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Hydrobiogeochemistry of forest ecosystems in Japan: major themes and research issues

Nobuhito Ohte,[1]* Naoko Tokuchi,[1] Hideaki Shibata,[2] Maki Tsujimura,[3] Tadashi Tanaka[3] and Myron J. Mitchell[4]

[1] *Graduate School of Agricultural Sciences, Kyoto University, Kyoto, Japan*
[2] *North Research Station, Hokkaido University Forests, Faculty of Agriculture, Hokkaido University, Hokkaido, Japan*
[3] *Institute of Geoscience, University of Tsukuba, Tsukuba, Japan*
[4] *College of Environmental Science and Forestry, State University of New York, New York, USA*

Abstract:

Hydrobiogeochemical information on forested watersheds in the Japanese archipelago is reviewed to qualitatively summarize the major factors affecting the hydrology and biogeochemistry of this region. Major features that distinguish Japanese forest watersheds from those of north central Europe and the northeast United States generally include higher temperature, greater precipitation and steeper topography. There have been three major themes in hydrobiogeochemistry research on Japanese forest ecosystems: (1) investigations of nutrient cycles with particular emphasis on the establishment and maintenance of forest ecosystems; (2) evaluations of streamwater chemistry as an output from the forest ecosystem; and (3) hydrological studies using biogeochemical tracers. High precipitation inputs during the growing season affect the seasonality of the streamwater NO_3^- concentration, resulting in different temporal patterns than those generally found in north central Europe and the northeast United States. The high alkalinity and pH of Japanese surface waters is due to the rapid weathering of relatively young soils that are generated by steep hillslopes. Warm temperatures and elevated soil moisture enhance high rates of mineral weathering. Hydrological studies using biogeochemical tracers have shown that the steep topography contributes to the highly heterogeneous movement of water within Japanese catchments. Copyright © 2001 John Wiley & Sons, Ltd.

KEY WORDS hydrology; biogeochemistry; forest ecosystem; Japan

INTRODUCTION

Geographic characteristics of Japanese forests

The Japanese archipelago extends from 45 °N, 150 °E to 2 °N, 122 °E. The climate is relatively humid and classified from Df (humid continental or subarctic) to Cfa (humid subtropical and hot summer) (Trewartha, 1968). The relative differences in temperature between the northern and southern parts are greater than the relative differences in precipitation, with highest inputs during the summer. The Japanese archipelago (except Hokkaido Island) is affected by the Asian monsoon system that results in the 'Baiu' rainy season from mid-June to mid-July. Typhoons hit the archipelago several times a year, mostly in late August and September. Lands facing the Sea of Japan and the mountainous regions in central Japan are covered by heavy snow in winter. The monthly precipitation and average air temperatures for the Moshiri experimental watersheds (Shibata, unpublished data), the Kiryu experimental watershed (Laboratory of Forest Hydrology, Kyoto University, unpublished data), the Hubbard Brook Experimental Forest (Long Term Ecological Research, 1998) and the Coweeta Hydrologic Laboratory (Swank and Vose, 1997; Long Term Ecological Research, 1998) are provided in Figure 1. The annual water budgets for these sites are summarized in Table I. The climates of Moshiri

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*Correspondence to: N. Ohte, Division of Environmental Science and Technology, Graduate School of Agricultural Sciences, Kyoto University, Kyoto 606-8502, Japan. E-mail: nobu@bluemoon.kais.kyoto-u.ac.jp

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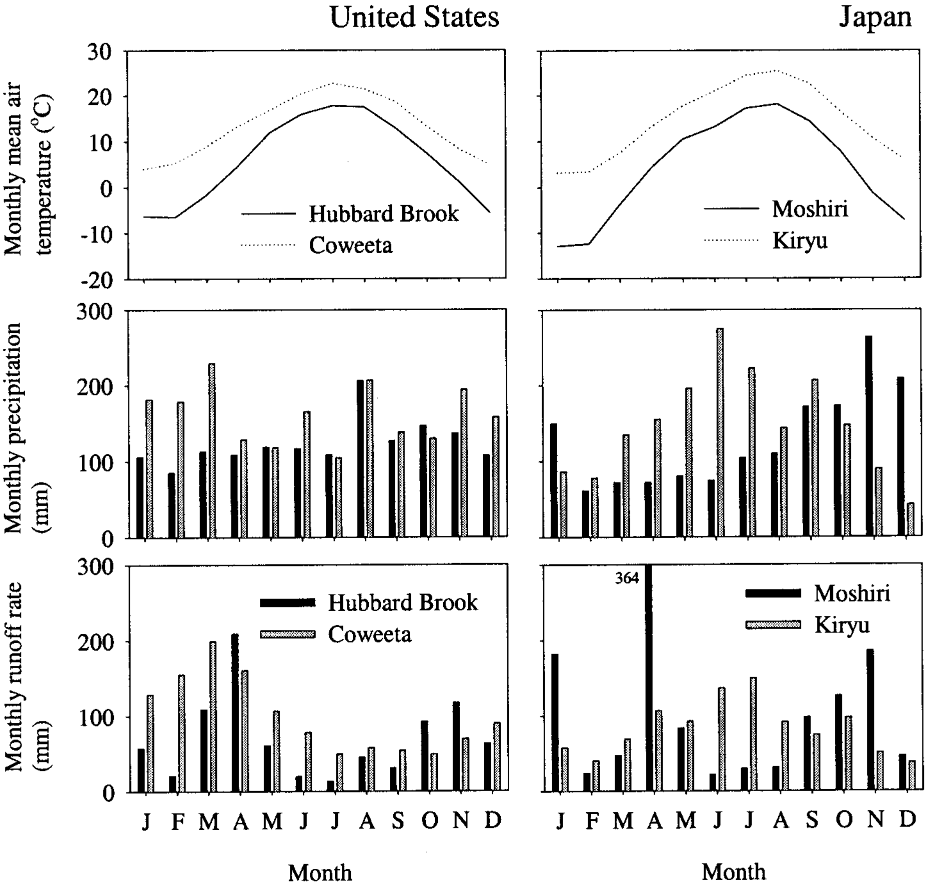

Figure 1. Seasonal variation in precipitation, air temperature and runoff rate of the Moshiri Experimental Watershed in northern Japan (1998, Shibata, unpublished data), the Kiryu Experimental Watershed in central Japan (averaged 1997–1999, Laboratory of Forest Hydrology, Kyoto University, unpublished data), the Hubbard Brook Experimental Forest (WS6) in the northeast United States (averaged 1991–1995, Long Term Ecological Research, 1998) and the Coweeta Hydrologic Laboratory (WS18) (averaged 1991–1995, Long Term Ecological Research, 1998; for runoff data, averaged 1990–1994, Swank and Vose, 1997)

and Kiryu are typical of the northern and central to southern parts of the Japanese archipelago, respectively. Although latitude, mean annual air temperature and annual precipitation at Moshiri and Kiryu are similar to those at Hubbard Brook and Coweeta, respectively (Table I), marked differences are found in seasonal patterns of precipitation and stream discharge.

