Effects of antecedent rain history on particulate phosphorus loss from a small forested watershed of Japanese cypress (Chamaecyparis obtusa)

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Antecedent moisture condition; Clockwise hysteresis loop; Diffuse pollution; Forested watershed; Hydrograph characteristics; Soil water repellency

Summary
This study aimed to clarify the effects of antecedent rain history on particulate phosphorus (PP) loss in a small mountainous watershed covered primarily with a plantation forest of Japanese cypress (Chamaecyparis obtusa). We analyzed stream discharge and PP concentration at 15–60 min intervals during 24 h in eight rain events with different discharge levels. The PP concentration versus stream discharge (PPC–Q) relationships exhibited clockwise hysteresis loops for each of the eight events monitored. Discharge could explain changes in PP concentration on the falling but not rising limb of the hydrograph. On the rising limb, a positive relationship between the rate of changes in discharge (dQ/dt) and the PP load (dL/dt) was found for each event. This indicates that a large amount of PP is strongly pulsed at times of rapidly increased discharge. These results suggest that dQ/dt is the driving force behind PP supply and the primary control on the clockwise hysteresis loop of PPC–Q relationship. There was a strong negative correlation between the antecedent precipitation index and the slope of the dL/dt versus dQ/dt relationship. This shows that a rapid increase in PP load occurs even with slight increases in discharge as antecedent moisture conditions become drier. The soil water repellency and rapid runoff response following dry conditions support that soil desiccation increases the PP supply associated with soil erosion via overland flow. Therefore, we concluded that the antecedent rain history affects the mobility of PP via soil desiccation. The findings of
Introduction

Phosphorus (P) is one of the major plant nutrients. Loss of P from a forested watershed is usually small because it is tightly conserved within forested ecosystems (Yanai, 1992). However, heavy rainfalls lead to a large P load from forested watersheds because of soil erosion (Hatch et al., 2001). In Japan, soil erosion has posed a serious problem in inadequately managed plantations of coniferous trees, which tend to have exposed soil surfaces because of canopy closure preventing understory growth (Fukuyama et al., 2005; Miura et al., 2002). Ide et al. (2007) suggested that inadequately managed plantations of Japanese cypress (Chamaecyparis obtusa), one of the most common plantation species in Japan, become major diffuse sources of particulate phosphorus (PP). Irregular P load pulsed by heavy rainfall may damage the ecological quality of downstream waters (Meyer and Likens, 1979). To develop effective nutrient management strategies (e.g., Nisbet, 2001), it is important to quantify the P load from plantations of coniferous trees in regions with high rainfall.

Most P from forested watersheds is exported in particulate form, i.e. PP, during intensive rain events, resulting in rapid and dramatic temporal variations in P load (Hatch et al., 1999; Ide et al., 2007; Meyer and Likens, 1979; Munn and Prepas, 1986). This makes it difficult to accurately estimate the P load because of the need for intensive water sampling during periods of highly fluctuating discharge. Accordingly, the P load during rain events is generally interpolated or extrapolated from the limited available data (e.g., Munn and Prepas, 1986).

Prediction of the P load has been based on stream discharge (e.g., Kronvang and Bruhn, 1996). However, the PP concentration does not depend solely on stream discharge (Prairie and Kalff, 1988) and can vary by several orders of magnitude at a given discharge level. This leads to a substantial margin of error in predictions of the P load. To improve the accuracy of load estimates, it is important to clarify the fluctuation characteristics of PP concentration.

Over the past few decades, attention has focused on soil erosion as the major source of diffuse P loss to the freshwater environment (e.g., Cosser, 1989; Culley and Bolton, 1983; Grant et al., 1996; Hatch et al., 2001; Kronvang, 1992; Kronvang et al., 1997; Mainstone and Parr, 2002; McKee et al., 2000; Vanni et al., 2001). The PP concentration versus stream discharge (PPC–Q) relationship during individual rain events usually exhibits differences between the rising and falling limbs of a hydrograph, i.e. a hysteresis loop. The dominance of clockwise hysteresis loops in previous studies has been interpreted as suggesting that a rainfall produced a sufficiently high discharge to mobilize nearby sources of PP along the channel and on nearby fields (Hatch et al., 1999; McKee et al., 2000; Meyer and Likens, 1979; Prairie and Kalff, 1988). These sources were postulated as being exhausted on the rising limb, resulting in lower concentrations of PP on the falling one. This suggests that the supply and transport processes of PP differ between rising and falling limbs. However, separation of the rising and falling limbs does not significantly improve the predictive relationship because the PPC–Q relationship is inherent in each event (Meyer and Likens, 1979).

