

**Effects of bedrock groundwater dynamics on
hydro-biogeochemical processes in granitic
headwater catchments**

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CHAPTER 1

Introduction

1.1 General background

1.1.1 The contribution of bedrock groundwater as a geographic source of stream water

Headwater catchments are source areas for water and solutes. Understanding runoff generation processes in headwater catchments is essential for taking measures to reduce damage from flood and drought in the lower reaches of rivers. It is also important for the protection of river ecosystems because runoff generation processes affect the transport of nutrients and pollutants from headwater catchments.

Studies of runoff generation processes in headwater catchments up until the 1980's assumed that the underlying bedrock was impermeable and investigated groundwater dynamics only within soil layers. This assumption is consistent with transient saturation at the soil-bedrock interface during rainfall, commonly observed in headwater catchments. However, it has been shown that water infiltrating into permeable bedrock layers significantly affects runoff generation processes in many headwater catchments (e.g., Anderson et al. 1997; Mulholland 1993; Onda et al. 2001). Water infiltrating from soil layers into bedrock layers on hillslopes in headwater catchments has been observed using dye-tracers (Noguchi et al. 1999; Frazier et al. 2002), hydrometric observations using tensiometers (Kosugi et al. 2006; Masaoka et al. 2016), and water budget observations (Tromp-van Meerveld et al. 2007). Water infiltrating into bedrock on hillslopes has been shown to recharge groundwater in bedrock layers (e.g., Katsura et al. 2009). Hereafter, the term “bedrock groundwater” is used to describe groundwater in bedrock layers.

The contribution of bedrock groundwater to stream water can be examined using end-member mixing analysis (EMMA). EMMA links the chemistry of geographic sources, or end-members (e.g., throughfall, groundwater, and soil water), to resulting stream water chemistry, assuming that stream water arises from a mixture of these sources (Hooper et al. 1990; Christophersen et al. 1990). EMMA produces a hydrograph separation of stream water into runoff components. In this study, “geographic source” is defined as an end-member calculated by EMMA.

Recent studies have shown that the contribution of bedrock groundwater to stream water changes with increasing spatial scales of catchments. Studies on the relationship between the contribution of

bedrock groundwater to stream water and catchment scale are listed in Table 1.1. In these studies, stream water was sampled simultaneously in many catchments of different scales and the geographic sources of the stream water were estimated using EMMA (Uchida and Asano 2010; Egusa et al. 2016; Fujimoto et al. 2016). Fujimoto et al. (2016) reported that the contribution of bedrock groundwater to baseflow stream water increased with catchment scale in volcanic-rock catchments ranging in size from 5.1 ha to 626.7 ha. Alternatively, Uchida and Asano (2010) showed that contributions of bedrock groundwater to baseflow stream water did not increase with catchment scale in granitic-rock catchments; they showed that the contribution of bedrock groundwater to baseflow stream water was more variable in catchments equal to or smaller than second-order catchments (approx. 10 ha) compared to catchments ranging in stream order from third to sixth. Egusa et al. (2016) also reported that catchments smaller than approx. 10 ha had greater variability than catchments larger than approx. 100 ha in a study site with sedimentary bedrock. These results indicate that the contribution of bedrock groundwater to baseflow stream water varies among neighboring or adjacent catchments smaller than approx. 10 ha, even within the same watershed. Bedrock groundwater dynamics have also been investigated using hydrometric observations from tensiometers and water level gages at hillslope- or zero-order catchment-scale. Table 1.2 lists study sites where bedrock groundwater dynamics were investigated using hydrometric observations. Hydrometric observations of bedrock groundwater dynamics were conducted on hillslopes or in zero-order catchments, mostly in the order of 10^{-2} – 10^{-1} ha, because it is difficult to install tensiometers and water level gages over large areas. The contribution of bedrock groundwater to stream water may change with increasing spatial scale from hillslope or zero-order catchment-scale to first- or second-order catchment-scale because the runoff processes in stream channels and riparian zones are added. To consider bedrock groundwater dynamics observed on hillslopes or in zero-order catchments in relation to stream runoff and chemistry in higher-order catchments, it is necessary to understand how the contribution of bedrock groundwater to stream water changes with increasing spatial scale from hillslope or zero-order catchment-scale to catchments larger than first-order. However, how the contribution of bedrock groundwater to baseflow stream water changes from hillslope or zero-order catchment-scale, in the order of 10^{-2} – 10^{-1} ha, to first- or second-order catchment-scale, in the order of 10^0 ha, has not yet been examined (Table 1.1).

As shown in Table 1.1, the relationship between bedrock groundwater contribution to stream water and catchment scale was examined only during baseflow periods; in contrast, considerable contribution of bedrock groundwater to stream water during storm flow periods has been reported for some headwater catchments. Table 1.3 lists studies that investigated the contribution of bedrock groundwater to stream water during storm flow periods using EMMA in a headwater catchment. During storm flow periods, the contribution of bedrock groundwater to stream water decreased with

increasing rainfall amount, but the contribution was not negligible in all catchments (Mulholland 1993; Fujimoto et al. 2008; Morel et al. 2009; Iwagami et al. 2010; Padilla et al. 2015). While the runoff of bedrock groundwater was relatively constant throughout storm flow periods in Fudoji, Japan, WB300 and WB0, USA, and Kervidy-Naizin, France (Mulholland 1993; Fujimoto et al. 2008; Morel et al. 2009), it increased more during secondary runoff peaks after rainfall than during baseflow periods and rainfall peaks in Ma and D catchments in Japan (Iwagami et al. 2010; Padilla et al. 2015). Onda et al. (2006) pointed out that the catchments where bedrock groundwater caused secondary runoff peaks were comprised of more permeable bedrock than the catchments where it did not cause secondary runoff peaks. These findings indicate that bedrock groundwater can play an important role as a geographic source of stream water during storm flow periods, although its response to rainfall differs depending on the geology of the catchment. Thus, how the contribution of bedrock groundwater to stream water changes from hillslope or zero-order catchment-scale, in the order of 10^{-1} ha, to first- or second-order catchment-scale, in the order of 10^0 ha, should be examined not only during baseflow periods but also during storm flow periods.

1.1.2 The effects of a perennial riparian groundwater body recharged by bedrock groundwater on stream flow paths during storm flow periods

In this study, “flow path” is defined as the route that each geographic source passes through before reaching a stream. Each geographic source contributes to stream water through various flow paths, such as hillslope subsurface flow, saturated overland flow in riparian zones, and channel precipitation. This means that not only geographic sources but also flow paths should be examined to understand the effects of bedrock groundwater on stream runoff and chemistry during storm flow periods. Observations of groundwater levels and soil water contents in hillslopes and zero-order catchments, shown in Table 1.2, revealed that bedrock groundwater seeped into the soil layer at the footslope or in the riparian zone before it contributed to stream water (Anderson et al. 1997; Masaoka et al. 2016). Seeps of bedrock groundwater at footslopes or riparian zones were also indicated by the observation of groundwater chemistry and groundwater levels in two granitic catchments in Shiga Prefecture, Japan (a 0.1-ha catchment in Fudoji (Uchida et al. 2003) and a 0.68-ha catchment in Kiryu (Katsuyama et al. 2005)). In these catchments, seeped bedrock groundwater recharged the perennial groundwater body within the riparian soil layer (“perennial riparian groundwater body”) (Anderson et al. 1997; Uchida et al. 2003; Katsuyama et al. 2005; Masaoka et al. 2016). Several flow paths have been shown to rapidly respond to rainfall, such as channel precipitation, hillslope subsurface stormflow, saturation overland flow, and rapid groundwater discharge in a riparian zone (e.g., Sidle et al. 2000; McGlynn and McDonnell 2003). The expansion of perennial riparian groundwater by bedrock groundwater may

increase saturation overland flow and rapid groundwater discharge in a riparian zone (“riparian runoff”). However, the contribution of bedrock groundwater to stream water does not necessarily increase due to increasing riparian runoff because throughfall precipitated on the riparian zone may discharge rapidly without mixing with bedrock groundwater stored there. Thus, examination of flow path is important for understanding the effects of bedrock groundwater on stream runoff and chemistry during storm flow periods.

1.1.3 The effects of deep percolation on nutrient budgets in headwater catchments

Bedrock groundwater that does not contribute to stream water leaves the catchment without being detected at a weir via deep percolation. Table 1.4 lists studies that have estimated the deep percolation of water by observing a water budget or a chloride budget. The ratio of deep percolation of water to precipitation was reported to be greater than 10% in headwater catchments comprised of igneous rock (Terajima et al. 1993; Katsuyama et al. 2010; Aishlin and McNamara 2011; Kormos et al. 2015), sedimentary rock (Stewart et al. 2007; Takagi 2013; Oda et al. 2013), and metamorphic rock (Oda et al. 2008; Swarowsky et al. 2011). Figure 1.1 shows the relationship between catchment area and the ratio of deep percolation of water to precipitation for the studies reviewed in Table 1.4. The ratio of deep percolation of water to precipitation was especially large in catchments in the order of 10^{-2} – 10^{-1} ha and tended to decrease with increasing catchment-scale. Data from Dry Creek, HIJ, Kiryu, and Oborasawa are shown with different symbols in Figure 1.1 because the ratio was observed in multiple catchments at these watersheds (Waichler et al., 2005; Graham et al., 2010; Katsuyama et al. 2010; Aishlin and McNamara 2011; Oda et al., 2013). When comparing the ratio of deep percolation of water to precipitation between catchments within the same watershed, the ratio also had a tendency to decrease with increasing catchment-scale in all study sites except for Oborasawa (Waichler et al., 2005; Graham et al., 2010; Katsuyama et al. 2010; Aishlin and McNamara 2011; Oda et al., 2013) (Figure 1.1). This corresponds to the result that the contribution of bedrock groundwater to streams increased with increasing catchment-scale in some study sites, as shown in Table 1.1. Although the ratio of deep percolation of water to precipitation tended to decrease with increasing catchment-scale, it was reported to be higher than 10% even in catchments larger than 100 ha (Waichler et al., 2005; Aishlin and McNamara 2011) (Figure 1.1). These results indicate that deep percolation is a significant component of water budgets for many forested headwater catchments.

Negligible water loss through deep percolation was the fundamental assumption used to evaluate a nutrient budget in a forest ecosystem by observation in small headwater catchments (Likens and Bormann 1995). In the Hubbard Brook Experimental Forest, where a nutrient budget was evaluated

for the first time, Bradley and Cushman (1956) conducted a detailed geological survey and confirmed the low permeability of the bedrock. Bormann and Likens (1967) assumed negligible water loss to deep percolation based on this result, and they evaluated the net gain or loss of nutrients in the forest ecosystem from measurements of its input by precipitation and its output in streamflow in the Hubbard Brook Experimental Forest. However, study sites where the low permeability of the bedrock has been confirmed by a geological survey are rare. Considerable water loss through deep percolation in many headwater catchments (Table 1.4; Figure 1.1) indicates that a nutrient budget might be evaluated based on a wrong assumption.

Among various nutrients, nitrogen (N) is one of the most important nutrients exported from the forest ecosystem to streams. Dissolved inorganic nitrogen (DIN) exported from forest ecosystems increases in response to disturbances, such as harvesting practices (Likens et al. 1970; Pardo et al. 1995; Swank et al. 2001) and insect defoliation (Swank et al. 1981; Houle et al. 2009; Tokuchi et al. 2013). Furthermore, it has been reported that elevated atmospheric N deposition from industrial and agricultural activities during the 20th century has also increased DIN export from forest ecosystems as a consequence of causing N saturation, which is defined as a condition that occurs when the availability of inorganic N is in excess of biotic demand (Aber et al. 1989; Ohrui and Mitchell 1997). Increasing DIN export from forest ecosystems can cause a deterioration of drinking water quality because a high nitrate level in drinking water can have toxic effects, such as methemoglobinemia (Scatena 2000). It can also affect aquatic ecosystems as a consequence of two phenomenon: surface water acidification and eutrophication (Stoddard 1994). To evaluate the N status in forest ecosystems, DIN budgets have been examined by observation of DIN input by precipitation and DIN output in streamflow at many forested headwater catchments around the world (Likens et al. 1970; Swank et al. 1981; Likens and Bormann 1995; Campbell et al. 2004; Tokuchi et al. 2013). However, the estimation of a DIN budget by observing DIN input by precipitation and DIN output in streamflow produces incorrect results if the deep percolation of DIN is not negligible in the catchment. To avoid evaluating N status in forest ecosystems based on a wrong estimation of the DIN budget, the effects of deep percolation on the DIN budget should be quantified.

Table 1.1 Studies that have investigated the relationship between contributions of bedrock groundwater to stream water and catchment scale using EMMA.

Catchment	Location	Geology	Elevation (m)	Smaller scale		Larger scale		References
				Study area (ha)	Contribution of bedrock groundwater (%)	Study area (ha)	Contribution of bedrock groundwater (%)	
Fudoji	Shiga, Japan	granite	300 – 600	approx. 0.1 – approx. 10	40 – 116	approx. 10 – 430	52 – 66	Uchida and Asano (2010)
Inokawa	Chiba, Japan	sedimentary	90 – 346	approx. 0.1 – approx. 10	0 – 100	approx. 100 – 510	50 – 100	Egusa et al. (2016)
Hosotani	Tottori, Japan	lava and pyroclastic flow	505 – 1448	5.1 – 191.6	37 – 40	221.2 – 332.3	50 – 57	Fujimoto et al. (2016)
Hontani	Tottori, Japan	lava and pyroclastic flow	520 – 1448	18.2 – 276.6	10 – 37	307.9 – 626.7	76 – 83	Fujimoto et al. (2016)

Table 1.2 Study sites where bedrock groundwater dynamics were investigated using hydrometric observation.

Catchment	Location	Geology	Study area (ha)	Maximum length of hillslope (m)	Average slope ($^{\circ}$)	Topography	References
PMRW	Georgia, USA	granite	—	20	13	hillslope	Tromp-van Meerveld et al. (2007)
M8, Maimai	New Zealand	conglomerate	—	50	35	hillslope	Gabrielli et al. (2012)
WS10, HJA	Oregon, USA	tuff and braccias	—	125	37	hillslope	Gabrielli et al. (2012)
Hirudani	Gifu, Japan	granite	0.0074	—	40	hillslope	Masaoka et al. (2016)
WS, Kiryu	Shiga, Japan	granite	0.024	—	—	hillslope	Kosugi et al. (2006)
CB1	Oregon, USA	sandstone	0.086	—	43	zero order catchment	Anderson et al. (1997)
Rivendell	California, USA	sandstone	0.4	125	32	zero order catchment	Salve et al. (2012)
Nishi'otafuku-Yama	Hyogo, Japan	granite	2.1	—	34	zero order catchment	Kosugi et al. (2011)

Table 1.3 Studies that have investigated the contributions of bedrock groundwater to stream water during storm flow periods using EMMA in a headwater catchment.

Catchment	Location	Geology	Elevation (m)	Study area (ha)	Contribution of bedrock groundwater to stream water (%)		References
					baseflow period	stormflow period ^a	
Fudoji	Shiga, Japan	granite	—	0.1	80	approx. 30 (46.5 mm)	Fujimoto et al. (2008)
Ma	Kumamoto, Japan	andesite and tuff	322–378	1	—	33 (106 mm) – 69 (36 mm)	Iwagami et al. (2010)
D	Miyazaki, Japan	sandstone and shale	—	1.45	87–88	73 (239 mm) – 88 (58 mm)	Padilla et al. (2015)
WB300	Tennessee, USA	dolomite	—	28.9	approx. 40	approx. 10 (45 mm)	Mulholland (1993)
WB0	Tennessee, USA	dolomite	—	38.4	approx. 60	approx. 40 (45 mm)	Mulholland (1993)
Kervidy-Naizin	Central Brittany, France	schist	93–135	500	—	approx. 5 (10.5 mm) – approx. 20 (8 mm)	Morel et al. (2009)

^a Values in parentheses are total rainfall during a storm event when contribution of bedrock groundwater to stream water was estimated.

Table 1.4 Studies that have estimated the deep percolation of water by observing a water budget or a chloride budget.

Study site	Catchment ID	Location	Geology	Study area (ha)	Average slope (°)	Gauge elevation (m)	Vegetation	Ratio of deep percolation of water to precipitation (%)	References
Akatsu	—	Aichi, Japan	Granite	0.255	—	340	forest	18	Terajima et al. (1993)
Coveeta	WS17	North Caroline, USA	metamorphic	13.5	57	742	forest	negligible	Ford et al. (2007)
Dry Creek	BG	Idaho, USA	Granodiorite	52	—	1698	grassland and forest	22	Aishlin and McNamara (2011)
Dry Creek	C1E	Idaho, USA	Granodiorite	858	—	1335	grassland and forest	27	Aishlin and McNamara (2011)
Dry Creek	C2E	Idaho, USA	Granodiorite	750	—	1158	grassland and forest	24	Aishlin and McNamara (2011)
Dry Creek	LG	Idaho, USA	Granodiorite	2693	—	1036	grassland and forest	14	Aishlin and McNamara (2011)
Dry Creek	TL	Idaho, USA	Granodiorite	2	—	1607	grassland and forest	34	Aishlin and McNamara (2011)
Fukuroyamasawa	—	Chiba, Japan	tuff and mudstone	0.802	25.5	126	forest	22	Oda et al. (2008)
HJA	WS1	Oregon, USA	andesite and breccias	96	—	460	forest	11	Waichler et al. (2005)
HJA	WS10	Oregon, USA	andesite and breccias	10.2	29	473	forest	22 ± 18	Graham et al. (2010)
HJA	WS2	Oregon, USA	andesite and breccias	60	—	530	forest	13	Waichler et al. (2005)
HJA	WS3	Oregon, USA	andesite and breccias	101	—	490	forest	13	Waichler et al. (2005)
Kiryu	A	Shiga, Japan	granite	0.086	—	—	forest	17	Katsuyama et al. (2010)
Kiryu	H	Shiga, Japan	granite	0.4	—	—	forest	39	Katsuyama et al. (2010)
Kiryu	K	Shiga, Japan	granite	5.99	—	190	forest	6	Katsuyama et al. (2010)

(continued)

Table 1.4 (continued)

Study site	Catchment ID	Location	Geology	Study area (ha)	Average slope (°)	Gauge elevation (m)	Vegetation	Ratio of deep percolation of water to precipitation (%)	References
Kiryu	M	Shiga, Japan	granite	0.68	—	—	forest	24	Katsuyama et al. (2010)
Kiryu	R	Shiga, Japan	granite	1.75	—	—	forest	7	Katsuyama et al. (2010)
MAT	—	Veracruz, Mexico	breccia, andesite and basalt	24.6	33	2020	forest	20	Muñoz-Villers et al. (2012)
Obara	—	Aichi, Japan	granite	0.087	—	270	forest	32	Terajima et al. (1993)
Oborasawa	1	Kanagawa, Japan	sedimentary	48	36	432	forest	-4	Oda et al. (2013)
Oborasawa	3	Kanagawa, Japan	sedimentary	7	36	435	forest	23	Oda et al. (2013)
Oborasawa	4	Kanagawa, Japan	sedimentary	5	36	470	forest	-50	Oda et al. (2013)
Pukemanga	—	North Island, New Zealand	greywacke and argillite	3	—	72	pasture	27	Stewart et al. (2007)
SEC	—	Veracruz, Mexico	breccia, andesite and basalt	11.9	31	2040	forest	25	Muñoz-Villers et al. (2012)
SFREC	—	California, USA	metamorphic	33	—	—	forest	14–22	Swarowsky et al. (2011)
Tano	—	Miyazaki, Japan	shale	0.41	28	146	forest	25	Takagi (2013)
Treeline	—	Idaho, USA	granite	1.5	—	1600	grassland	34 ± 12	Kormos et al. (2015)
USC	—	Idaho, USA	basalt	26	—	1840	grassland	6–10	Chauvin et al. (2011)

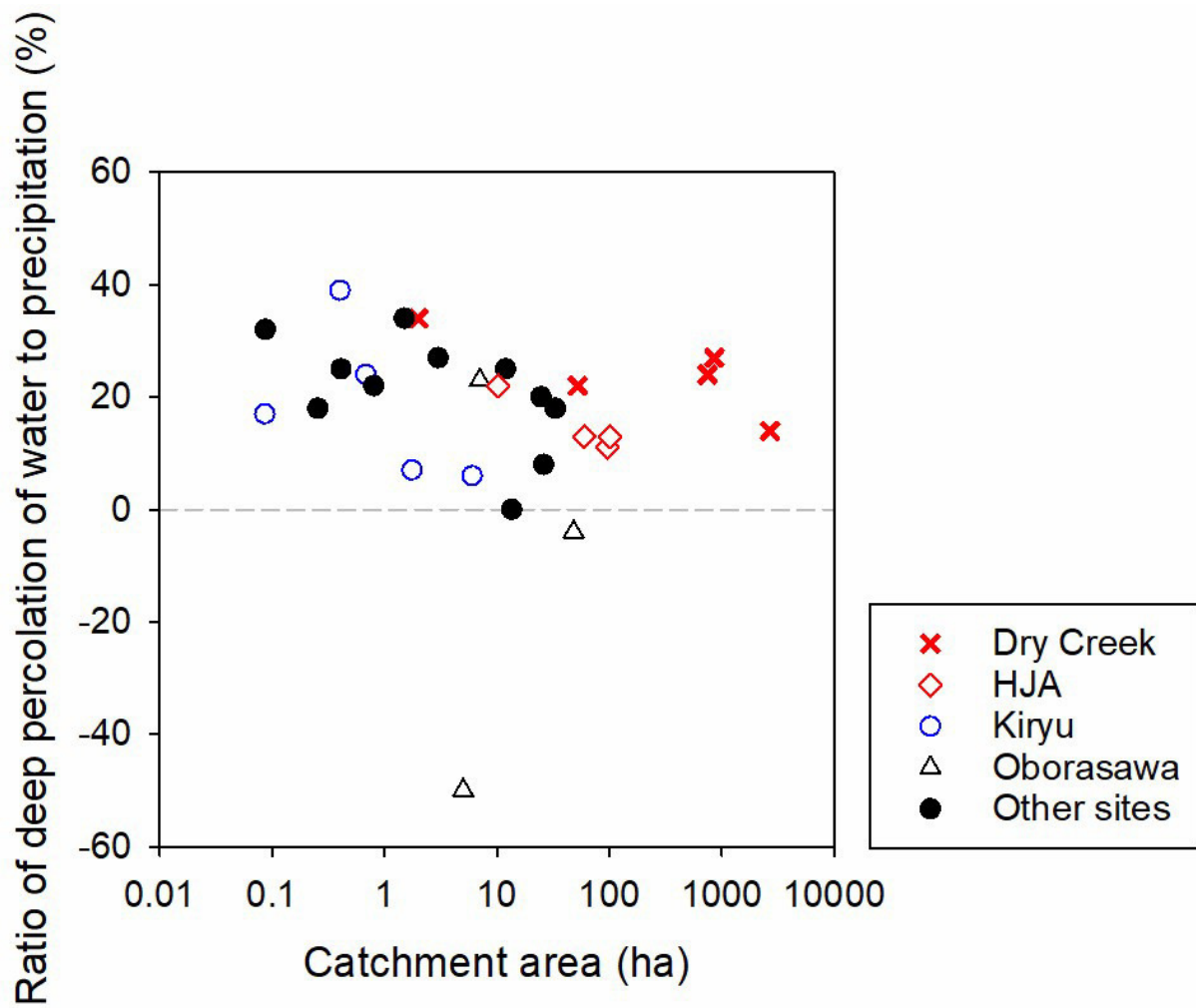


Figure 1.1 Relationship between catchment area and ratio of deep percolation of water to precipitation.

1.2 The objective and procedure of this study

To understand how bedrock groundwater dynamics observed at hillslope scale relate to stream runoff and chemistry observed in higher-order catchments, it is important to bridge the gap of the spatial scale between a hillslope or zero-order catchment, in the order of 10^{-2} – 10^{-1} ha, and a higher-order catchment, in the order of 10^0 ha. Bedrock groundwater dynamics may also relate to a nutrient budget due to the large deep percolation of water in small headwater catchments. Detailed hydrological and hydrochemical observations in neighboring or nested headwater catchments smaller than the order of 10^1 ha should be an effective approach for examining these themes. The objective of this study was to clarify the effects of bedrock groundwater dynamics on hydro-biogeochemical processes in headwater catchments in the order of 10^{-2} – 10^0 ha. The study focused on the change of geographic source and flow path of stream water with increasing spatial scale from zero- to second-order catchments, as well as the effects of deep percolation on the nutrient budget. To achieve this objective, continuous hydrometric observations and monthly and event-based sampling of throughfall, groundwater, and stream water were conducted in a 5.99-ha granitic second-order catchment and its four sub-catchments, with areas ranging between 0.09 and 1.75 ha in the Kiryu Experimental Watershed (KEW), Japan. The structure of this study is shown in Figure 1.2.

In Chapter 2, the characteristics of the study sites and the procedures used for the hydrometric observations, water sampling, and chemical analyzes are described.

The aim of Chapter 3 is to answer the following questions. (1) How does the contribution of bedrock groundwater to stream water differ between different-sized headwater catchments during baseflow and stormflow periods? (2) Why does bedrock groundwater contribution to storm runoff vary with catchment scale? To achieve the aim, the geographic source of stream water during baseflow and storm flow periods was estimated using end-member mixing analysis (EMMA) for the five granitic headwater catchments in the KEW.