In the northeast United States seasonal changes in precipitation are rather small but discharge rates are at a minimum during the summer for catchments both with (e.g. Hubbard Brook) and without (e.g. Coweeta) snowmelt. In marked contrast, in watersheds in central and southern Japan, highest discharge rates occur during the summer growing season due to high precipitation inputs. The evapotranspiration rate in Kiryu is almost twice that of Hubbard Brook. Common forest types in Japan include coniferous and broadleaf evergreen forests (Figure 2). The forest types are primarily distributed according to temperature regime.

The Japanese archipelago is located at the junction of four major tectonic plates (Eurasian, Philippine Sea, North American and Pacific Ocean), resulting in high orogenic activity. Most of the Japanese archipelago is dominated by the Quaternary orogeny (UNESCO, 1976) and the local topography is undulating and dissected, with steep hillslopes being common. Kaizuka (1969) noted that the rate of orogenic uplift and average denudation in the Japanese archipelago is 10 times greater than continental regions not affected by

Table I. Location, annual mean air temperature and water budget of the Moshiri Experimental Watershed, the Kiryu Experimental Watershed, the Hubbard Brook Experimental Forest (WS6) and the Coweeta Hydrologic Laboratory (WS18)

	Longitude (deg. min)	Latitude (deg. min)	Elevation (range in m)	Mean annual air temperature (°C)	Annual precipitation (mm year^{-1})	Annual discharge (mm year^{-1})	Precipitation − discharge (mm year^{-1})	Period of record
Japan								
Moshiri	142°00′E	43°46′N	290–540	2·5	1540	1240	300	1998[a]
Kiryu	135°59′E	34°58′N	200–265	14·3	1776	1004	772	1997–1999[b]
United States								
Hubbard Brook (WS6)	71°45′W	43°56′N	490–775	5·5	1289	837	452	1991–1995[c]
Coweeta (WS18)	83°25′W	35°03′N	726–993	13·1	1933	1198[d]	735	1991–1995[c]

[a] Unpublished data by Shibata.
[b] Unpublished data by Laboratory of Forest Hydrology, Kyoto University.
[c] Long Term Ecological Research (1998).
[d] Averaged 1990–1994 by Swank and Vose (1997).

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Figure 2. Potential natural vegetation map of Japan. Redrawn from Sidei and Kira (1977). (Reproduced by permission of the University of Tokyo Press.)

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the Quaternary orogeny. Denudation rates in orogenic regions also increase with an increase in local relief and slope (Ruxton and McDougall, 1967). Several studies in the orogenic mountains of Japan (Shimokawa, 1984; Iida, 1993) suggest that the return period of the slope failure ranges from 10^2 to 10^3 years.

Surface erosion and landslides in forested watersheds in Japan retard the development of mature soil profiles and may help maintain the high base status of the soils. The majority of Japanese forest soils are cambisols (FAO-UNESCO, 1974) that are characterized by low amounts of weathered minerals and high base saturation.

Themes in hydrobiogeochemical research in Japan

There are three major research themes in the hydrobiogeochemistry of Japanese forest ecosystems. The first and historically most important theme has been the study of nutrients important for establishing and maintaining forest ecosystems, including analyses of C and N relationships. The biogeochemical studies of forest ecosystems in Japan have also included international efforts such as the International Biological Program (IBP) from 1967 to 1972 (Shidei and Kira, 1977).

The second major theme has been investigation of the hydrobiogeochemistry of surface waters. There has been some overlap in the research on surface waters and forest biogeochemistry (e.g. Iwatsubo and Tsutsumi, 1968; Kawasoe and Yoshimoto, 1981; Ohte *et al.*, 1995). In the early 1980s, not only the scientific community, but also the Japanese public recognized acid rain as a severe environmental problem. Interest in acid rain has provided additional impetus for hydrobiogeochemical research. Many field experiments and catchment-scale hydrochemical studies have been conducted to evaluate the impacts of acidic deposition on forest ecosystems (Satake, 1999).

The third theme has been the use of biogeochemical tracers for understanding hydrological processes. The first symposium for isotope utilization in hydrology was held in Tokyo by the International Atomic Energy Administration (IAEA) in 1963, and tracer use for hydrology research in Japan began in 1970 (Tanaka, 1982). In the 1990s, stable isotopic tracer techniques have been used extensively in the analyses of hillslope hydrology (e.g. Matsutani *et al.*, 1993; Tsujimura and Tanaka, 1996; Tanaka and Ono, 1998; Tsujimura *et al.*, 1999).

In this paper we review the hydrobiogeochemical studies in Japanese forest ecosystems and compare these results with some findings from Europe and North America. By focusing on the unique characteristics of the climatic and geologic features of Japan, we will evaluate how these factors affect hydrobiogeochemistry. This review will focus on the three major themes of hydrobiogeochemical research in Japan outlined above.

BIOGEOCHEMICAL PROCESSES AND NUTRIENT CYCLING

Studies on nutrient cycling in forest watersheds

Japanese soil chemists and soil biologists have studied forest nutrient cycling, emphasizing the evaluation of N dynamics. During the IBP the flow and pools of nutrients were analysed in subpolar to warm temperate and subtropical forests on the Japanese archipelago (Shidei and Kira, 1977). Information on N dynamics in forest ecosystems was compiled for representative forest types in central Japan. Investigations were carried out on broadleaf deciduous forests at Ashiu, Kyoto, mixed broadleaf deciduous and evergreen forests at Kamigamo, Kyoto and evergreen coniferous forests at Kiryu, Ohtsu (Tsutsumi, 1962; Iwatsubo and Tsutsumi, 1967, 1968; Kawahara *et al.*, 1968; Tsutsumi *et al.*, 1968; Harada *et al.*, 1969; Kawahara, 1971; Katagiri and Tsutsumi, 1975, 1976, 1978). Studies of the role of mineral weathering, including reactions both within and outside the rooting zone, have also been conducted (Tsutsumi, 1989; Iwatsubo, 1996).

Iwatsubo and Tsutsumi (1968) evaluated nutrient cycles in a forested watershed in Kyoto. The total input of NO_3^-–N was 2·7 kg ha^{-1} $year^{-1}$ and the output 0·3 kg ha^{-1} $year^{-1}$ (averaged during 1965–1967). In contrast, for undisturbed watersheds at the Hubbard Brook Experimental Forest (HBEF), the annual input of NO_3^-–N was 19·7 kg ha^{-1} $year^{-1}$ and the output 17·1 kg $ha^{-1}year^{-1}$ (averaged during 1963–1974; Likens *et al.*, 1977). These fluxes were much higher than those found for later periods at the HBEF. For example, from 1992–1993 for Watershed 6, input was 7·3 kg ha^{-1} year^{-1} and output was 0·66 kg ha^{-1} year^{-1} (Likens and Bormann, 1995). These results suggest that in Kyoto during the 1960s, N input from atmospheric deposition was low, although the site was near urban and industrial areas that might be expected to be sources of N deposition.