Paustian and Beschta (1979) found a positive relationship between the suspended sediment (SS) concentration and rate of change in discharge (dQ/dt) during individual rain events. Beschta (1987) compared two rain events with similar peak discharges but contrasting dQ/dt and found higher levels of SS concentration in an event with higher dQ/dt values. Terajima et al. (1997) showed that dQ/dt indicates the erosive force generating sediment load in subsurface flow. Since changes in the PP load are generally similar to those in the SS load (Mainstone and Parr, 2002), these results suggest that a large amount of PP is strongly pulsed when stream discharge is rapidly increased. Thus, dQ/dt is one of factors characterizing the PPC–Q relationship for each event.

As well as the above factor, the seasonal effect is essential to correctly characterize the PPC–Q relationship (Wall and Webb, 1982). Previous studies report that the amounts of released fine sediment and associated P (PP) vary depending on seasonal changes in hydrological conditions (McKee et al., 2000; Meyer and Likens, 1979; Munn and Prepas, 1986; Nistor and Church, 2005; Paustian and Beschta, 1979; Sidle and Campbell, 1985). Studies in North America found that high levels of SS occurred in a rain event soon after a long-term dry season, while a reduction in SS concentration was observed during successive rain events during the rainy season (Beschta, 1978; Paustian and Beschta, 1979; Sidle and Campbell, 1985). Bowes et al. (2005) indicated that the largest clockwise hysteresis loop for PP would occur in a late summer/autumn event following long dry periods, and then diminish through the wet winter and spring periods. These results suggest that the loss of PP in a rain event varies in response to antecedent rain history rather than seasonal changes in hydrological conditions, resulting in an event-specific PPC–Q relationship. However, little information is available about how antecedent rain history affects PP loss for each event in a forested watershed. Therefore, there is no model presently able to fully explain seasonal effects on PP loss.

Previous studies have considered that PP is accumulated in stream channels during antecedent dry periods because of the deposition of PP and sorption of dissolved P by bed-sediment (Bowes and House, 2001; Dorioz et al., 1998; Hill, 1981). The following rain event can remobilize the accumulated PP, associated with channel erosion. Therefore, it is possible that antecedent rain history affects the PP loss for each event via the accumulation of PP in stream channels.

Antecedent rain history is reflected in the antecedent moisture conditions in the forested watershed. Doerr
et al. (2003) and Shakesby et al. (2000) showed that water repellency is generally most severe when soils dry out, which in turn can lead to enhanced Hortonian overland flow and associated increases in soil erosion. Water repellent soils are being identified in an increasing number of natural environments in different climatic conditions around the world (Dekker et al., 2005). The soil in a plantation forest of Japanese cypress generally represents water repellency, probably because the litter includes large amounts of hydrophobic organic matter (Kobayashi and Shimizu, 2007). Therefore, it is speculated that antecedent rain history affects PP loss via soil water repellency, as well as by remobilization of PP accumulated in stream channels. However, few studies have investigated the effects of antecedent rain history on the PP loss in forested watersheds with water repellent soil. In this study, to clarify these effects, we investigated stream discharge and PP concentration in rain events in a small mountainous watershed covered with a plantation forest of Japanese cypress.

**Methods**

**Site description**

The study site (Ochozu Experimental Watershed: OEW) was a mountainous watershed located approximately 15 km east of Fukuoka City in western Japan (33°38’N, 130°32’E; Fig. 1). A mountain stream in the study area flows into an enclosed sea area, Hakata Bay, via the Tatara River. The watershed area is 9.5 ha. The length of the main stream is 265 m and the mean catchment width 358 m. The mean stream gradient (tangent) is 0.22 and the mean slope gradient (tangent) is 0.37. Located in the Asian monsoon region, the OEW receives frequent rainfall. From October 2001 to September 2003, the average annual precipitation was 1879 mm and air temperature 16.2 °C. Compared to Fukuoka City (Japan Meteorological Agency, 2006), the average annual precipitation in the OEW is approximately 300 mm higher and average air temperature approximately 1 °C lower.

The predominant forest soil is yellow-brown, and the underlying bedrock consists of serpentinite and chlorite schist. The soil texture is clay loam. The saturated hydraulic conductivity of the forest soil (depth: 10–50 cm) on the east-facing hillslope (Fig. 1; as marked by a rectangular symbol) ranged from $3.3 \times 10^{-6}$ to $4.6 \times 10^{-3}$ cm s$^{-1}$ (Higashii, 2005 unpublished data).

Approximately 46% of the OEW is covered by a plantation forest of Japanese cypress (*Chamaecyparis obtusa*) planted in 1957 along a stream channel. The tree density is approximately 1900 trees ha$^{-1}$ (Enoki, 2006 unpublished data). The tree height ranges from 8.5 to 19.0 m (average: 14.2 m, $n = 64$) and diameter at breast height ranges from 8.9 to 27.2 cm (average: 20.2 cm, $n = 64$) (Fujiyama et al., 2005). The ridge area is covered by a mixed forest of deciduous and evergreen trees such as *Myrica rubra* and *Clethra barbinervis*. Except for a small area, the trees in this watershed have not been pruned or thinned since 1993. Thus, the canopy of the plantation forest is closed and vegetation on the forest floor is scarce along the stream channel. The soil surface is covered by a duff-layer of leaf litter in the ridge area but is exposed in the riparian area.

