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Importance of frequent storm flow data for evaluating changes in stream water chemistry following clear-cutting in Japanese headwater catchments

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ABSTRACT

This study investigated the changes in stream water chemistry during low flow and storm flow following clear-cutting in a paired forest catchments. Regular grab samples, obtained mainly during low flow, showed that the Cl^- concentration in stream water decreased, the NO_3^- concentration increased, and the Na^+, SO_4^{2-}, K^+, Mg^{2+}, and Ca^{2+} concentrations did not change significantly after clear-cutting. Highly frequent samplings revealed that solute concentrations at high flow changed to a larger degree after cutting than those at low flow. The NO_3^- concentration at high flow changed earlier and to a greater degree than that at low flow. In addition, Na^+, K^+, and Ca^{2+} also increased at high flow just after clear-cutting, in association with NO_3^- flushing. The mean annual NO_3^- export after clear-cutting, as estimated by the highly frequent samplings including concentrations during storm flow, was more than 60 kg ha^{-1} and 1.5-fold higher than that calculated by only weekly sampling data. The largest difference was found 8 months after clear-cutting, when the methods differed by 40 kg ha^{-1} yr^{-1} and the estimate by the regular measurements with high-frequency sampling data was three-fold higher than that with the weekly regular sampling. These results show that high-frequency observation following forest cutting is important for accurate estimation of output fluxes.

1. Introduction

Deforestation and natural disturbances have significant effects on stream chemistry (Likens et al., 1970; Neal et al., 1998; Swank et al., 2001). In particular, increases in the NO_3^- concentration in stream water have been found in many catchments after forest cutting. The NO_3^- concentration begins to increase about 1 year after forest cutting and declines slowly for 3 to 5 years to a pre-cutting level (Binkley and Brown, 1993; Burns and Murdoch, 2005; Neal et al., 1998). Changes in NO_3^- are largely the result of reduced uptake by vegetation and changes in microbial processes such as N-mineralisation and nitrification (Likens et al., 1970; Burns and Murdoch, 2005). In addition, changes in concentrations of other chemicals and acidity following forest cutting have been reported due to decreased input of dry deposition resulting from loss of foliage areas (Reynolds et al., 1995; Feller, 2005; Cummins and Farrell, 2003) and changes in weathering rates accompanied by increased N-mineralisation and nitrification (Lawrence et al., 1987).

These previous studies focused mainly on stream water during baseflow conditions, and samples were collected weekly to monthly. Several studies have observed stream chemistry during storm flow following forest cutting and showed differences at high flow. For example, Burns and Murdoch (2005) reported that the stream NO_3^- concentration during storm flow increased more than 100-fold relative to pre-cutting values within 4 months after completion of clear-cutting, and increased 10-fold during baseflow at a study site in eastern North America. Furthermore, in northeastern America, Wang et al. (2006) found that the first response of stream water chemistry following cutting was an increase in Ca^{2+} at high flow due to soil disturbances and sediment movement associated with skidding. However, few studies have focused on stream water chemistry during storm flow following forest cutting.

In recent years, the hydrologically induced mobilisation of NO_3^- from forested watershed has attracted increased research interest (Creed et al., 1996; Rusjan et al., 2008). For example, Cirmo and McDonnell (1997) noted that differences in the nitrogen (N) source may be illustrative of hydrologic flow path variations thought to exist between “baseflow” and “event flow” in complex riparian zones. Stream water flushing NO_3^- pulses were usually observed in autumn and spring, and were associated with greater amounts of precipitation in autumn and snow melt in spring (McHale et al., 2002; Inamdar et al., 2006). The “flushing hypothesis” is a conceptual explanation of mechanisms responsible for the commonly observed peak in solute concentration on the rising limb (Hornberger et al., 1994). The flushing of NO_3^- occurs with the mobilisation of NO_3^- stores in the upper soil layer during storm

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events. Therefore, stream water NO_3^- is controlled by biogeochemical conditions in the soil layer and by catchment hydrology.

Forest cutting may have large impacts on N dynamics in root zones, with the lack of plant uptake of N being the principal cause of increased NO_3^- export (Burns and Murdoch, 2005). Therefore, chemical information obtained from stream water during high flow, to which soil water contributes, is necessary to accurately evaluate the impact of deforestation on nutrient export.

High-frequency sampling of stream water chemistry, especially during storm events, is critical for quantifying exports of solutes (Inamdar, et al., 2006; Rusjan et al., 2008). In previous studies, the export of solutes from forest catchments was calculated more accurately by discharge-proportional sampling than by grab sampling (Schleppi et al., 2006; Ide et al., 2007). Kirchner et al. (2004) also noted that high-frequency sampling conducted over longer time periods will yield new insights into the hydrologic and biogeochemical processes regulating stream chemistry. However, few studies have evaluated the chemical export from clear-cut catchments with high-frequency sampling. We hypothesised that a more significant impact of forest cutting would be found during high flow rather than during low flow, and that frequent sampling of data during storm flow following clear-cutting would provide insights on solute transport. The objectives of this study were (1) to evaluate the impact of deforestation on stream water chemistry using a seven-year time series of stream water chemistry during baseflow following clear-cutting and intensive observation during nine storm events, and (2) to investigate how clear-cutting affects stream water chemistry.

2. Methods

2.1. Site description

This study was conducted in a pair of small headwater catchments (Fukuroyamasawa Experimental Watershed, Catchment A, 0.8 ha; Catchment B, 1.1 ha) at the Tokyo University Forest in Chiba, Chiba Prefecture, Japan (35°12′N, 140°06′E) (Fig. 1). Both catchments were located less than 10 km from the ocean and underlain by Tertiary formation sedimentary rock. The altitude ranged from 126 to 230 m above sea level. The annual mean temperature was 14 °C (1994–2003), and mean annual precipitation was 2230 mm (1994–2003). The average slope of the main stream was 25.5° in Catchment A and 23.5° in Catchment B. The mean soil depth was 2.9 m in Catchment A and 2.2 m in Catchment B (Shiraki et al., 1999).

A mature mixed plantation of *Cryptomeria japonica* and *Chamaecyparis obtusa* planted from 1928 to 1931 covered both catchments until March 1999. The mean height of the trees was approximately 20 m. The trees were clear-cut in April 1999 in Catchment B. To prevent soil surface disturbance as much as possible, logging from catchment B was done using skyline, and most branches and leaves were piled by hand and left in approximately 40 places in the catchment. These piles of litter were roughly estimated at 4.7×10^4 kg (dry weight) in 1999 (Hashimoto and Suzuki, 2004; Hotta et al., 2007). Seedlings of the same species, with heights of less than 1 m, were then planted in April 2000. In 2007, the mean height of the planted stands was 7.3 m. Catchments A and B are referred to as the control and clear-cut catchments, respectively, in the following sections.

The mean annual discharge for the interval before clear-cutting (1994–1998) was 758 mm in the control catchment and 709 mm in the clear-cut catchment. After cutting (1999–2003), these values were 959 mm and 1224 mm, respectively. Hence, the annual discharge of the clear-cut catchment increased by approximately 300 mm after cutting. This increase was considered to be largely caused by a reduction in evapotranspiration (Maita et al., 2005; Maita and Suzuki, 2007).

2.2. Samplings and chemical analyses

Rainfall amount was measured using both a 0.5-mm tipping bucket rain gauge and a funneled bottle collector 21 cm in diameter at a meteorological station (Shinta Weather Station) located 200 m from Fukuroyamasawa Experimental Watershed. The discharge rate was measured continuously by 90° V-notch weirs, the locations of which are marked as Weir A (control catchment) and Weir B (clearcut catchment) in Fig. 1. Precipitation samples were collected for chemical analysis at Shinta Weather Station located 200 m from Fukuroyamasawa Experimental Watershed between April 1998 and December 2006. The samples were collected in a polyethylene plastic bottle attached to a funnel with a 21-cm-diameter aperture. Throughfall samples were collected using the same type of bottle at the control catchment. Throughfall samples at the control catchment were collected weekly or biweekly between April 1998 and December 2006 at point TA in Fig. 1. Stemflow samples were collected in the control catchment using a plastic bottle and a wrapping hose that was placed around the stem of a *C. japonica* at a height of 1.3 m, as described previously (Kuraji et al., 2001). These samples were collected weekly or biweekly from April 1998 to December 2006 at point SA in Fig. 1. In the control catchment, throughfall and stemflow samples were collected at one fixed point in the middle of the forest because trees were being planted manually at the time of investigation, and the trees and nature of the canopy were similar across the whole catchment. Groundwater samples were collected from groundwater. Perennial groundwater samples for chemical analysis were collected from a tube attached to the bottom of two observation wells at the clear-cut catchment from 1998 to 2006. At points B6 and B7, wells of 270- and 150-cm depths, respectively, were installed (B6-270 and B7-150). Soil water samples were collected from three porous cups installed at depths of 20, 30, and 60 cm.