Landscape scale heterogeneity within a catchment

During the 1980s, several field experiments evaluated nutrient budgets of catchments (Haibara and Aiba, 1982; Arimitsu, 1982). Since the late 1980s, interest has shifted towards evaluating the spatial heterogeneity and/or distributions of N dynamics (Takeda, 1994). The interpretation of the spatial variation of the N dynamics within steep topography has often emphasized how the distribution of soil water influences hydrological processes, with distinct differences in the soil moisture conditions between upper and lower hillslope positions. Tokuchi *et al.* (1993) and Ohte *et al.* (1997) conducted *in situ* lysimeter experiments with controlled moisture

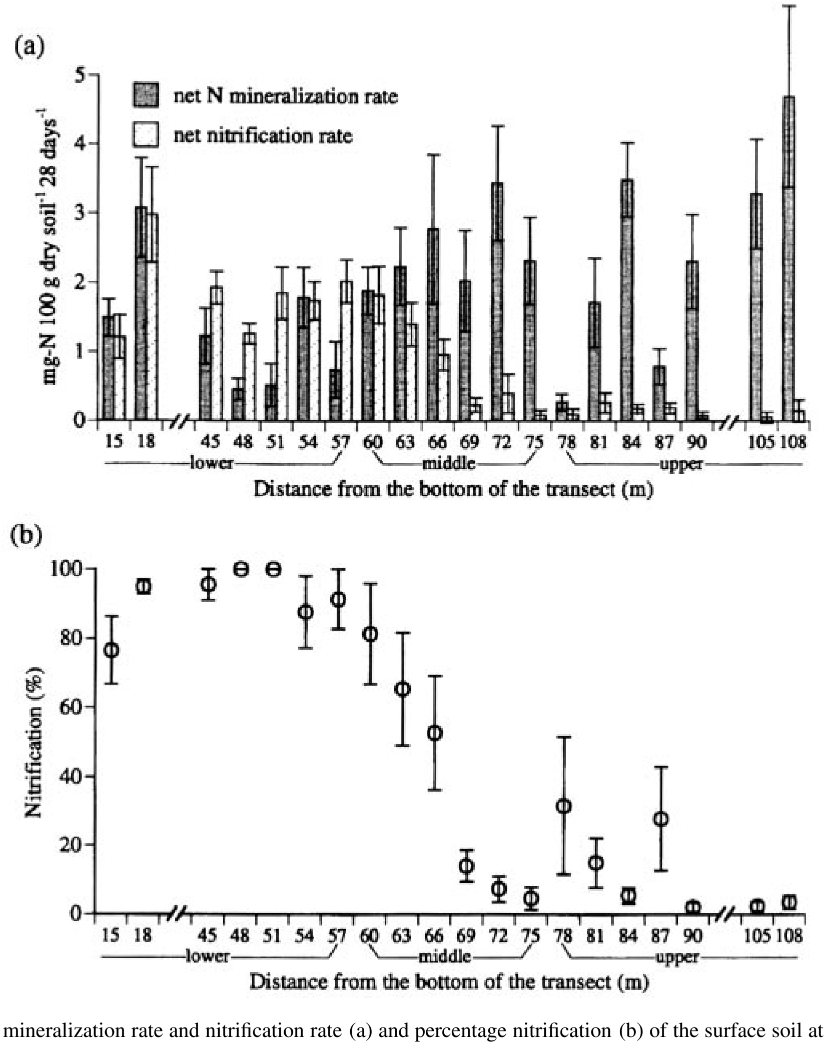

Figure 3. Net nitrogen mineralization rate and nitrification rate (a) and percentage nitrification (b) of the surface soil at 20 sampling locations on a slope in a *Cryptomeria japonica* D. Don plantation in central Japan (Hirobe *et al.*, 1998). (Reprinted from Hirobe *et al.*, *European Journal of Soil Biology*, **34**. Hirobe M, Tokuchi N, Iwatsubo G, Spatial variability of soil nitrogen transformation patterns along a forest slope in a *Cryptomeria japonica* D. Don plantation, 123–131. Copyright 1998, with permission from Elsevier Science.)

content in a coniferous forest and confirmed the presence of the optimum moisture conditions for nitrification. Hirobe *et al.* (1998) reported that the spatial distribution of the nitrification potential along the hillslope was strongly influenced by the C/N ratio in addition to the soil moisture gradient (Figure 3).

Many other studies focusing on N dynamics along the hillslope have been conducted in coniferous plantations (Toda *et al.*, 1987; Kutsuna *et al.*, 1988; Toda *et al.*, 1991; Toda and Haibara, 1994; Takahashi *et al.*, 1994; Enoki *et al.*, 1997). Takeda (1994) proposed a conceptual model that explained how the spatial distribution of decomposition processes and the N dynamics along a slope control plant biodiversity and soil types.

Nitrogen dynamics and variations in flow paths in the riparian zone are important for controlling N drainage losses from catchments (Cirmo and McDonnell, 1997; Ohrui and Mitchell, 1998b). Konohira *et al.* (1997a,b) and Koba *et al.* (1997) used information on ^{15}N values of soil solutions, subsurface water and streamwater to show that the inorganic N leached from the soil layer at the hillside was denitrified in the riparian zone. Koba *et al.* (1997) showed that temporal patterns of denitrification corresponded to seasonal variation of the groundwater level. The applications of the stable isotopes to N dynamics, such as the measurement of the gross nitrification and the natural abundance of ^{15}N in soil profiles in forest ecosystems, was advanced by soil biologists in Japan during the 1990s (Koba *et al.*, 1998; Tokuchi *et al.*, 1998a).

Nitrogen saturation of Japanese forests

Recently there has been increasing attention focused on N saturation, especially in North America and Europe (Aber *et al.*, 1989; Stoddard, 1994). In Japan atmospheric input of N is elevated, especially in urban areas. Several studies have reported that the dominant anion in Japanese precipitation is NO_3^- rather than SO_4^{2-} (Figure 4; Tonooka, 1997), despite substantial SO_2 emission inputs from east Asia (Table II). Air pollutants generated in Japan also contribute to acidic deposition (Katoh *et al.*, 1990; Nashimoto, 1992; Igawa *et al.*, 1998; Hatakeyama, 1999; Maruta *et al.*, 1999). Although there are many other potential causes for forest decline in Japan, it has been suggested that air pollution is a contributing factor (Okita, 1996). Studies on the causal relationship between ozone concentration and forest decline have been conducted near Tokyo (e.g. Hatakeyama and Murano, 1996; Wakamatsu, 1996). However, the visible evidence of forest decline resulting from disturbances of N cycling due to excessive atmospheric N inputs and freezing reported elsewhere (Nihlgård, 1985; Fincher *et al.*, 1989; Thornton *et al.*, 1994) have not been found in Japan.