Chapter 4 aims to address the following questions. (1) Does a perennial riparian groundwater body recharged by bedrock groundwater affect the dominant flow path of peak runoff? (2) How does response time and runoff source relate to the dominant flow path of peak runoff? To achieve this objective, the dominant flow path of peak runoff was examined by observing rainfall, runoff, and groundwater level in the five granitic headwater catchments of the KEW. Then, the relationships of the dominant flow path of peak runoff to geographic source revealed in Chapter 3 and response time are discussed.

The objective of Chapter 5 is to clarify the effects of deep percolation of water on the dissolved inorganic nitrogen (DIN) budget in granitic headwater catchments. To achieve this aim, bedrock

groundwater dynamics were examined using groundwater level and the groundwater chemistry in a zero-order catchment without a perennial riparian groundwater body and the neighboring first-order catchment of the KEW. The geographic source clarified in Chapter 3 was also taken into consideration for examining bedrock groundwater dynamics. Then, the annual DIN budget was estimated, considering deep percolation, in the two catchments.

Finally, Chapter 6 summarizes the results from each of the preceding chapters. General conclusions from this study are then presented.

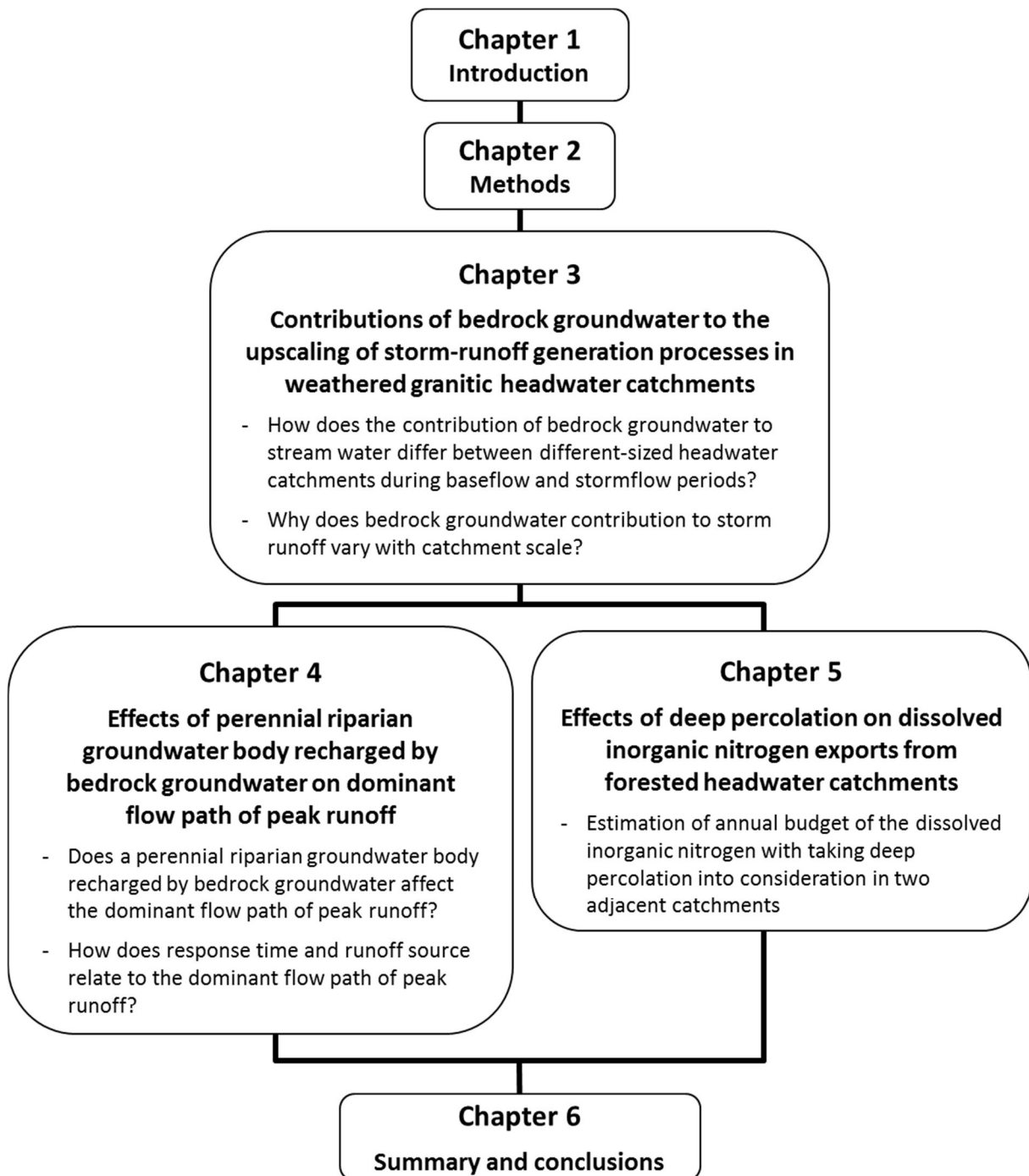


Figure 1.2 Structure of this study.

1.3 Terminology

Some of the terms used in this paper have special meanings that are defined herein:

- Bedrock—The layer beneath the soil layer with an N_4 value in excess of 100 (see Chapter 2). The N_4 value is the number of blows required for a 4-cm penetration of a cone penetrometer (cone diameter 19.5 mm, mass 1.17 kg, and fall distance 20 cm).
- Bedrock infiltration and deep seepage—Based on the concept of Graham et al. (2010), two components are defined: bedrock infiltration (water or solutes that infiltrate into the bedrock) and deep percolation on the catchment scale (a portion of the bedrock infiltration that does not reemerge into a stream channel as bedrock return flow, but leaves the catchment undetected at the weir) (see Chapter 5).
- Bedrock groundwater—Groundwater in the bedrock layer.
- Geographic source—End-member calculated by EMMA.
- Flow path—The route that each geographic source passes through before reaching a stream.
- Highly weathered bedrock groundwater (HBGW)—Groundwater in highly weathered bedrock located several meters below the soil surface. Highly weathered bedrock corresponds to bedrock with a D_H – D_L weathering class defined by the Japanese Society of Soil Mechanics and Foundation Engineering (1985). Rock classified as D_H – D_L is entirely weathered, is characteristically a yellowish-brown color, and is able to be crushed by finger pressure (Japanese Society of Soil Mechanics and Foundation Engineering, 1985; Katsura et al. 2009).
- Moderately weathered bedrock groundwater (MBGW)—Groundwater in moderately weathered bedrock located about 10 meters below the soil surface. Moderately weathered bedrock corresponds to bedrock with a C_L weathering class defined by the Japanese Society of Soil Mechanics and Foundation Engineering (1985). Rock classified as C_L is entirely weathered but retains a rock fabric and structure, plagioclase is generally altered, and quartz is left unweathered. It is light yellowish-brown to yellowish-brown in color and can be partly crushed by finger pressure (Japanese Society of Soil Mechanics and Foundation Engineering, 1985; Katsura et al. 2009).
- Riparian zone—Gently sloped area located near a stream channel.
- Hillslope—Steeply sloped area located away from a stream channel.
- Riparian groundwater—Groundwater located in a soil layer in a riparian zone.
- Hillslope groundwater—Groundwater located in a soil layer on a hillslope.
- Riparian runoff—Saturation overland flow and rapid groundwater discharge in a riparian zone during rainfall (see Chapter 4).
- Riparian buffering—Definitions of riparian buffering are diverse; however, in this study, the term

represents the volumetric buffering of hillslope runoff by groundwater in the vicinity of a stream, following McGlynn and McDonnell (2003) and Jencso et al. (2010) (see Chapter 3).

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CHAPTER 2

Methods

2.1 Site description

The study area was the Kiryu Experimental Watershed (KEW). Figure 2.1 shows the location of KEW. KEW is located in the southern part of Shiga Prefecture, central Japan ($34^{\circ}58'N$, $136^{\circ}00'E$; 190–255 m above mean sea level). KEW is included in the Tanakami Mountain. Hydrological observations have continued since 1967 in KEW.

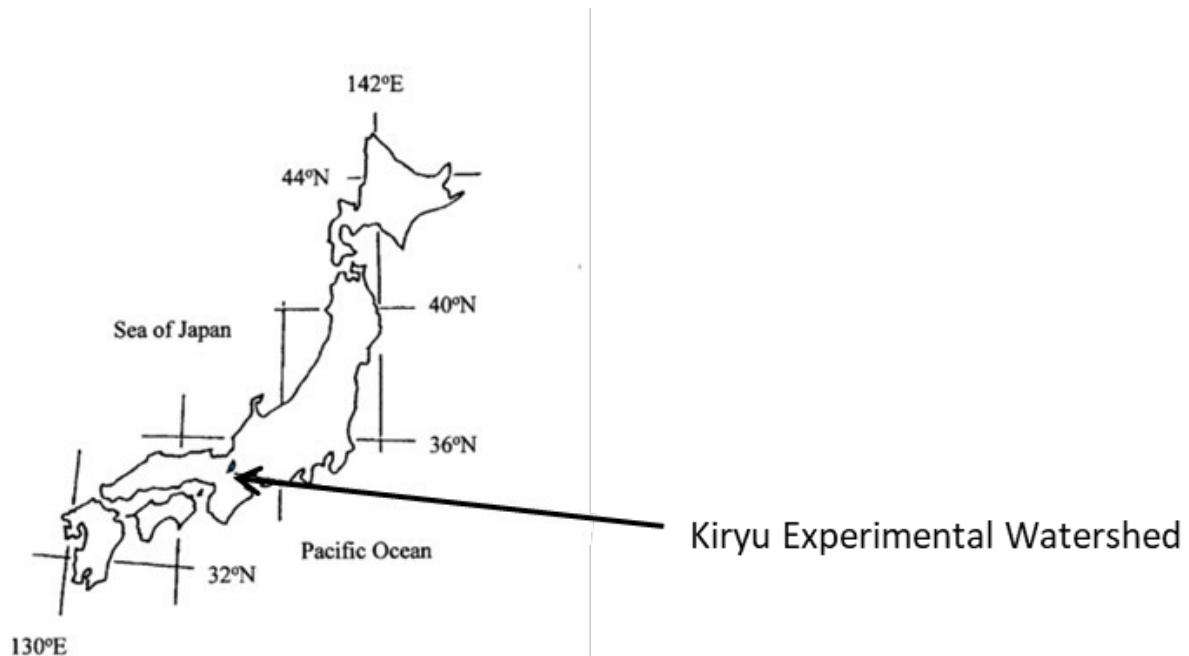


Figure 2.1 Location of the study site.

Figure 2.2 shows the topography of KEW. To investigate the differences in the effects of bedrock groundwater dynamics on hydro-biogeochemical processes between neighboring catchments, five hydrological observation nests were established within KEW: Catchment K (5.99 ha), which corresponded to the entire KEW watershed and four sub-catchments (R, 1.75 ha; M, 0.68 ha; H, 0.40 ha; and A, 0.086 ha) within Catchment K.

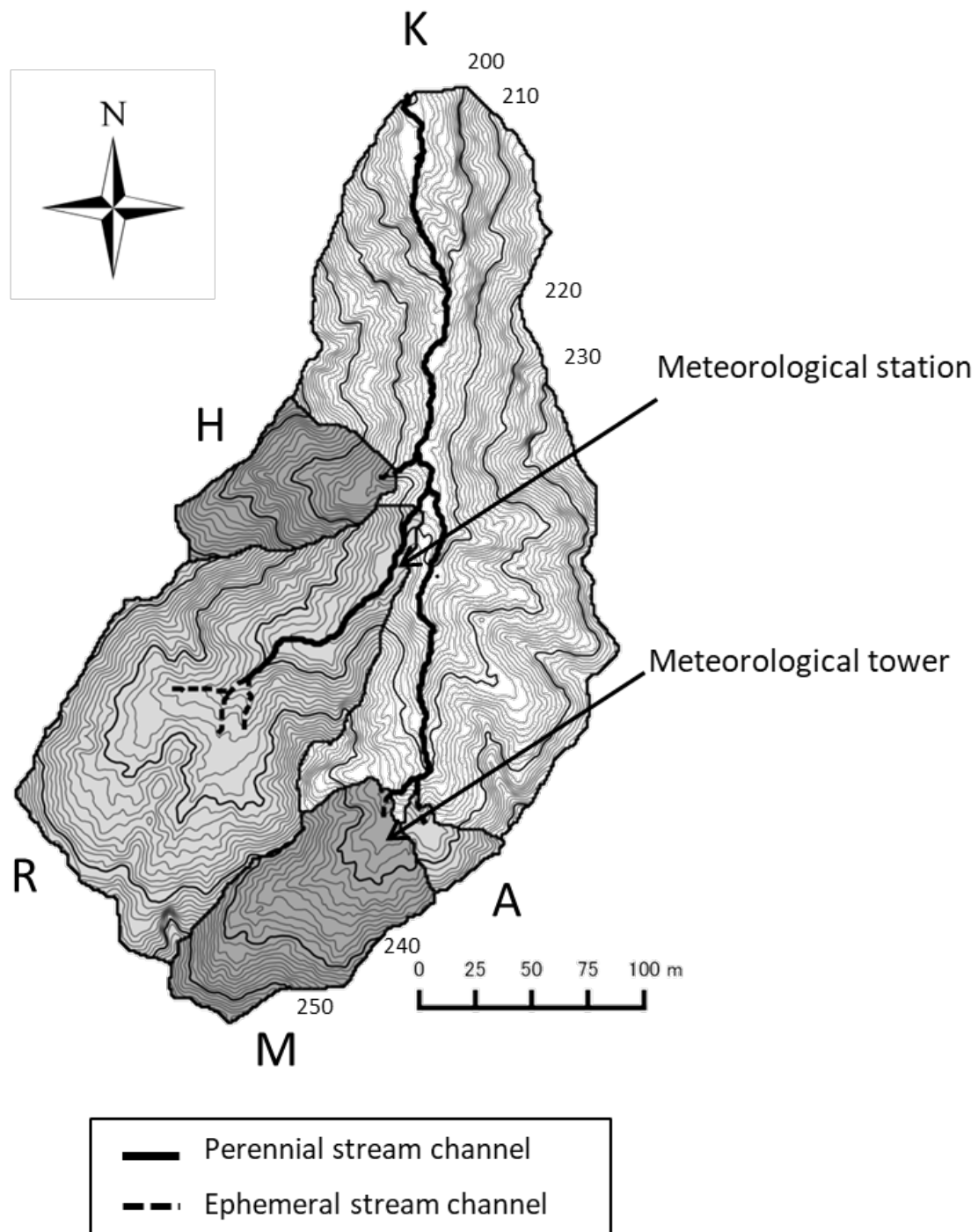


Figure 2.2 Topography of the study site. Interval between contours is 1 m.

The vegetation of KEW consists mainly of Japanese cypress (*Chamaecyparis obtusa*). Japanese cypress trees are mainly about 60 years old in Catchments M and A while there exists Japanese cypress trees with ages of about 100 years old in Catchments R and H. There also exists Japanese

cypress trees with ages of about 30 years old around the second-order stream channel in Catchment K. Some stonemasonry dams were constructed on the site ca. 100 years ago to prevent sediment discharge at a time when KEW was deforested and suffered severe soil erosion. After revegetation, soil erosion substantially decreased. Because the dams are highly permeable, they do not prevent the passage of water. Mean annual precipitation and mean air temperature observed at a local meteorological station (Figure 2.2) from 2005 to 2013 were 1677.8 mm and 13.4°C, respectively.

The catchment is underlain by Cretaceous biotite granite, which is referred to as Tanakami granite. The N_4 value is the number of blows required for a 4-cm penetration using a cone penetrometer (cone diameter: 19.5 mm, mass: 1.17 kg, fall distance: 20 cm). In this study, the bedrock layer was defined as having an N_4 value in excess of 100, in accordance with Katsuyama et al. (2005). The shallow bedrock occurred at depths less than roughly 5 m on hillslopes, had the properties of saprolite, and was highly weathered and permeable (Katsura et al. 2009). The saturated hydraulic conductivity of soils 15 and 30 cm below the surface was $5.07 \times 10^{-4} \text{ m s}^{-1}$ with a standard deviation of $2.16 \times 10^{-4} \text{ m s}^{-1}$ ($n = 4$), whereas that of rocks 0 to 17 cm below the soil–bedrock interface was $1.04 \times 10^{-6} \text{ m s}^{-1}$ with a standard deviation of $0.11 \times 10^{-6} \text{ m s}^{-1}$ ($n = 3$) (Katsura et al. 2006); hence, the soil and the weathered bedrock had clearly distinguishable hydraulic properties in this catchment. Soil physical properties are homogeneous in KEW such that the saturated hydraulic conductivity of soil was on the order of 10^{-4} m s^{-1} in Catchment M (Ohte et al. 1989) and Catchment A (Katsura et al. 2006), as well as on the hillslopes near the meteorological station and the weir in Catchment K (Ohta et al. 1983). Soil type is decomposed granite. The soil profile has no apparent structure and organic matter is not mixed to any noticeable degree. The A_0 horizon has a Moder humus form without a remarkable H horizon at both the upper and lower parts of the hillslope. According to the observation at Catchment M, the mineral soil consists of an A and an AC horizon thinner than 2 cm and a B horizon has not been well established at the upper part of the hillslope, while there is an A horizon of about 7 cm and a B horizon of about 30 cm (Ohte et al. 1995). Detailed descriptions of the chemical characteristics of the soil were given by Ohte et al. (1997) and Asano et al. (1998).

The geomorphic factors of the five catchments are shown in Table 2.1. Catchment K is a second-order catchment, Catchment R is a first-order catchment, and Catchments M, H, and A are zero-order catchments. Although stream channels exist between the spring and the weir in Catchments A, H, and M, I regarded these catchments as zero-order catchments because the spring point fluctuates and surface flow does not occur at almost all areas in the channel during dry winter season.

The slope angle and the topographic index (TPI, Beven and Kirkby, 1979) were calculated based on 1-m digital terrain models (DTMs). The topographic index has the form

$$\text{TPI} = \ln(\alpha/\tan\beta) \quad (2.1)$$

where α is the upslope area per unit contour length contributing to a grid cell and $\tan\beta$ is the local slope angle at the cell. Areas with high TPI will tend to saturate first and will therefore indicate potential subsurface or surface contributing areas (Beven 1997). The spatial distributions of the slope angle and the topographic index are shown in Figure 2.3. Frequency distributions of the slope angle and the topographic index in each catchment are also shown in Figure 2.4. Differences in the slope angle and TPI between catchments are relatively small, although mean slope angle is smaller and mean TPI is larger in Catchments M and R than in Catchments A, H, and K (Table 2.1). Mean soil depth is the largest in Catchment M and the smallest in Catchment H (Table 2.1).

Size of perennial groundwater body within riparian soil layer (“perennial riparian groundwater body”) is different between the five catchments. In zero-order Catchments A, H, and M, riparian aquifers exist only around springs because they hardly have a stream channel (Figure 2.2; Table 2.1). Springs are recharged by bedrock groundwater and subsurface flow at the soil-bedrock interface from hillslopes (Katsuyama et al., 2005). The zero-order catchments have sedimentary zones just above the spring that are formed by bedrock depression in Catchment M (Katsuyama et al. 2005) and water-permeable check dams in Catchments A and H. Previous studies suggested that bedrock groundwater exfiltrated into the soil layer throughout the year within the sedimentary zone in Catchment M (Katsuyama et al. 2005), but not in Catchment A (Katsuyama et al. 2004). Annual deep percolation of water was larger, and the mean residence time (MRT) of stream water was shorter in Catchment H, whereas annual deep percolation of water was less and the MRT of stream water was longer in Catchments A, R, and K (Katsuyama et al. 2010). The streamflow was perennial in Catchments K, R, M, and A. In contrast, runoff from Catchment H ceased during the dry season when there was only a small amount of rainfall. This suggests that the size of perennial riparian groundwater body is different due to bedrock groundwater dynamics between the zero-order catchments: it is the largest in Catchment M, followed by Catchments A and H. In Catchments R and K, riparian aquifers exist along the stream channel throughout the year because they are first- and second-order catchments, respectively.

Table 2.1. Area and geomorphic factors.

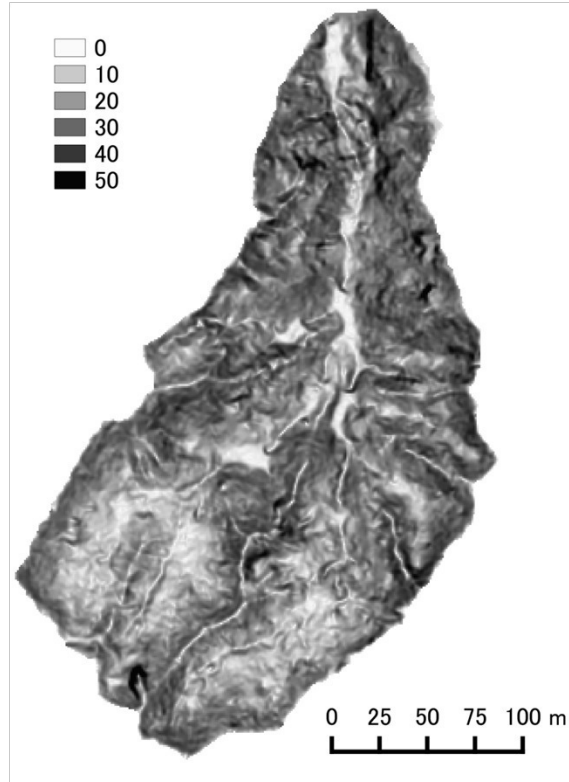
	A	H	M	R	K
Area (ha)	0.09	0.40	0.68	1.75	5.99
Stream Order	0	0	0	1	2
Channel length (m)	15	5	12	125	320
Mean slope angle (°)	23.0	26.5	21.4	22.7	25.3
Mean TPI ^a	2.50	2.49	2.68	2.69	2.59
Ratio of riparian area to catchment area (%) ^b	4.2	0.6	0.7	6.1	6.8
Mean soil depth (cm) ^c	81	51	151	65	65

^a TPI means topographic index.

^b See Section 4.4.2 for estimation of the ratio of riparian area to catchment area.

^c Mean soil depths are data from Nagano and Katsuyama (unpublished data).

(a) Slope angle (°)



(b) Topographic index

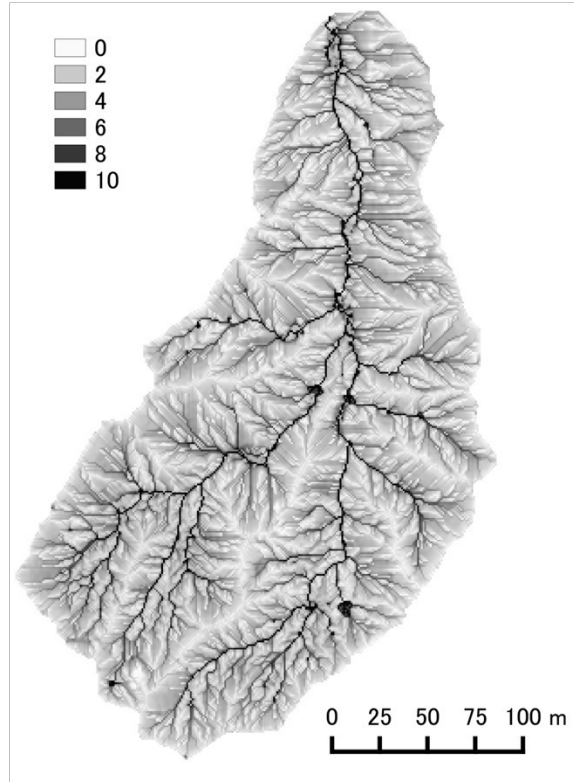


Figure 2.3 Spatial distributions of the slope angle (a) and the topographic index (b) in KEW.

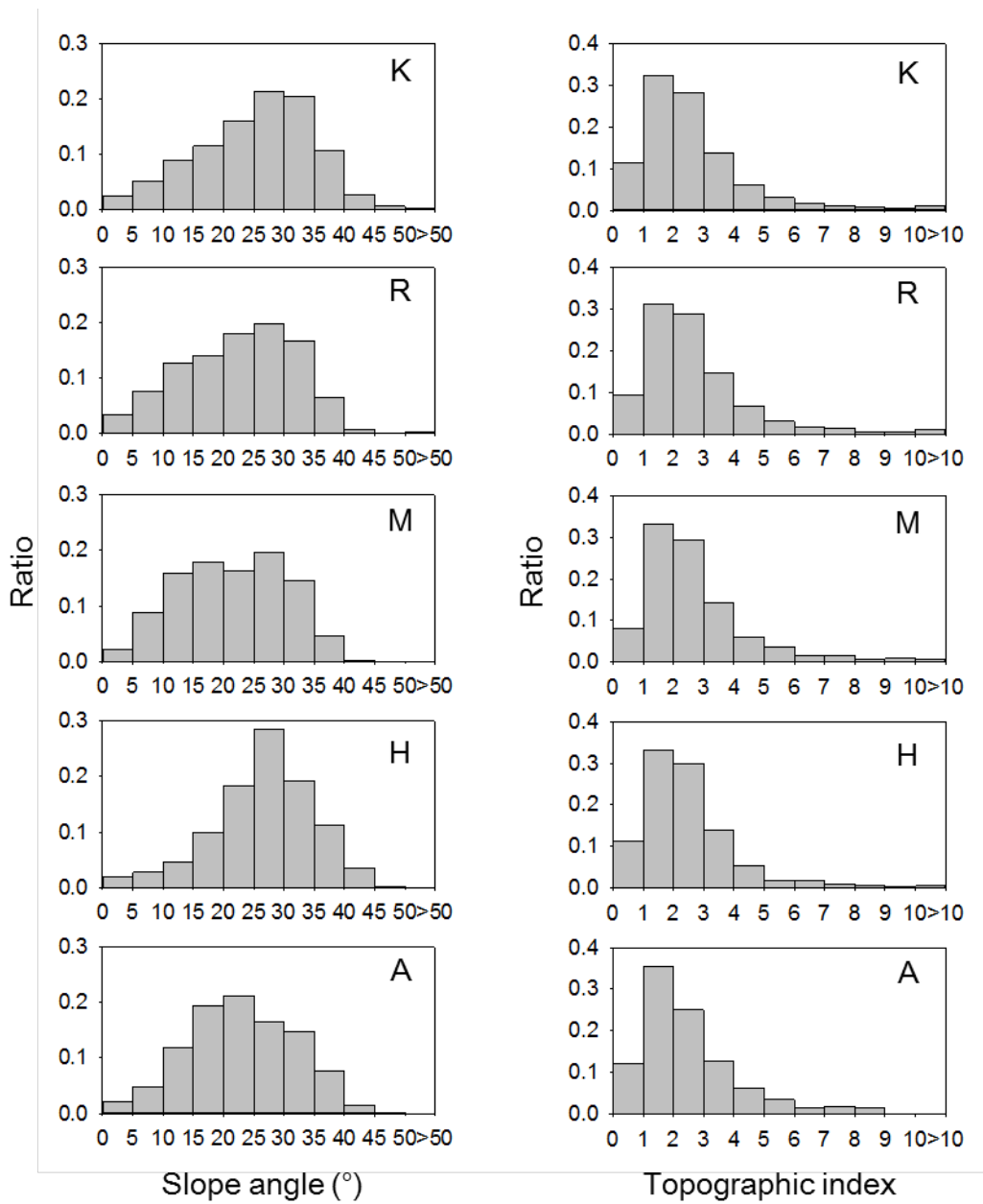


Figure 2.4 Frequency distributions of the slope angle and the topographic index in each catchment.