**Observations**

*Molarity of Ethanol Droplet* test

To estimate the in situ hydrophobicity of the soil surface, we undertook a ‘Molarity of an Ethanol Droplet’ (MED) test on the east-facing slope of the OEW (Fig. 1), using procedures according to Doerr (1998). The MED test makes use of the known surface tensions of standardized solutions of ethanol in water. Drops of these dilutions are applied to a soil surface and their instant infiltration behavior is observed. A droplet with a higher surface tension than that of the soil surface will remain on the surface for some time, whereas a droplet with a lower surface tension will immediately infiltrate the soil. Drops (0.05 ml) of prepared solu-

![Figure 1](map.png)  
**Figure 1** Map of the Ochozu Experimental Watershed (OEW).
tions were applied onto the soil surface using a micropipiette. Increasing ethanol concentrations were used until drop penetration occurred within 3 s. The extent of water repellency was categorized by the ethanol concentrations according to Doerr et al. (2003) (Table 1). The MED test was conducted at four sites on the slope (the upper, middle, lower, and bottom sites) on three independent dry days with contrasting antecedent dry periods (June 2005, 13 days; October 2005, 5 days; December 2005, 2 days).

Hydrological surveys and water sampling
Meteorological stations were installed on the west and east ridges; precipitation was measured by tipping-bucket rain gauges at each site. A compound weir of triangular and rectangular notches was placed at the end of the OEW. Water levels were recorded at 10 min intervals using a hydraulic pressure sensor (OSASI Tech. Inc., Kochi, Japan, PC-001).

Intensive sampling at 15–60 min intervals during 24 h periods was also conducted manually with an automatic sampler (ISCO Inc., NE, USA, ISCO-6712) on eight separate days from October, 2001 to September, 2003. In some cases the sample bottles in the automatic sampler were not sufficiently filled with stream water due to a blockage of the aspiration tube. In such cases the concentration data of the sample bottles were excluded from data analyses because the concentration was extremely high.

Chemical analyses
The samples of stream water were stored at 5 °C in the dark on the day of sampling until analysis in the laboratory. We then analyzed the water samples for total phosphorus (TP), dissolved phosphorus (DP) and suspended sediment (SS). Particulate phosphorus (PP) was calculated by subtracting DP from TP. For TP analysis, water samples were digested using potassium peroxodisulfate (K2S2O8) (Menzel and Corwin, 1965) and then TP was measured using molybdenum blue (ascorbic acid) absorptiometry (Murphy and Riley, 1962). For DP analysis, water samples were filtered through glass fiber filters with a nominal pore size of 1.2 μm (Whatman, GF/C), followed by the same methods as those used in the TP analysis. The detection limits of TP and DP for our analytical conditions were both 0.4 μg-P l⁻¹. For SS, water samples were filtered (Whatman, GF/C) and the residue oven-dried at 105 °C for 2 h and then weighed.

Data analyses

Antecedent precipitation index
The antecedent precipitation index (API) was calculated for the rain events to determine the antecedent moisture con-
\[ C = a + bQ \]

where \( C \) is the PP concentration [mg l\(^{-1}\)], \( Q \) is the stream discharge [l s\(^{-1}\)], and \( a \) and \( b \) are empirical parameters. An analysis of covariance (ANCOVA) was performed among the rain events of each limb.

**Relationship between rates of changes in discharge and PP load**

To examine the prediction that a large amount of PP is strongly pulsed at times of rapidly increased discharge, rate of changes in discharge (\( \frac{dQ}{dt} \)) and load (\( \frac{dL}{dt} \)) were calculated as follows:

\[
\frac{dQ}{dt} \approx \frac{Q_i - Q_{i-1}}{t_i - t_{i-1}} 
\]

\[
\frac{dL}{dt} \approx \frac{L_i - L_{i-1}}{t_i - t_{i-1}} = \frac{C_iQ_i - C_{i-1}Q_{i-1}}{t_i - t_{i-1}} 
\]

where \( i \) is the order of the time-series data, \( t_i \) is the time [s], and \( L \) is the PP load [mg-P l\(^{-1}\)].

For an increasing rate of discharge (\( \frac{dQ}{dt} > 0 \)), a regression analysis was conducted for each rain event using the following equation:

\[
\frac{dL}{dt} = a' + b' \frac{dQ}{dt} 
\]

where \( a' \) and \( b' \) are empirical parameters.