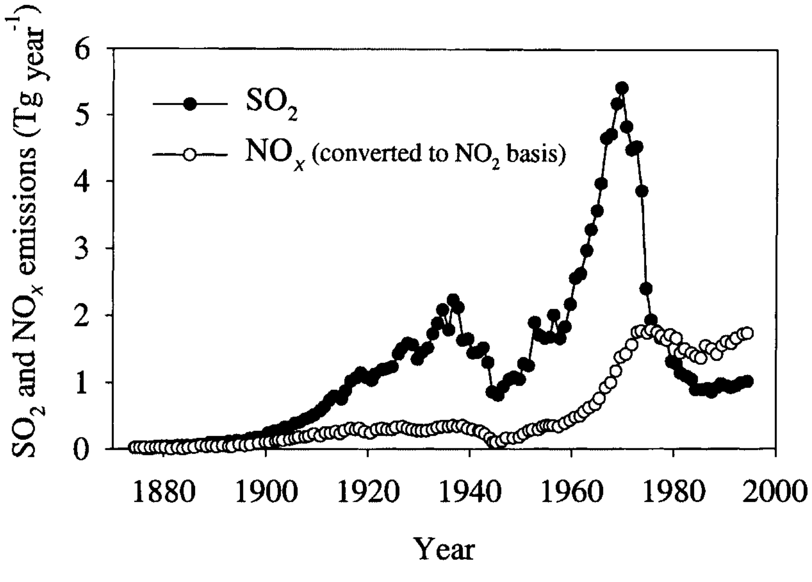

Figure 4. Anthropogenic SO_2 and NO_x emissions and heat consumption in Japan since 1874 (Tonooka, 1997). (Reproduced by permission of Chuohoki Publishing Co. Ltd.)

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Table II. Anthropogenic SO_2 and NO_x emissions in eastern Asia 1990 (Department of Global Environment, 1997)

Region	SO_2 emission (Gg year^{-1})	NO_x emission (Gg year^{-1})
Japan	989	1602
China	20 951	6722
Taiwan (1991)	583	599
South Korea	1611	925
North Korea	676	0
Russia (1989)	247	89
Mongolia	62	40
Total	25 119	9977

Note: Estimated values do not include emissions by air and marine crafts.

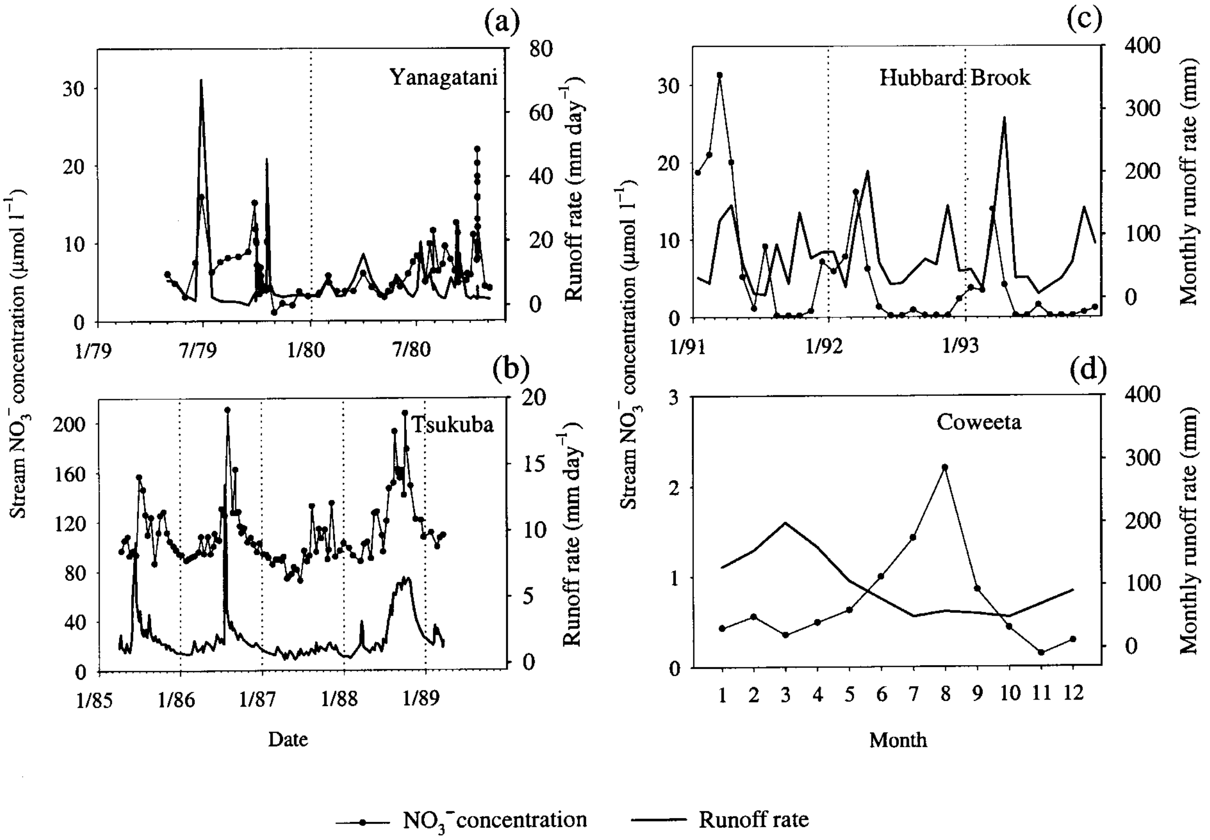

Figure 5. Seasonal variations in stream NO_3^- concentrations and runoff rate at forested watersheds. (a) Yanagatani watershed in western Japan (Igaki, 1983); (b) Tsukuba watersheds in eastern Japan (Hirata and Muraoka, 1993); (c) Hubbard Brook Experimental Forest in the northeast United States (based on Mitchell *et al.*, 1996); (d) Coweeta Hydrologic Laboratory (NO_3^- concentrations are mean monthly values from 1972 to 1984, runoff data are averaged from 1990 to 1994; Swank and Vose, 1997)

Although many forested watersheds in Japan show high levels of N retention, some catchments near metropolitan Tokyo exhibit high N drainage losses (Mitchell *et al.*, 1997). For these catchments, atmospheric inputs of inorganic N exceeded 10 kg N ha^{-1}year^{-1} and their N output by drainage water exceeded inputs in bulk deposition. Mitchell *et al.* (1997) also pointed out that the seasonality in stream NO_3^- of Japanese watersheds is generally characterized by high concentrations in summer through early fall. This pattern was observed in watersheds over a wide range of atmospheric inputs and drainage losses (Igaki, 1983 (Yanagatani in Figure 5a); Hirose *et al.*, 1988; Tokuchi *et al.*, 1998b; Hirata and Muraoka, 1993 (Tsukuba in Figure 5b); Baba and Okazaki, 1998).

In contrast, watersheds in the northeast United States generally show a marked decrease in stream NO_3^- concentrations during the growing season, and high concentration during spring snowmelts (Stoddard, 1994; Mitchell *et al.*, 1996 (Hubbard Brook in Figure 5c)). Despite the differences in seasonality of NO_3^- loss between the northeast United States and Japan, both regions generally show a positive relationship between discharge rate and NO_3^- concentration (e.g. Tokuchi *et al.*, 1993; Ohrui *et al.*, 1995 for the Japanese watersheds; McHale *et al.*, 2000 for the northeast United States). For Japanese watersheds, the two factors which contribute to the high NO_3^- losses during the growing season include: (1) high rates of N mineralization and nitrification in the warm, wet summer that generate high concentrations of NO_3^- in forest floor and mineral soil water; and (2) high precipitation inputs in the summer that result in movement of N solutes to surface waters from both soil water and groundwater. Subsurface flow can transport dissolved N from the forest floor and soil to surface waters when the groundwater level is elevated during periods of high precipitation inputs (e.g. Muraoka and Hirata, 1988; Katsuyama *et al.*, 2000). A similar mixing concept was used by Kato *et al.* (1995) to explain the changes of spring NO_3^- concentration at the headwater catchment in the region near Tokyo.