2.2 Hydrological and hydrochemical observations

Rainfall and runoff rates from each catchment were observed from 2005 to 2013. Observation facilities of the five catchments are shown in Figure 2.5. Rainfall was measured using a 0.5 mm tipping-bucket rain gauge at the meteorological station. Runoff rates were monitored using a

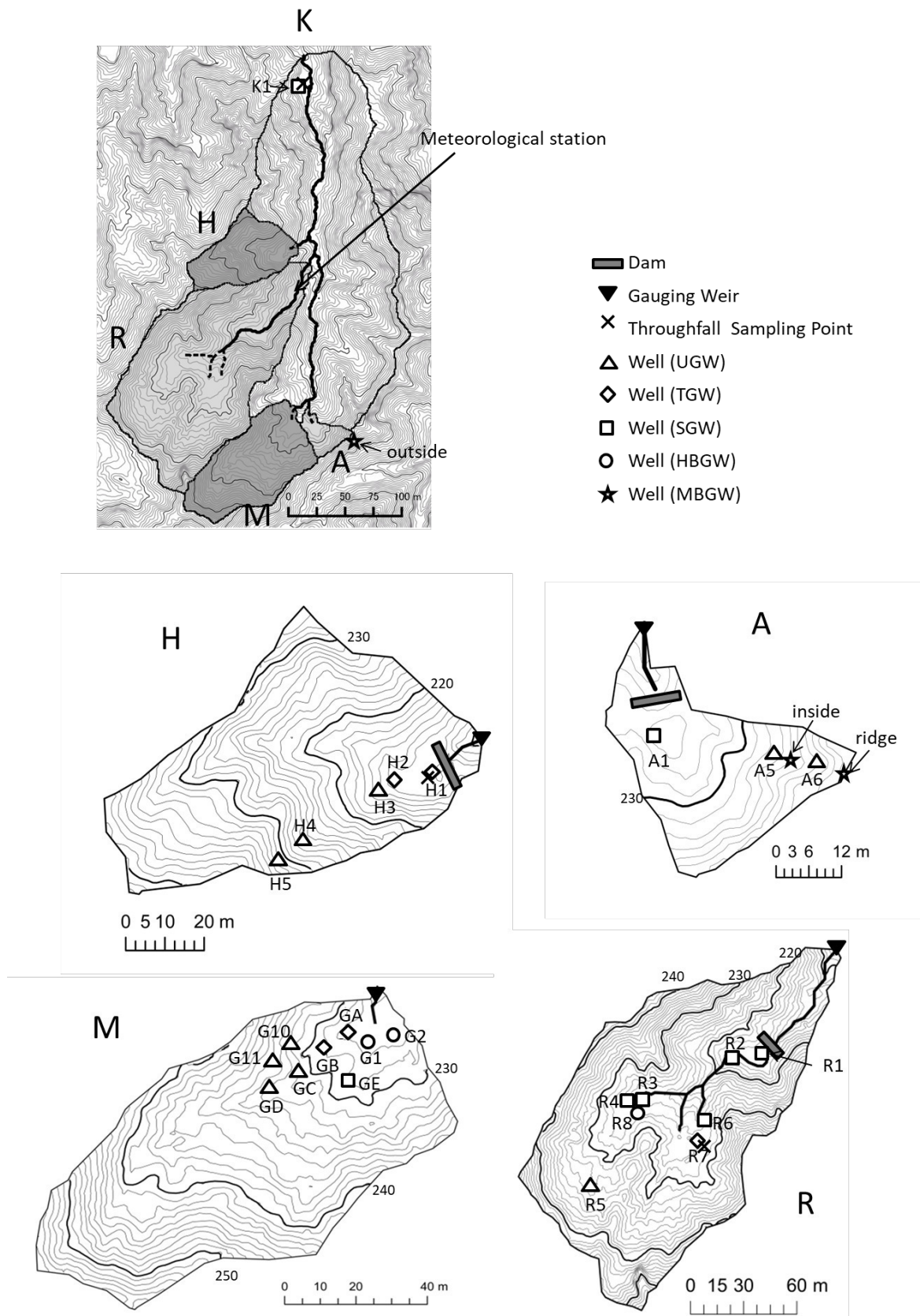


Figure 2.5 Observation facilities of the five catchments.

90° V-notch weir in Catchment K and 30° V-notch weirs in the other catchments. Stage height was recorded every 10 minutes and the discharge rate was calculated using the measured relationship between stage height and the actual discharge rate from the weir. The weir pool was cleaned, and a reference stage height was checked at least once a month using a hook gauge. Data gap was not filled.

Groundwater level was monitored from 2011 to 2013 using a pressure-type water gauge in Catchment A and capacitance-type water gauges in the other catchments. Although the type of water gauge is different between the catchments, difference in it does not affect the results because all water gauges were calibrated every few months by actual groundwater level of the well observed using a multimeter. In the subcatchments, groundwater level was monitored just above the spring point (A1 in Catchment A, H1 in Catchment H, G1 in Catchment M, and R7 in Catchment R; Figure 2.5). In Catchment K, it was monitored at the sedimentary zone just above the weir (K1; Figure 2.5). The well depths of A1, H1, G1, R7, and K1 were 173, 194, 83, 167, and 91 cm, respectively. Soil depths of A1, H1, G1, R7, and K1 measured by a cone penetrometer or soil auger were 175, 194, 470, 84, and 91 cm, respectively. Observation intervals of rainfall, runoff rate, and groundwater level were 10 minutes.

Wells were also installed within soil layer to manually record the groundwater level and collect groundwater samples. Location of each well is shown on Figure 2.5. All wells except for G1-250 and G2-400 were made from PVC pipes with a diameter of 6 cm, and were installed into holes drilled with a hand auger. These wells except for R8 contained small holes around their periphery throughout the full length. A stainless steel pipe with a diameter of 6 cm was driven into the ground with a hammer at G1-250. R8 and G1-250 was screened only at 197-cm and 250-cm depth from the ground surface, respectively. At G2-400, a tension lysimeter was installed into a 400-cm-deep hole drilled with a cone penetrometer. A rod attached by plastic cups with the height of 5 or 6 cm (Figure 2.6) was immersed in 24 wells (all wells except for K1, G1-250 and G2-400 on Table 2.2). Groundwater level was manually recorded during baseflow period on a monthly basis using cups at the 24 wells from August 2010 to December 2012. Groundwater level was manually monitored by recording the height of cups which was filled by groundwater (Ohte et al. 1995).

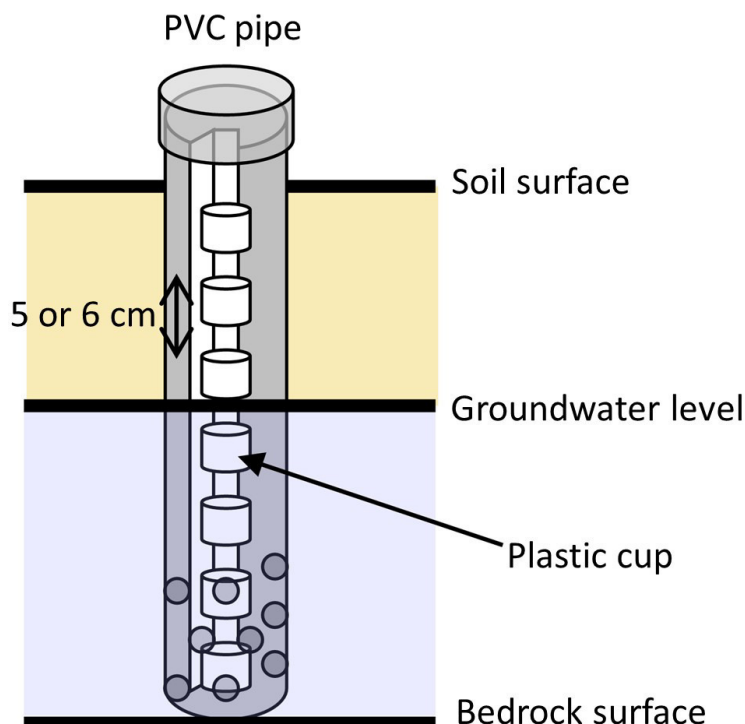


Figure 2.6 The structure of observation wells.

The wells were grouped into four categories depending on whether saturated conditions occurred at the time when observations were made (Ohte et al. 1995; Katsuyama et al. 2005) and the sampling depth. The grouping of the wells was conducted based on results of the groundwater level observation shown on Chapter 4. Well depth, soil depth, and type of the observation wells are shown on Table 2.2. The first category was the unsaturated zone during baseflow period, which was located on hillslopes and saturated only occasionally during rainstorms. Saturation levels declined immediately after rainstorms and the wells were unsaturated during baseflow periods. Wells at H3, H4, and H5 in Catchment H, A5 and A6 in Catchment A, GC, GD, G10 and G11 in Catchment M, and R5 in Catchment R correspond to this zone. Hereafter, groundwater collected in this zone is referred to as groundwater at unsaturated zone during baseflow period (UGW). The second category was the transient saturated zone, which was situated at the interface between unsaturated zone during baseflow period and shallow saturated zone. Although saturated conditions were always present during the wet season, the zone became unsaturated during the dry season. Wells at H1 and H2 in Catchment H, GA and GB in Catchment M, and R7 in Catchment R correspond to this zone. Hereafter, groundwater collected in this zone is referred to as transient saturated zone groundwater (TGW). The third category was the shallow saturated zone, which was saturated all year round and was sampled from the whole

depth of groundwater using the wells with hailes throughout full length. Wells at A1 in Catchment A, G1-75, G2-177, and GE in Catchment M, R1, R2, R3, and R6 in Catchment R, and K1 correspond to this zone. Hereafter, groundwater collected in this zone is referred to as shallow saturated groundwater (SGW). Katsuyama et al. (2005) used SiO_2 as a tracer, and showed that groundwater in the highly weathered bedrock recharged groundwater at the deep riparian soil layer year round at KEW. Thus, the groundwater sampled from riparian soil layer deeper than 1.9 m using the wells without hailes throughout full length (R8, G1-250 and G2-400) was regarded as highly weathered bedrock groundwater (HBGW).

In addition to groundwater in the soil layer, groundwater in the moderately weathered bedrock (MBGW) was manually drawn from three boreholes that were 12 m (outside), 15 m (inside), and 20 m (ridge) deep at and near Catchment A (Figure 2.5). These wells were drilled using a hydraulic-feed-type boring machine and screened at the bottom of the borehole. The wells had a diameter of 6.6 cm. Their depth below the bedrock surface were 11 m, 14 m, and 20 m at outside, inside, and ridge, respectively. More detailed characteristics of the wells of MBGW are provided by Katsura et al. (2009).

Grab samples of stream water were taken monthly from July 2010 to December 2012. The stream water from monthly sampling was treated as baseflow stream water except for that collected when the 24-h antecedent rainfall was greater than 10 mm. Stream water samples were also collected during three rainfall events using auto samplers (American Sigma Models 900, 900Max, and SD900: Hach, Loveland, CO, USA). Samplers were activated manually before storm events. The streamwater-sampling interval was 1 or 2 h on the hydrograph rising limb and between 1 and 4 h on the recession limb. The observed rainstorm characteristics are summarized in Table 2.3. Three rainfall periods with different total amounts of rainfall were chosen. Event 1 (total rainfall: 67.4 mm) was caused by a seasonal rain front, Event 2 (total rainfall: 180.7 mm) was caused by a typhoon, and Events 3-5 (total rainfall: 272.3 mm) was caused by a combination of a typhoon and a seasonal rain front. Although Events 3, 4, and 5 was the continuous rainfall, they were treated as separated events because individual events were separated by intervals of at least 24 h without precipitation. The total rainfall of 272.3 mm was the largest 8-day precipitation recorded in the last 5 years at KEW. Stream water samples were collected in three catchments (H, R, and K) during Events 1 and 2, and in all five catchments during Event 3, 4, and 5.

Table 2.2 Well depth, soil depth, and type of the observation wells.

Well	Well depth (cm)	Soil depth (cm)
UGW		
A5	95	80
A6	96	80
H3	59	59
H4	104	104
H5	27	27
R5	174	174
GC	217	82
GD	179	30
G10	25	26
G11	49	36
TGW		
H1	194	194
H2	77	77
R7	83	84
GA	76	74
GB	82	32
SGW		
A1	173	175
R1	186	186
R2	43	43
R3	44	44
R6	51	51
G1-75	75	470
G2-177	177	446
GE	185	350
K1	91	91
HBGW		
R8	197	352
G1-250	250	470
G2-400	400	446

Table 2.3 Rainstorm characteristics.

	Event 1	Event 2	Events 3-5
Observed period	10-11/6/2011	1-8/9/2011	15-22/6/2012
Total rainfall (mm)	67.4	180.7	272.3
Maximal rainfall intensity (mm/hr)	17.4	14.9	20.9
5 day antecedent rainfall (mm)	1.0	12.9	4.1
10 day antecedent rainfall (mm)	16.1	34.5	71.6

Throughfall was sampled using bottles with funnels (diameter, 18 cm). Throughfall samples were collected at H1 in Catchment H, R7 in Catchment R, and K1 (Figure 2.5) at monthly intervals on the same days that stream water was collected. They were also collected during the five rainfall events at K1. Because the KEW watershed has uniform vegetation and a small area (5.99 ha), I regarded the throughfall samples at this site as representative of the entire watershed.

Groundwater samples were collected monthly on the same day as stream water and throughfall. Groundwater samples which were collected immediately after the rainfall events (27/6/2011 for Event 1; 26/9/2011 for Event 2; 26/6/2012 for Events 3-5) were used for the analysis of each rainfall event. Groundwater was sampled from the observation wells (Figure 2.5). A tension lysimeter was used at G2-400 to take a groundwater sample from 400-cm depth, while groundwater samples at the other locations were taken from cups immersed in the wells (Ohte *et al.*, 1995; Katsuyama *et al.*, 2005). Groundwater samples were taken from cups immersed in the wells showing the greatest rise in water level. Thus, groundwater that occasionally rose during and after rainfall on the hillslope could be sampled.

2.3 Chemical analysis

The samples were filtered through a cellulose acetate filter with 0.45- μm pores in a laboratory within 24 h of collection for monthly sampling and 5 days for storm sampling. Samples for the analysis of solute concentrations were sealed in polyethylene bottles, and refrigerated until analysis. The solute concentrations were analyzed in a laboratory at Kyoto University. Cl^- , NO_2^- , NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , Mg^{2+} , and Ca^{2+} concentrations were measured using ion chromatography (ICS-90 and ICS-1100, Dionex, Sunnyvale, CA, USA), and SiO_2 concentrations were measured using inductively coupled plasma (ICP) emission spectrometry (SPS1500VR, Seiko Instruments Inc., Chiba, Japan; iCAP6300, Thermo Scientific, Waltham, MA, USA). Samples for isotope analysis were sealed in glass vials. They were analyzed for $\delta^{18}\text{O}$ via cavity ring-down spectrometry (L2120-I, Picarro Inc., Santa Clara, CA, USA)

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CHAPTER 3

Contributions of bedrock groundwater to the upscaling of storm-runoff generation processes in weathered granitic headwater catchments

3.1 Introduction

The effect of spatial scale on discharge and water quality is a significant unresolved issue in the hydrological sciences. The importance of hillslopes and small headwaters should not be underestimated in studies conducted at larger catchment scales because groundwater–surface water interactions in such areas are an important component of catchment functioning and are potentially vulnerable to the effects of environmental change (Tetzlaff and Soulsby 2008; Jencso et al. 2009; Jencso et al. 2010). However, realistic hillslope-scale complexity and heterogeneity have yet to be incorporated into the hydrological models used for larger river basins (McDonnell 2003; Sivapalan 2003). To predict discharge and water quality at larger scales based on our knowledge of small-scale catchments, we need to understand how the runoff processes change with an increase in catchment scale.

Some studies have revealed that bedrock groundwater affects the upscaling of baseflow runoff processes. It has been pointed out that contribution of bedrock groundwater to stream water changed with increasing catchment scale as reviewed in Section 1.1.1 (Table 1.1). Shaman et al. (2004) also suggested that the contribution of bedrock groundwater to stream runoff increased as basin size increased by examining low-flow runoff from 11 nested catchments in the Neversink River watershed, USA. In addition, Katsuyama et al. (2010) reported an increasing contribution of bedrock groundwater with upscaling from zero- to second-order catchments, resulting in a lengthening of the mean residence time of baseflow stream water in Japanese granitic headwater catchments.

Although bedrock groundwater plays an important role in the upscaling of baseflow runoff processes, previous studies of the upscaling of storm-runoff generation processes have not considered the effects of bedrock groundwater. Sidle et al. (2000) monitored storm runoff at several nested scales from a hillslope to a first-order basin in Hitachi Ohta Experimental Watershed, Japan, and suggested that upscaling of storm-runoff generation processes should be considered on the basis of the linkages of landscape units such as riparian zones, linear hillslopes, and zero-order basins. Additionally, McGlynn et al. (2004) found a significant relationship between catchment size and runoff lag times, as

well as tracer lag times, in a 280-ha catchment in Maimai, New Zealand, and suggested that the spatial distribution of landscape units has an important role in the upscaling of runoff production and solute transport processes. In mesoscale catchments (1.5–25,800 ha), Didszun and Uhlenbrook (2008) reported that an increasing number of landscape units affected the upscaling of storm-runoff generation processes in the Dreisam catchment, Germany. These studies focused on the influence of the spatial organization of landscape units, and were not concerned with the effects of bedrock groundwater.

Some studies have shown that bedrock groundwater affects the hydrological connectivity of landscape units. The concept of riparian buffering is crucial in understanding this connectivity. The definitions of riparian buffering are diverse, but in this study we use the term to represent the volumetric buffering of hillslope runoff by groundwater in the vicinity of a stream, following McGlynn and McDonnell (2003) and Jencso et al. (2010). Katsuyama et al. (2005) suggested that groundwater infiltrating into bedrock on a hillslope recharged the riparian reservoir year round, while saturated throughflow within the hillslope soil layer occurred only during storms. Additionally, Gabrielli et al. (2012) suggested that exfiltration of bedrock groundwater into a riparian or stream zone resulted in a larger runoff ratio at the catchment-scale than at the hillslope-scale. Thus, bedrock groundwater can increase the riparian buffering potential due to recharge of the riparian reservoir. Bedrock groundwater may therefore affect the upscaling of storm-runoff generation processes by changing the connectivity of landscape units. However, previous studies have considered only the effects of bedrock groundwater on upscaling during baseflow conditions.

The aim of this chapter is to answer the following questions: (1) How does the bedrock groundwater contribution to storm-flow stream water differ between different-sized headwater catchments? (2) Why do bedrock groundwater contributions to storm runoff vary with catchment scale? These questions were addressed using a chemical mixing model. Detailed hydrological and hydrochemical measurements were performed in five headwater catchments that ranged from zero to second order, where annual bedrock infiltration differed (Katsuyama et al. 2010) despite their similar geology.

3.2 Data collection methods

3.2.1 Site description

To investigate the spatial-scale effects of storm-runoff generation processes, five hydrological observation nests were established within Kiryu Experimental Watershed (KEW) (Figure 2.2 and Table 2.1): Catchment K (5.99 ha), which corresponded to the entire KEW watershed, and four sub-catchments (R, 1.75 ha; M, 0.68 ha; H, 0.40 ha; and A, 0.086 ha) within Catchment K. Catchment K is a second-order catchment, Catchment R is a first-order catchment, and Catchments M, H, and A

are zero-order catchments. Perennial riparian groundwater body does not exist in Catchment H while it exists in Catchments M and A. Size of the perennial riparian groundwater body is larger in Catchment M than Catchment A. In Catchments R and K, perennial riparian groundwater body exists along the stream channel because they are first- and second-order catchments, respectively. Annual bedrock infiltration was larger, and the mean residence time (MRT) of stream water was shorter in Catchment H, whereas annual bedrock infiltration was less and the MRT of stream water was longer in Catchments A, R, and K (Katsuyama et al. 2010). Details of study site are shown in Section 2.1.

3.2.2 Hydrological and hydrochemical observations

In this chapter, the data of rainfall and runoff rates from each catchment observed from 2010 to 2012 are used. The water chemistry data of groundwater, throughfall, and stream water are also used. Groundwater was classified into five categories: groundwater at unsaturated zone during baseflow period (UGW), transient saturated zone groundwater (TGW), shallow saturated groundwater (SGW), highly weathered bedrock groundwater (HBGW), and groundwater in the moderately weathered bedrock (MBGW). Groundwater, throughfall, and stream water was collected monthly from July 2010 to December 2012. The stream water from monthly sampling was treated as baseflow stream water except for that collected when the 24-h antecedent rainfall was greater than 10 mm. Stream water in each catchment and throughfall at K1 (Figure 2.5) were also collected during three rainfall periods (Table 2.3). Groundwater samples which were collected immediately after the rainfall events (27/6/2011 for Event 1; 26/9/2011 for Event 2; 26/6/2012 for Event 3-5) were used for the analysis of each rainfall event. Detailed methods of hydrological observation, water sampling, and observed rainfall periods are shown in Section 2.2.

3.2.3 Chemical analysis

In this chapter, the Na^+ , Cl^- , SO_4^{2-} , Mg^{2+} , Ca^{2+} and SiO_2 concentrations of groundwater, throughfall, and stream water are used. Methods of chemical analysis are shown in Section 2.3.

3.3 Analytical methods

3.3.1 End-member mixing analysis (EMMA)

The selection of solutes as tracers was made from the whole potential end-member and stream water data set. First, the solutes' ability to distinguish between end-members was confirmed. Nonparametric Kruskal-Wallis tests were performed using IBM SPSS Statistics 21 (IBM Inc., Armonk, NY, USA) to evaluate differences in concentration between at least one pair of potential end-members (Ali et al.

2010). Second, conservative tracers were evaluated using bivariate plots of solutes against each other. Tracers were considered acceptable if stream water samples were bounded by potential end-members (Liu et al. 2004; Williams et al. 2006).

EMMA was performed to identify the potential geographic sources of stream water, following the procedures of Christophersen and Hooper (1992) and Hooper (2003). All of the stream water geochemical data—including both monthly and storm event samplings—were standardized using their means and standard deviations. Principal components analysis (PCA) was completed for six conservative solutes (SiO_2 , Na^+ , Cl^- , SO_4^{2-} , Mg^{2+} , and Ca^{2+}) of all the stream samples to determine the dimensionality of the data set using IBM SPSS Statistics 21 (IBM Inc., Armonk, NY, USA). The main purpose of PCA is to identify a lower-dimensional space in which most of the observations can be assumed to lie within a specified accuracy. I used the common rule of accepting all principal components with an eigenvalue greater than or equal to 1 (“rule of 1” (e.g. Hooper 2003)). Then, residual analysis was conducted to confirm the dimensionality of the mixing space. For an individual observation of stream water chemistry, a residual is defined by subtracting the original value from its orthogonal projection. According to Hooper (2003), a “good” mixing space is characterized by a random pattern of residuals plotted against the concentration of the original sample. Additional eigenvectors were therefore retained until there was no structure to the residuals. Because a space with n dimensionality is determined by at least $n + 1$ points, at least one more end-member than the dimensionality was needed to explain the stream water mixture. After the number of end-members was defined, I selected end-members. Each solute concentration of a potential end-member was standardized using the mean and standard deviation of the concentrations of all the stream water including both monthly and storm event sampling. They were then projected into the mixing space by multiplying the standardized values by the matrix of retained eigenvectors. When the solutes mix conservatively, the mixture is a convex combination of the end members. Thus, end-members were selected by determining potential end-members, which form a convex polygon to bound most of the stream chemistry (Christophersen and Hooper 1992).

3.3.2 Hydrograph separation

For stormflow samples, the EMMA model was used to calculate the contributions of each end-member by solving the mass-balance equations. To eliminate the effects of any seasonal changes, only end-member samples collected during or immediately after each event were used for hydrograph separations. In some cases, stream observations were outside the mixing domain defined by the selected end-members, which resulted in negative fractions of relative contributions. When this problem occurred, negative fractions were forced to zero, and the other fractions were resolved by a

geometrical approach (Liu et al. 2004; Williams et al. 2006).