**Results**

**Soil water repellency**

From the upper to the bottom sites of the hillslope (Fig. 1), ‘very strong — extreme’ water repellency of the soil surface was observed on the day when the antecedent dry period was 13 days (Table 2). In contrast, water repellency was measured as ‘none’ on the day when the antecedent dry period was 2 days. Potential evaporation (\( E_p \)) was highest among the three days on the day of ‘very strong — extreme’ water repellency, whereas \( E_p \) was lowest on the day of ‘none’. On the day when ‘very strong’ water repellency was observed, the antecedent dry period was 5 days and the \( E_p \) value was between those of the other days. The rank of soil water repellency on the three days corresponded to that of API, depending on the day number (e.g., API\(_{10}\), API\(_{20}\); Table 2).

**Hydrological properties**

The average annual specific discharge in the OEW was 933 mm during the observation period (from October 2001 to September 2003). Approximately 50% of the precipitation (1879 mm) became outflow at the watershed outlet (Fig. 2). Frequency of a daily specific discharge of less than 5 mm day\(^{-1}\) accounted for 89% (652 days) of the days during the observation period (Fig. 3a). The sum of the discharge on these days accounted for 36% of the annual discharge (Fig. 3b). Thus, the sum of the discharges of more than 5 mm day\(^{-1}\) accounted for 64% of the annual discharge although these days accounted for just 11% (78 days) of the observation period.

The response time decreased with increasing rainfall intensity (Fig. 4). When data were divided on the basis of API\(_{10}\), response times can be over 350 min at rainfall intensities ranging from 0 to 10 mm 30 min\(^{-1}\) on API\(_{10}\) of more than 12.4 mm. On API\(_{10}\) of less than 12.4 mm, the response time was less than 200 min. The bin-average of response time on API\(_{10}\) of less than 12.4 mm was significantly lower than that of API\(_{10}\) of more than 12.4 mm.

### Table 2 Results of ‘molarity of an ethanol droplet (MED)’ test, antecedent dry periods, potential evaporation, and API

<table>
<thead>
<tr>
<th>Date</th>
<th>MED (mol l(^{-1}))</th>
<th>Classifications of water repellency</th>
<th>Antecedent dry period (day)</th>
<th>Potential evaporation ( E_p ) (mm day(^{-1}))</th>
<th>API(_{10}) (mm)</th>
<th>API(_{20}) (mm)</th>
<th>API(_{30}) (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>June 1, 2005</td>
<td>4.09–6.14</td>
<td>Very strong — extreme</td>
<td>13</td>
<td>4.6</td>
<td>0.0</td>
<td>6.1</td>
<td>12.6</td>
</tr>
<tr>
<td>October 11, 2005</td>
<td>3.07–4.09</td>
<td>Very strong</td>
<td>5</td>
<td>2.8</td>
<td>7.1</td>
<td>7.1</td>
<td>7.3</td>
</tr>
<tr>
<td>December 20, 2005</td>
<td>0</td>
<td>None</td>
<td>2</td>
<td>1.3</td>
<td>12.4</td>
<td>44.2</td>
<td>55.5</td>
</tr>
</tbody>
</table>

Figure 2  Hyeto-hydrograph of the Ochozu experimental watershed during the observation period. Vertical axis of the discharge represents a logarithmic scale. Upper bars show daily precipitation. Circles represent the timings of the eight rain events monitored. (Modified Ide et al., 2007)
on API10 of more than 12.4 mm at rainfall intensities ranging from 0 to 10 mm 30 min

Characteristics of rain events

PP and SS concentrations and discharge data were obtained for eight rain events during the observation period (Fig. 2). These events are referred to as Ev.1 to Ev.8 in order by date (Table 3). The maximum and minimum values of daily discharge differed by a factor of 10, while the maximum and minimum values of maximum peak discharge differed by a factor of 100. The heaviest rainfall occurred in Ev.2. Based on precipitation data (1961–2005) in Fukuoka City (Japan Meteorological Agency, 2006), Ev.2 had a probable annual maximum daily precipitation of once in a decade.

There was strong positive correlation between concentrations of PP and SS for all rain events (r = 0.92, p < 0.001; Fig. 5). The slope of the regression line shows that the P content of SS averaged 0.03% throughout the eight events.

For all eight events, the maximum peaks of PP concentration occurred prior to the maximum peak discharge, and the concentrations rapidly decreased after the maximum peaks (Fig. 6); these trends are indicative of the flushing effect. In some rain events the maximum peak discharge was preceded by a small peak discharge (Ev.2, Ev.7 and Ev.8). In these instances the maximum peak PP occurred prior to the small peak discharge.