Regular samples of stream water were collected manually weekly or biweekly from April 1998 to December 2006 at Weir A and Weir B in Fig. 1. Stream water samples during storm events were collected in the control and clear-cut catchments using an automatic sampler (American Sigma, New York, Model 900) at a time interval of 10 to 120 min in nine events: July 16, 1998 (Storm 1); October 27, 1999 (Storm 2); July 7, 2000 (Storm 3); September 6, 2000 (Storm 4); October 20, 2000 (Storm 5); July 10, 2002 (Storm 6); June 16, 2006 (Storm 7); July 5, 2006 (Storm 8); and September 27, 2006 (Storm 9) (Table 1). The storm events were the largest of the top 10% of all storms by size during the observation period.

Samples were filtered using 0.2-μm membrane filters, and concentrations of Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+} were determined by ion chromatography (Shimadzu, Kyoto, Japan, LC-10A).

2.3. Methods of flux calculation

The input nutrient loads in the forest catchment were calculated by multiplying the volume-weighted concentration of input by the water volume. The input before forest cutting was calculated by the mix of throughfall and stemflow inputs, and the input after cutting was calculated by precipitation input. Annual stemflow volume and throughfall volume were 5% and 79% of annual precipitation, respectively, and the water volume of canopy interception was calculated by the annual mean ratio as 17% precipitation (Tanaka et al., 2005).

Output loads were calculated by integrating hourly fluxes of solutes. First, the hourly concentrations were estimated. The regression method (Schleppi et al., 2006) was applied to estimate the hourly concentration because discharge was measured more frequently. The equation was expressed as follows:

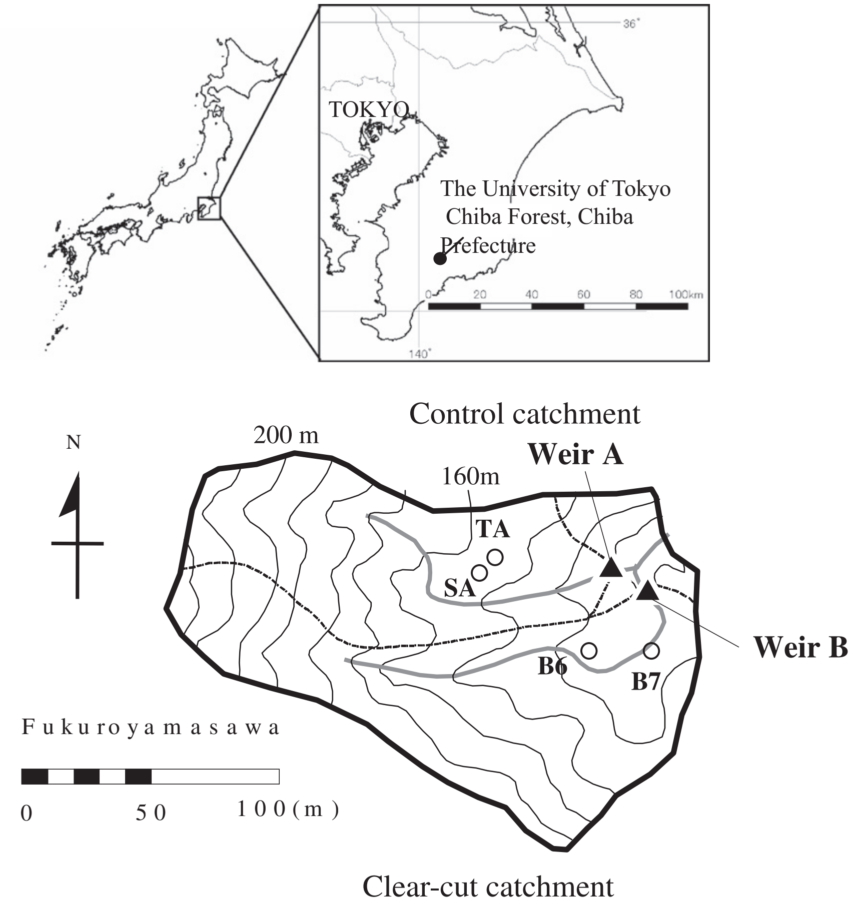

Fig. 1. Location of the Fukuroyamasawa Experimental Watershed. The dashed line denotes the border between catchments A (control) and B (clear-cut). SA, TA are sampling points of stemflow and throughfall. B6 is sampling point of soilwater and groundwater. B7 is sampling point of groundwater.

Table 1
Characteristics of storm events.

Event number	Date	Total rainfall (mm)	Maximum rainfall (mm/h)
1	1998/7/16	124	31
2	1999/10/27–29	126	22
3	2000/7/7–10	223	29
4	2000/9/6–8	168	60
5	2000/10/20–23	60	12
6	2002/7/10–12	110	23
7	2006/6/16–17	63	17
8	2006/7/5–6	80	24
9	2006/9-26–28	178	62

$$C(t) = a \log Q(t) + b \tag{1}$$

$$L = \sum C(t)Q(t) \tag{2}$$

where $C(t)$ is the solute concentration at time t, $Q(t)$ is the discharge at time t (mm hr^{-1}), a and b are empirical parameters, and L is the load of solutes (kg ha^{-1}).

When output fluxes were calculated only by weekly sampled stream water concentrations, the fluxes were calculated by multiplying the average measured solute concentration at the beginning and end of the interval of grab samplings by the discharge during that period (Martin et al., 2000; Rosen et al., 1996). Annual fluxes were determined by the sum of fluxes of each observed period.

Output fluxes were calculated by both methods from April 1998 to April 1999 as before cutting, and from April 1999 to December 1999, 2000, 2002, and 2006 to show the trend after cutting when intensive stream flow observation was conducted.

3. Results

3.1. Input ion concentration

The mean concentrations of Cl^-, NO_3^-, SO_4^{2-}, Na^+, K^+, Mg^{2+}, and Ca^{2+} in throughfall were approximately two to three times higher than those in precipitation (Fig. 2). Concentrations of NH_4^+ in precipitation were similar to those in throughfall ($p > 0.05$). Those in stemflow were similar or higher than those in throughfall except NO_3^- and NH_4^+. These results show that the concentrations of most of solutes increased while passing through the canopy. The concentrations of nutrients decreased significantly ($p < 0.001$) after cutting. The variations in solute concentrations in throughfall and stemflow were larger than those in precipitation. The difference was caused by the variation in dry deposition input, which is delivered by winds and accumulates on the canopy.

3.2. Changes in stream water, groundwater, and soil water chemistry after clear-cutting

3.2.1. Stream water (regular sampling)

The solute concentrations from regular samplings in both catchments from 1998 to 2006 are shown in Fig. 3. Streamwater anion chemistry in the clear-cut catchment was different from that in

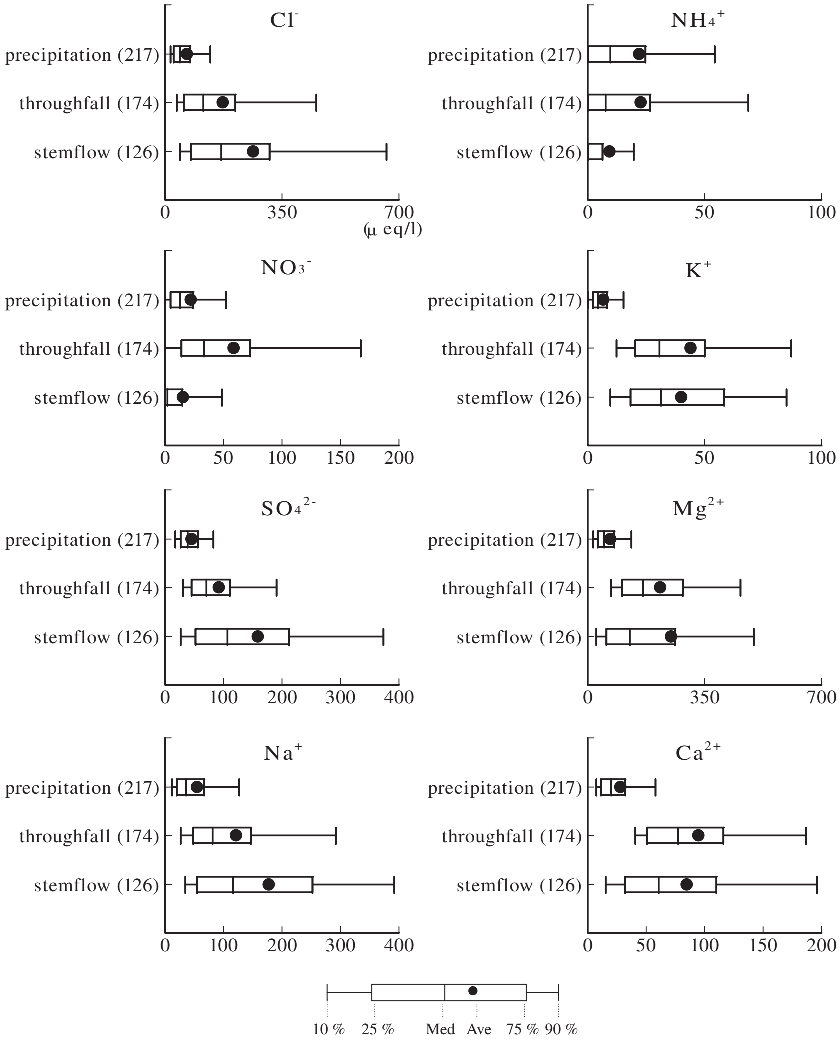

Fig. 2. Concentrations of solutes in precipitation, throughfall, and stemflow from 1998 to 2006. Values in parentheses mean the sample numbers.

the control catchment ($p < 0.01$) during pre-cutting period, whereas cation chemistry was not different from that in the control catchment ($p > 0.01$). Some factors, such as hydrological processes, can result in different streamwater chemistry in nearby catchments, but the effect of clear-cutting on streamwater chemistry was clear in this case.