A different seasonal pattern in stream NO_3^- concentration has been found at Coweeta in the southeast United States (Swank and Vose, 1997) (Figure 5d). Although the lowest flow rate occurs during the summer as for the other sites in the eastern United States (Figure 1), this low flow coincides with the highest NO_3^- concentrations which are substantially less than typically found in the northeast United States. These higher concentrations may be due to high rates of N mineralization and nitrification under warm climate conditions in summer, or possibly contributions from groundwater as found in the Catskill mountains (Burns *et al.*, 1998).

Stoddard (1994) suggested that high concentrations and small seasonal changes in NO_3^- concentration might be indicative of an advanced stage of N saturation. Some watersheds ~100 km from metropolitan Tokyo have high NO_3^- concentrations (>100 $\mu mol_c\ l^{-1}$) and do not exhibit seasonality in stream NO_3^- (Ohrui and Mitchell, 1997). Using mixing analysis, Ohrui and Mitchell (1999) suggested that bedrock fissures were a major contributor to stream water under baseflow conditions. Although the contribution of soil water having higher NO_3^- concentration than groundwater increased stream NO_3^- concentration during high discharge, the NO_3^- concentration of groundwater was always >100 $\mu mol_c\ l^{-1}$. Sites with high atmospheric N inputs and high rates of N mineralization may be especially likely to produce groundwaters with high NO_3^- concentrations that contribute NO_3^- to surface waters. Burns *et al.* (1998) also suggested the importance of groundwater contribution to NO_3^- concentrations in a Catskill mountain stream in the United States, and showed that the high NO_3^- groundwater from bedrock fractures consistently contributes to sustain the stream NO_3^- concentration in the summer.

STREAMWATER CHEMISTRY

Acid rain and hydrobiogeochemistry

Before the 1980s, most Japanese forest hydrologists focused on the physical aspects of water movement. Although several studies used chemical tracers (e.g. Nakamura, 1971; Tanaka *et al.*, 1984), biogeochemical relationships were not evaluated (e.g. Iwatsubo and Tsutsumi, 1968). Researchers in Japan have recently recognized the importance of integrating hydrological and biogeochemical measurements within forested catchments, especially for understanding and predicting the effects of acid rain.

The geographical distribution of acid rain has recently been described (1989–1997) (Figure 6). The mean pH of the precipitation is similar to that found in high atmospheric deposition regions of Europe and North America (Department of Global Environment, 1997). Studies of the impact of acid rain on forest ecosystems

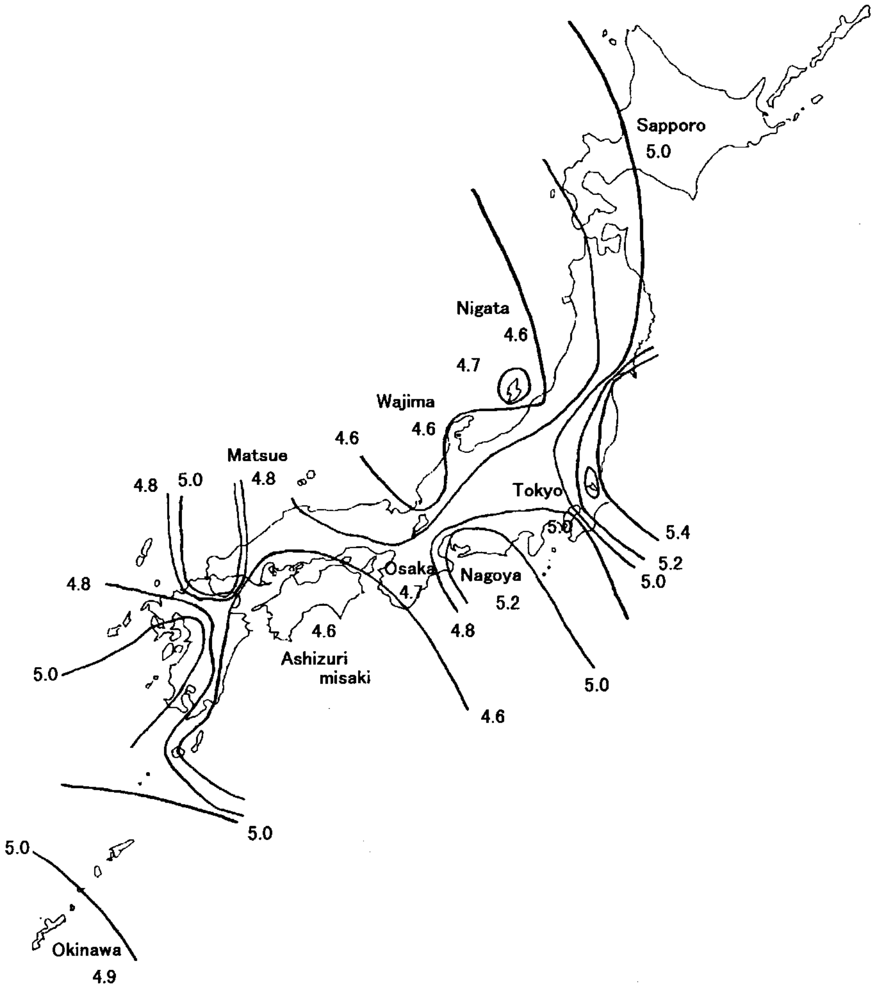

Figure 6. Geographical distribution of rainwater pH in Japan. Contour lines are drawn using annual average data at 46 points covering the Japanese archipelago. Monitoring by the National Environmental Agency of Japan has been conducted since 1989 at 29 points, with 17 points added in 1993. Averaging was done using the data before 1996. The averaging period at each point is detailed in Department of Global Environment (1997)

in Japan can be categorized into three areas: (1) effects of acidic substances in wet and dry depositions on plant physiology; (2) effects on forest decline through soil acidification; and (3) factors affecting freshwater acidification and the evaluation of acid neutralization at the watershed scale (Satake, 1999). Several examples of forest decline have been reported in the suburban forest of the Kanto district (near Tokyo, northeast Japan). Some researchers have confirmed a causal relationship with atmospheric pollutants (Katoh *et al.*, 1990; Nashimoto, 1992). Igawa *et al.* (1998), Hatakeyama (1999) and Maruta *et al.* (1999) suggested the ozone effects on foliar damage at high elevation forests were similar to those found for red spruce in the eastern United States. Igawa *et al.* (1998) also reported high nitrate concentrations in acidic fog water in the same region.