Relationship between PP concentration and discharge on rising and falling limbs

The determination coefficient of the PPC–Q relationship on a double logarithmic scale for all rain events was 0.21 (Fig. 7). The PP concentration varied by up to two orders of magnitude at discharge levels ranging from 4 to 6 l s

Empirical parameters, determination coefficients and probabilities for regression equations of PPC–Q relationships in the eight events were determined (Table 4). For the falling limb of Ev.4, the latter part of data where both PP concentration and discharge increased was excluded from the regression analysis. On the rising limb, the deter-
ministration coefficients were generally low and the regression coefficients \( b \) in Eq. (5)) for just three events were statistically significant \((p < 0.05\) in all cases). On the falling limb, the determination coefficients were relatively high and all values of \( b \) were significant \((p < 0.05\) in all cases) and positive. On each limb, the regression lines of the PPC–Q relationships were significantly different among the rain events (ANCOVA, \( p < 0.001\) for the rising limb; test of homogeneity of regression in ANCOVA, \( p < 0.05\) for the falling limb). There was no significant correlation between \( b \) and API on the falling limb (Fig. 9a and b).

### Relationship between rates of changes in discharge and PP load

For each of the eight events, the \( dL/dt \) versus \( dQ/dt \) (\( dL/dt–dQ/dt \)) relationships were positive (Table 5). The regression coefficients \((b' \) in Eq. (8)) for just three events were statistically significant \((p < 0.05\) in all cases). However, the determination coefficients of the \( dL/dt–dQ/dt \) relationships were higher than those of the PPC–Q relationships on the rising limb except for Ev.1 and Ev.6 (Tables 4 and 5). In the case of \( dL/dt > 0 \), the determination coefficients were much higher except for Ev.6 whereas the values of \( b' \) were not significantly different from those in all data \((dL/dt < 0 + dL/dt > 0) \) (\( u \)-test, \( p = 0.8747\)). There was a strong negative correlation between \( b' \) (the case of \( dL/dt > 0 \)) and API on a logarithmic scale \((r = -0.91, p = 0.0017\) for \( API_{10} \); Fig. 10a). Also, the correlation was statistically significant \((p < 0.05\) regardless of the day number of API (Fig. 10b). This negative relationship was retained without the data of Ev.2, in which \( b' \) was much larger and API was much smaller than in the other seven events \((r = -0.72\) to \(-0.39\) in the \( API_{9} \) to \( API_{30} \) range).

### Discussion

**Effects of antecedent dry period on soil water repellency**

The results of the MED tests (Table 2) suggested that soil water repellency may increase with the length of the antecedent dry period, although this was based on only three observations. Since the day of the ‘very strong – extreme’ water repellency was during summer, potential evaporation \((E_p)\) was high and thus the soil surface might dry readily. On the other hand, since the day of non-water-repellency was during winter, \( E_p \) was low and thus the soil surface could be expected to have been moister than during summer conditions. These results support the suggestion of Walsh et al. (1994) that soil water repellency is most severe in dry summer and least in wet winter conditions. Therefore, dry conditions lead to strong water repellency of the soil surface in the OEW as well as previous studies (Doerr and Thomas, 2000; Kobayashi and Shimizu, 2007).

**Runoff characteristics of PP on rising and falling limbs**

As rapid and brief increases in discharge resulted in a small number of water samples on the rising limb where the case of \( dQ/dt > 0 \) was commonly observed, not all of \( dL/dt–dQ/dt \) relationships in the eight events were significant \((p < 0.05\) despite this limitation, there was a stronger correlation between \( dL/dt \) and \( dQ/dt \) than between the PP concentration and discharge on the rising limb (Tables 4 and 5). Hydrograph characteristics such as \( dQ/dt \) help to explain the supply and transport processes of PP primarily on the rising limb (e.g., Bowes et al., 2005). Therefore, the \( dL/dt–dQ/dt \) relationship is a better indicator of the PP
supply and transport processes than the PPC–Q relationship on the rising limb. The positive $dL/dt–dQ/dt$ relationships in the case of $dL/dt > 0$ for each of the eight events (Table 5) show that a large amount of PP is strongly pulsed when a rapid increase in discharge occurs. The values of $b$’ in the case of $dL/dt > 0$ differed little from those in all data ($dL/dt < 0 + dL/dt > 0$), suggesting that the positive $dL/dt–dQ/dt$ relationships for each event could be robust even if sampling frequency increased. Unfortunately, the concentration data during peak discharges in Ev.1 were excluded from the data analysis because the sample bottles in the automatic sampler were not sufficiently filled with stream water, possibly because of the blockage of the aspiration tube. However, the value of $b$’ in Ev.1 should have remained almost unchanged even if all concentration data during peak discharges had been obtained, because $dL/dt$ can be expected to show a roughly constant relationship to $dQ/dt$ during individual events.