In the control catchment, the solute concentrations from the period of cutting in the clear-cut catchment (1999–2006) did not change largely compared with those in the pre-cutting period (1998) ($p > 0.05$). In the clear-cut catchment, Cl^- and NO_3^- concentrations changed significantly ($p < 0.001$) after cutting. The mean Cl^- concentration before cutting was 243 μeq L^{-1} and began to decrease 1 year after cutting. The Cl^- concentrations gradually decreased for 6 years and then remained stable at approximately 100 μeq L^{-1} in 2006. The mean Cl^- concentration in the clear-cut catchment in 2006 was less than the mean concentration in the control catchment in the same period, indicating that the decline was caused by clear-cutting.

The mean NO_3^- concentration in the clear-cut catchment was 0 μeq L^{-1} before cutting, which was lower than that in the control catchment (26 μeq L^{-1}), but suddenly began to increase at 6 months after cutting and reached a peak at 280 μeq L^{-1} in April 2000 (1 year after clear-cutting). After the peak, the NO_3^- concentration decreased gradually for 6 years; however, the mean concentration in 2006 was 12 μeq L^{-1}, still larger than the pre-cutting level.

The mean SO_4^{2-} and Na^+ concentrations in the clear-cut and control catchments before the cutting period were 160 and 219 μeq L^{-1} and 318 and 373 μeq L^{-1}, respectively, and the mean concentrations in 2006 were 118 and 201 μeq L^{-1} and 256 and 354 μeq L^{-1}, respectively. This result shows that the SO_4^{2-} and Na^+ concentrations slightly decreased after cutting ($p < 0.01$). The concentrations of, K^+, Mg^{2+}, and Ca^{2+} in stream water did not change following clear-cutting ($p > 0.05$).

3.2.2. High-frequency sampling during storm events

The high-frequency sampling included storm events, although regular samplings were mainly during baseflow. Solute concentration changes during two storm events (1999 and 2000) are shown in Fig. 4. During the storm events, the NO_3^- concentration increased with discharge. Within 1 year following clear-cutting, the peak NO_3^- concentration in the clear-cut catchment was 700 μeq L^{-1} and much higher than the NO_3^- concentration in the control catchment. Concentrations of K^+, Na^+, and Ca^{2+} also increased with discharge in the clear-cut catchment, although these concentrations decreased in the control catchment. In 2000, the NO_3^- concentration at peak flow was less than 200 μeq L^{-1}, which was close to that in the control catchment. The concentration changes of other solutes during storm events were also similar to concentrations in the control catchment.

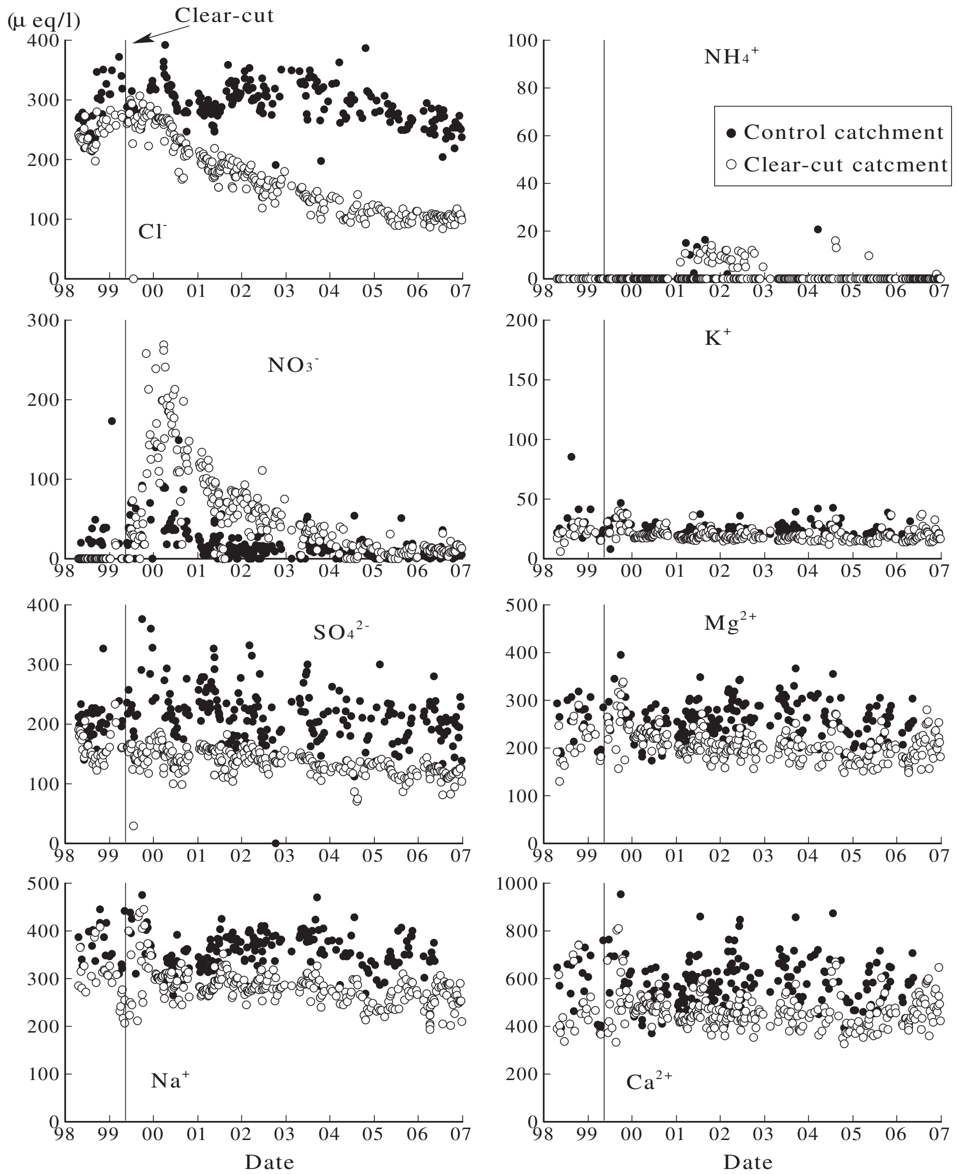

Fig. 3. Solute concentrations by regular samples of stream water in control and clear-cut catchments from 1998 to 2006.

Fig. 5 presents the relationships between solute concentration and discharge following clear-cutting in 1999, 2000, 2002, and 2006 from the high-frequency stream water sampling. In a single logarithmic plot, regression equations of all solutes were linear and constant in each period. A negative linear relationship was observed between the solute concentrations and discharge, with the exception of NO_3^-; a positive linear relationship was observed between NO_3^- concentration and discharge. In the control catchment, the relationships were similar from 1998 to 2006 (Fig. 5).

Cl^- concentrations during low flow ($Q < 0.1$ mm hr^{-1}) and high flow (1 mm hr$^{-1} < Q$) in 2000 were higher than those in 2002 ($p < 0.01$). Also, Cl^- concentrations during low flow in 2002 were also higher than in 2006 ($p < 0.01$) (Fig. 5). Low-flow Cl^- concentrations decreased gradually, and there was a similar decline in streamwater Cl^- concentrations from grab samplings. In contrast, Cl^- concentrations during high flow (1 mm hr$^{-1} < Q$) converged to 80 μeq L^{-1} in 2002, which is the same level as that in 2006. Cl^- concentrations during high flow in 2006 were not different from high-flow Cl^- concentrations in 2002 ($p > 0.05$), although Cl^- concentrations during high flow in 2000 were significantly higher than those in 2002 ($p < 0.01$). These findings showed that the relationship between discharge and Cl^- concentration changed year to year after cutting, and the Cl^- concentrations at high flow decreased and converged earlier than at low flow.

The relationship between NO_3^- concentration and discharge also changed following cutting, particularly at 8 months after cutting In the 8 months after cutting (April–December 1999), the NO_3^- concentration increased to 700 μeq L^{-1} during high flow (1 mm hr$^{-1} < Q$), which was approximately 10-fold higher than that at high flow in the control catchment; however, the mean concentration during low flow was 60 μeq L^{-1}, which was similar to the low-flow NO_3^- concentration in the control catchment ($p > 0.05$). One year after forest cutting, the concentration at high flow rapidly decreased (approximately 200 μeq L^{-1}) and then gradually arrived at the pre-cutting level in 7 years.