The measurements of acid buffering capacity and estimation of critical loads have been conducted using experimental approaches such as the application of artificial acid rain to soil columns by Japanese scientists (e.g. Ishizuka *et al.*, 1991; Sato and Ohkishi, 1993). Using various forest soils in Japan, they evaluated the rapid acid-neutralizing capacity which is mainly regulated by exchange reactions, and suggested that there were

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some regions exhibiting high sensitivity of the surface soil to acidic deposition. Currently in Japan, however, there has been little indication that acidic deposition has contributed to the acidification of surface waters. However, it has been suggested by the Japanese Department of Global Environment (1997) that freshwater acidification might become a problem at current levels of deposition. Particular attention needs to be given to evaluating the relationship between geochemistry and catchment buffering potential (Satake, 1999).

Catchment scale studies on hydrobiogeochemistry

The number of case studies on the hydrobiogeochemistry of catchments has substantially increased since the 1980s. In the relatively cold regions such as Hokkaido and the coastal regions of the Japan Sea, investigations have focused on the chemical changes of streamwater during snowmelt. Japanese researchers have evaluated the effects of mobile anions on the flushing of solutes from the forest floor during snowmelt (Sato *et al*., 1992; Suzuki, 1995a,b), including investigations on Hokkaido Island (e.g. Nakagawa and Iwatsubo, 1995; Kobayashi *et al*., 1999; Nakagawa and Iwatsubo, 1999). Soils on Hokkaido are generally andosols dominated by volcanic ash with high concentrations of base cations. Therefore, the streamwater alkalinity of these regions is higher than for acid-sensitive regions of Europe and the northeast United States that have climates similar to Hokkaido (Ohte and Tokuchi, 1999). Effects of acid snow brought by seasonal wind from the northwest in winter have received considerable attention in the communities in the coastal regions of the Japan Sea. The International Congress of Acid Snow and Rain (Niigata University, Niigata, Japan) discussed the effect of acid snow on the environment (Aoyama *et al*., 1997).

In the cool and warm temperate regions of Japan, catchment scale experiments have focused on how the variations of forest types and geology affect hydrobiogeochemistry (e.g. Fukushima and Matsushige, 1995; Ikeda and Miyanaga, 1995). Ohrui *et al*. (1995) and Ohrui and Mitchell (1996, 1998a) discussed the effects of the variations of dominant tree species (broadleaf deciduous or evergreen conifer) and age variation of planted coniferous forests. Asano *et al*. (1998, 1999, 2000) examined the changes in proton budget that accompany forest succession. Several researchers have examined the spatial and temporal variations of solutes along hydrological pathways, such as the unsaturated/saturated zones and vertical/lateral water movement (Muraoka and Hirata, 1988; Shimada *et al*., 1993; Ohte *et al*., 1991a, 1995; Ohte and Tokuchi, 1997; Ohrui and Mitchell, 1999). Shimada *et al*. (1993) demonstrated that SiO_2 in streamwater of a granitic headwater catchment in central Japan is generated mainly during vertical infiltration through the unsaturated soil layer rather than by contact with bedrock.

Evaluations of the buffering of forested catchments

Monitoring of stream and river water chemistry has also been conducted by geochemists to provide elemental mass balances (e.g. Yoshioka *et al*., 1984; Yoshioka and Itoh, 1985; Hirose *et al*., 1994; Sakurai *et al*., 1998). Since the beginning of the 1990s, more emphasis has been placed on evaluating the contribution of chemical weathering in relatively shallow soil layers (e.g. Ikeda and Miyanaga, 1995; Ohte *et al*., 1995; Ohrui and Mitchell, 1998a). Proton budget estimates have been used to quantify the linkage between buffering processes and elemental cycling in forest ecosystems (Nanbu *et al*., 1994; Shibata *et al*., 1998a,b; Tokuchi and Ohte, 1998; Asano *et al*., 1999). Tokuchi and Ohte (1998) compared the proton budgets of Japanese forested catchments with those of Europe and the northeast United States. They concluded that the dominant processes in Japanese watersheds were linked to N transformations and cation uptake by plants rather than inputs from outside the ecosystem such as were found for some watersheds in Europe and the northeast United States.

Sato and Takahashi (1996) demonstrated that the cation/Si ratios of solution in laboratory weathering experiments were the same as those of streamwater, suggesting that acid buffering potential was provided primarily by mineral weathering without cation losses from the exchange sites. Using climatological indexes, Hirose *et al*. (1988) and Nakagawa and Iwatsubo (1999) summarized the geographical variations of inorganic ion concentrations of streamwater in 34 forested watersheds in Japan. The pH of all sites ranged from 6·7 to 7·4 with some indications of episodic acidification. In the seasonally snow covered region, Suzuki (1995b)

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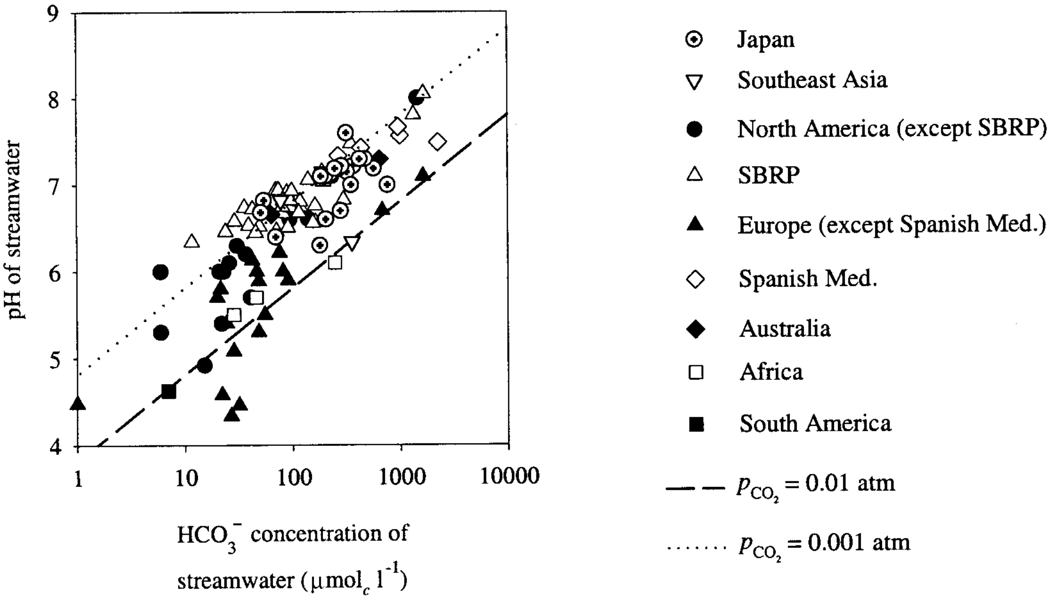

Figure 7. Relationship of HCO$_3^-$ concentration and pH of streamwater of the world vegetated catchments. The diagonal dotted lines indicate the theoretical relationship for constant pCO$_2$ that is derived by the equilibrium equation of the dissolution reaction of CO$_2$. SBRP: Southern Blue Ridge Province. Med.: Mediterranean. Site descriptions and references are listed in Ohte and Tokuchi (1999)