3.4 Results

3.4.1 Runoff responses to rainstorms

Runoff varied significantly among the five catchments. Table 3.1 and Figure 3.1 show the annual runoff and the hydrographs from the outlet of each catchment from 2010 to 2012, respectively. Compared to Catchment K, the annual runoff was smaller in Catchments H and M, and larger in Catchments A and R. Catchment H had the smallest baseflow, and runoff ceased during the dry winter season. In this catchment, the hydrological responses to rainstorms were smaller under drier conditions, but became similar to or larger than the other catchments under wet conditions. In contrast, the runoffs from Catchments A, M, R, and K were perennial. Among the five catchments, the smallest hydrological responses to rainstorms under wet conditions were observed for catchment M. Catchments A, R, and K had a larger runoff than Catchments H and M, and Catchment R had the largest peak runoff. Catchment K had a moderate flow compared to the other catchments.

Table 3.1. Annual precipitation and runoff from each catchment.

Year	Precipitation (mm)	Runoff (mm)				
		K	R	M	H	A
2010	1909.3	970.7	—	656.3	383.0	1386.1
2011	1905.2	972.9	1231.5	712.2	335.4	—
2012	1892.0	909.0	1118.0	684.4	412.4	—

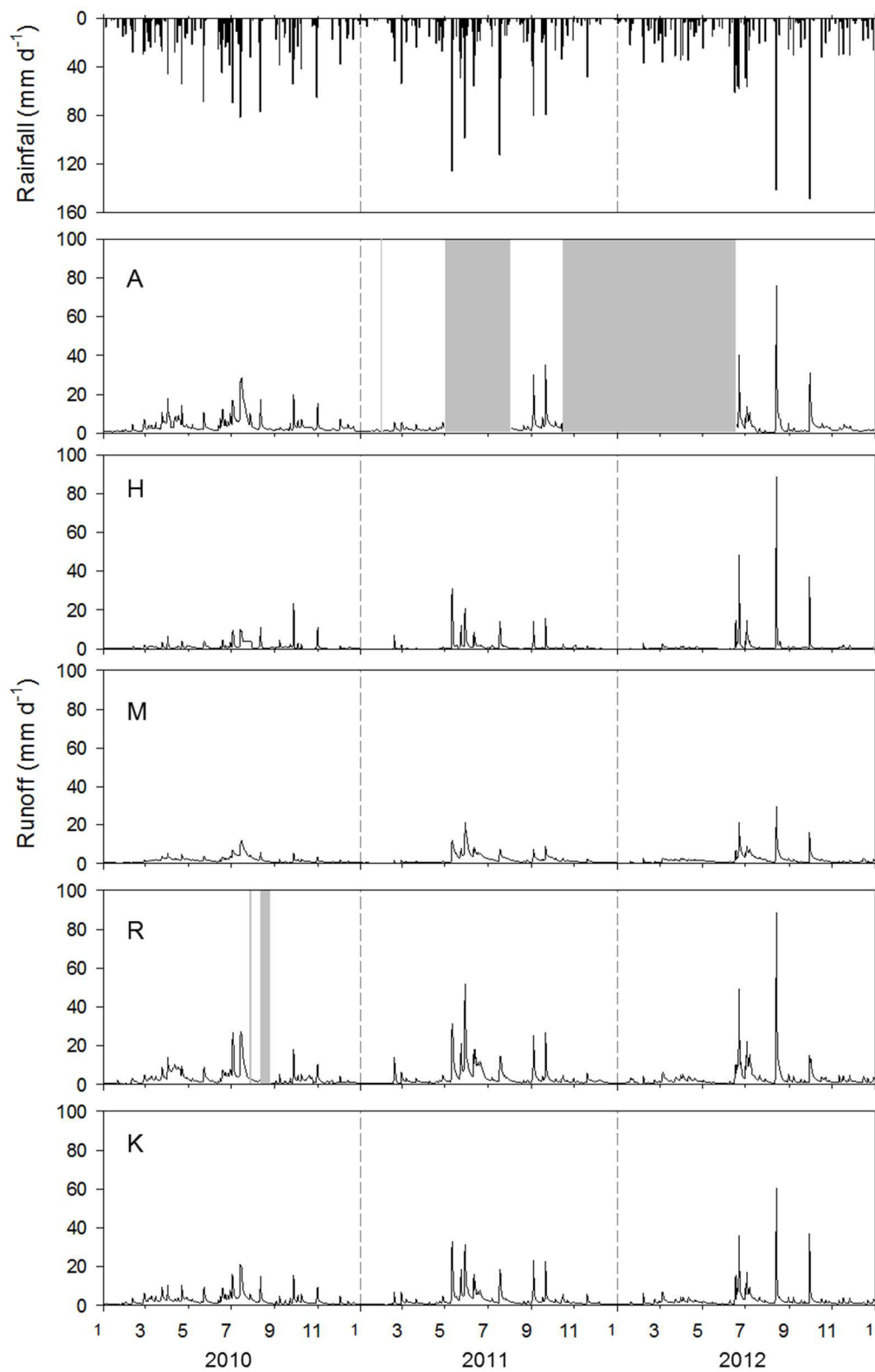


Figure 3.1 Hydrographs from each catchment. For runoff, grey shading indicates periods when data were not available.

3.4.2 Tracer selection

The spatial distributions of the six solutes in the potential end members are shown in Figure 3.2. The concentrations of SiO_2 , Na^+ , Cl^- , and SO_4^{2-} were lowest in throughfall and highest in HBGW or MBGW. The concentrations of SiO_2 , Na^+ , and SO_4^{2-} increased gradually from UGW to HBGW via TGW. The concentrations of Mg^{2+} were highest in UGW. The concentrations of Ca^{2+} were also higher in UGW than throughfall, TGW, SGW, and HBGW, although MBGW had the highest concentrations of Ca^{2+} . The nonparametric Kruskal-Wallis tests confirmed that the concentrations of all six solutes (SiO_2 , Na^+ , Cl^- , SO_4^{2-} , Mg^{2+} , and Ca^{2+}) were significantly different ($p < 0.001$) among at least one pair of the six categories of water samples collected during monthly sampling (i.e., throughfall, UGW, TGW, SGW, HBGW, and MBGW). Therefore, these six solutes could potentially be used to distinguish between end members.

Conservative tracers were evaluated using bivariate plots of solutes. Three examples of the bivariate plots are shown in Figure 3.3. Almost all stream water chemistry was bounded by potential end members for each combination of solutes. The existence of scatter; i.e., lack of a linear structure, for the bivariate plots of Cl^- - Mg^{2+} and SiO_2 - Ca^{2+} (Figure 3.3b, 3.3c) suggests that the dimensionality of the stream water chemistry data was at least two (Hooper 2003). A total of 15 bivariate plots were evaluated, including all potential combinations of paired solutes for SiO_2 , Na^+ , Cl^- , SO_4^{2-} , Mg^{2+} , and Ca^{2+} and it was confirmed that most stream water chemistry was bounded by potential end members for all solute combinations.

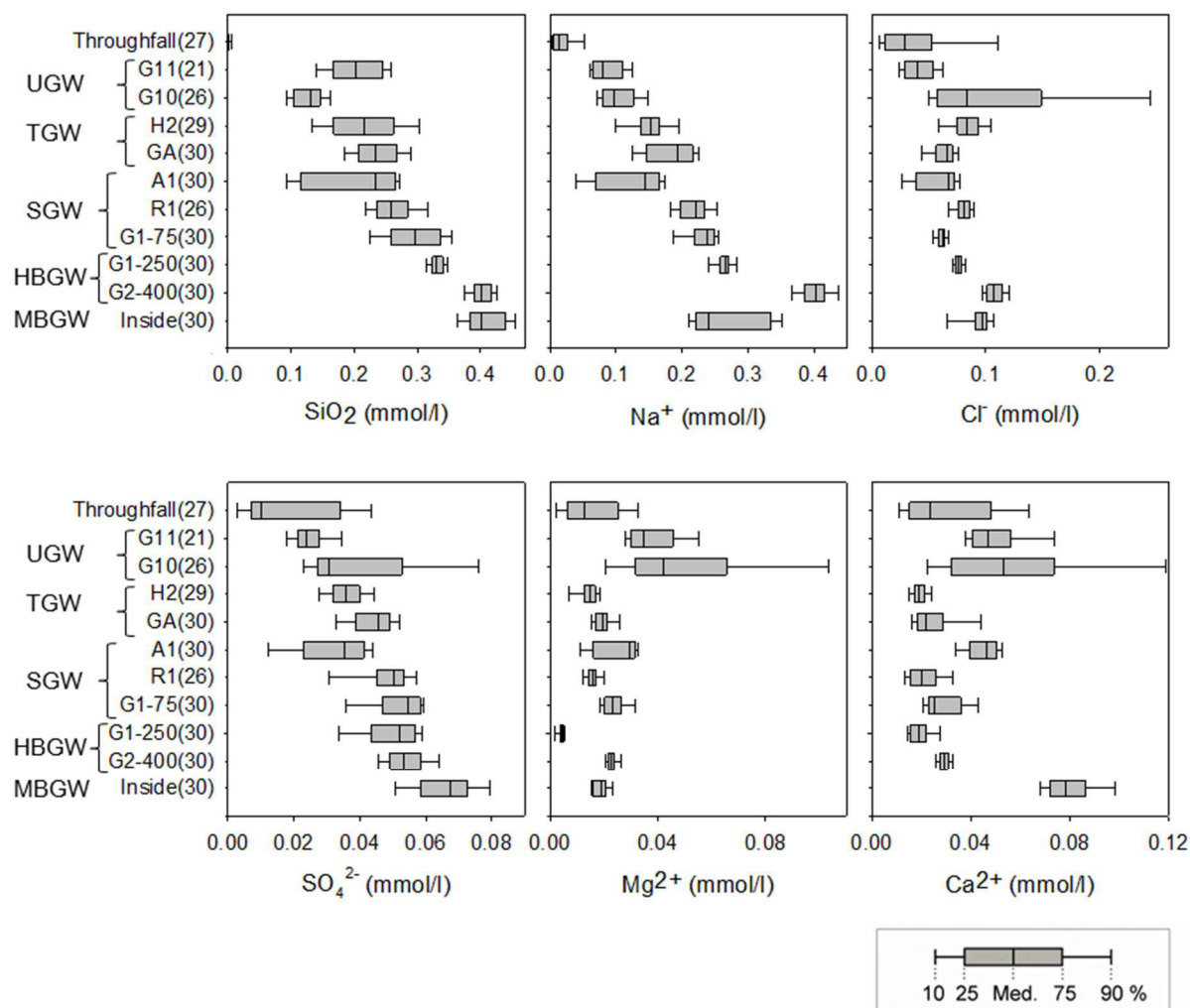


Figure 3.2 Spatial distributions of six solutes in throughfall and groundwater. All the samples were collected in monthly sampling. Values in parentheses are number of samples.

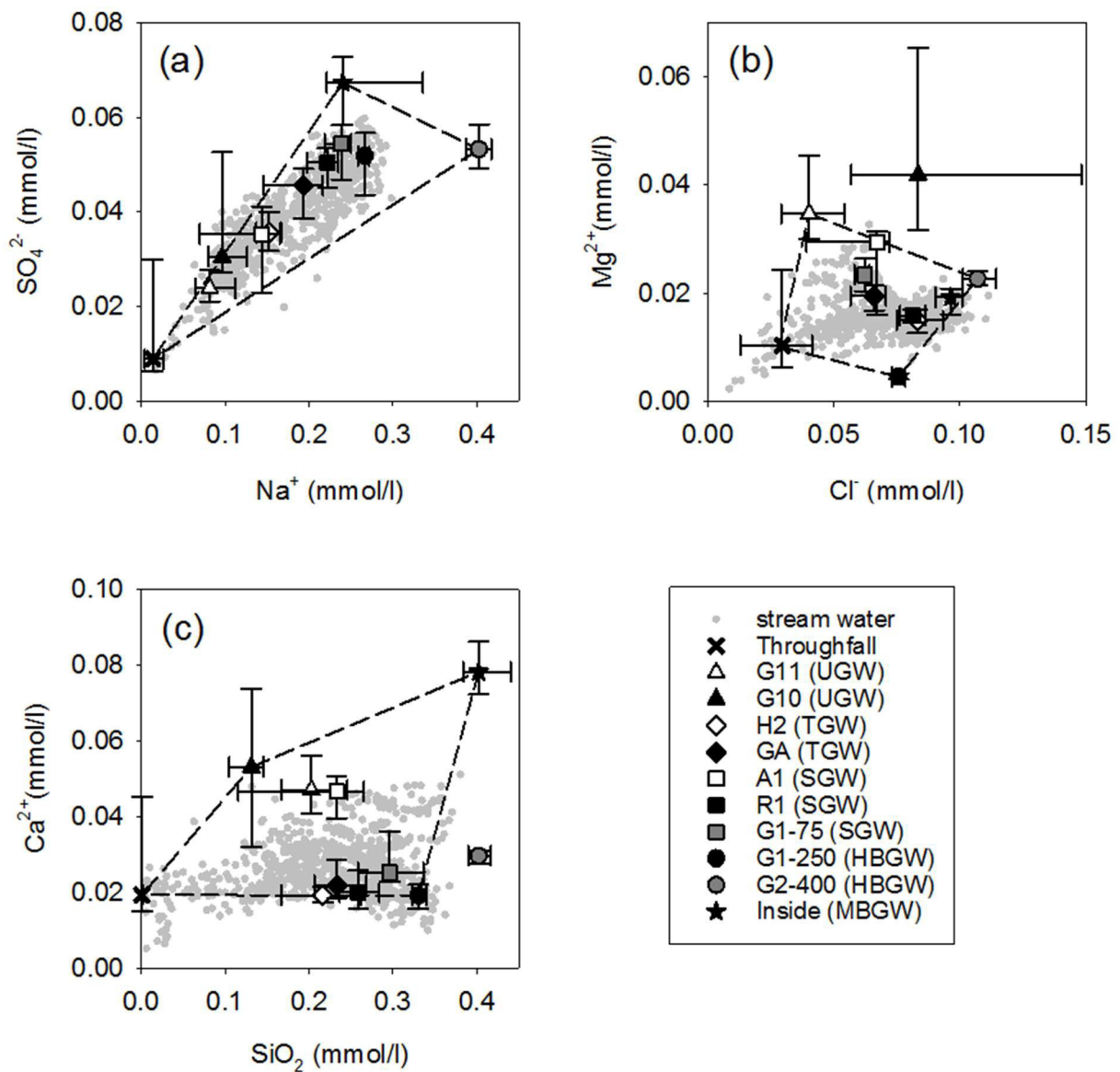


Figure 3.3 Conservative tracers evaluated by plotting medians and quartiles of potential end members along with solute concentrations of all the stream water samples in five catchments. A total of 15 mixing diagrams were evaluated. Here I show the results using (a) Na^+ and SO_4^{2-} , (b) Cl^- and Mg^{2+} , and (c) SiO_2 and Ca^{2+} .

3.4.3 End-member mixing analysis (EMMA)

I estimated the dimensionality of the stream water chemistry data using the six conservative solutes. The PCA results are shown in Table 3.2, which indicates that the first two components should be

retained if the rule of 1 is applied. To further evaluate mixing-space dimensionality, I examined the residuals of the predicted concentration against the observed concentration for each solute. Figure 3.4 illustrates the residuals for individual solutes given a mixing space of two and three dimensions. The residual pattern of Mg^{2+} concentrations displayed a significant linear trend ($r^2 = 0.42$) for a two-dimensional mixing space, although this linear trend was not found for a three-dimensional mixing space ($r^2 = 0.01$). Therefore, retention of a third eigenvector is indicated, suggesting that at least four end members are required.

End members were selected using a mixing diagram defined by the first three principal components (Figure 3.5). All potential end members that were collected monthly were also projected onto the mixing diagrams. Throughfall clearly bounded stream water concentrations on the negative PC1 axis. UGW bounded a positive PC2 axis and negative PC3 axis. PC1 and PC3 increased from UGW to MBGW via TGW, SGW, and HBGW. PC2 decreased from UGW to HBGW, but increased from HBGW to MBGW. Chemical differences between the categories of water samples, such as UGW and HBGW, were sufficiently larger than the error bars in individual sampling sites. Thus, I selected throughfall, UGW, HBGW, and MBGW as end members. Comparing the two UGW sampling wells, groundwater in G11 fell on the line between throughfall and groundwater in G10, indicating that, in the context of EMMA, groundwater in G11 was a mixture of throughfall and groundwater in G10. I also compared the two HBGW sampling wells. Groundwater in G2-400 had a larger PC2 value than groundwater in G1-250, suggesting that groundwater in G2-400 was a mixture of groundwater in G1-250 and MBGW because G2-400 is deeper than G1-250 and reaches the bedrock surface, in contrast to G1-250. Differences in groundwater chemistry between wells in the same category were sufficiently smaller than the differences between the categories. Therefore, I used G10 and G1-250 to represent UGW and HBGW, respectively.

Table 3.2 Results for PCA for all the stream water data.

Principal component	Eigenvalue	Variance explained (%)	Principal component loadings					
			Na^+	Mg^{2+}	Ca^{2+}	Cl^-	SO_4^{2-}	SiO_2
1st	3.35	55.9	0.93	0.22	0.31	0.82	0.9	0.93
2nd	1.25	20.9	-0.19	0.75	0.78	0.05	-0.21	-0.08
3rd	0.79	13.1	0	-0.62	0.51	0.25	-0.28	0.03
4th	0.37	6.1	-0.11	0.09	-0.19	0.51	-0.1	-0.2
5th	0.14	2.4	-0.22	0.01	-0.06	0.03	-0.08	0.29
6th	0.1	1.7	0.18	0.06	-0.06	-0.01	-0.24	0.07

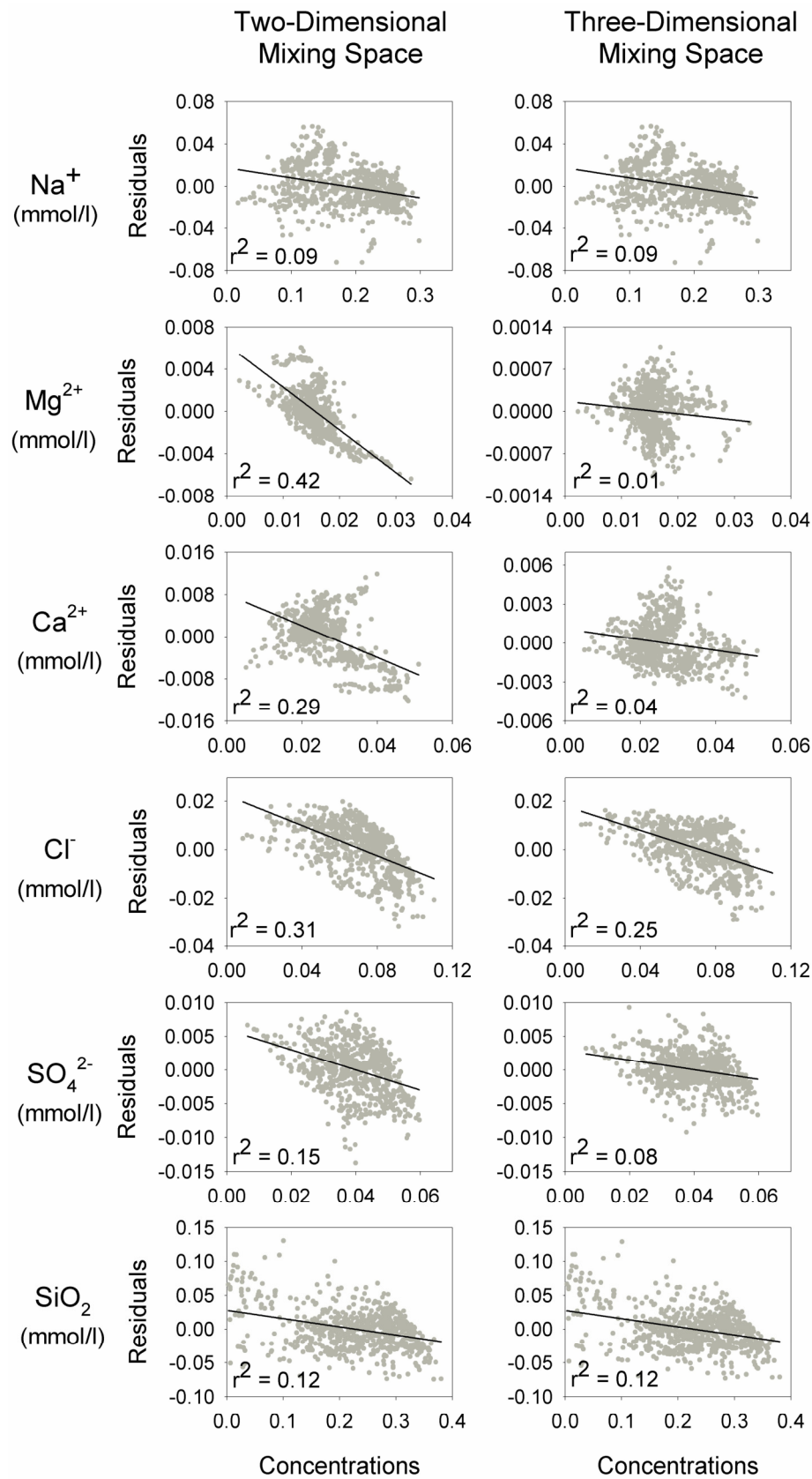


Figure 3.4 Residual plot for the mixing space of two and three dimensions.

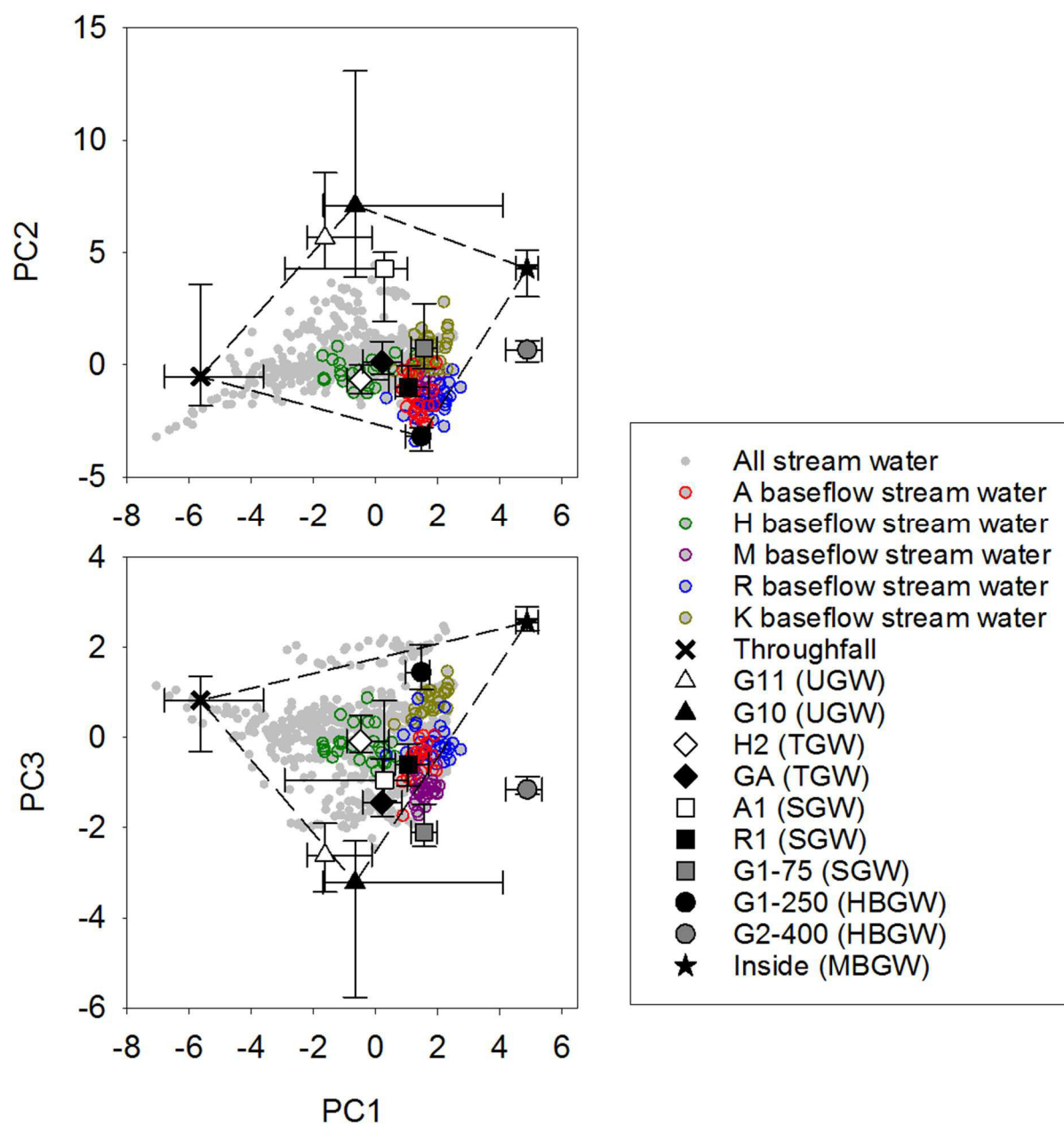


Figure 3.5 Mixing diagrams defined by the first three principal components for all stream water chemistry. For potential end members, symbols indicate the median values and error bars represent the quartiles.