The relationships between discharge and concentration of SO_4^{2-} did not change significantly after cutting and were similar to those of the control catchment ($p > 0.05$). In control catchment and clear-cut catchment in the period of before cutting, Na^+, Mg^{2+}, and Ca^{2+} concentrations in stream water clearly decreased with discharge ($p < 0.05$). The concentrations of Na^+, Mg^{2+}, and Ca^{2+} increased with discharge within 1 year after cutting ($p < 0.05$), and then in 2000

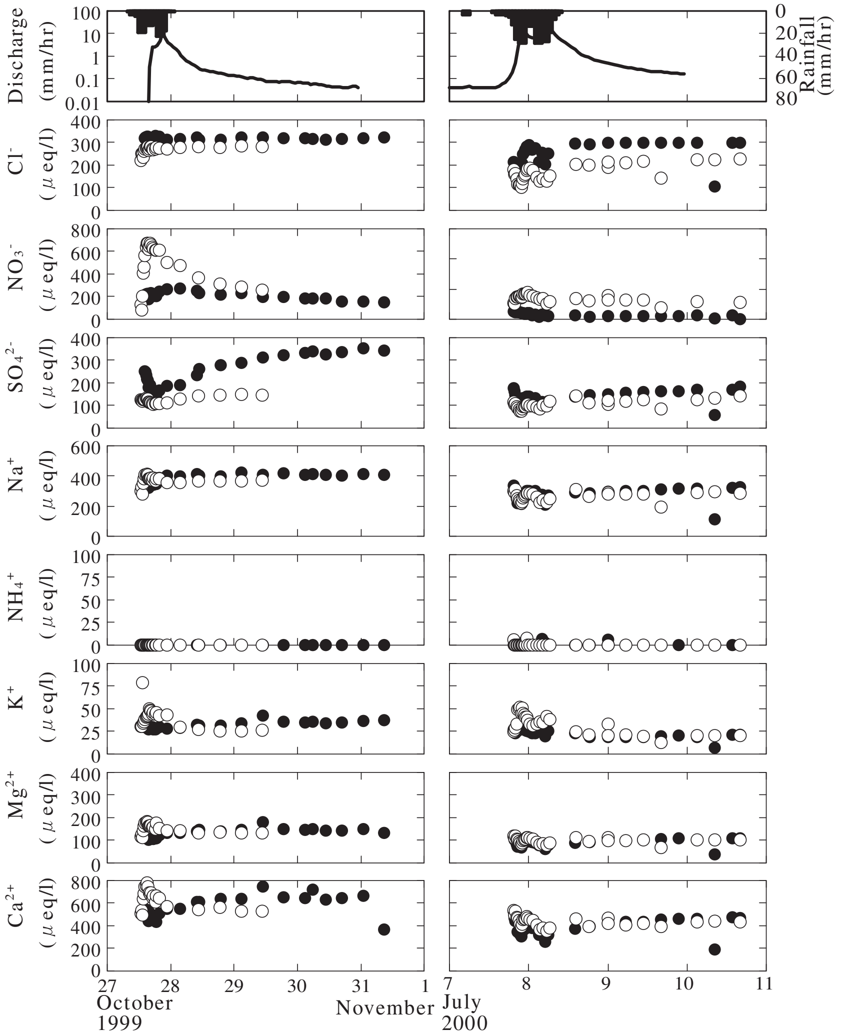

Fig. 4. Solute concentration changes shown by hydrographs for the clear-cut (○) and control (●) catchments during the storm events on October 27, 1999, and July 7, 2000.

the concentrations decreased with discharge as pre-cutting period ($p < 0.05$). Also, the relationship between K^+ concentrations and discharge changed after cutting. K^+ concentrations during high flow increased with discharge in 1999 and 2000 ($p < 0.01$), although K^+ concentrations during low flow and high flow were similar ($p > 0.05$) before cutting (Fig. 5). These results show that clear-cutting leads to the release of cations during high flow.

3.2.3. Groundwater and soil water chemistry

Concentration changes in groundwater at B6-270 and B7-150 are shown in Fig. 6. At both B6-270 and B7-150, Cl^- concentrations began to decrease 1 year after cutting. The Cl^- concentrations gradually decreased for 6 years and then remained stable at approximately 100 $\mu eq\,L^{-1}$ in 2006. These concentration changes were similar to those in stream water as shown by regular grab samplings. The NO_3^- concentration in B6-270 increased after cutting, but soon decreased after the peak, and the mean concentration in 2001 was 13.8 $\mu eq\,L^{-1}$. The NO_3^- concentration in B7-150 also increased after cutting and then decreased gradually as the stream water NO_3^- concentration changed. The changes in NO_3^- concentrations differed between the two groundwaters ($P < 0.001$). B6-270 had deeper groundwater in which denitrification occurred, and the NO_3^- concentrations might have been lower than those in B7. On the other hand, Na^+, K^+, NH_4^+, Mg^{2+}, and Ca^{2+} concentrations in groundwater did not change after cutting ($P > 0.05$).

Concentration changes in soil water in the clear-cut catchment are shown in Fig. 7. The Cl^- concentrations in soil water at the three depths gradually decreased, similar to the groundwater and stream water concentrations. The NO_3^- concentration in B6-20 cm peaked at 700 $\mu eq\,L^{-1}$ at the end of 1999 and then decreased to 200 to 300 $\mu eq\,L^{-1}$ in 2000. Soil water NO_3^- concentrations at other depths (30 and 60 cm) also increased following clear-cutting. The peak NO_3^- concentration of approximately 300 $\mu eq\,L^{-1}$ was similar to that in stream water at peak during baseflow. However, the timing of the NO_3^- peak concentration in soil water was earlier than that in the stream water during baseflow. At the time of NO_3^- peak concentration in soil water, concentrations of Na^+, K^+, Mg^{2+}, and Ca^{2+} in soil water were also significantly higher than concentrations during the succeeding period ($p < 0.001$). Conversely, soil water SO_4^{2-} concentrations in 1999 were not different compared with the succeeding period ($p > 0.05$). NO_3^- and cation concentrations in the soil water increased just after cutting, although Cl^- decreased and SO_4^{2-} concentrations did not change during the same period. Cl^- is considered conservative (Peters and Ratcliffe, 1998), and its source is mainly wet and dry deposition. Although Cl^- concentrations in soilwater decreased after cutting due to decreased inputs of dry deposition, Na^+, NO_3^-, SO_4^{2-}, K^+, Mg^{2+}, and Ca^{2+} concentrations did not change or increased soon after the cutting period in soil water and groundwater despite a decrease in dry deposition inputs. These results showed that forest cutting made