In the case of $dQ/dt > 0$, the PP load at time $t$ ($L_t$) can be regarded as an integrated value of the PP load increment ($\Delta L$), which is supplied (produced) and transported from the initial increase in discharge to time $t$

$$\Delta L = \int_0^t \frac{dL}{dt} \, dt = L_t - L_0$$

$$L_t = L_0 + \Delta L$$

where $L_0$ is the PP load immediately prior to the increase in discharge [mg-P s$^{-1}$]. According to Eq. (9), $dQ/dt$ affects the PP load via $\Delta L$. Consequently, on the rising limb, changes in the PP load and concentration are not expressed as a simple function of discharge (e.g., Eq. (5)).

Since discharge $Q$ is expressed as a product of flow area $A$ and flow velocity $v$ ($Q = A \cdot v$), $dQ/dt$ is equal to $dA/v dt$. When $A$ is constant, $dL/dt$ is expressed as a function of $dv/dt$. Newton’s law ($F = m \cdot dv/dt$, where $F$ is the body...
force, \( m \) is the body mass, and \( \frac{dv}{dt} \) is acceleration) physically supports the proposal that \( \frac{dv}{dt} \) is the driving force of the fluvial PP supply via its effect in generating soil erosion (Terajima et al., 1997). This concept is applicable to the PP loads derived from soil macropores in an unsaturated zone and thus \( A \) is constant. Since the stream in the OEW is an open channel, the expansion of \( A \), in addition to \( \frac{dv}{dt} \), will contribute to the PP supply by enhancing erosion of the channel bed and bank. These results suggest that \( \frac{dQ}{dt} \) is the driving force behind the PP supply. In terms of the rising limb, the frequency of PP supply should be higher. Thus, the PP load is higher at a given discharge level than that on the falling limb.

On the falling limb, the PP concentration with respect to each event decreased depending on the discharge (Table 4). Since the case of \( \frac{dQ}{dt} < 0 \) is commonly observed on the falling limb, the frequency of increases in the PP load (\( \frac{dL}{dt} > 0 \)) should be low. Walling et al. (2000) reported that the particle size of sediment load either remained con-

**Figure 7** Relationship between particulate phosphorus concentration and stream discharge for eight rain events. Vertical and horizontal axes are both logarithmic scales. Solid line represents the regression line (\( y = 0.0028x^{0.46}, R^2 = 0.21 \)).

**Figure 8** Relationship between particulate phosphorus concentration and stream discharge for each of eight rain events. Note that the scale varies for the different rain events.
stant or decreased on the falling limb, reflecting the absence of any major changes in sediment sources. These results suggest that supply of PP into stream water is largely absent on the falling limb; the transport of PP in the stream may be dominant over the supply (Terajima et al., 1997).

The clockwise hysteresis loop of the PPC–Q relationship can be attributed to the fact that DL addition, which leads to marked fluctuations in the PP load, occurs primarily on the rising limb whereas on the falling limb DL is largely absent. Thus, supply and transport processes of PP differ between rising and falling limbs.

### Effects of antecedent rain history on PP loss

The regression coefficient ($b'$) in Eq. (8) is equal to $dL/dQ$ at a given $dQ/dt$ level, leading to a rapid increase in PP load even with a slight increase in discharge. Thus, $b'$ can be defined as follows:

$$b' = \frac{dL}{dQ}$$

Table 4  Parameters, determination coefficients, and probabilities for the regression equations between PP concentration and discharge for the rising and falling limbs of each rain event

<table>
<thead>
<tr>
<th>Event</th>
<th>Rising limb</th>
<th>Falling limb</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$a$ ($\times 10^{-2}$ mg-P l$^{-1}$)</td>
<td>$b$ ($\times 10^{-3}$ mg-P s$^{-1}$)</td>
</tr>
<tr>
<td>Ev.1</td>
<td>1.58</td>
<td>1.54</td>
</tr>
<tr>
<td>Ev.2</td>
<td>9.61</td>
<td>–0.03</td>
</tr>
<tr>
<td>Ev.3</td>
<td>0.18</td>
<td>0.60</td>
</tr>
<tr>
<td>Ev.4</td>
<td>0.08</td>
<td>1.47</td>
</tr>
<tr>
<td>Ev.5</td>
<td>3.13</td>
<td>–0.20</td>
</tr>
<tr>
<td>Ev.6</td>
<td>1.22</td>
<td>0.37</td>
</tr>
<tr>
<td>Ev.7</td>
<td>1.31</td>
<td>–0.05</td>
</tr>
<tr>
<td>Ev.8</td>
<td>1.39</td>
<td>–0.17</td>
</tr>
</tbody>
</table>

Table 5  Parameters, determination coefficients, and probabilities for the regression equations between rate of changes in PP load ($dL/dt$) and discharge ($dQ/dt$) for each rain event