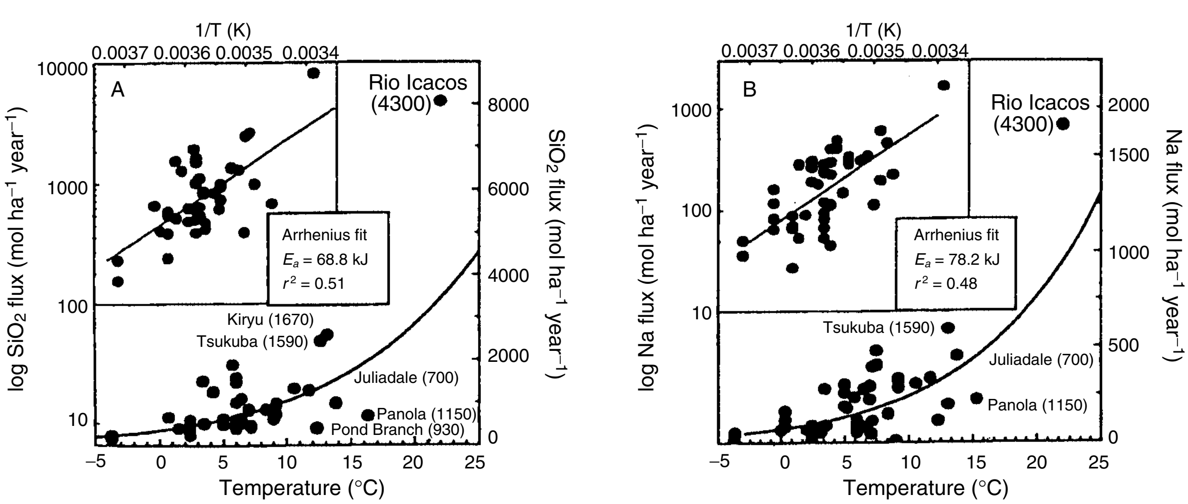

Figure 8. Relationships between watershed SiO$_2$ and Na weathering fluxes, and the annual mean air temperature. Inserts are Arrhenius plots of logarithmic values of fluxes vs. 1/[temperature (K)]. Diagonal lines correspond to linear regression fits to the equation of Arrhenius relationship: $r_T = Ae^{(-Ea/RT)}$ with activation energies and correlation coefficients indicated. Corresponding plots of data on linear scales are shown in the outer portions of the figures, with indicated annual precipitation (mm) for selected watersheds (White and Blum, 1995). (Reprinted from *Geochimica et Cosmochimica Acta*, **59**. White AF, Blum AE, Effects of climate on chemical weathering in watersheds, 1729–1747. Copyright 1995, with permission from Excerpta Medica Inc.) Kiryu and Tsukuba are Japanese catchments. Rio Icacos, Panola and Pond Branch are located in the United States. Juliadale is a Rhodesian catchment

showed that stream pH decreased from ~6·5 in summer to 6·0 during the snowmelt period in a region 130 km from the Tokyo metropolitan area. This decrease in pH was associated with an increase of NO$_3^-$ and SO$_4^{2-}$ concentrations in stream water. Although Baba and Okazaki (1998), working near Tokyo, found decreases of stream pH with increased NO$_3^-$ concentration during the growing season, the pH was always >6·4 during their five-year study. The pH and alkalinity of surface waters in the Japanese archipelago are generally higher than those found in many catchments in Europe and the northeast United States (Figure 7). White and Blum (1995) indicated that the weathering rates in the Japanese forest watersheds are extremely high. They emphasized the effects of precipitation and temperature as controlling factors of weathering for watersheds underlain by granitic rock types (Figure 8). Ohte and Tokuchi (1999) suggested that in addition to climatic controls, tectonic activity and topography are important in regulating HCO_3^- in watersheds throughout the world. Weathering reactions are particularly important for watersheds with soils having high base concentration, such as calcic and dolomitic soils and volcanic ash. Ohte and Tokuchi (1999) developed a conceptual model that describes the high HCO_3^- discharge in Japanese watersheds based upon the orogenically active geology and the complex, steep topography.

In this hydrobiogeochemical system with high weathering rates and high alkalinity solutions, the soil CO_2 environment strongly controls the pH of subsurface water and groundwater (Ohte *et al.*, 1995; Sato and Takahashi, 1996). Based upon the field observations in the Kiryu Experimental Watershed, seasonal changes of CO_2 concentration in forest soil were at a maximum at the middle of the growing season (July). High temperature and high precipitation in the summer enhanced plant and microbial respiration on the forest floor. Most of the CO_2 was derived from the A_0 and A horizons, but the highest concentrations (1·5%) were found at 150 to 180 cm due to diffusive transport (Hamada *et al.*, 1996).

Evaluating the effects of acid rain in Japan using the critical load concept has been proposed (Department of Global Environment, 1997). Shindo *et al.* (1995) reviewed previous works regarding the critical load estimates based on a steady-state mass balance model. They discussed its limitations for long-term prediction of acidification and emphasized the importance of the development of a dynamic simulation technique for the Japanese environment (Shindo and Fumoto, 1998; Shindo, 1999). Application of previously developed hydrochemical models such as MAGIC (Cosby *et al.*, 1985) and ILWAS (Gherini *et al.*, 1985) has been carried out for several forest watersheds in eastern Japan (Miyanaga and Ikeda, 1994).

HYDROLOGY USING BIOGEOCHEMICAL TRACERS

Several studies on the short-term variations of streamwater chemistry during a storm event have focused on temporal changes in flow paths (Muraoka and Hirata, 1988; Shimada *et al.*, 1993; Tsuboyama *et al.*, 1994; Katsuyama *et al.*, 1998; Ohrui and Mitchell, 1999). The *in situ* tracer experiments done by Tsuboyama *et al.* (1994) clearly indicated the role of preferential flow in storm runoff generation.

Utilization of chemical tracers for catchment hydrology in Japan began in the 1970s. Changes in conductivity, alkalinity and composition of stable isotopes of hydrogen and oxygen have been used to trace

Table III. Basin area, relief ratio and proportion of pre-event water to total discharge of typical Japanese forested watershed

Basin	Basin area (km^{-2})	Relief ratio ($m\ m^{-1}$)	Pre-event water (%)	Reference
Koitogawa	10·300	0·04	80	Nakamura (1971)
Tama	0·020	0·23	86	Tanaka *et al.* (1984)
Uryu	1·280	0·12	88	Kobayashi (1986)
Watarase-gawa 1	0·013	0·60	42	Ohrui *et al.* (1992)
Watarase-gawa 2	0·018	0·45	77	Ohrui *et al.* (1992)
Inabu	0·016	0·46	88	Ichiyanagi and Kato (1998)
Inabu	0·534	0·25	85	Ichiyanagi and Kato (1998)
Ohmiya	0·003	0·63	20	Harada *et al.* (1999)
Yodagiri 1	0·053	0·84	64	Tsujimura *et al.* (1999)
Yodagiri 2	0·063	0·67	98	Tsujimura *et al.* (1999)

