(0.76)
Typhoon data			
Nitrate [μM]	23(0.36)	39(0.30)	108(0.19)
Phosphate [μM]	0.42(0.24)	0.36(0.22)	1.05(0.54)
TSM [g/l]	2.05(1.49)	1.61(1.53)	2.50(1.39)

846 * Values in parentheses represent coefficients of variation.

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848 **Table 3.** The power regression equation of typhoon samples taken for three study watersheds, and
849 the runoff depth, calculated DIN load, DIP load for the typhoon events in 2007 and 2008.

Rating curve,			
Load[kg/day]	Pristine	Moderately-cultivated	Intensively-cultivated
=aQ[m ³ /sec] ^b			
DIN load [kg-N/day]	29Q ^{0.97} (0.86)*	117Q ^{0.79} (0.86)	189Q ^{0.90} (0.95)
DIP load [kg-P/day]	0.85Q ^{1.11} (0.99)	0.85Q ^{1.13} (0.99)	1.18Q ^{1.23} (0.94)
2007 typhoons			
(Pabuk, 7-10 Aug; Sepat, 17-20 Aug; Wipha, 17-20 Sep; Krosa, 5-10 Oct)			
Runoff [mm]	1,253(33) [@]	1,195(30)	1,395(50)
DIN load [kg-N]	8,043(44)	45,709(20)	63,985(48)
DIP load [kg-P]	384(52)	1,114(47)	1,526(84)
2008 typhoons			
(Kalmaegi, 17-20 Jul; Fung-Wong, 27 Jul-1 Aug; Sinlaku, 12-18 Sep; Jangmi, 27 Sep-1 Oct)			
Runoff [mm]	1,655(48)	1,769(48)	1,384(55)
DIN load [kg-N]	10,645(64)	67,071(41)	66,940(70)
DIP load [kg-P]	500(66)	1,642(60)	1,315(80)

850 * Values in parentheses represent coefficient of determination, i.e. R²; [@] Values in parentheses stand
851 for the percentage of annual amount.

852

853 **Table 4.** Different time frames (annual, typhoon, and non-typhoon period) of the observed runoff,
854 calculated DIN yield (DIN_y), DIP yield (DIP_y) and DIN_y:DIP_y ratios for the three study watersheds.
855 DIN_y and DIP_y estimated by the model proposed by Smith et al. (2003) are also in the table.

		Pristine	Moderately-cultivated	Intensively-cultivated
Runoff [mm]	Annual	3779, 3447*	3897, 3686	2765, 2497
	Typhoon	1253, 1655	1195, 1769	1395, 1384
	Non-typhoon	2526, 1792	2702, 1917	1370, 1113
DIN _y [kg-N/ha/yr]	Annual	8.6, 7.9	29, 22	43, 30
	Typhoon	3.8, 5.1	6.2, 9.1	20, 21
	Non-typhoon	4.8, 2.8	23, 13	22, 9.0
DIP _y [kg-P/ha/yr]	Annual	0.35, 0.36	0.32, 0.37	0.59, 0.53
	Typhoon	0.18, 0.24	0.15, 0.22	0.49, 0.43
	Non-typhoon	0.17, 0.12	0.17, 0.15	0.10, 0.10
DIN _y :DIP _y [molar ratio]	Annual	108, 97	410, 262	323, 254
	Typhoon	93, 94	182, 181	186, 225
	Non-typhoon	126, 103	613, 382	1036, 369
Model estimation (Smith et al., 2003)				
DIN _y [kg-N/ha/yr]		3.7, 3.4	3.8, 3.6	2.9, 2.7
DIP _y [kg-P/ha/yr]		0.47, 0.43	0.49, 0.46	0.37, 0.34

856 *Two numbers in one cell stand for values in 2007 and 2008, respectively.

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