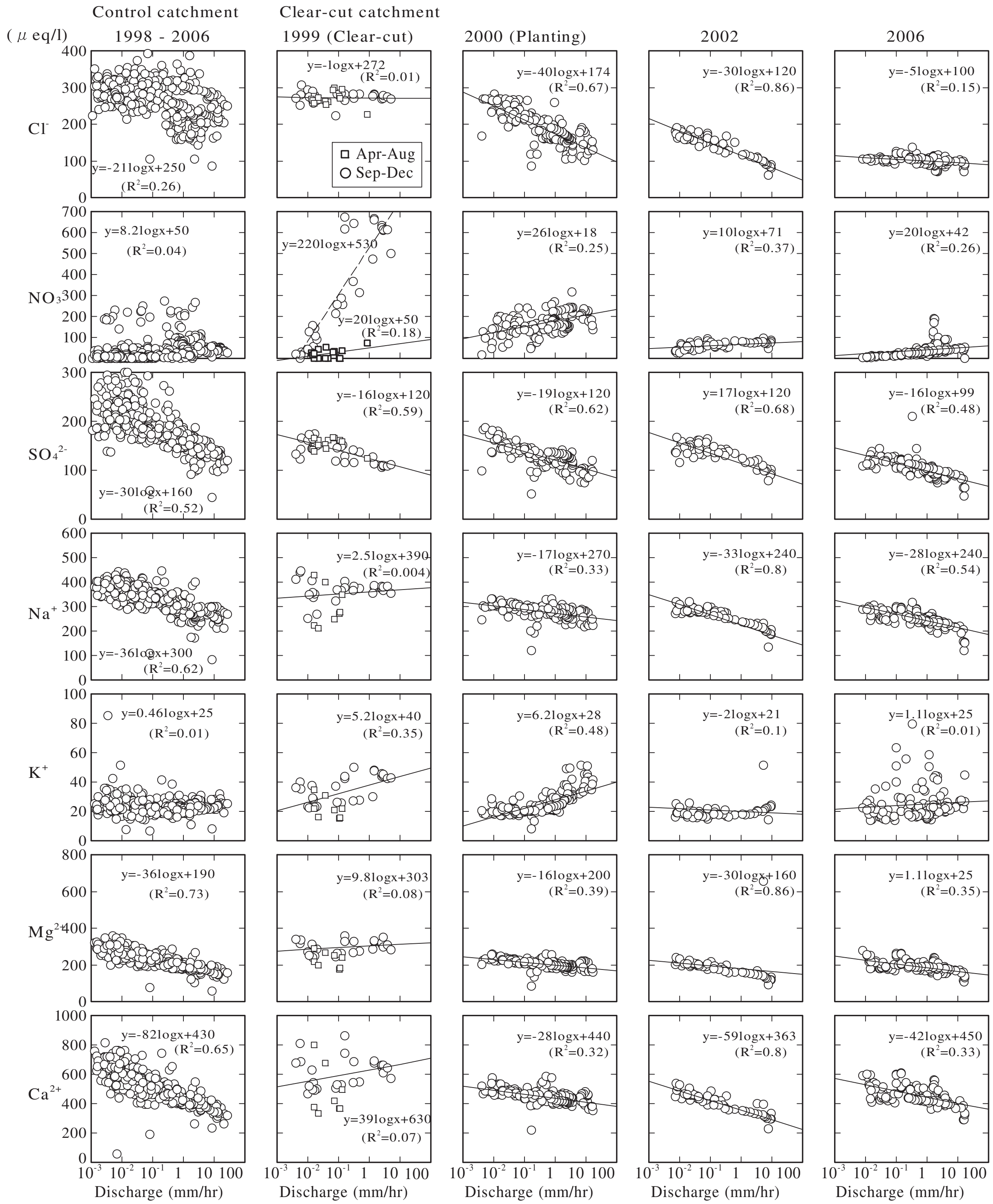

Fig. 5. The relationships between discharge and concentration in control and clear-cut catchments from 1998 to 2006. In the clear-cut catchment, the relationships in 1999, 2000, 2002, and 2006 were separately plotted.

effect on biogeochemical condition in soil, such as increase weathering-derived Na^+, K^+, Mg^{2+}, and Ca^{2+} and reduced uptake of nutrients by plants.

3.3. Change in chemical flux following clear-cutting

Following clear-cutting, the inputs in the clear-cut catchment decreased to less than half of those in the control catchment (Table 2). In particular, the mean input of K^+ largely decreased from approximately 20 to 6 kg ha^{-1} y^{-1}. The decrease may have been due to declines in dry deposition and leaching from canopy leaves.

The solute outputs were calculated by regression methods using high-frequency sampling data (Table 3). Before cutting, both catchments had similar outputs of solutes (Table 3). The outputs of solutes changed after clear-cutting, particularly for Cl^-, NO_3^-, Na^+, K^+, Mg^{2+}, and Ca^{2+}. The output of Cl^- decreased following cutting, and the difference in output between clear-cut and control catchments increased gradually, with the clear-cut catchment showing half the

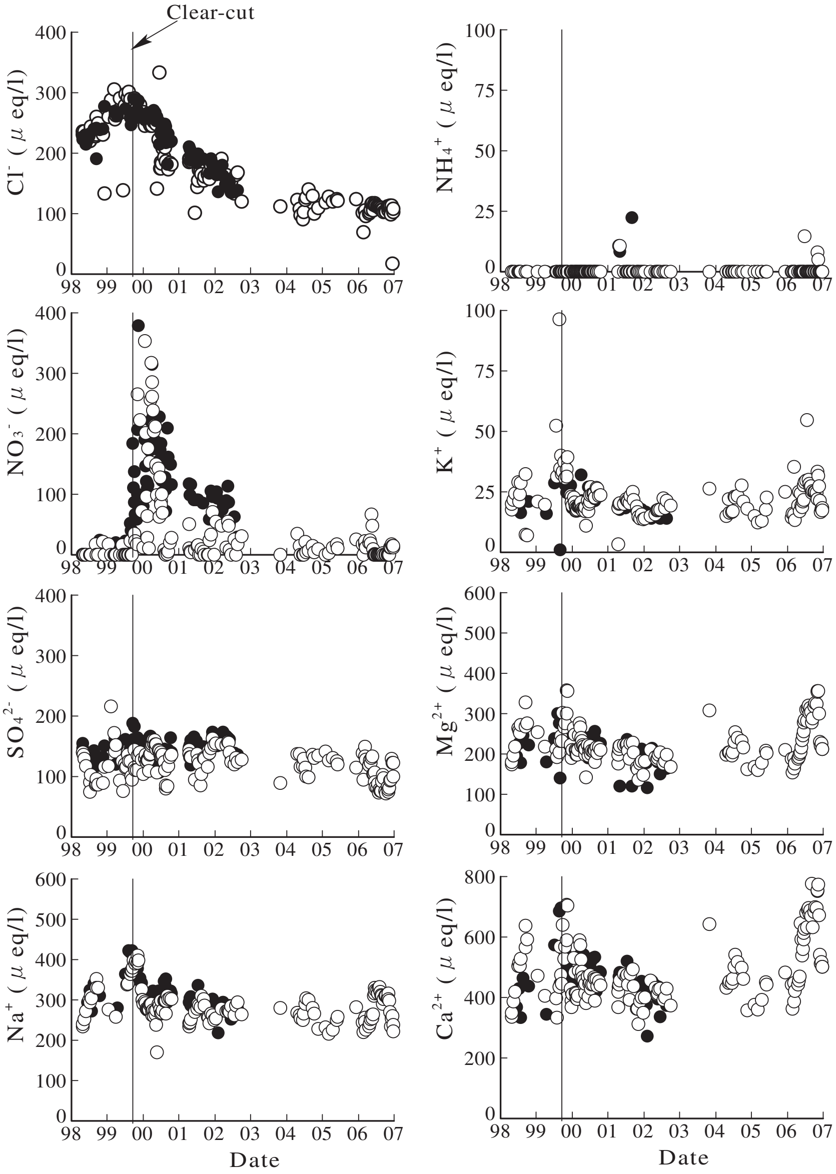

Fig. 6. Solute concentrations of groundwater at B6-270 (○) and B7-150 (●) in the clear-cut catchment from 1998 to 2006.

output of the control catchment in 2006. Output of NO_3^- increased rapidly following forest cutting. In particular, the NO_3^- output within 3 years after cutting was three to four times larger than that of the control catchment. The exports of Na^+, K^+, Mg^{2+}, and Ca^{2+} also increased approximately two-fold compared with those of the control catchment in the 8 months after cutting, and the outputs were similar to or slightly higher than those of the control catchment in the following years.

Output fluxes calculated by the regression method using high-frequency sampling data were different from the fluxes calculated by only regular sampling data in clear-cut and control catchments (Figs. 8 and 9). The differences between high-frequency samplings and regular samplings were mainly found in the export of NO_3^-, K^+, Na^+, Mg^{2+}, and Ca^{2+} just after clear-cutting. The difference in NO_3^- export between these methods was large just after forest cutting; the mean difference in annual export of NO_3^- was more than 20 kg ha^{-1} during the 3 years after cutting. In particular, the largest difference in NO_3^- between the two methods was found 8 months after cutting; the difference was 40 kg ha^{-1} y^{-1} and the estimate by the high-frequency sampling method was three-fold larger than that by the regular grab sampling method (Fig. 8). Outputs of K^+, Na^+, Mg^{2+}, and Ca^{2+} calculated by high-frequency data were larger in the first year after cutting, although in the control and clear-cut catchments during the period before cutting, the outputs of these cations were lower than calculated by regular sampling data sets.

4. Discussions

4.1. Differences in solute concentrations and exports between regular and frequent samplings

Chemical exports calculated by high-frequency samplings were different from exports calculated by regular grab samplings. In the control catchment, exports estimated by high-frequency samplings were higher than those by regular grab samplings in NO_3^-, and lower in other solutes. Some previous studies reported similar results (Schleppi et al., 2006; Ide et al., 2007). However, in the clear-cut catchment, chemical exports calculated from high-frequency sampling were significantly different from those calculated by regular grab samplings following clear-cutting. Within 1 year after cutting, clear differences were found in NO_3^-, K^+, Na^+, Mg^{2+}, and Ca^{2+} exports between sampling methods. In particular, the NO_3^- export