<table>
<thead>
<tr>
<th>Event</th>
<th>All data</th>
<th>The case of $dL/dt &gt; 0$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$a'$ ($\times 10^{-4}$ mg-P s$^{-2}$)</td>
<td>$b'$ (mg-P l$^{-1}$)</td>
</tr>
<tr>
<td>Ev.1</td>
<td>–1.55</td>
<td>0.090</td>
</tr>
<tr>
<td>Ev.2</td>
<td>–40.80</td>
<td>0.238</td>
</tr>
<tr>
<td>Ev.3</td>
<td>–0.04</td>
<td>0.016</td>
</tr>
<tr>
<td>Ev.4</td>
<td>–0.05</td>
<td>0.022</td>
</tr>
<tr>
<td>Ev.5</td>
<td>–0.39</td>
<td>0.090</td>
</tr>
<tr>
<td>Ev.6</td>
<td>0.04</td>
<td>0.016</td>
</tr>
<tr>
<td>Ev.7</td>
<td>–0.23</td>
<td>0.021</td>
</tr>
<tr>
<td>Ev.8</td>
<td>–0.03</td>
<td>0.018</td>
</tr>
</tbody>
</table>

* Both cases of $dL/dt > 0$ and $dL/dt < 0$.

Figure 9  (a) Relationship between parameter $b$ (see Table 4) and API10, and (b) correlation coefficients for relationship between $b$ and API on a logarithmic scale. Horizontal axis in (a) is a logarithmic scale. Solid line in (a) represents the regression line ($y = –0.0002\log x + 0.0006$, $R^2 = 0.07$). Gray zone in (b) represents a confidence interval at the 5% level.
be used as an indicator of the mobility of PP to runoff for each event. This analysis is similar to that in Asselman (1999) that plotted maximum and minimum SS concentrations against discharge on the rising limb and used the slope of the line as an indicator of the sediment availability. The strong negative correlation between $b$’ and API (Fig. 10) suggested that the mobility of PP increased as the antecedent moisture conditions became drier. Although this negative relationship appears to be based on Ev.2, the result showed that it did not depend solely on Ev.2. This result was consistent with previous studies in which sediment availability and mobility of PP are large when antecedent conditions are dry (Asselman, 1999; Bowes et al., 2005; Nistor and Church, 2005). During Ev.2 following a dry period of more than 1 week, rapid increases in the PP concentration were recorded prior to the early peak discharge (Fig. 6). This is attributable to a greater increase in mobility of PP during the longer antecedent dry period. Long antecedent dry periods would strengthen the soil water repellency in the OEW. Therefore, the increase in mobility of PP can be explained according to the hypothesis that Hortonian overland flow readily occurs because of an increase in soil water repellency caused by soil desiccation (Doerr et al., 2003).

The scaly leaf litter of Japanese cypress separates into small pieces within a few months and is thus easily removed from the forest floor by rainfall and/or wind soon after defoliation (Hattori et al., 1992). As the OEW is covered primarily by a plantation forest of Japanese cypress with scarce understory vegetation, the litter layer is poorly developed and the soil surface is noticeably exposed along the stream channel. Therefore, surface crusting such as a skin seal or an erosion crust is formed on the surface by raindrop impact, resulting in low permeability (Onda and Yamamoto, 1998). Hortonian overland flow can easily occur upon such soil during rain events. The ‘very strong — extreme’ water repellency of the soil surface under dry conditions (Table 2) would also contribute to the occurrence of Hortonian overland flow. Soil water repellency can accelerate rapid runoff responses by Hortonian overland flow generated near a stream channel (Doerr et al., 2003). Therefore, the relatively short response time following dry conditions (Fig. 4) supports the occurrence of Hortonian overland flow, which causes surface erosion (Shakesby et al., 2000) and thus increases the PP supply to the stream water. Also, soil water repellency in itself can enhance rainsplash erosion (Terry and Shakesby, 1993). Accordingly, the negative relationship between $b$’ and API (Fig. 10) reflected the fact that soil water repellency affects the supply processes of PP, depending on soil moisture conditions.

As well as soil water repellency, slaking might also explain the negative relationship between $b$’ and API. For a dry soil, slaking caused by the compression of air trapped within an aggregate can efficiently break down and disperse aggregates in response to a sudden rain event (Le Bissonnais et al., 1989). McDowell and Sarpley (2002) reported that antecedent moisture conditions greatly affect erosion and PP transport potential via slaking. Since the potential for slaking increases with longer antecedent dry periods, a large amount of PP supply would then occur during the course of an event due to soil detachment and transport processes. The sediment source in a watershed would be primarily the stream channel bed and bank (Walling, 1983). Therefore, it is plausible that the channel bank, within which the available sediment and associated P are stored immediately prior to the beginning of an event, is rapidly inundated by the rising water table in response to a sudden rainfall. Consequently, slaking occurs, resulting in the supply of PP to the stream water. Although we did not measure slaking, it is possible that an increase in the mobility of PP derived from soil desiccation can be explained by soil water repellency and by slaking.