Differences in baseflow stream water chemistry in the five catchments were evaluated using the three-dimensional mixing space (Figure 3.5). Baseflow stream water points in Catchments A, M, and R were located near HBGW. However, the baseflow stream water chemistry shifted from these points toward TGW (i.e., a low value of U1) in Catchment H and toward MBGW (i.e., a high value of U1, U2, and U3) in Catchment K. This means that the contribution of TGW and MBGW to the baseflow stream water was largest in Catchments H and K, respectively.

Mixing diagrams for individual storms are shown in Figure 3.6. End members collected during or immediately after each event were projected onto the mixing diagrams. Almost all stream water points occurred within the convex polygon delineated by four end members in all events. The results of hydrograph separations using the results of EMMA are shown in Figure 3.7, 3.8, and 3.9. The relative contribution of throughfall increased during rainfall in all catchments, and took a longer time to decrease after a rainfall peak in Catchment H than in Catchments A, M, and R. In Catchment H, the relative contribution of UGW decreased when the accumulated rainfall exceeded 200 mm during Events 3-5. Conversely, in Catchments A and M, UGW accounted for a large portion of streamwater after the rainfall peak. In Catchments R and K, the ratio of UGW increased after rainfall in all events, but the increasing rate was much smaller than in Catchments A and M. The relative contribution of HBGW decreased during rainfall in all catchments. After the rainfall peak, it recovered most rapidly in Catchment R and most slowly in Catchment H. In Catchment K, the relative contribution of MBGW was larger than for other catchments during most of the observation period, but it decreased as rainfall continued.

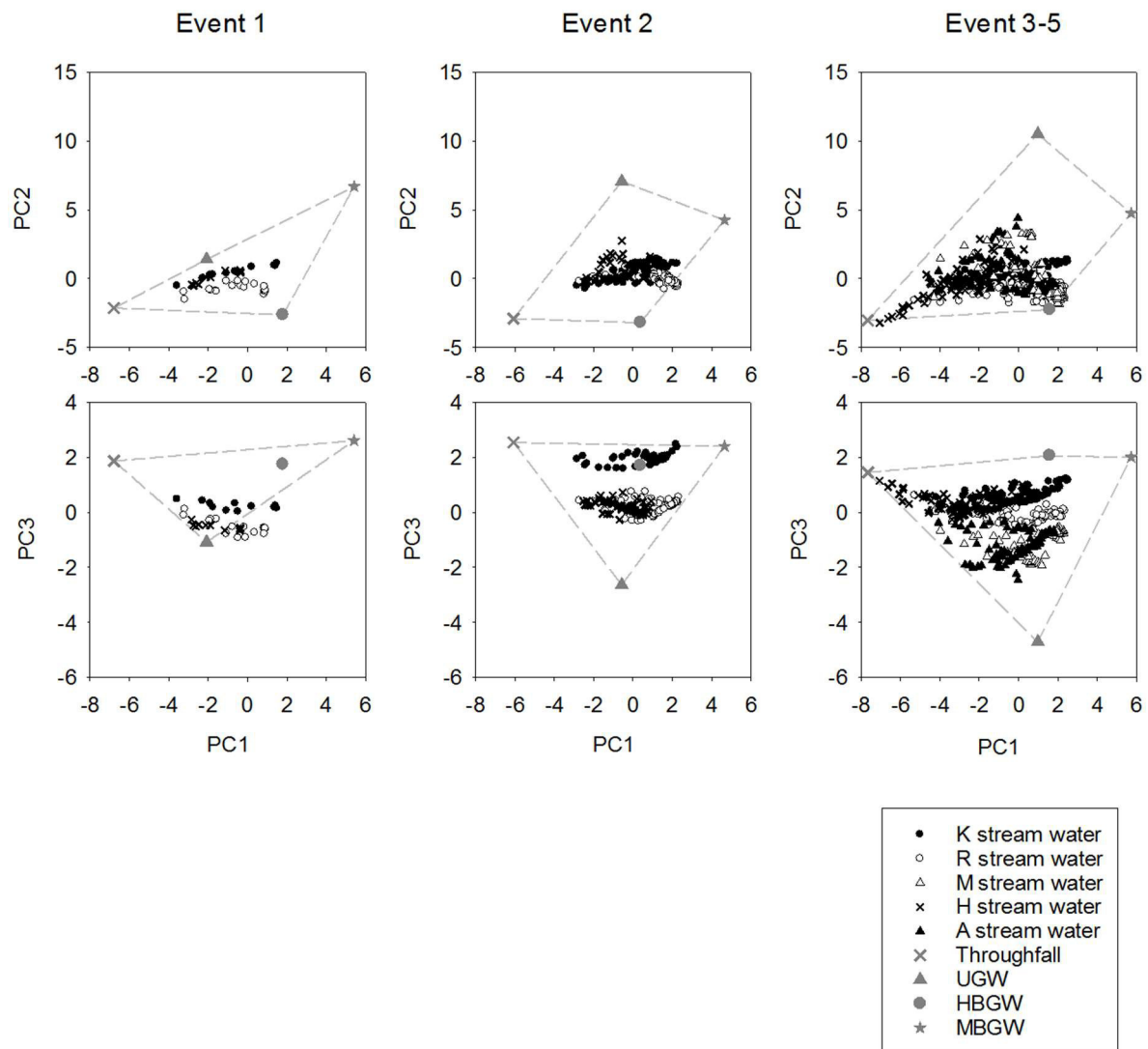


Figure 3.6 Mixing diagrams for individual storms. End members are the samples collected during each rainfall event.

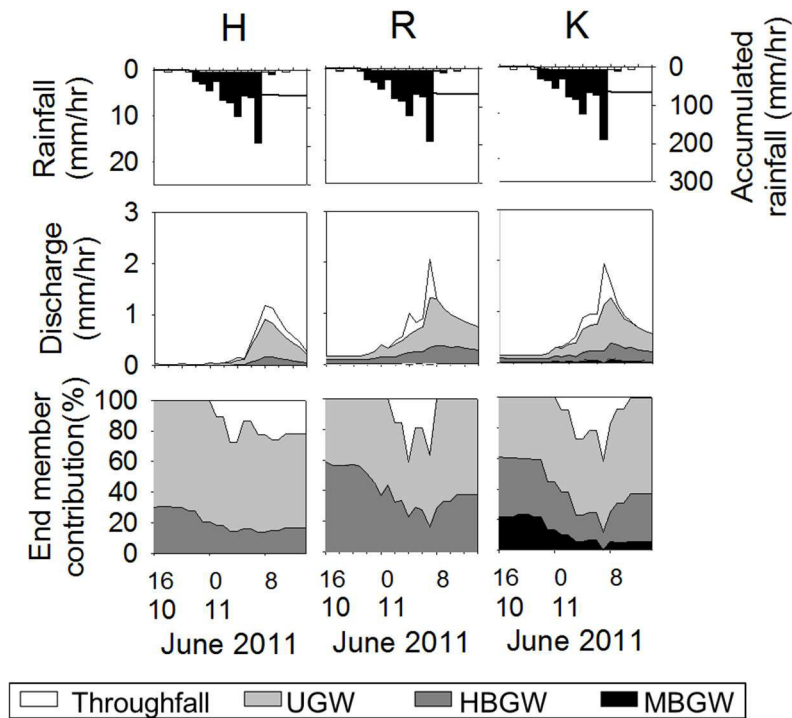


Figure 3.7 Hydrograph separations for Event 1.

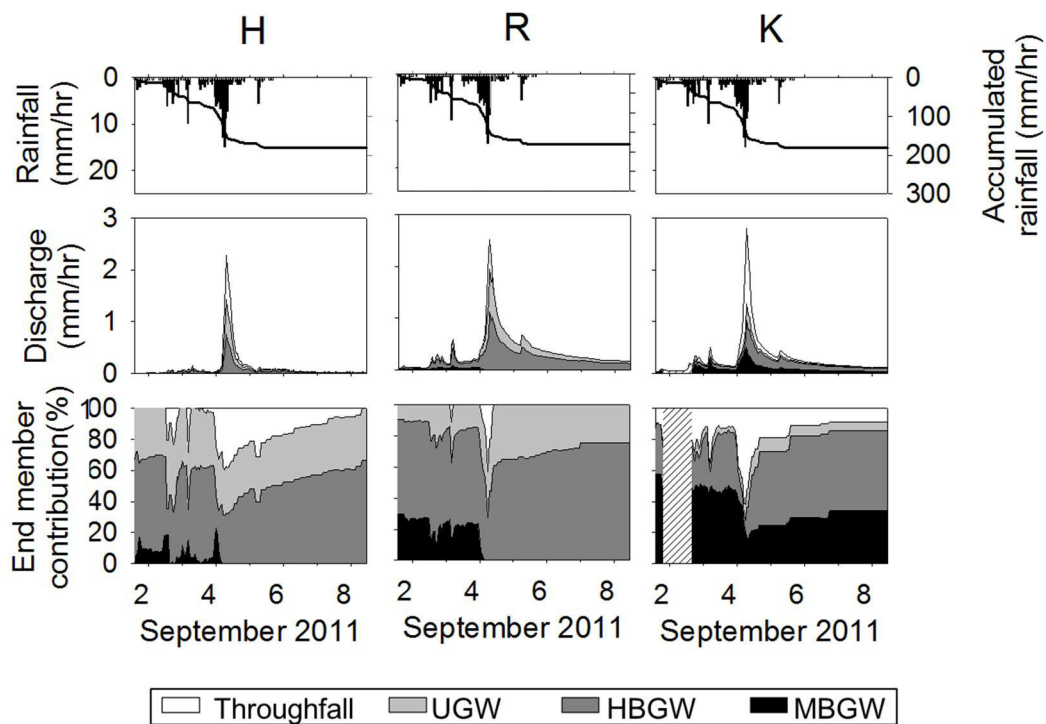


Figure 3.8 Hydrograph separations for Event 2. Slanted line indicates periods when data were not available.

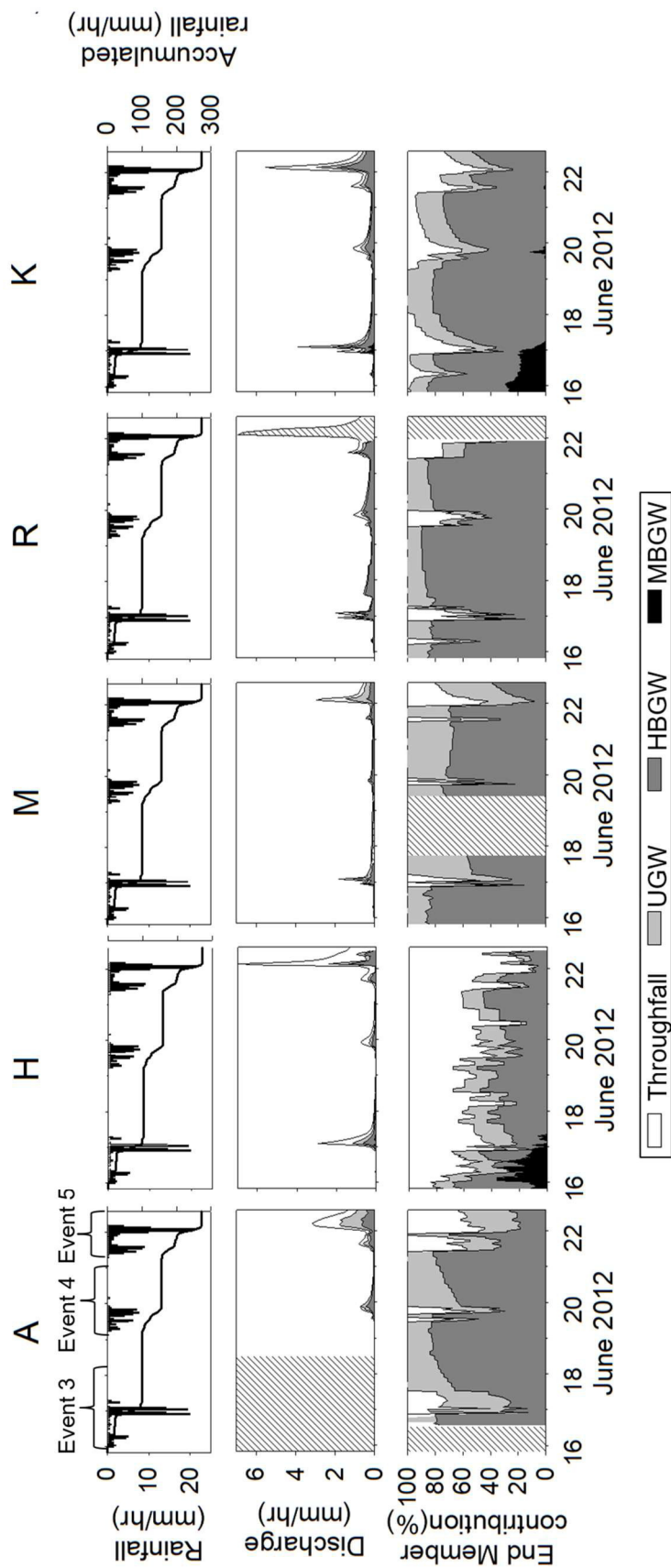


Figure 3.9 Hydrograph separations for Events 3-5. Slanted line indicates periods when data were not available

3.5 Discussion

3.5.1 Comparison of bedrock groundwater contribution into baseflow stream water among catchments

Kosugi et al. (2006) estimated that ca. 35% to 55% of annual precipitation infiltrated into bedrock in a 0.024-ha hillslope zone at Catchment A following a water budget analysis. They also conducted hydrometric observations using soil and bedrock tensiometers and confirmed an infiltrating gradient into the bedrock. Thus, considerable bedrock infiltration occurs through hillslope zones in KEW.

I estimated the geographic sources of stream water during baseflow period from the EMMA results. The contribution of TGW into baseflow stream water was significant in Catchment H, whereas base-flow stream chemistry was similar to that of HBGW in Catchments A, M, and R (Figure 3.5). Catchment H had the smallest annual runoff among the five catchments (Figure 3.1, Table 3.1). After examining the water budget and mean residence time of stream water, Katsuyama et al. (2010) suggested that a larger amount of water is lost from the catchment as deep percolation in Catchment H than A, M, and R. Thus, my results indicate that the contribution of HBGW into baseflow stream water was smaller in Catchment H than in Catchments A, R, and M because of the smaller HBGW discharge within the catchment. In contrast to these four sub-catchments, the contribution of MBGW to baseflow stream water was higher in Catchment K (Figure 3.5). This indicates that MBGW discharged in the area of Catchment K was not covered by the four sub-catchments; i.e., the second-order channel. By measuring runoffs from 19 tributaries, Onda et al. (2006) suggested that an increasing contribution of deep bedrock groundwater occurred with a decrease in altitude during dry periods in Japanese serpentinite catchments. My results suggest that deep bedrock groundwater is discharged slowly at the foot of the mountain, and is added to the baseflow stream water as the fourth end member when upscaling from first- to second-order catchments.

3.5.2 Comparison of bedrock groundwater contribution into stormflow stream water among catchments

Hydrograph separations were performed to determine the differences in the bedrock groundwater contribution into stormflow stream water among catchments. The hydrograph separations were based on the assumption that the concentrations of end-members were both temporally and spatially invariant. I have confirmed that temporal chemical changes within each end member were considerably smaller than the differences in chemistry between end members by projecting all the monthly sampled potential end members into the mixing space (Figure 3.5). I also confirmed that each

end member had the distinguishable chemistries of each category (e.g., MBGW had higher levels of SiO_2 and Ca^{2+} , HBGW had higher levels of SiO_2 and lower Ca^{2+} , UGW had lower levels of SiO_2 and higher Ca^{2+} , and throughfall had lower concentrations of all solutes) (Figure 3.2). Therefore, I considered the effects of temporal and spatial variability in end members to be too small to affect the patterns of geographic source composition in each catchment.

The results of hydrograph separations were compared among the three zero-order catchments. In Catchments A and M, HBGW was dominant during light rainfall, but it decreased, with an increased contribution from UGW, after heavier rainfall (Figure 3.9). Katsuyama et al. (2001) used Na^+ and SO_4^{2-} concentrations to show the increasing contribution of hillslope transient groundwater as the saturated zone spread during rainfall in Catchment M. My findings are consistent with the results of this study. In contrast, in Catchment H, the contribution of UGW and HBGW decreased and throughfall became dominant during Events 3-5 (Figure 3.9). Dusek et al. (2012) reported that more than 60% of subsurface stormflow from hillslopes consisted of pre-event water during most rainfall events, but the contribution of event water significantly increased under continuous rainfall conditions. McGlynn and McDonnell (2003) argued that the riparian reservoir buffered hillslope runoff during small rainfall events, but hillslope runoff displaced riparian groundwater, and became the dominant contributor to the total runoff as rainfall continued. Because the riparian reservoir is recharged by bedrock groundwater during the baseflow period in KEW (Katsuyama et al. 2005), the lower bedrock groundwater contribution to base-flow stream water in Catchment H indicates that the riparian reservoir maintained by bedrock groundwater is smaller in H than in A and M. Thus, saturated throughflow with a high proportion of throughfall under continuous rainfall conditions discharged directly in Catchment H, while it was displaced by shallow riparian groundwater, which is a mixture of UGW and HBGW, in Catchments A and M. As mentioned above, riparian buffering greatly affected the runoff processes in the zero-order catchments despite their extremely short stream channels (Table 2.1). This is probably because a large amount of water was stored in the soil sedimentation area just above the spring point. Although the sedimentation area is formed by a bedrock depression in Catchment M (Katsuyama et al. 2005), it is formed by a soil saving dam in Catchments A and H (Figure 2.2). Therefore, Catchments A and H might have less riparian buffering potential if the dams were not present.

The results of hydrograph separations were compared between zero- and first-order catchments. During rainfall, the contribution of HBGW returned to its pre-event level after peak discharge more rapidly in Catchment R than in the zero-order catchments (Figures 3.7, 3.8, and 3.9), indicating that Catchment R has a higher buffering potential than the zero-order catchments. Based on the results of hydrographs and the mean residence time of streamwater, Katsuyama et al. (2010) suggested that

larger amounts of bedrock groundwater are discharged in Catchment R than in the zero-order catchments (A, H, and M). Additionally, the first-order catchment R has a considerably larger riparian area ratio than the zero-order catchments (Table 2.1). Therefore, a larger amount of bedrock groundwater can be stored in the riparian zone, resulting in Catchment R having a larger riparian buffering potential than the zero-order catchments.

Stream water in the second-order Catchment K received a greater contribution from MBGW than the zero- and first-order catchments during baseflow (Figure 3.5). During rainstorms, the proportion of MBGW decreased in Catchment K (Figures 3.7, 3.8, and 3.9), indicating a smaller contribution of the MBGW component to storms than the other components. Bedrock groundwater dynamics during rainfall differ depending on the bedrock geology. Prior researchers have reported that the contribution of bedrock groundwater decreased during storm events in some catchments (e.g., Walker Branch Watershed in the USA, which is underlain by dolomite (Mulholland 1993), and CB1 catchment in the USA, which is underlain by sandstone (Anderson et al. 1997)), and increased in other catchments (e.g., Ma catchment in Japan, which is underlain by andesite and tuff (Iwagami et al. 2010)). Onda et al. (2006) showed that bedrock flow dominated storm runoff in shale catchments, but subsurface stormflow through the soil layer was dominant in catchments underlain by granite bedrock with a lower primary permeability than shale. Therefore, my results showing a decreasing MBGW contribution during storms could be attributed to the catchment geology being insufficiently permeable to allow a rapid MBGW response to storms.

3.5.3 Implication of bedrock groundwater for upscaling of storm-runoff generation processes

A conceptual model explaining why bedrock groundwater contributions to storm runoff vary with catchment scale is presented based on my findings at KEW. A large portion of rainfall infiltrates into bedrock on the hillslopes. The groundwater that infiltrates into bedrock recharges the perennial riparian groundwater body in zero- to second-order catchments. Exfiltration of bedrock groundwater increases with increasing catchment order and catchments with a greater exfiltration of bedrock groundwater have greater riparian buffering potential. In addition, the recharge by deeper bedrock groundwater flow is larger in second-order channels or the riparian zone along them than in zero- and first-order catchments during the baseflow period. This model indicates that bedrock groundwater influences the upscaling of storm-runoff generation processes by affecting the linkages of landscape units such as hillslopes, riparian zones, and stream channels. The importance of the hydrological connectivity between landscape units for upscaling of storm-runoff generation processes has also been reported in previous studies (e.g. Sidle et al. 2000; McGlynn et al. 2004; Jencso et al. 2009; Jencso et

al. 2010). These studies related the hydrological connectivity to geomorphic factors, such as the size of the upslope accumulated area (Jencso et al. 2009) and the riparian: hillslope area ratios (Jencso et al. 2010). Also, Inamdar and Mitchell (2007) suggested that soil depth should be considered in addition to topographic characteristics when the riparian buffering potential is determined in a watershed. Linkages of landscape units may be controlled largely by topography and soil depth in catchments with poorly permeable bedrock. However, they will also be controlled by bedrock groundwater flow in catchments with permeable bedrock.

My results regarding the changes in the contribution of bedrock groundwater with scale can be attributed to the characteristics of KEW, such as the lack of a balanced water budget due to the highly permeable bedrock and the relatively small area (5.99 ha). Although the water budget will be balanced in catchments with a large area, some studies have reported that bedrock groundwater affects the upscaling of baseflow runoff processes even in catchments larger than KEW. For example, Mulholland (1993) performed EMMA using Ca and SO₄, and revealed that the relative contribution of bedrock groundwater increased when upscaling from a 28.9- to a 38.4-ha catchment in Walker Branch Watershed, USA. In addition, Shaman et al. (2004) examined low-flow runoff in the Neversink River watershed, USA, suggested that the relative contribution of bedrock groundwater to baseflow stream water varied in catchments of less than 8 km². Thus, my results will be applicable to larger catchments than KEW. My results in catchments in which the water budget is not balanced suggest that the changes in the bedrock groundwater contribution into riparian zones or stream channels should be considered when predicting the hydrological and stream chemical response to rain storms in larger catchments.

3.6 Conclusions

End-member mixing analysis (EMMA) was applied to determine geographic source composition during storm events as well as during baseflow periods in neighboring or nested five catchments that ranged in stream order from zero to second. Throughfall, hillslope groundwater, highly weathered bedrock groundwater, and moderately weathered bedrock groundwater were identified as end-members. The perennial riparian groundwater body was recharged by highly weathered bedrock groundwater and/or moderately weathered bedrock groundwater. During baseflow periods and recession periods after rainfall, the contribution of highly weathered bedrock groundwater to stream water was the smallest in the zero-order catchment without perennial riparian groundwater, followed in order by the zero-order catchments with perennial riparian groundwater, and the first-order catchment. In the second-order catchment, bedrock groundwater deeper than that found in the zero- and first-order catchments contributed to stream water during baseflow periods and recession periods

after rainfall. The change of stream geographic source with increasing catchment scale was attributed to the storage of highly weathered bedrock groundwater and/or moderately weathered bedrock groundwater within the perennial riparian groundwater body during the baseflow period. From these findings, it was shown that bedrock groundwater dynamics affect the change of stream geographic source with increasing catchment scale during not only baseflow but also storm flow periods by stored within perennial riparian groundwater body. Thus, when we compare stream water chemistry or discharge among small catchments, we should consider whether the water budget of each catchment is balanced. Moreover, my result suggests that when the results of hillslope- or headwater-scale observations are incorporated into models operated at the watershed scale, not only the changes in geomorphic factors but also three-dimensional water movement, including bedrock groundwater dynamics, must be considered.

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CHAPTER 4

Effects of perennial riparian groundwater body recharged by bedrock groundwater on dominant flow path of peak runoff

4.1 Introduction

Headwater catchments are important source areas for runoff generation. To understand the stream runoff generation processes or the effects of land use on runoff and stream chemistry, detailed hydrological and hydrochemical observations have been made in headwater catchments worldwide. In many studies, small headwater catchments were selected as the study sites because the heterogeneity of rainfall characteristics and bedrock geology can be ignored in this case, and groundwater dynamics can be observed in detail. The scale of observed catchments was especially small, and many studies have been conducted in headwater catchments with areas less than 1 ha in regions with complex geology and topography, such as Japan (e.g. Asano et al. 2002; Haga et al. 2005; Katsuyama et al. 2005; Sidle et al. 2000; Uchida et al. 1999). Observations in small headwater catchments revealed the existence of various runoff processes such as infiltration excess overland flow, saturation excess overland flow, saturated throughflow, preferential flow, and bedrock groundwater flow (e.g. McDonnell 2013; McGuire and Likens 2011). Recently, it was pointed out that catchment classification by runoff characteristics is necessary for generalizing the knowledge obtained from the observations in headwater catchments because the dominant runoff process differs between catchments (McDonnell and Woods 2004; Wagener et al. 2007; Barthold and Woods 2015). For instance, Barthold and Woods (2015) conducted a literature survey and meta-analysis and classified headwater catchments with stream orders less than second-order based on the response time, stream time source, soil physical properties, rainfall properties, geology, groundwater status, and occurrence of overland flow. Although they compared catchments in various regions, it was reported that small headwater catchments have a large variability in runoff and stream chemistry even within the same region (Woods et al. 1995; Temnerud et al. 2007; Asano et al. 2009; Fujimoto et al. 2016). The differences in runoff processes between catchments in different regions and the variability in runoff processes among catchments in the same region should be considered separately. Thus, it is important to identify the factors that affect variability in runoff processes among catchments in the same region.