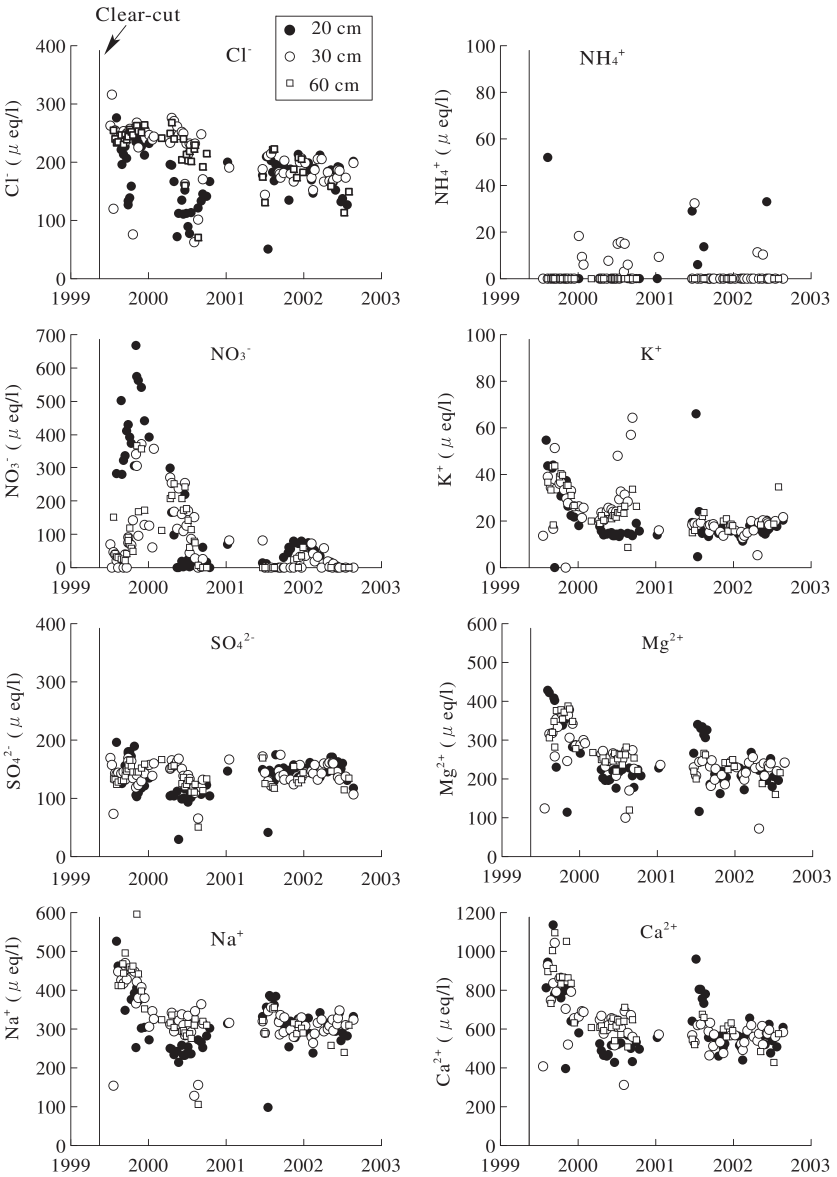

Fig. 7. Solute concentrations of soil water at B6-20 cm, B6-30 cm, and B6-60 cm in the clear-cut catchment from 1999 to 2002.

Table 2
Input of nutrients in control and clear-cut catchments in 1998, 1999, 2000, 2002, and 2006.

	Rainfall (mm)	Cl^- (kg ha^{-1} yr^{-1})	NO_3^-	SO_4^{2-}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}
Control									
1998/4–1999/4	2324	151.8	55.7	66.9	40.0	14.6	13.5	8.5	16.7
1999/4–1999/12	1606	104.9	38.5	46.2	27.6	10.1	9.3	5.9	11.5
2000	2151	70.0	41.0	52.1	32.8	5.9	20.1	8.0	22.1
2002	2363	140.9	56.2	81.2	72.8	3.4	29.0	15.5	28.4
2006	2780	90.5	31.3	63.1	46.5	9.8	33.7	11.3	32.7
Clear-cut									
1998/4–1999/4	2324	151.8	55.7	66.9	40.0	14.6	13.5	8.5	16.7
1999/4–1999/12	1606	28.9	14.0	21.9	19.0	4.2	2.0	2.2	4.6
2000	2151	38.8	16.7	35.6	21.6	10.0	4.3	4.4	12.2
2002	2363	56.8	17.8	40.2	30.8	8.4	3.5	5.3	9.6
2006	2780	62.5	18.5	37.4	36.1	3.6	6.7	7.1	15.5

calculated by high-frequency samplings was three times higher than that calculated by regular sampling following clear-cutting.

The difference was attributable to high flushing of NO_3^- just after cutting. This study showed that during high flow, the stream water NO_3^- concentration was 100-fold higher than that during low flow in 1999 (Fig. 4). The high release of NO_3^- during storm flow was found only by the high-frequency sampling. The NO_3^- concentration during high flow was close to the soil water NO_3^-

Table 3
Output of nutrients in control and clear-cut catchments in 1998, 1999, 2000, 2002, and 2006.

	Rainfall (mm)	Discharge (mm)	Cl^- (kg ha^{-1} yr^{-1})	NO_3^-	SO_4^{2-}	Na^+	K^+	Mg^{2+}	Ca^{2+}
Control									
1998/4-1999/4	2324	910	79.0	17.9	72.6	62.5	8.0	22.2	81.8
1999/4-1999/12	1606	696	57.5	20.8	50.4	44.7	6.2	15.5	57.1
2000	2151	768	65.1	28.1	58.2	51.0	6.8	17.8	65.8
2002	2363	895	76.3	13.7	68.6	59.8	7.9	21.0	77.5
2006	2780	1448	119.2	37.4	103.2	92.3	12.9	31.7	117.2
Clear-cut									
1998/40-1999/4	2324	867	60.4	6.7	54.8	64.3	6.6	21.1	82.1
1999/4-1999/12	1606	860	82.9	59.6	51.4	71.0	12.7	31.5	107.8
2000	2151	996	64.1	107.6	59.0	62.9	10.5	24.3	88.2
2002	2363	1147	50.0	49.3	67.0	63.3	9.2	25.1	85.2
2006	2780	1589	55.8	44.3	73.3	86.3	15.7	35.0	139.3

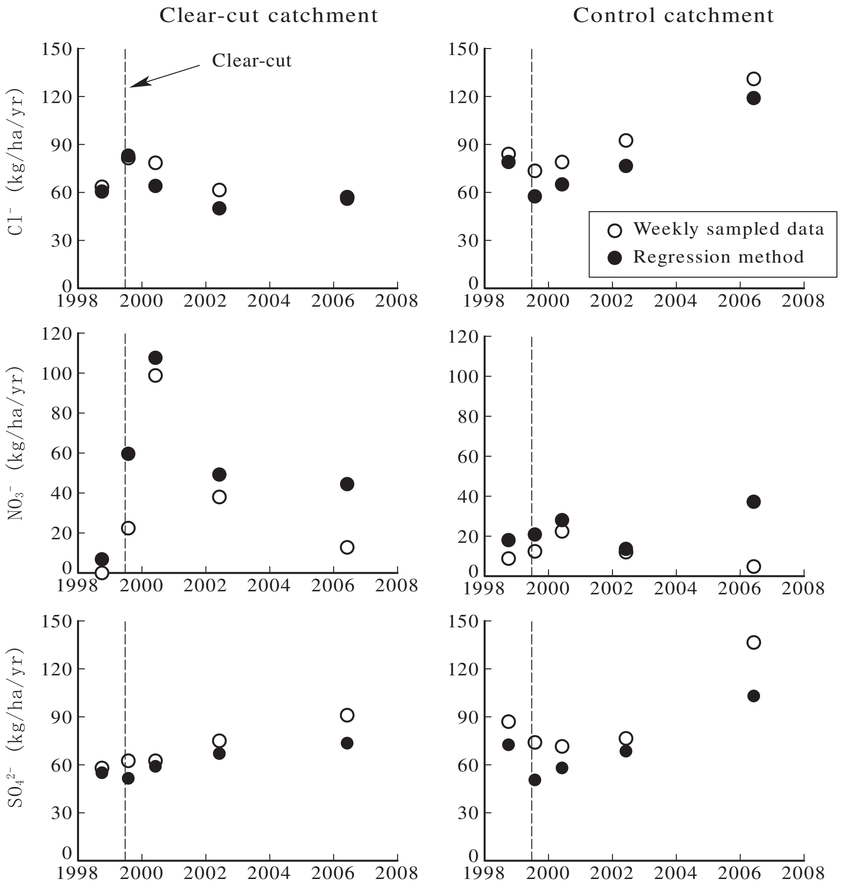

Fig. 8. Difference in output fluxes of Cl^-, NO_3^-, and SO_4^{2-} between the estimation methods (○ = calculated by regular sampling data, and ● = regression method using high-frequency sampling data) from 1998 to 2006 in control and clear-cut catchments.

concentration at B6 (Fig. 7). The concentration change was similar to that in stream water during baseflow and groundwater. This indicates that the source of NO_3^- during baseflow was groundwater, and the source of flushing NO_3^- during storm flow just after clear-cutting was mainly soil water. Increased NO_3^- in soil water after clear-cutting has been reported by many studies and has been attributed to biological mineralisation of organic matter, combined with reduced nutrient uptake because of the death of root systems (Likens et al., 1970; Bormann and Likens, 1979; Cummins and Farrell, 2003; Neal et al., 1998; Feller, 2005). These results also indicate that clear-cutting has a significant impact on N flushing because a large amount of NO_3^- accumulated in the upper soil layer and the NO_3^- was transported to stream water with lateral flow during storm events.