The negative relationship between $b$’ and API (Fig. 10) also appears to be indicative of remobilization of within-channel PP accumulated during antecedent periods because API is reflected in antecedent flow conditions. Previous studies suggest that increases in sediment availability could be explained in part by within-channel sediment deposition in low flow conditions (Asselman, 1999; Nistor and Church, 2005). Since changes in the PP concentration were similar to those in the SS concentration in the OEW (Fig. 5), PP would be deposited in the stream channel in low flow conditions. As well, DP would be sorbed onto the upper surface of fine bed-sediment and some of the bioavailable soluble P would be assimilated into biomass in low flow conditions, thereby converting it from DP to PP (Bowes and House, 2001). An early rain event following low flow conditions

**Figure 10** (a) Relationship between parameter $b$’ (see Table 5) and API$_{10}$, and (b) correlation coefficients for relationship between $b$’ and API (Fig. 10) reflected the fact that soil water repellency affects the supply processes of PP, depending on soil moisture conditions.

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can quickly entrain and transport P-rich surface bed-sediment in a stream channel, possibly leading to a rapid increase in PP load even with a slight increase in discharge. Therefore, the dominant sources of the rapid increased PP load in the OEW are likely to be the remobilization of within-channel PP accumulated during an antecedent period and soil erosion via soil water repellency and slaking. Based on sediment ‘fingerprinting’ studies, Walling and Amos (1999) demonstrated that SS could represent sediment delivered directly to the channel rather than remobilization of within-channel sediment deposits because of bare soil in a cultivated field within their study watershed of River Piddle, UK. Given the bare soil surface along the stream channel in our OEW, it is predicted that the remobilization of within-channel PP may have contributed less to the rapid increase in PP load during rain events soon after long dry periods than the PP supply associated with soil erosion. Further investigations will be required to clarify how much each of the remobilization of within-channel PP or soil erosion contributes to rapid increases in PP load.

On the falling limb, the regression coefficient for the PPC–Q relationship (b) is equal to C/Q and was thus regarded as the decreasing rate of the PP concentration in response to discharge fluctuations. Given that the supply of PP into stream water is largely absent on the falling limb, the non-significant correlation between b and API (Fig. 9) suggested that antecedent moisture conditions did not affect the transport process of PP on the falling limb. Therefore, the antecedent moisture conditions affected the supply and transport processes of PP primarily on the rising limb.

Inter-event differences in the hydrograph characteristics, which determine dQ/dt, and the mobility of PP were able to explain the prominent flushing effects recorded during Ev.2, Ev.7, and Ev.8. Such a flushing effect may occur in the case where discharge rapidly increases in the early stage of an event (e.g., Ev.7 and Ev.8), where b’ is high, or where both conditions are met (e.g., Ev.2). During Ev.2, when the highest levels of PP were observed immediately prior to the early peak discharge, it is likely that soil desiccation during antecedent dry periods enhanced the flushing effect on the rising limb.

In summary, the antecedent moisture conditions affected the increasing rate of PP load during the rising limb. Soil desiccation during dry periods would enhance the mobility of PP. Therefore, differences in PP concentration at a given discharge level between rising and falling limbs generally tend to be wider during an early event following a long dry period than in subsequent events.

Conclusions

In this study, the PPC–Q relationship for each rain event exhibited a clockwise hysteresis loop, which is the most common hysteresis loop generally observed. The results revealed that the clockwise hysteresis loop could be attributed to differences in the supply and transport processes between the rising and the falling limbs. On the rising limb, dQ/dt > 0 was commonly observed and the dQ/dt determined the increasing rate of the PP load (dL/dt), suggesting that the PP supply was associated with soil erosion and remobilization of within-channel PP. On the falling limb, the PP concentration decreased with discharge. These results suggested that dQ/dt is a crucial determinant affecting the PP supply and the primary control on the clockwise hysteresis loop of the PPC–Q relationship for each event.

Our data suggested that antecedent moisture conditions determined the slope of the regression line between dL/dt and dQ/dt for each event. Thus, the PP load increment (dL) can become large even with slight increases in discharge as antecedent moisture conditions become drier. The effects of antecedent rain history on the PP loss in previous studies can be interpreted to suggest that longer antecedent dry periods enhance the flushing effect on the rising limb and thereby the clockwise hysteresis loop becomes wider during an early event than in subsequent events. The results of the MED tests and the relatively short response times following dry conditions support the proposal that the strong water repellency of a soil surface caused by soil desiccation can induce Hortonian overland flow and thus soil erosion. Therefore, we concluded that soil desiccation during antecedent dry periods would enhance the mobility of PP during rain events in a plantation of Japanese cypress under current management practices.

When predicting the P load from a forested watershed based on discharge, predictive accuracy would be improved by considering the effects of antecedent rain history on the mobility of PP.

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References


Effects of antecedent rain history on particulate phosphorus loss from a small forested watershed