Some studies pointed out that the organization of landscape units is a key factor that determines water and solute transport from headwater catchments during peak flow (Sidle et al. 2000; McGlynn and McDonnell 2003b; Jencso et al. 2010). Stream channel and riparian zones respond to rainfall in a

different manner as compared to hillslopes (Kendall et al. 1999; Sidle et al. 2000; McGlynn et al. 2003). In many cases, the dominant storm runoff process on hillslopes is subsurface stormflow (Weiler et al. 2006; Penna et al. 2011). Contribution to stream runoff from hillslope subsurface stormflow is likely to be delayed from peak rainfall (Tani 1997; Sidle et al. 2000; Haga et al. 2005). In contrast, channel precipitation rapidly responds to rainfall. Saturation overland flow (Hewlett and Hibbert 1967) and rapid groundwater discharge (e.g. Sklash and Farvolden, 1979) in the riparian zones (riparian runoff) also contributes to stream runoff almost simultaneously with peak rainfall. In addition to runoff generation, groundwater chemistry and biogeochemical processes also differ among hillslopes and riparian zones (Hooper et al. 1998; McGlynn and McDonnell 2003a; Inamdar and Mitchell 2007; Jencso et al. 2010). These results indicate that it is important to understand whether the dominant flow path of peak runoff in a headwater catchment is hillslope subsurface flow or riparian runoff and channel precipitation for the catchment classification.

In Chapter 3, I revealed that bedrock groundwater dynamics caused the differences in runoff source between neighboring headwater catchments during baseflow period. Dominant runoff source was also different during recession period after rainfall because of the different storage amount of bedrock groundwater in perennial riparian groundwater body between the catchments. In contrast, contribution of throughfall was dominant in the all catchments during large rainfall (Figure 3.9). However, similar runoff source does not mean the same flow path between the neighboring catchments. There are several flow paths that throughfall mixes to stream water such as channel precipitation, riparian runoff, and hillslope subsurface flow. Riparian runoff should be larger in catchments with larger perennial riparian groundwater body than in catchments with smaller perennial riparian groundwater body. In our study site, bedrock groundwater also recharged the perennial riparian groundwater body as shown in Chapter 3. Thus, I hypothesized that perennial riparian groundwater body recharged by bedrock groundwater affect dominant flow path of storm runoff.

Dominant flow path of peak runoff may relate to the response time and runoff source. Previous studies indicated that the response time differs depending on whether the dominant runoff process is infiltration excess overland flow or subsurface flow (Dunne 1978) and on the geology (Onda et al. 2001) between headwater catchments. In contrast, Haga et al. (2005) suggested that response time reflects the landscape unit that contributes the most to peak runoff in a catchment. They showed that hillslope subsurface stormflow was dominant and peak runoff was delayed by more than 24 h from peak rainfall during large storms; however, channel precipitation or riparian runoff were dominant, and peak runoff was delayed by less than 2 h from the peak rainfall during small storms in the Mizugaki Research Watershed, Japan. Thus, the runoff response time may reflect the differences in the dominant flow path of storm runoff between catchments. With regard to the runoff source, the reason why runoff

source was similar in spite of different flow path should be examined if dominant flow path of peak runoff was different between the catchments to understand the runoff processes in headwater catchments.

The objective of this chapter is to address the following questions: (1) Does perennial riparian groundwater body recharged by bedrock groundwater affect dominant flow path of peak runoff? (2) How does response time and runoff source relate to the dominant flow path of peak runoff? These questions will be addressed by assessing results from long-term runoff monitoring with groundwater level observations, hydrochemical observations, and terrain analyses in a second-order catchment and its four neighboring subcatchments. In addition to geographic source revealed in Chapter 3, I examined time source as runoff source in this chapter.

4.2 Study area

Observations were conducted in five hydrological observation nests within Kiryu Experimental Watershed (KEW) (Figure 2.2 and Table 2.1): Catchment K (5.99 ha), which corresponded to the entire KEW watershed, and four sub-catchments (R, 1.75 ha; M, 0.68 ha; H, 0.40 ha; and A, 0.086 ha) within Catchment K. Catchment K is a second-order catchment, Catchment R is a first-order catchment, and Catchments M, H, and A are zero-order catchments. Perennial riparian groundwater body does not exist in Catchment H while it exists in Catchments M and A. Size of the perennial riparian groundwater body is larger in Catchment M than Catchment A. In Catchments R and K, perennial riparian groundwater body exists along the stream channel because they are first- and second-order catchments, respectively. Annual bedrock infiltration was larger, and the mean residence time (MRT) of stream water was shorter in Catchment H, whereas annual bedrock infiltration was less and the MRT of stream water was longer in Catchments A, R, and K (Katsuyama et al. 2010). Details of study site are shown in Section 2.1.

4.3 Methods

4.3.1 Data collection methods

In this chapter, I use the data of rainfall and runoff rates from each catchment observed from 2005 to 2013. I also use the groundwater level data monitored from 2011 to 2013 in each catchment. In the subcatchments, groundwater level was monitored just above the spring point (A1 in Catchment A, H1 in Catchment H, G1 in Catchment M, and R7 in Catchment R; Fig. 2.5). In Catchment K, it was monitored at the sedimentary zone just above the weir (K1; Fig. 2.5). In addition, I use the water chemistry data of stream water collected during Events 3, 4, and 5 (Table 2.3). Detailed methods of

hydrological observation, water sampling, and observed rainfall events are shown in Section 2.2.

In this chapter, I use $\delta^{18}\text{O}$ and Na^+ concentrations of throughfall, and stream water. Methods of chemical analysis are shown in Section 2.3.

4.3.2 Analysis of hydrological data

Individual storm events were separated by intervals of at least 24 h without precipitation. I defined peak rainfall or peak runoff rate as the maximum value of rainfall or runoff rate during a storm event. I used the antecedent precipitation index from the peak rainfall (API_{10}) as an index of antecedent moisture conditions:

$$\text{API}_{10} = \sum_{i=1}^{10} (P_i / i) \quad (4.1)$$

where P_i is the total rainfall amounts i days prior to the time of peak rainfall. I analyzed events with more than 10 mm of total rainfall and intensity greater than $2 \text{ mm } 10 \text{ min}^{-1}$. I extracted peak rainfall, API_{10} , and response times from each event in each catchment. Since Barthold and Woods (2015) used the response time calculated by numerous methods following Dingman (2008), three types of methods for calculating this parameter were used in this study. Peak-lag-to-peak, lag-to-peak, and centroid-lag-to-peak times were calculated following Dingman (2008). The peak-lag-to-peak time was the time from the peak rainfall intensity to the peak discharge. The lag-to-peak time was the time from the beginning of precipitation to the peak discharge. The centroid-lag-to-peak time was the time from the centroid of precipitation to the peak discharge. The centroid of precipitation was determined using the following equation:

$$t_{wc} = \frac{\sum_{i=1}^n W_i \times t_i}{\sum_{i=1}^n W_i} \quad (4.2)$$

where t_{wc} = centroid of precipitation, W_i = precipitation for period i , and t_i = time for period i . When the same peak rainfall intensity or peak runoff rate occurred several times in an event, the last time of the peak was used to calculate lag times. The Steel–Dwass test was performed to examine the differences in response times among the catchments. Statistical analyses were performed using R3.2.3 (R Development Core and Team, 2015).

4.3.3 Analysis of hydrochemical data

Stream runoff from each catchment was separated into new and old water components based on the well-known two-component hydrograph separation approach (Pinder and Jones 1969; Sklash and Farvolden 1979). Throughfall samples for each event were used for new water, and stream samples at 1–2 h before each event were used for old water.

4.3.4 Terrain analysis

Each catchment was divided into hillslopes and riparian zones using a digital elevation model (DEM), following Jencso et al. (2010) and Camporese et al. (2014). First, the actual stream channel was mapped based on field observations during the base flow period. Then I estimated riparian zones as the cells where the vertical distance to the channel was less than a certain threshold. The threshold was determined based on groundwater level observations. Hillslopes were mapped as areas except for riparian zones. I conducted terrain analysis using SAGA GIS Version 2.1.2 (<http://www.saga-gis.org/>).

4.4 Results

4.4.1 Annual hydrographs and groundwater level fluctuations

Figure 4.1 shows the hydrographs and groundwater fluctuations from 2012 to 2013 in the five catchments. Groundwater level was always observed within the soil layer in wells in Catchments A, M, and K, though it was not observed during the dry season in Catchments H and R. The groundwater level in Catchments M and K exceeded 50 cm below the soil surface at almost all times and had smaller fluctuations than in the other catchments. The peak runoff rate during large events was larger in Catchments H, R, and K than in Catchments A and M.

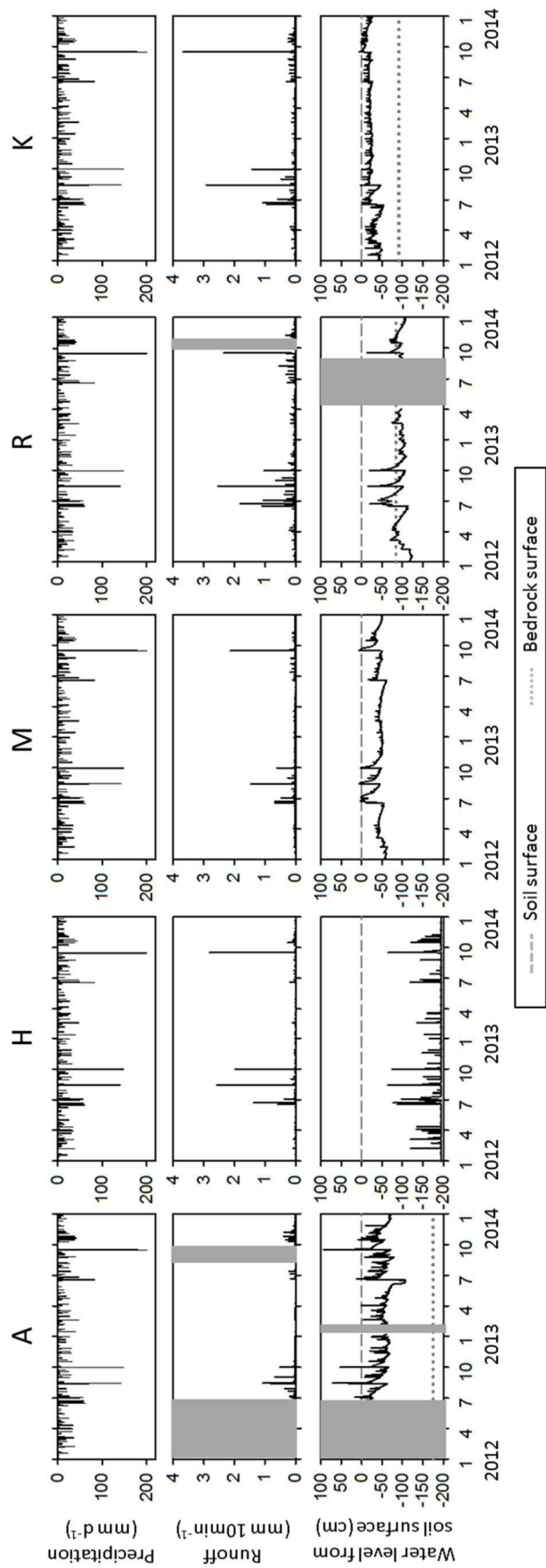


Figure 4.1 Hydrographs and groundwater fluctuations. Grey shading indicates periods when data were not available.

4.4.2 Riparian area derived by DEM

The summary of manual groundwater level observations is presented in Table 4.1. Spatial distribution of groundwater occurrence derived from the data of automatic (Figure 4.1) and manual (Table 4.1) observation was mapped on Figure 4.2. The threshold value of elevation for estimating the riparian zone was set as 1 m. This is because the riparian zone estimated by using 1 m as the threshold corresponded well to the area where the groundwater level was shallower than 50 cm below the soil surface at most observation times (Figure 4.2). The ratio of the riparian area to the catchment area was estimated at 4.2%, 0.6%, 0.7%, 6.1%, and 6.8% in Catchments A, H, M, R, and K, respectively.

Table 4.1 Summary of manual groundwater level observation.

Well	Well depth (cm)	Soil depth (cm)	Frequency of observation time (%)	
			groundwater level > -50 cm	groundwater level > bedrock surface
A5	95	80	0	0
A6	96	80	0	0
H2	77	77	3	41
H3	59	59	0	0
H4	104	104	0	0
H5	27	27	—	0
R1	186	186	0	100
R2	43	43	100	100
R3	44	44	100	100
R5	174	174	0	0
R6	51	51	100	100
R8	197	352	0	100
G2	177	446	11	100
GA	76	74	3	86
GB	82	32	41	24
GC	217	82	0	7
GD	179	30	0	3
GE	185	350	3	100
G10	25	26	3	3
G11	49	36	7	7

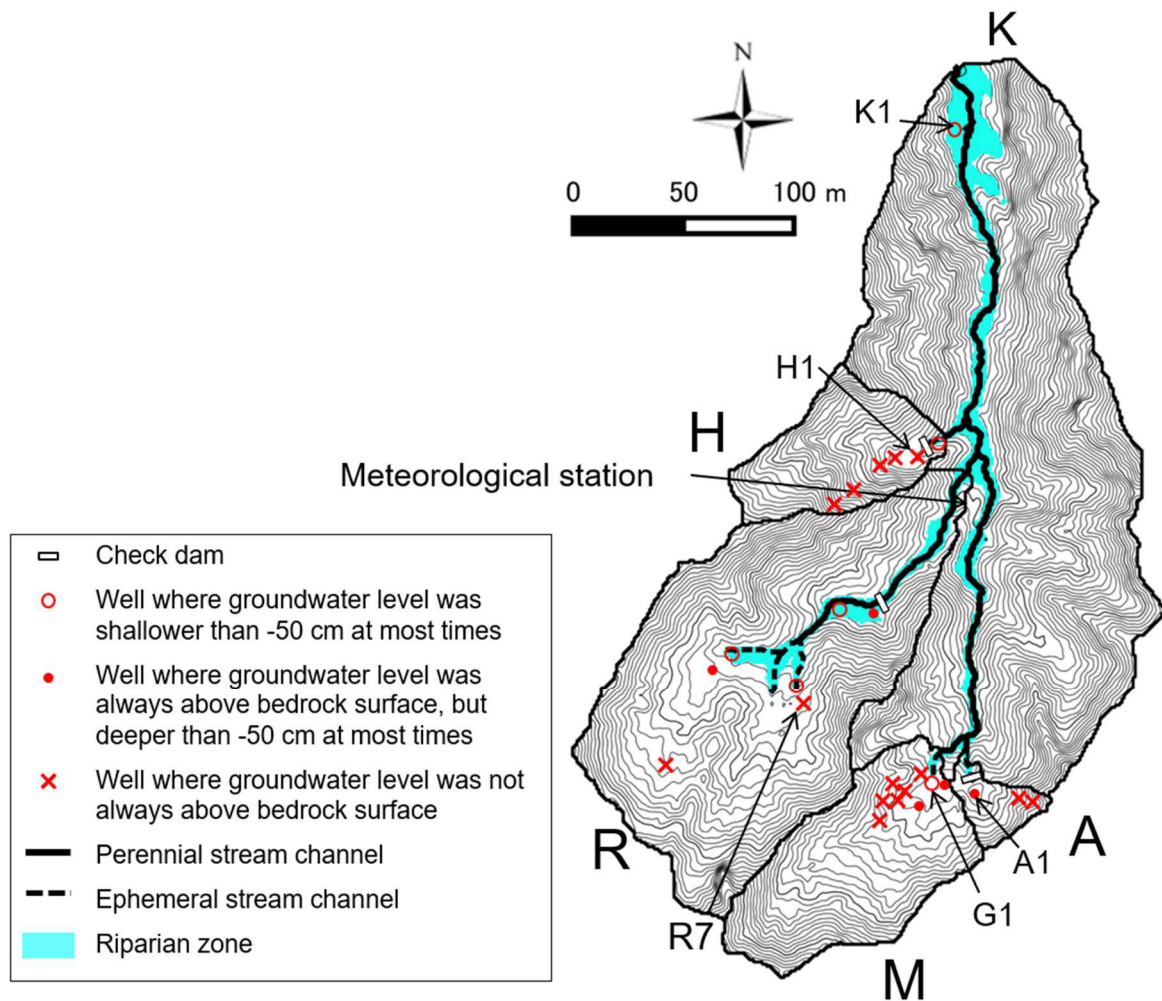


Figure 4.2 Spatial distribution of groundwater during baseflow period and riparian zone in KEW.

4.4.3 Hydrographs and groundwater level fluctuations during rainfall events

To consider the runoff and groundwater level response during rainfall events, the hydrographs and groundwater fluctuations during three events are shown in Figure 4.3. These events had similar peak rainfall rates of 5–8 mm per 10 min, but values of API_{10} were 9.5, 41.4, and 101.9 mm in Events A, B, and C, respectively. In Catchment H, the peak runoff was the largest during Event C, and it was the smallest among the five catchments during Events A and B. Table 4.2 presents the values of peak-lag-to-peak time and the lag time from peak rainfall to peak groundwater level in each catchment during each event. Peak runoff occurred almost simultaneously with peak rainfall and preceded peak groundwater level in Catchments M, R, and K during all events. However, the timings of the peak

runoff lagged behind those of peak rainfall and peak groundwater level in Catchments A and H during Events B and C, though the timings of the peak runoff showed the same pattern in all catchments during Event A. These results indicate that the timing of peak runoff relative to peak groundwater level changed with antecedent moisture conditions in Catchments A and H.

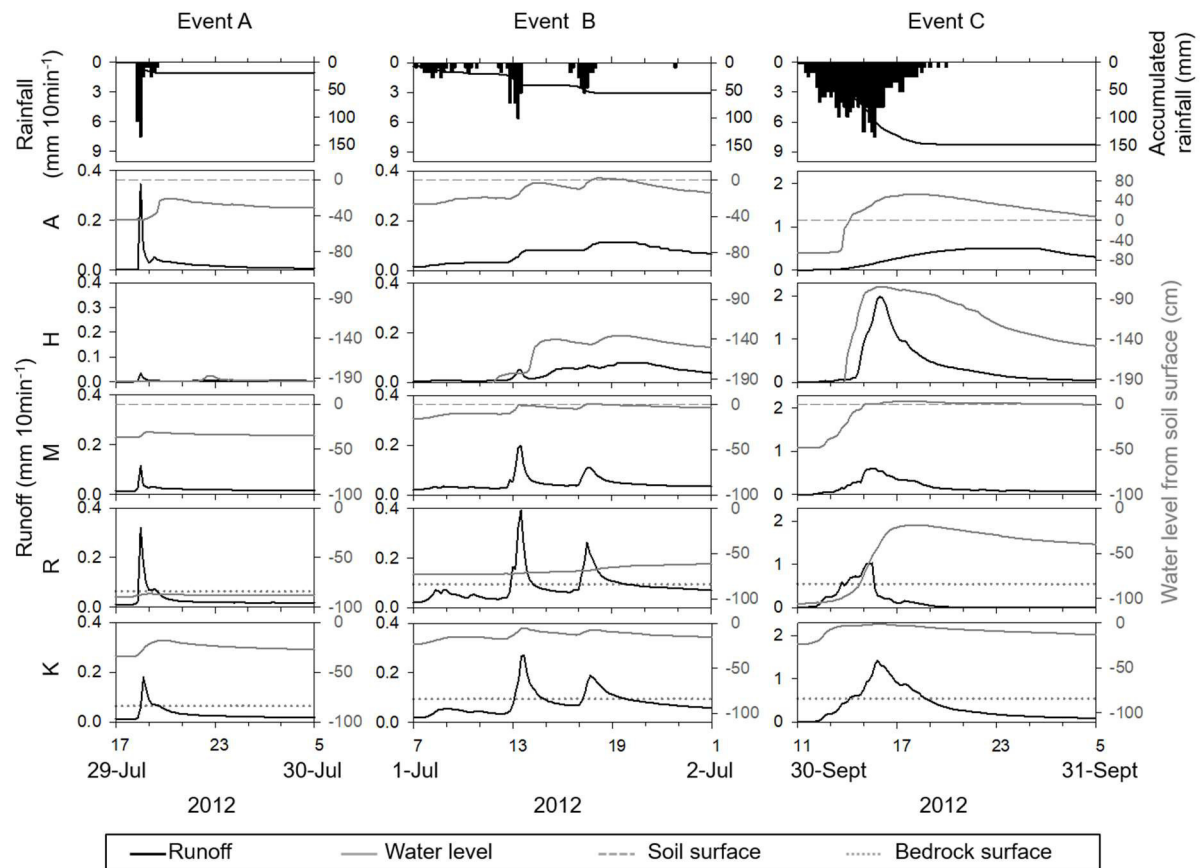


Figure 4.3 Hydrographs and groundwater fluctuations during storm events.

Table 4.2 Peak-lag-to-peak time and lag time from peak rainfall to peak groundwater level during events.

	Peak-lag-to-peak (h)					Lag time from peak rainfall to peak groundwater level (h)				
	A	H	M	R	K	A	H	M	R	K
Event A	0.0	0.0	0.0	0.0	0.2	1.7	4.3	0.5	0.5	1.5
Event B	7.2	7.5	0.2	0.2	0.3	5.0	6.2	4.5	15.2	0.3
Event C	9.7	0.3	0.0	-0.2	0.2	2.7	0.3	1.8	3.0	0.3

4.4.4 Response time

To examine the relation between response time and antecedent rainfall conditions, the frequency distribution for peak-lag-to-peak time in each catchment during events with low (<30 mm) and high (≥ 30 mm) API_{10} are shown in Figure 4.4. The frequency of events with short peak-lag-to-peak times (<1 h) was similar between the events with low API_{10} and high API_{10} in Catchments M, R, and K. In contrast, events had short peak-lag-to-peak times more frequently when API_{10} was less than 30 mm than when it exceeded 30 mm in Catchments A and H. To summarize the differences in response time among the catchments, the median values of the response times for three calculation methods are listed in Table 4.3. When API_{10} was smaller than 30 mm, the median values of peak-lag-to-peak times were less than 2 h in all catchments and did not significantly differ among Catchments H, M, R, and K ($p < 0.05$). In contrast, when API_{10} exceeded 30 mm, the median values exceeded 2 h in Catchments A and H but remained less than 2 h in Catchments M, R, and K. The peak-lag-to-peak time was significantly different in Catchments A and H as compared to Catchments M, R, and K when API_{10} exceeded 30 mm. Further, for $API_{10} > 30$ mm, the centroid-lag-to-peak and lag-to-peak times exceeded 2 h in all catchments, and those in Catchment H did not significantly differ from those in Catchments M, R, and K. This indicates that the results of response time change depending on the calculation methods.

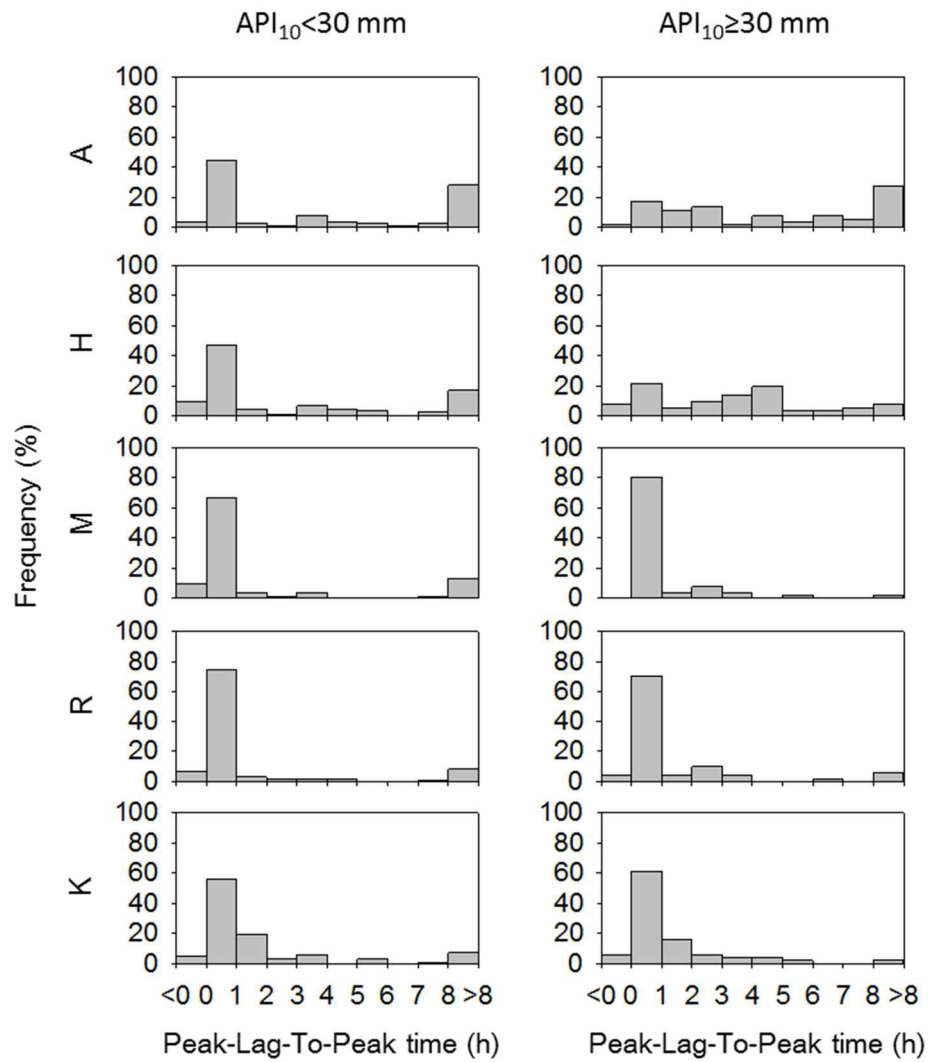


Figure 4.4 Frequency distribution for Peak-Lag-To-Peak time in each catchment during events with low (<30 mm) and high (≥30 mm) API₁₀. Number of events were 99 for API₁₀<30 mm and 51 for API₁₀≥30 mm.