The high-frequency sampling data also showed increases in exported K^+, Na^+, Mg^{2+}, and Ca^{2+} within 1 year after clear-cutting, while the regular grab samplings showed no export changes in these solutes after cutting. These increases in K^+, Na^+, Mg^{2+}, and Ca^{2+} exports within 1 year after clear-cutting were also attributable to increases in flushing exports during storm events. The increases in Na^+, K^+, Mg^{2+}, and Ca^{2+} concentrations during high flow were associated with large NO_3^- release during high flow after cutting. Flushing concentrations increased because of the transportation of accumulated cations with NO_3^- from the upper soil layer to stream water during storm events. The slight increases in Na^+, K^+, Mg^{2+}, and Ca^{2+} concentrations and exports were due to increased nitrification in the upper soil layer following forest cutting. The NO_3^- produced by nitrification is accompanied by H^+ ions,

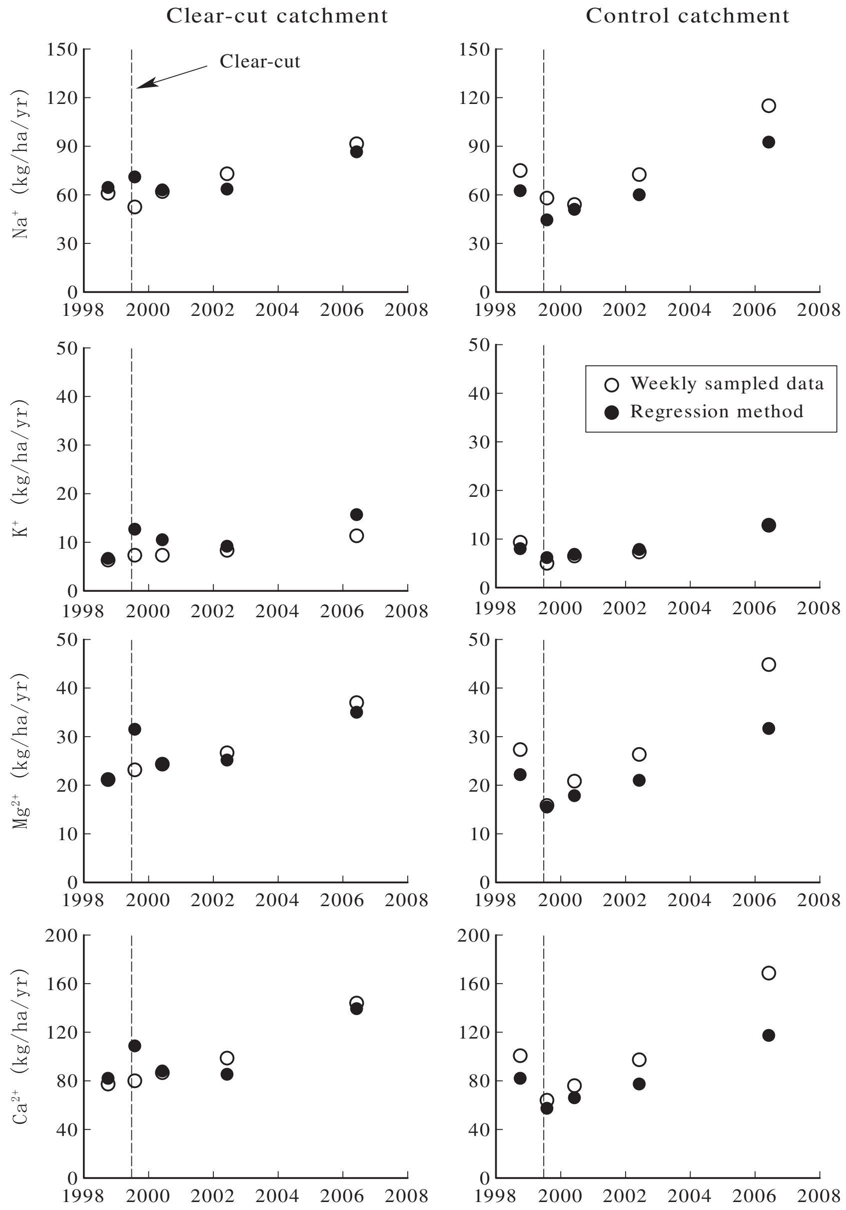

Fig. 9. Difference in output fluxes of Na^+, K^+, Mg^{2+}, and Ca^{2+} between the estimation methods (○ = calculated by regular sampling data, and ● = regression method using high-frequency sampling data) from 1998 to 2006 in control and clear-cut catchments.

and thus nitrification also influences the weathering rates and leaching of other cations from soil (Lawrence et al., 1987; Feller, 2005). The fact that soil water concentrations of these cations and NO_3^- increased during the same period (Fig. 7) supports this hypothesis.

The difference in exports of Cl^- and SO_4^{2-} calculated by high-frequency sampling was small compared with that calculated by regular grab sampling following cutting. There were small differences in the concentrations of these solutes between soil water and groundwater following cutting. Cl^- is considered to be conservative (Peters and Ratcliffe, 1998), and its source is mainly wet and dry deposition. Therefore, the decrease in input after cutting (Fig. 2) had an effect on the difference in concentrations between soil water and groundwater in the period after cutting. In 2000 and 2002, the Cl^- exports calculated by high-frequency samplings were smaller (Fig. 8); in this period, soil water had been replaced by low-Cl^--concentration water, but groundwater had not been replaced by low-concentration water. However, the difference in the Cl^- concentration between soil water and groundwater was small because the increase in the concentration is only in the evaporative concentration. The source of SO_4^{2-} was weathering rock, and the concentrations in soil water and groundwater were stable.

These high-frequency sampling results indicate that catchment hydrological processes, such as water source or direct flow rates during storm flow, affect chemical export following cutting. For example, in these study catchments, the direct flow rate was more than 90% (Maita et al., 2005) and the impact of export during storm flow was larger than that during baseflow. In such high-flush catchments, high-frequency sampling is important for accurate estimation of chemical export and evaluation of the influence of forest cutting.

4.2. Hydrological effect on NO_3^- export following clear-cutting

This study evaluated the dynamics of NO_3^- flushing during high flow and subsequent long-term high release of NO_3^- during baseflow following clear-cutting. High-frequency samplings showed that high flushing of NO_3^- during high flow occurred within 1 year after cutting, which was before the NO_3^- concentration peak during baseflow in stream water, and then the NO_3^- concentration during high flow rapidly decreased after 2000. The NO_3^- flushing following cutting may have been due to transportation of shallower soil water NO_3^- with lateral flow during storm events. The soil water NO_3^- concentration increased due to promoted nitrification and lack of plant uptake. The subsequent decrease in the stream water NO_3^- concentration during high flow was caused by the decrease in the soil water NO_3^- concentration with the recovery of plant uptake after planting in April 2000. In general, the more rapidly an area revegetates after clear-cutting, the more rapidly the uptake of N is renewed (Martin et al., 2000; Feller, 2005; Mou et al., 1993). The planting within 1 year after cutting may have had a significant effect on the rapid recovery of N uptake and restriction of NO_3^- release to stream water.

Furthermore, the time lag between nitrification due to disturbance and NO_3^- loss in stream water could have been caused by the flux of water from surface soils to deeper soil horizons (Vitousek et al., 1982). In this study, the time lags between clear-cutting and the NO_3^- flushing peak and subsequent baseflow NO_3^- concentration change could have been due to water infiltration processes. The time lag was reasonable for the stream water Cl^- concentration change after cutting, which may represent the residence time of water in the catchment because Cl^- is conservative and can serve as a tracer (Peters and Ratcliffe, 1998; Oda et al., 2009). The time lag was approximately 1 year between clear-cutting and the stream Cl^- concentration decrease during baseflow (Fig. 3) and may show the time for infiltration through the soil layer to groundwater.

The subsequent long-term high NO_3^- concentration period during baseflow was also affected by infiltration processes, such as the groundwater residence time, because there is less bioactivity under the root zone than in the shallower soil layer. The high NO_3^- concentration in soil water infiltrated vertically, and then the groundwater NO_3^- concentration gradually increased after cutting. The NO_3^- concentration in groundwater then gradually decreased because low-concentration water infiltrated from the upper layer and mixed with the groundwater after planting. The stream water NO_3^- concentrations of regular samples increased 1 year after cutting and gradually decreased for 5 to 6 years (Fig. 3). This result agreed with those of previous studies that reported that the increase begins approximately 1 year after a disturbance and continues for some 3 years, with later values declining slowly to premanipulation levels (Neal et al., 1998; Cummins and Farrell, 2003; Gundersen et al., 2006). Compared with the long-term period of decline in the stream NO_3^- concentration after the peak and the stream water Cl^- decline in the clear-cut catchment, the time of decline in the NO_3^- concentration during baseflow was similar to the Cl^- concentration decline; however, the NO_3^- concentration decline was earlier than the Cl^- concentration decline. The small difference indicated that the time lag for the post-peak decrease in the stream water NO_3^- concentration was mainly caused by hydrological processes, but was also influenced by biological effects such as denitrification in the groundwater. The rapid decrease in the deeper groundwater (B6-270) NO_3^- concentration to approximately $0\ \mu eq\ L^{-1}$ after the increase in the NO_3^- concentration (Fig. 6) supports the significance of the denitrification effect on NO_3^- concentration changes. These findings suggest that the timing of NO_3^- release and the period of high NO_3^- exports were affected by the residence time of water in the catchment.

5. Conclusion

High-frequency monitoring of stream water chemistry following clear-cutting during storm flow in addition to weekly grab sampling provides valuable insights into solute cycles in catchments and solute transport through catchments.