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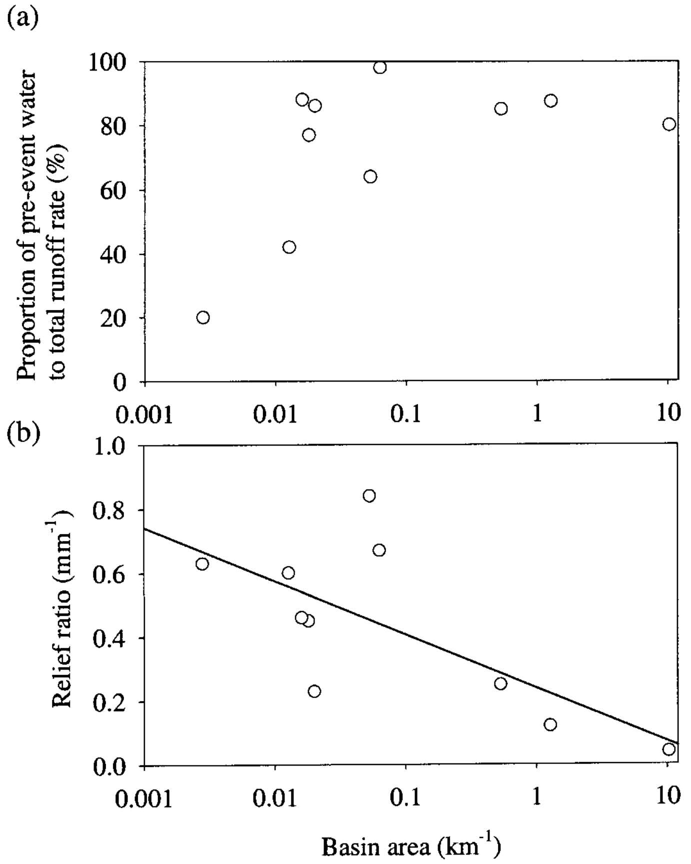

Figure 9. (a) Relationships between basin area and proportion of pre-event water to total runoff rate. (b) Relationships between geographical area and relief ratio of typical Japanese watersheds. A negative correlation between logarithmic value of basin area and relief ratio is significant ($r^2 = 0.485$, $p < 0.05$, slope = -0.1676, intercept = 0.2393). Date sources are listed in Table III

flow paths and to estimate water residence time (Nakamura, 1971; Tanaka *et al.*, 1984; Kobayashi, 1986; Matsubaya *et al.*, 1990; Ohrui *et al.*, 1992; Ichiyanagi and Kato, 1998; Tsujimura and Tanaka, 1998; Tanaka and Ono, 1998; Harada *et al.*, 1999; Tsujimura *et al.*, 1999). Examples of hydrograph separation using chemical tracers done in Japanese forested catchments are summarized in Table III and Figure 9. The proportion of the pre-event water ranges widely from 13 to 98%. Although the data are limited, correlations with topographic factors can be found. The proportion of the pre-event water increases with an increase in basin size and with a decrease in the relief ratio of the basin (Figure 9). The proportion of event water contribution to stream runoff tends to be larger in steep mountainous basins. Buttle (1994) reviewed the relationship of basin size and pre-event water proportions at the event discharge for Scandinavia, North America and Australia, and concluded that the proportion of pre-event water was independent of basin size. Pearce (1990) stated that this proportion decreases with an increase in basin size from 0·003 to 2·8 km^2 in the Maimai catchment in New Zealand. In Japan the relationship between basin size and pre-event water may be due to landscape features including steep hillslopes with thin soil layers in forested basins draining into small flat valleys. Therefore, during storm events, the growth of the saturated zone at the valley bottom does not expand the source area for overland flow, but rather contributes to the transport of subsurface water (pre-event water) from hillslopes (Sidle *et al.*, 2000). Therefore an increase in basin size may proportionally increase the contributing area of subsurface and groundwater flow from the hillslope, and specifically, the pre-event water contribution increase with watershed area.

Matsutani *et al.* (1993) estimated water residence time for a headwater catchment using variations of tritium concentration. Their results indicated a highly heterogeneous movement of the stored water in the catchment, although the catchment is relatively small (0·14 km^2) and hilly (relief ratio = 0·38). The estimated residence times of soil water and groundwater were ~4 months and 19 years, respectively.

Hillslope hydrologists recognize that the information provided by hydrograph separation is not sufficient for describing water pathways (Tsujimura and Tanaka, 1996). As Bonell and Fritsch (1997) noted, simultaneous applications of multiple tracers that have spatially different distributions and/or intensive hydrometric observations are required.

CONCLUDING REMARKS

In Japan, the seasonal pattern that is characterized by high precipitation in summer differs from that found in most of Europe and the northeast United States. Both seasonal variations of N dynamics and changes in the dominant flow path should also be evaluated, including determination of how these patterns change with respect to slope position and soil depth within Japanese catchments. New conceptual models for evaluating N saturation in Japanese forested watersheds are needed.

Quantitative information on the fluxes and seasonal patterns of C losses in drainage waters (e.g. POC, DOC and DIC) is quite limited for Japanese forest watersheds. Although it is apparent that DIC plays a major role in regulating surface water chemistry, some reports suggest that DOC in soil solution may play an important role in regulating N dynamics (e.g. Hart *et al.*, 1994). The evaluation of DOC dynamics along hydrological pathways in forest catchments may be important not only for quantifying carbon cycling, but also for understanding the spatial and temporal variations of biotic regulation of N dynamics (Konohira, 1999).

Although there is little direct evidence indicating the direct effects of air pollution (SO_x, NO_x and O_3) upon physiological functions or surface water acidification, further research in this area is required (Department of Global Environment, 1997). The prediction of freshwater acidification in Japan requires better estimates of acid neutralization of catchments (Shindo, 1999). Determination of mineral weathering rates within catchments over extended periods is also needed (Shindo and Fumoto, 1998).

Because surface waters of most Japanese catchments have high acid-neutralizing capacity and pH often close to neutrality, it is not likely that acidic deposition will lead to marked increases in acidity of surface waters. However, further evaluation of the generation of nitrate in surface waters is needed in combination with monitoring of the acid buffering processes. High NO_3^- discharge with high leaching rates and associated cations may contribute to eutrophication of large riverine systems. In Japan studies of eutrophication of freshwater and coastal marine systems has focused on impacts of anthropogenic nutrient loads and artificial alteration of flow regimes on aquatic ecosystems (e.g. Gohda, 1985; Sudoh, 1996; Uchiyama *et al.*, 2000). The effects of N saturation of forest watersheds on freshwater and marine coastal regions need to be evaluated further.

The three critical research areas that need to be addressed by Japanese hydrobiogeochemists include: (1) quantification of the influence of steep slopes and the abundance of highly weatherable minerals on hydrobiogeochemistry; (2) determination of how the seasonality of precipitation inputs and the temperature regime affect nutrient cycling in Japanese forest ecosystems; and (3) evaluation of how different forest management practices affect hydrobiogeochemistry. More case studies, especially for the cool temperate and subtropical regions in Japan, that include the measurement of both hydrologic variables (e.g. soil moisture content and groundwater level) and biogeochemistry (nutrient pools and fluxes) are needed. The standardization of the monitoring methods for catchment hydrology and biogeochemistry is required to develop more comprehensive databases. The development of Long Term Ecological Research projects (Risser, 1991) should be fostered. Such developments will be useful for comparing the hydrology and biogeochemistry of Japan with other regions of the world.

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