Table 4.3 Median values of response times during events with low (<30 mm) and high (≥ 30 mm) API₁₀.

Calculation method of response time	API ₁₀	Catchment				
		A	H	M	R	K
Peak-lag-to-peak (h)	<30	1.5 a	0.3 ab	0.2 b	0.3 b	0.8 a
Peak-lag-to-peak (h)	≥ 30	4.5 a	3.5 a	0.2 c	0.3 bc	0.5 b
Centroid-lag-to-peak (h)	<30	3.1 a	1.5 ab	0.5 b	0.1 b	0.8 b
Centroid-lag-to-peak (h)	≥ 30	7.6 a	5.1 ab	3.2 b	3.2 b	3.3 b
Lag-to-peak (h)	<30	13.8 a	10.0 ab	7.3 ab	6.8 b	7.7 ab
Lag-to-peak (h)	≥ 30	23.2 a	18.8 a	17.2 a	20.3 a	17.5 a

Number of events were 99 for API₁₀<30 mm and 51 for API₁₀ ≥ 30 mm.

Medians not sharing the same letter are significantly different at $P < 0.05$ (Steel-Dwass test).

4.4.5 Time source of stream water

Figure 4.5 depicts the $\delta^{18}\text{O}$ and Na^+ concentrations of stream water in five catchments and throughfall from June 15 to June 22, 2015. This period was separated by three rainfall events, namely, Events 3, 4, and 5 (see Chapters 2 and 3). Fluctuations of the stream $\delta^{18}\text{O}$ differed among catchments. During Event 3, $\delta^{18}\text{O}$ increased after rainfall in Catchments A and H, while it decreased in Catchments M, R, and K. $\delta^{18}\text{O}$ decreased to values less than the pre-event level during the peak rainfall of Event 4 in Catchments R and K but not in the other catchments. During Event 5, $\delta^{18}\text{O}$ remained low for a longer time after rainfall in Catchments A and H than in Catchments R and K. Hydrograph separation could not be conducted during the second half of Event 3 and Event 4 because the $\delta^{18}\text{O}$ of throughfall did not significantly differ from that of stream water. Na^+ concentrations were significantly different between throughfall and base flow stream water during all events. The concentrations decreased during rainfall but increased to the pre-event level after rainfall during all events in Catchments M, R, and K. In contrast, they took longer to recover to the pre-event level in Catchments A and H than in Catchments M, R, and K. Fluctuations of the new water ratio estimated by $\delta^{18}\text{O}$ is shown in Figure 4.6. The total new water ratios during the first half of Event 3 were 39%, 24%, 22%, and 29% in Catchments H, M, R, and K, respectively. During Event 5, they were 38%, 54%, 43%, and 38% in Catchments A, H, M, and K, respectively. Catchment H had the highest new water ratio, whereas Catchments A, M, R, and K had similar new water ratios in both events. After the events, the new water ratio remained high for a longer period in Catchments A and H than in Catchments M, R, and K.

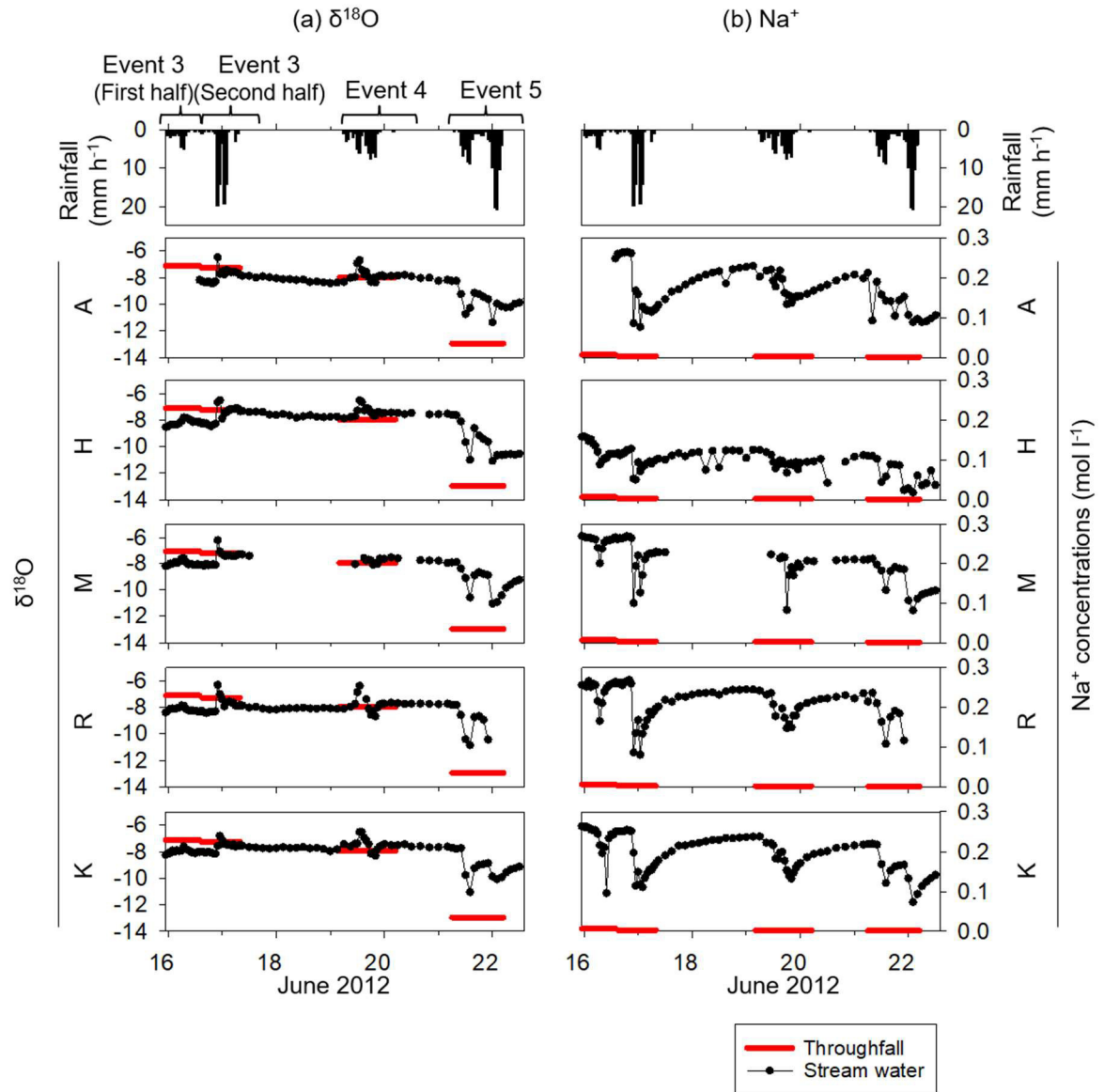


Figure 4.5 $\delta^{18}\text{O}$ (a) and Na^+ (b) concentrations of stream water in five catchments and throughfall during Event 3, 4, and 5.

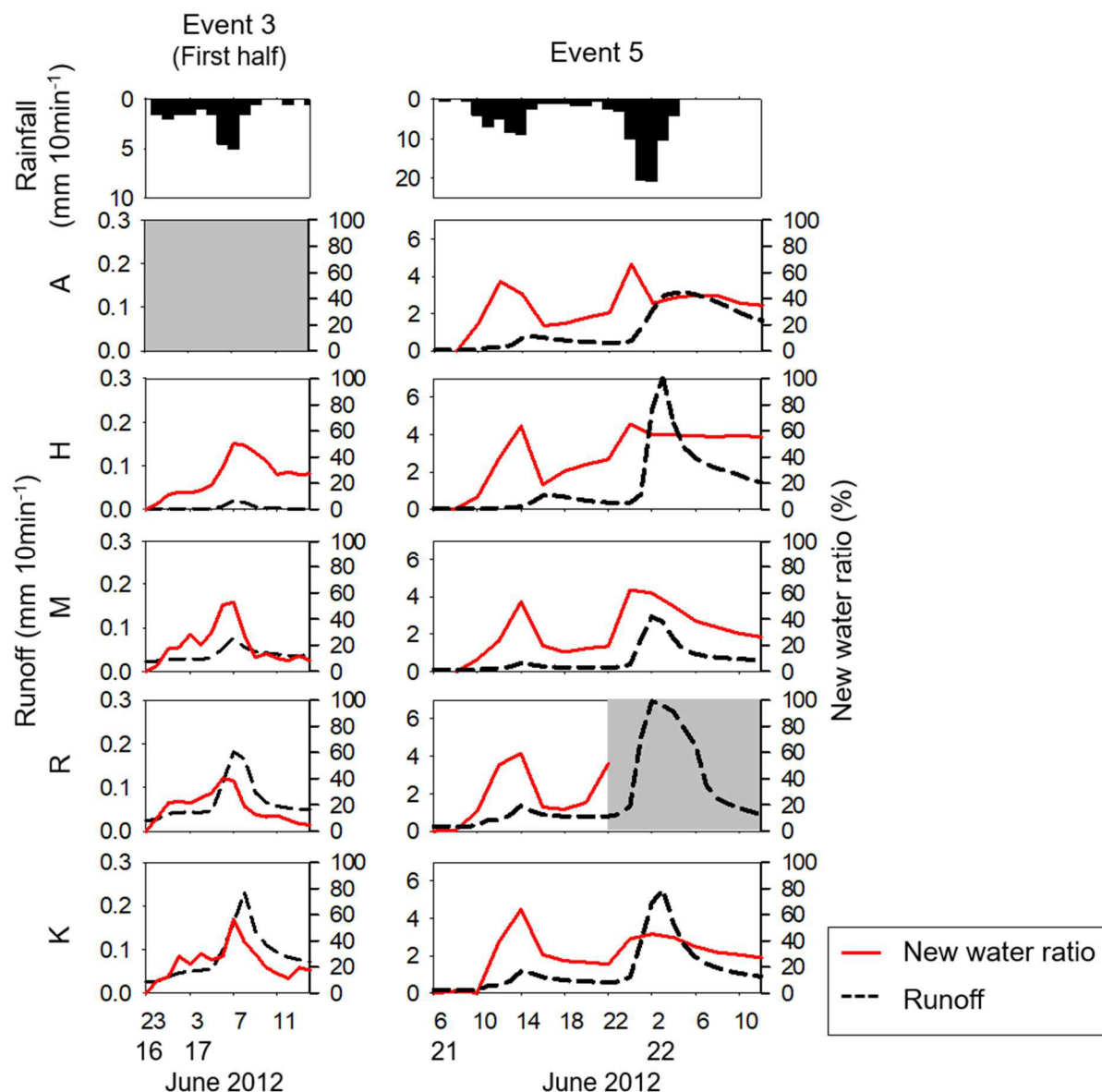


Figure 4.6 Hydrograph and new water ratio during the first half of Event 3 and Event 5. Grey area indicates periods when data was unavailable.

4.5 Discussion

4.5.1 Is the dominant flow path of peak runoff different between neighboring headwater catchments in KEW?

The timing of peak runoff lagged behind that of peak groundwater levels and peak rainfall when the

antecedent moisture condition became wet, though it was faster than that of the peak groundwater level when the antecedent moisture condition was dry in Catchments A and H (Figure 4.3; Table 4.2). In Catchments A and H, channel precipitation and riparian runoff should not be delayed from the peak rainfall because channel and riparian zones only exist near the weir (Figure 4.2). It has been reported that the subsurface flow was activated on hillslopes and became the dominant flow path instead of channel precipitation and riparian runoff as the catchments became wet in many headwater catchments (McGlynn and McDonnell 2003b; Haga et al. 2005; Penna et al. 2011). Thus, the results indicate that the dominant flow path of peak runoff was subsurface storm flow from hillslopes during large rainfall events and channel precipitation or riparian runoff during small rainfall events in Catchments A and H. In contrast to Catchments A and H, the peak runoff occurred earlier than that of peak groundwater level regardless of the antecedent moisture condition in Catchments M, R, and K (Figure 4.3; Table 4.2). This result indicates that the dominant flow path of peak runoff was channel precipitation and riparian runoff regardless of the antecedent moisture condition in Catchments M, R, and K.

4.5.2 Effects of perennial riparian groundwater body recharged by bedrock groundwater on dominant flow path of peak runoff

The riparian area was estimated by taking actual stream channel and spatial distribution of groundwater level into consideration. The riparian area ratio was larger in Catchments R and K than in Catchments A and H, but smaller in Catchment M than in Catchment A (Table 2.1). This does not coincide with the differences in the dominant flow path between catchments. This discrepancy may be caused by smaller hillslope runoff in Catchment M than in Catchment A because of larger soil depth and TPI as well as smaller slope angle (Table 2.1). Another factor that can relate to the discrepancy is the large perennial riparian groundwater body in Catchment M. Katsuyama et al. (2005) reported that 3–8 m of sediment deposits on bedrock depression and groundwater perennially exist within the soil layer at the footslope and riparian zone. They also found that the hillslope above GE (Figure 2.2) did not contribute to peak runoff because the groundwater ridge occurred during large rainfall in the sedimentary zone. These results indicate that the size of perennial riparian groundwater body is a key factor to determine the dominant flow path of peak runoff. In Chapter 3, it was shown that perennial riparian groundwater body was recharged by highly weathered bedrock groundwater and/or moderately weathered bedrock groundwater. Thus, the bedrock groundwater dynamics and soil depth as well as topography relate to the difference in the dominant flow path between catchments.

4.5.3 Relation between response time and dominant flow path of peak runoff

The peak-lag-to-peak time was significantly different between the catchments where hillslope

runoff was dominant and the catchments where riparian runoff and channel precipitation was dominant during large runoff (Table 4.3). This means that peak-lag-to-peak time reflects the difference in dominant flow path of peak runoff between neighboring catchments. In contrast, the centroid-lag-to-peak and lag-to-peak times were not always significantly different between these two types of catchments (Table 4.3). This result was caused because the time from the start or centroid of rainfall to the peak rainfall largely varied depending on rainfall events.

Although the catchments where storm runoff was evaluated by separating hillslope runoff and riparian runoff are scarce, some studies have mentioned the differences in runoff response times between hillslope runoff and riparian runoff in headwater catchments. It was shown that the peak-lag-to-peak time became longer when the dominant flow path changed from hillslope runoff to riparian runoff in a granitic catchment with an area of 0.65 ha (Haga et al. 2005). Penna et al. (2011) also reported that the runoff peaked prior to soil moisture content at hillslope when riparian runoff was dominant, whereas it peaked after the hillslope moisture content when the hillslope runoff was dominant in a 14-ha catchment with underlying dolomite. Storm hydrograph data provided by Sidle et al. (2000) showed that the matrix flow from hillslope peaked after stream runoff from a 2.48-ha catchment with underlying metamorphic rock. Moreover, McGlynn and McDonnell (2003) showed that riparian runoff peaked prior to hillslope runoff by quantifying runoff from each landscape unit in a 2.6-ha catchment with underlying conglomerate. These results suggest that peak-lag-to-peak time of hillslope runoff is slower than riparian runoff regardless of catchment geology. In contrast, McGlynn et al. (2004) pointed out that the timing of peak runoff was delayed in a 280-ha catchment than its subcatchments because the groundwater response in the downstream valley bottom floodplains was delayed from rainfall peak, whereas the headwater riparian zone responded rapidly to rainfall. These results suggest that the slower peak-lag-to-peak time of hillslope runoff than riparian runoff may not occur in larger catchments.

4.5.4 Relation between runoff source and dominant flow path of peak runoff

The new water ratio may have an error because throughfall samples could not be collected with high frequency (Figure 4.5). Nevertheless, the isotope fluctuation data reflect the runoff characteristics of each catchment because they clearly differed between the catchments in spite of the catchments being close to each other (Figure 4.5). Catchment H had the highest new water ratio in both events, while Catchments A, M, R, and K had similar new water ratios (Figure 4.6). In contrast, Catchments A and H had the common feature that the new water ratio continued to be high after rainfall as compared to riparian runoff-dominated catchments (Figure 4.6). In these catchments, Na^+ concentrations remained low for a longer period after rainfall than in Catchments M, R, and K (Figure 4.5). This result also

suggests that hillslope runoff-dominated catchments have higher new water ratios after rainfall than riparian runoff-dominated catchments. In Catchments A and H, new water precipitating in a channel or riparian zone should be drained immediately after rainfall because the channel and riparian zone only exist near the weir (Figure 4.2). This indicates that the new water that contributed to stream water after rainfall in Catchments A and H was included in hillslope runoff. Thus, the results suggest a delayed response of hillslope runoff with significant amount of new water caused a high new water ratio after rainfall in the hillslope runoff-dominated catchments. High new water ratio in hillslope runoff suggests significant contribution of throughfall to hillslope runoff. Dominant contribution of throughfall in all the catchments during large rainfall (Chapter 3) should be attributed to high new water ratio in not only channel precipitation and riparian runoff but also hillslope runoff.

Many studies regarded the new water ratio to be smaller in hillslope runoff than in riparian runoff (McGlynn and McDonnell 2003b; McGlynn et al. 2004; Penna et al. 2016; Saffarpour et al. 2016). However, the present results showed that the new water ratio of stream water in hillslope runoff-dominated catchments was not necessarily smaller than that in riparian or channel-runoff dominated catchments and continued to be high after a rainfall event (Figure 4.6). Uchida et al. (2006) suggested that soil drainable porosity controls the new water ratio of discharged water from wet steep hillslopes. They indicated that hillslope runoff had a larger new water ratio owing to the larger soil drainable porosity in the Fudoji Catchment than in the Maimai Catchment. Since the Fudoji Catchment is located within 10 km from KEW and has the same geology, the high new water ratio in hillslope runoff in the catchments considered in this study should be attributed to a larger soil drainable porosity. Thus, similar runoff source during rainfall peak between catchments with the different dominant flow path may be related to soil physical property in KEW.

4.5.5 Implication for catchment classification

The dominant flow path of peak runoff differed among the neighboring headwater catchments as seen from the present results. Differences among the landscape unit that contribute the most to peak runoff between nested catchments were also reported by McGlynn et al. (2004). They showed that riparian runoff dominated runoff response in 2.6-ha headwater catchments, whereas channel network dominated runoff response at the 280-ha catchment scale. Moreover, Didszun and Uhlenbrook (2008) pointed out that differences in hydrological response units caused a difference in storm runoff response between nested catchments. The catchments smaller than those considered in these previous studies were examined in the present study. The dominant runoff—hillslope runoff or riparian runoff and channel precipitation—was different between neighboring catchments. This means that small headwater catchments should be classified into hillslope runoff-dominated catchments and channel

precipitation- or riparian runoff-dominated catchments when we generalize the observation-based facts.

Devito et al. (2005) proposed to hierarchically classify catchments based on climate, bedrock geology, surficial geology, soil type and depth, and topography. Surficial geology, soil, and topography affect the differences in runoff characteristics among catchments with the same climate and bedrock geology. The present results suggest that the landscape unit that contributes the most to peak runoff is determined by not only topography but also by soil depth and bedrock groundwater dynamics (Section 4.5.2). The peak-lag-to-peak time was useful for assessing which landscape unit controlled catchment runoff as a result of the integration of these factors (Section 4.5.3). In the meta-analysis, Barthold and Woods (2015) used runoff response time calculated by numerous methods with the stream time source, soil physical properties, rainfall properties, and geology. However, the present study suggests that unlike the other indices, runoff response time is useful for classifying catchments in the same region. The present results also show that the lag times estimated from the three methods differed and that only the peak-lag-to-peak time could significantly distinguish the differences in the landscape unit that contributes the most to peak runoff between neighboring catchments (Section 4.5.3). Thus, when classifying headwater catchments, an effective method will be to first assess the differences in flow paths between catchments in the same region based on the peak-lag-to-peak time and then compare the catchments in different regions based on the other indices such as stream time source, soil physical properties, rainfall properties, and geology.

4.6 Conclusions

The dominant flow paths of peak runoff were examined in the neighboring or nested five headwater catchments were compared. In the two zero-order catchments, where a perennial riparian groundwater body was absent or small, the dominant flow path of peak runoff changed from channel precipitation or saturation overland flow and rapid groundwater discharge in the riparian zones (riparian runoff) to hillslope runoff, as antecedent moisture conditions became wet. In contrast, the dominant flow path for peak runoff remained as channel precipitation or riparian runoff even during large rainfall events in the zero-order catchment with a large perennial riparian groundwater body and in the first- and second-order catchments. The size of the perennial riparian groundwater was related to the differences in the dominant flow path of peak runoff between the catchments. In Chapter 3, it was shown that the perennial riparian groundwater body was recharged by highly weathered bedrock groundwater and/or moderately weathered bedrock groundwater. The lag time from peak rainfall to peak runoff was longer in the hillslope runoff-dominated catchments compared to channel precipitation- or riparian runoff-dominated catchments. These results indicate that bedrock groundwater dynamics affect the

dominant flow path of peak runoff by stored within the perennial riparian groundwater body and the effects are reflected by the lag time from peak rainfall to peak runoff.

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CHAPTER 5

Effects of deep percolation on dissolved inorganic nitrogen exports from forested headwater catchments

5.1 Introduction

Nitrogen is an essential nutrient for plant growth, development and reproduction. Dissolved inorganic nitrogen (DIN) export via stream from forest ecosystems is small under conditions lacking disturbance because nitrogen (N) is a limiting nutrient to most forest ecosystems. However, DIN export from forest ecosystems increases in response to the disturbances such as harvesting practices (Likens et al. 1970; Pardo et al. 1995; Swank et al. 2001) and insect defoliation (Swank et al. 1981; Houle et al. 2009; Tokuchi et al. 2013). Furthermore, it has been reported that elevated atmospheric N deposition due to industrial and agricultural activities during 20th century also increases DIN export from forest ecosystems as a consequence of causing N saturation, which is defined as a condition that occurs when the availability of inorganic N is in excess of biotic demand (Aber et al. 1989; Ohrui and Mitchell 1997). Increasing DIN export from forest ecosystems can cause a deterioration of drinking water because a high nitrate level in drinking water can have toxic effects such as methemoglobinemia (Scatena 2000). It can also affect the aquatic ecosystem as a consequence of two phenomena: surface water acidification and eutrophication (Stoddard 1994). Thus, it is necessary to evaluate N budget in forest ecosystems.

“Small watershed approach” is a common method to evaluate N budget in forest ecosystems (Likens and Bormann 1995). Small watershed approach is a method to evaluate the net gain or loss of a nutrient in the forest ecosystem from measurements of its input in precipitation and its output in streamflow in forested headwater catchments based on the assumption of negligible water loss as groundwater. In Hubbard Brook Experimental Forest where small watershed approach originated, negligible water loss as groundwater was confirmed by a geological survey, water budget, and chloride budget (Likens and Bormann 1995). Although N cycle may be affected by transport as dissolved organic N and gaseous exchange such as N fixation and denitrification, DIN output in streamflow can sufficiently reflect N status in forest ecosystems (Aber et al. 2002). Thus, DIN budget was evaluated by the observation of DIN input in precipitation and DIN output in streamflow at many forested headwater catchments around the world (Likens et al. 1970; Swank et al. 1981; Likens and Bormann 1995; Campbell et al. 2004; Tokuchi et al. 2013).

Based on a concept by Graham et al. (2010), we define two components, the bedrock infiltration

and the deep percolation at the catchment scale as follows; the bedrock infiltration is water or solutes that infiltrate into the bedrock. On the other hand, the deep percolation is a partial of the bedrock infiltration which does not re-emerge into the stream channel as bedrock return flow, but leaves the catchment not detected at the weir. Negligible water loss through deep percolation was the fundamental assumption in small watershed approach. However, previous studies have revealed that deep percolation of water cannot be ignored in granitic forested headwater catchments in Kiryu Experimental Watershed (KEW), Japan (e.g. Kosugi et al. 2006; Katsuyama et al. 2010). Conceptual diagram of runoff generation processes in KEW revealed by previous studies is shown in Figure 5.1. In KEW, highly weathered bedrock layer and moderately weathered bedrock layer exist between soil layer and weakly weathered bedrock layer: Weakly weathered bedrock has almost no effective porosity, whereas highly weathered bedrock has enough effective porosity to allow matrix flow (Katsura et al. 2009). Katsuyama et al. (2004) observed water budget and showed that approximately 49% of the annual rainfall infiltrated into highly weathered bedrock on a 0.024 ha hillslope plot in KEW (Arrow 2 in Figure 5.1). Bedrock infiltration of water at the hillslope was also confirmed by tensiometric observations (Kosugi et al. 2006). Katsuyama et al. (2005) observed groundwater level and groundwater chemistry in a sub-catchment in KEW and found that water infiltrating into bedrock recharged groundwater body within highly and moderately weathered bedrock (Point c in Figure 5.1). They also found that a partial of bedrock groundwater re-emerged into riparian soil layer as bedrock return flow (Arrow 3 in Figure 5.1) and discharged as stream water (Arrow 5 in Figure 5.1) after mixing with saturated throughflow from hillslope soil layer (Arrow 4 in Figure 5.1). Bedrock groundwater that did not re-emerge as bedrock return flow leaves the catchment via bedrock as deep percolation (Arrow 7 in Figure 5.1): Katsuyama et al. (2010) reported that annual deep percolation of water calculated from annual water budget was ranged from 5.9 to 39.4% of annual precipitation in five sub-catchments in KEW. In Chapter 3, I performed end-member mixing analysis (EMMA) and pointed out that a partial of deep percolating water discharged into outside the catchment (Arrow 8 in Figure 5.1; hereafter, it is referred to as “interbasin bedrock groundwater discharge”).