The trend in the relationship between discharge and concentration showed that concentrations at high flow, rather than at low flow, changed significantly after cutting. High release of NO_3^- in high-flow stream water just after clear-cutting indicated that high levels of NO_3^- were accumulated in the shallow soil. The trends in Cl^- and NO_3^- concentrations at high and low flows following clear-cutting also suggest that hydrology, such as residence time through the soil water and groundwater, controls the timing of flushing NO_3^- release from the upper soil layer during storm events and the slower release of N to the stream over the year or over succeeding years. High NO_3^- flushing also caused a high release of cations, such as Na^+, K^+, Mg^{2+}, and Ca^{2+}. These results were obtained only by the high-frequency observations. When monitoring stream water chemistry following cutting during baseflow and storm flow, the annual mean NO_3^- export after cutting was 1.5-fold greater than that calculated by only weekly sampling data. This study showed that high NO_3^- release during high flow just after cutting would be missed if water was collected only during low flow. The frequent sampling data during storm events was important to accurately estimate the export of solutes during the immediate post-cutting period and during high-precipitation periods. This study also showed that both hydrological processes (water sources and residence time) contributed to the time lag of the stream water change after cutting, and is important to accurately forecast the impact of forest cutting on nutrient export.

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References

Binkley, D., Brown, T.C., 1993. Forest practices as nonpoint sources of pollution in North-America. Water Resources Bull 29, 729–740.

Bormann, F.H., Likens, G.E., 1979. Pattern and Process in a Forested Ecosystem. Springer-Verlag, New York, Inc..

Burns, D.A., Murdoch, P.S., 2005. Effects of a clearcut on the net rates of nitrification and N mineralization in a northern hardwood forest, Catskill Mountains, New York, USA. Biogeochemistry 72, 123–146.

Cirmo, C.P., McDonnell, J.J., 1997. Linking the hydrologic and biogeochemical controls of nitrogen transport in near-stream zones of temperate-forested catchments: A review. Journal of Hydrology 199, 88–120.

Creed, I.F., Band, L.E., Foster, N.W., Morisson, I.K., Nicolson, J.A., Semkin, R.S., Jefferies, D.S., 1996. Regulation of nitrate N release from temperate forests: a test of the N flushing hypothesis. Water Resources Research 32, 3337–3354.

Cummins, T., Farrell, E.P., 2003. Biogeochemical impacts of clearfelling and reforestation on blanket-peatland streams II. Major ions and dissolved organic carbon. Forest ecology and management 180, 557–570.

Feller, M.C., 2005. Forest harvesting and streamwater inorganic chemistry in western North America: A review. Journal of the American Water Resources Association 41 (4), 811–875.

Gundersen, P., Schmidt, I.K., Raulund-Rasmussen, K., 2006. Leaching of Nitrate from temperate forests – Effects of air pollution and forest management. Environmental Review 14, 1–57.

Hashimoto, S., Suzuki, M., 2004. The impact of clear-cutting on soil temperature: a comparison between before and after cutting, and between clear-cut and control sites. Journal of Forest Research 9, 125–132. doi:10.1007/s10310-003-0063-x.

Hornberger, G.M., Bencala, K.E., McKnight, D.M., 1994. Hydrological controls on dissolved organic carbon during snowmelt in the Snake River near Montezuma, Colorado. Biogeochemistry 25, 147–165.

Hotta, N., Kayama, T., Suzuki, M., 2007. Analysis of suspended sediment yields after low impact forest harvesting. Hydrological processes 21, 3565–3575.

Ide, J., Nagafuchi, O., Chiwa, M., Kume, A., Otsuki, K., Ogawa, S., 2007. Effects of discharge level on the load of dissolved and particulate components of stream nitrogen and phosphorus from a small afforested watershed of Japanese cypress (*Chamaecyparis obtusa*). Journal of Forest Research 12 (1), 45–56.

Inamdar, S.P., Leary, N.O., Mitchell, M.J., Riley, J.T., 2006. The impact of storm events on solute exports from a glaciated forested watershed in western New York, USA. Hydrological Processes 20 (16), 3423–3439.

Kirchner, J.W., Feng, X., Neal, C., Robson, A.J., 2004. The fine structure of water-qualoty dynamics: the (high-frequency) wave of future. Hydrological Processes 18, 1353–1359.

Kuraji, K., Tanaka, Y., Tanaka, N., Karakama, I., 2001. Generation of stemflow and chemistry in a mature Japanese cypress forest. Hydrological Processes 15, 1967–1978.

Lawrence, G.B., Fuller, R.D., Driscoll, C.T., 1987. Release of Aluminium following whole-tree harvesting at the Hubbard Brook Experimental Forest, New Hampshire. Journal Environmental Quality 16, 383–390.

Likens, G.E., Bormann, F.H., Johnson, N.M., Fisher, D.W., Pierce, R.S., 1970. Effects of forest cutting and herbicide treatment on nutrient budgets in Hubbard Brook Watershed-Ecosystem. Ecological Monographs 40, 23–47.

Maita, E., Suzuki, M., Ohta, T., 2005. Change in the annual discharge after clear-cutting a 70-year-old mixed plantation of Sugi and Hinoki in a Tertiary Formation Area. Journal of the Japanese Forest Society 87, 124–132 (in Japanese and English summary).

Maita, E., Suzuki, M., 2007. Change in the monthly water yield after clear-cutting in Fukuroyamasawa, Chiba. Journal of the Japanese Forest Society 89 (4), 278–287 (in Japanese and English summary).

Martin, C.W., Hornback, J.W., Likens, G.E., Buso, D.C., 2000. Impacts of intensive harvesting on hydrology and nutrient dynamics of northern hardwood forests. Canadian Journal of Fisheries and Aquatic Sciences 57, 19–29.

McHale, M.R., McDonnell, J.J., Mitchell, M.J., Cirmo, C.P., 2002. A field-based study of soil water and groundwater nitrate release in an Adirondack forested watershed. Water Resources Research 38, 1031–1047.

Mou, P., Fahey, T.J., Huges, J.W., 1993. Effects of soil disturbance on vegetation recovery and nutrient accumulation following whole-tree harvesting of a northern hardwood ecosystem. *Journal of* Applied Ecology 30, 661–675.

Neal, C., Reynolds, B., Wilkinson, J., Hill, T., Neal, M., Hill, S., Harrow, M., 1998. The impacts of conifer harvesting on runoff water quality: a regional study foe Wales. Hydrology and Earth System Sciences 2, 323–344.

Oda, T., Asano, Y., Suzuki, M., 2009. Transit time evaluation using a chloride concentration input step shift after forest cutting in a Japanese headwater catchment. Hydrological Processes 23, 2705–2713.

Peters, N.E., Ratcliffe, E.B., 1998. Tracing hydrologic pathways using chloride at the Panola Mountain Research Watershed, Georgia, USA, *Water, Air*. And Soil Pollution 105, 263–275.

Reynolds, B., Stevens, P.A., Hughes, S., Parkinson, J.A., Weatherley, N.S., 1995. Stream chemistry impacts of conifer harvesting in Welsh catchments. Water Air Soil Pollution 79, 147–170.

Rosen, K., Aronson, J., Eriksson, H.M., 1996. Effects of clear-cutting on streamwater quality in forest catchments in central Sweden. Forest Ecology and Management 83, 237–244.

Rusjan, S., Brilly, M., Mikos, M., 2008. Flushing of nitrate from a forested watershed: An insight into hydrological nitrate mobilization mechanisms through seasonal high-frequency stream nitrate dynamics. Journal of Hydrology 354, 187–202.

Schleppi, P., Walder, P.A., Stahli, M., 2006. Errors of flux integration methods for solutes in grab samples of runoff water, as compared to flow-proportional sampling. Journal of Hydrology 319, 266–281.

Shiraki, K., Liu, R.G., Karakama, I., Shuin, Y., Ohta, T., 1999. Water balance in Fukuroyamasawa Experimental Watershed in University forest in Chiba, The university of Tokyo. Bulletin of Tokyo University Forest 102, 71–86 (in Japanese and English summary).

Swank, W.T., Vose, J.M., Elliott, K.J., 2001. Long-term hydrologic and water quality responses following commercial clearcutting of mixed hardwoods on a southern Appalachian catchment. Forest Ecology and Management 143, 163–178.

Tanaka, N., Kuraji, K., Shiraki, K., Suzuki, M., Suzuki, M., Ohta, T., Suzuki, M., 2005. Throughfall, stemflow and rainfall interception at mature *Cryptomeria japonica* and *Chamaecyparis obtuse* stands in Fukuroyamasawa watershed. Bulletin of Tokyo University Forest 113, 197–240 (in Japanese and English summary).

Vitousek, P.M., Gosz, J.R., Grier, C.C., Mellilo, J.M., Reiners, W.A., 1982. A comparative analysis of potential nitrification and nitrate mobility in forested ecosystems. Ecological Monograph 52, 155–178.

Wang, X., Burns, D.A., Yanai, R.D., Briggs, R.D., Germain, R.H., 2006. Changes in stream chemistry and nutrient export following a partial harvest in the Catskill Mountains, New York, USA. Forest Ecology and Management 223, 103–112.