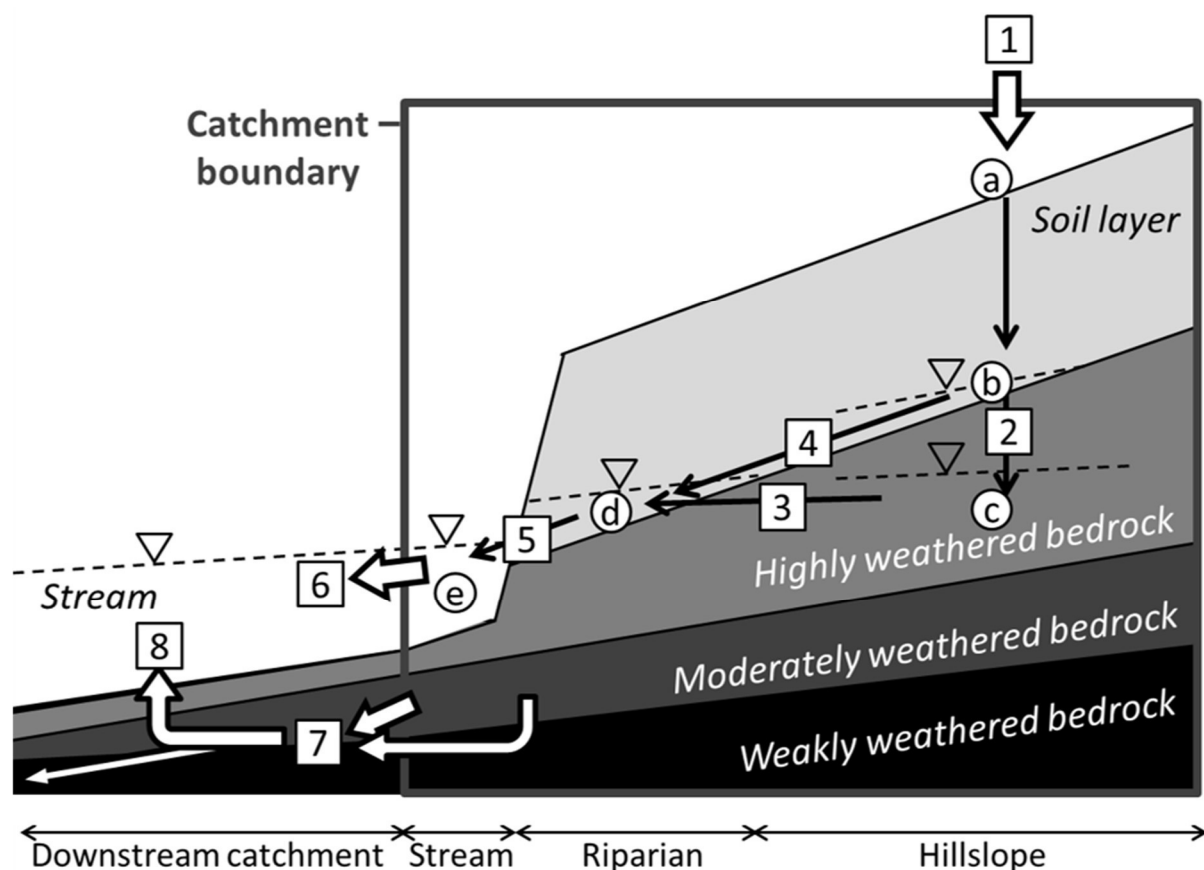


Figure 5.1 Conceptual diagram of runoff generation process in the Kiryu Experimental Watershed. Alphabets in the figure indicate water sampling points: a, throughfall; b, hillslope groundwater; c, bedrock groundwater; d, riparian groundwater; e, stream water. Numbers of arrows indicate different kinds of water flow: 1, precipitation input; 2, bedrock infiltration; 3, bedrock return flow; 4, saturated throughflow; 5, discharge; 6, stream output; 7, deep percolation; 8, interbasin bedrock groundwater discharge.

Considerable amount of deep percolation was also reported in many other forested headwater catchments as reviewed in Section 1.1.3 (Table 1.4; Figure 1.1). Oda et al. (2013) observed the chloride mass balance at three forested headwater catchments underlain by Cenozoic sedimentary rock, and showed that deep percolation of water ranged from -51 to 23% of annual precipitation. Stewart et al. (2007) studied the long-term water balance in a small (3 ha) headwater catchment with greywacke/argillite bedrock in New Zealand, and reported that the deep percolation of water corresponded to 51% of the precipitation. Verry (2003) analyzed the data of stream discharge response to tree harvesting treatment in 32 forested headwater catchments and showed that annual deep percolation of water was 45% of streamflow in average. Thus, deep percolation is a significant

component of water and nutrient budgets for many forested headwater catchments.

In addition, water deep percolating in upper catchments significantly contributes to water or solute budgets in downstream catchments as interbasin bedrock groundwater discharge at many sites around the world (Winograd 1962; Maxey 1968; White 1979; Jones 2002; Genereux et al. 2005; Güler and Thyne 2006). Genereux et al. (2005) investigated the water and chemical budgets and showed that interbasin bedrock groundwater discharge accounted for about two thirds of the water input and about 97% of the input of six solutes (Cl^- , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} , Ca^{2+}) in a lowland catchment in Costa Rica. Shaman et al. (2004) suggested that the interbasin bedrock groundwater discharge increased with increasing catchment size by examining low-flow runoff from 11 nested catchments in Neversink River watershed, where Burns et al. (1998) found that bedrock groundwater with high NO_3^- concentrations discharges as perennial spring. Since interbasin bedrock groundwater discharge has different flowpath and residence time from stream discharge, it may have a significant impact on the response of downstream drinking water quality and aquatic ecosystems to upstream forest disturbances. Thus, it is necessary to discuss where deep percolating DIN discharges.

To clarify the effects of deep percolation on DIN export, the deep percolation of DIN was quantified based on water budget and spatial distributions of DIN concentrations along with the water flow path and where deep percolating DIN discharged was discussed in two neighboring catchments where a different deep percolation of water was reported by Katsuyama et al. (2010), and different bedrock groundwater contributions to stream water during both baseflow and storm flow periods were found in Chapter 3. To identify the causes of differences in deep percolation between the catchments, I discussed the water re-emerged into soil layer from bedrock (i.e. bedrock return flow) using Na^+ as a tracer of water flow path, because it is the dominant cation of chemical weathering and its concentration increases with depth during vertical infiltration in our study area (Ohte et al. 1995).

5.2 Material and methods

5.2.1 Study area

The water and DIN budgets were examined in two subcatchments within KEW: Catchments H (0.40 ha) and R (1.75 ha) (Figure 2.2). Catchments H and R are zero- and first-order catchments, respectively. Although the streamflow was perennial in catchment R, runoff from catchment H ceased during the dry season when there was only a small amount of rainfall. Katsuyama et al. (2010) reported that the mean annual deep percolation of water was larger and the mean residence time of stream water was shorter in Catchment H than in catchment R. Perennial riparian groundwater body does not exist in Catchment H while it exists along the stream channel in Catchment R. Details of the study site are shown in Section 2.1.

5.2.2 Data collection methods

In this chapter, the data of rainfall and runoff rates from each catchment observed from 2010 to 2012 were used. The groundwater level data monitored from 2011 to 2012 in each catchment were also used. Groundwater level was monitored just above the spring point (H1 in Catchment H and R7 in Catchment R; Figure 2.5). In Catchment K, it was monitored at the sedimentary zone just above the weir (K1; Figure 2.5). In addition, the water chemistry data of groundwater, throughfall, and stream water were used. Groundwater was classified into five categories: groundwater at unsaturated zone during baseflow period (UGW), transient saturated zone groundwater (TGW), shallow saturated groundwater (SGW), highly weathered bedrock groundwater (HBGW), and groundwater in the moderately weathered bedrock (MBGW). Groundwater, throughfall, and stream water was collected monthly from August 2010 to December 2012. The stream water from the monthly sampling was treated as baseflow stream water, except for that collected when the 24-hour antecedent rainfall was greater than 10 mm. Stream water was also collected during three rainfall periods (Table 2.3). Detailed methods of hydrological observation, water sampling, and observed rainfall periods are shown in Section 2.2.

In this chapter, I use the Na^+ concentrations of groundwater, throughfall, and stream water. Methods of chemical analysis are shown in Section 2.3.

5.2.3 Analytical methods

Annual deep percolation of water in each catchment was estimated by subtracting the annual runoff and evapotranspiration from annual precipitation. I used evapotranspiration estimated by Matsumoto et al. (2011) using the eddy covariance method from data collected at the meteorological tower in KEW (Figure 2.2). The eddy covariance method is currently recognized as one of the most direct and reliable methods of measuring evapotranspiration from forest catchments (e.g. Baldocchi and Ryu 2011). Since long-term evapotranspiration data estimated by the eddy covariance method are not available for many sites, it is my advantage that I could estimate deep percolation of water using such a reliable annual evapotranspiration data. Over the period of 2001 to 2008, the annual evapotranspiration ranged from 715 to 780 mm (Matsumoto et al. 2011). I used this range (715-780 mm) as the range of annual evapotranspiration.

The DIN input in throughfall was estimated by multiplying DIN concentrations of throughfall by the throughfall flux for each monthly sample. The DIN output was divided into two categories: DIN output in streamflow and deep percolation of DIN.

$$L = L_S + L_D \quad (5.1)$$

where L is the total DIN output ($\text{g-N ha}^{-1} \text{ yr}^{-1}$), L_S is the DIN output in streamflow ($\text{g-N ha}^{-1} \text{ yr}^{-1}$), and

L_D is the deep percolation of DIN ($\text{g-N ha}^{-1} \text{ yr}^{-1}$). L_S was estimated using a regression equation. I used the L-Q equation (Ide et al. 2007; Kaneko et al. 2007) as the regression equation:

$$L_S(t) = a Q_S(t)^b \quad (5.2)$$

$$L_S = \Sigma L_S(t) \quad (5.3)$$

where $L_S(t)$ and $Q_S(t)$ are the DIN output in streamflow ($\text{g-N ha}^{-1} \text{ h}^{-1}$) and stream runoff (mm h^{-1}) at time t , respectively, and a and b are empirical parameters. The parameters a and b were determined from both monthly and event-based sampling data in each catchment using IBM SPSS Statistics 21 (IBM Inc., Armonk, NY, USA). The range of L_D was estimated by multiplying the range of annual deep percolation of water by the range of mean DIN concentrations in bedrock groundwater, because deep percolating water leaves the catchment as bedrock groundwater (Figure 5.1).

5.3 Results

5.3.1 Annual water budget

Table 5.1 lists the annual water budget in each catchment from 2010 to 2012. In 2010, annual runoff and deep percolation of water from Catchment R were not available because the discharge data from R were gapped for 18 days. The annual runoff in Catchment R ranged from 1118.0 to 1231.5 mm and corresponded to 59–65% of the annual precipitation. In contrast, the annual runoff in Catchment H ranged from 335.4 to 412.4 mm, which corresponded to 18–22% of annual precipitation and was considerably smaller than in Catchment R. The annual deep percolation of water from 2010 to 2012 was 699.6–854.8 mm and -106.3–59.0 mm, which corresponded to 37–45% and -6–3% of annual precipitation in Catchment H and R, respectively.

Table 5.1 Annual water budget in Catchments H and R.

Year	Precipitation (mm)	Runoff (mm)		Deep percolation (mm)			
		H	R	H		R	
				Min	Max	Min	Max
2010	1909.3	383.0	—	746.3	811.3	—	—
2011	1905.2	335.4	1231.5	789.8	854.8	-106.3	-41.3
2012	1892.0	412.4	1118.0	699.6	764.6	-6.0	59.0

5.3.2 Spatial distributions of Na^+ and DIN concentrations in monthly sampling data

Seasonal variations of Na^+ and DIN concentrations in UGW, TGW, SGW, stream water, and bedrock groundwater were shown in Figure 5.2 and 5.3, respectively. Na^+ concentrations of UGW in Catchments H and R were lower than 4 mg L^{-1} at all times. Stream water and SGW (R1 and R6) in Catchment R had Na^+ concentrations higher than 4 mg L^{-1} , while stream water in Catchment H and TGW (H1 and R7) in Catchments H and R had Na^+ concentrations lower than 4 mg L^{-1} at most of the sampling time. Na^+ concentrations of bedrock groundwater were higher than 4 mg L^{-1} at all times. DIN concentrations higher than 1 mg-N L^{-1} were observed only at UGW and TGW in Catchments H and R. Stream water in Catchment R had lower DIN concentrations than that in Catchment H at all the time when its runoff was smaller than 0.2 mm h^{-1} .

The mean DIN concentrations in throughfall, groundwater, and baseflow stream water are shown in Figure 5.4. In both Catchments H and R, the mean DIN concentration was highest in UGW (H5 in H and R5 in R) and gradually decreased during the runoff process from hillslope to stream via the riparian zone. The mean DIN concentration of baseflow stream water was 0.36 mg-N L^{-1} in H, while it was 0.15 mg-N L^{-1} in R. The mean DIN concentrations in bedrock groundwater ranged from 0.09 to 0.36 mg-N L^{-1} .

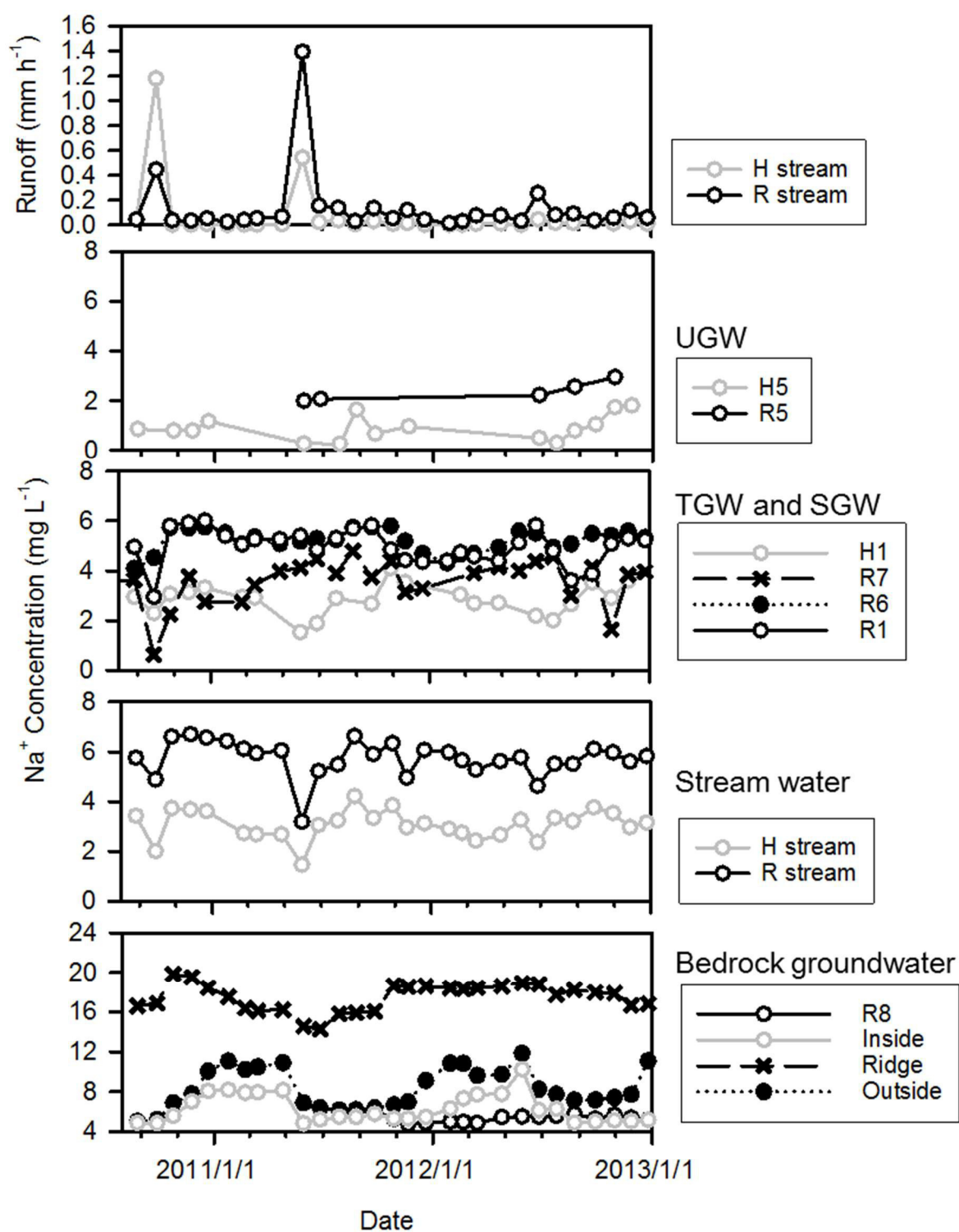


Figure 5.2 Runoff at every monthly sampling time and seasonal variations of Na^+ concentrations in groundwater, riparian groundwater, stream water, and bedrock groundwater.

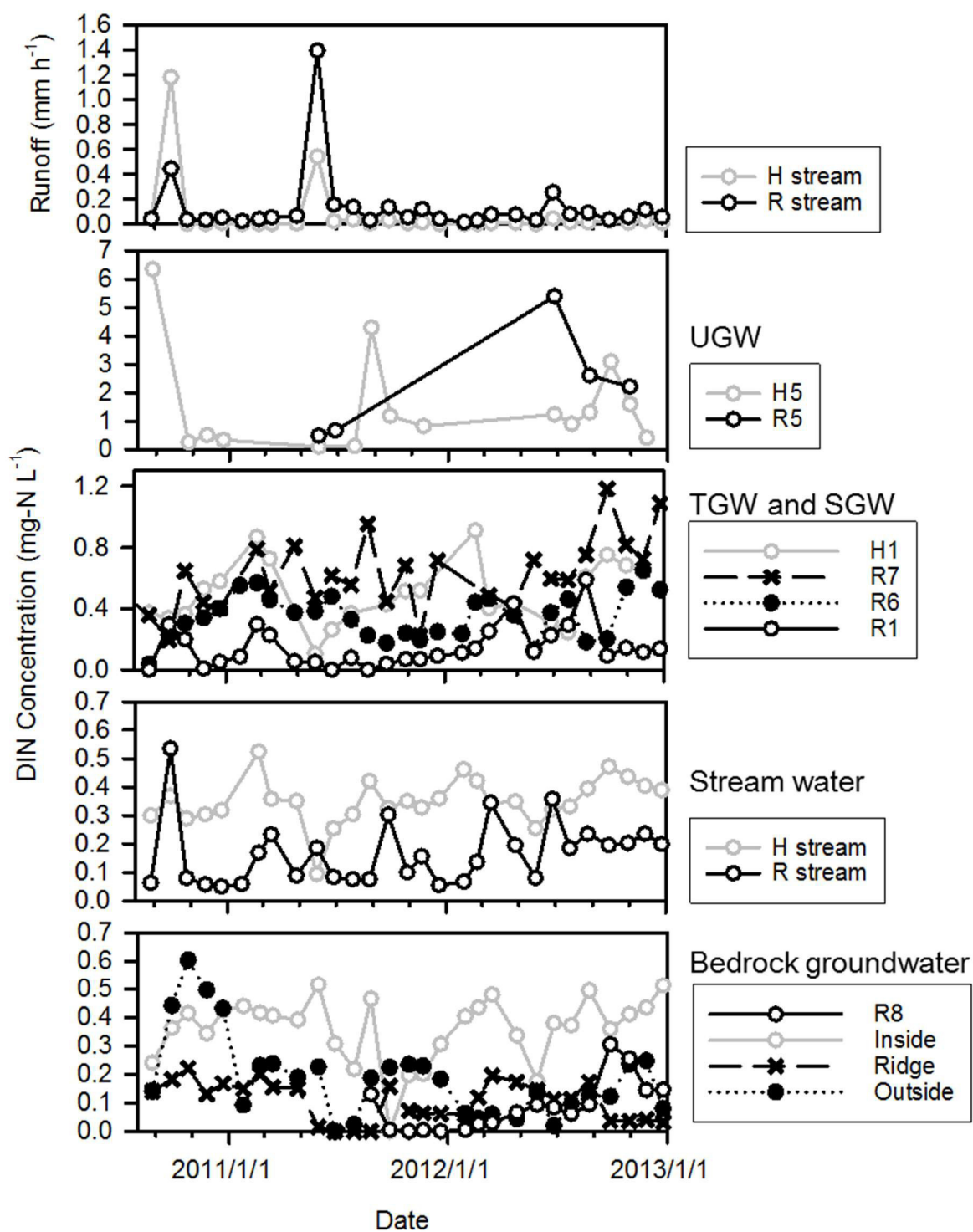


Figure 5.3 Runoff at every monthly sampling time and seasonal variations of DIN concentrations in groundwater in the soil layer, stream water, and bedrock groundwater.

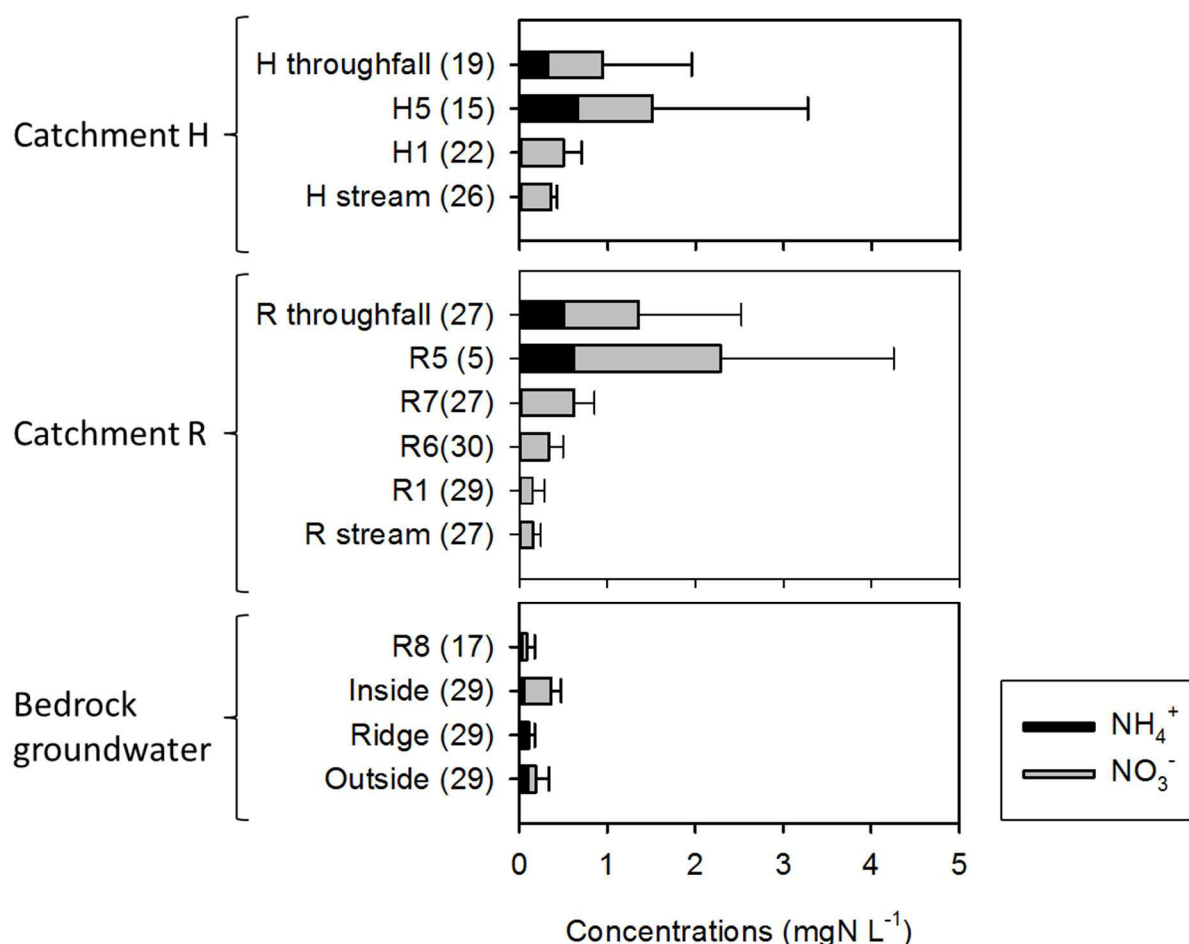


Figure 5.4 Mean concentrations of DIN (NH_4^+ and NO_3^-) in throughfall, groundwater, and baseflow stream water. Error bars represent standard deviations of DIN concentrations. Values in parentheses are number of samples.

5.3.3 Stream DIN concentrations during rainfall events

Storm flow DIN concentrations in Catchments H and R are shown in Figure 5.5. There was little NH_4^+ detected in both catchments throughout the entire period. In all rainfall events, the NO_3^- concentrations before rainfall were approximately 0.3 mg-N L^{-1} in Catchment H and 0.1 mg-N L^{-1} in Catchment R. Before accumulated rainfall reached 100 mm, the NO_3^- concentration in Catchment R remained lower than 0.3 mg-N L^{-1} in all the events. However, when accumulated rainfall exceeded 100 mm during Events 2 and 3, the NO_3^- concentration in Catchment R was simultaneously elevated to

more than 0.4 mg-N L^{-1} with the rising riparian groundwater level. On the other hand, the NO_3^- concentration after Events 1, 2, 4, and 5 in Catchment H was similar to or less than the concentration before rainfall, although it elevated to more than 0.4 mg-N L^{-1} during Events 2 and 3.

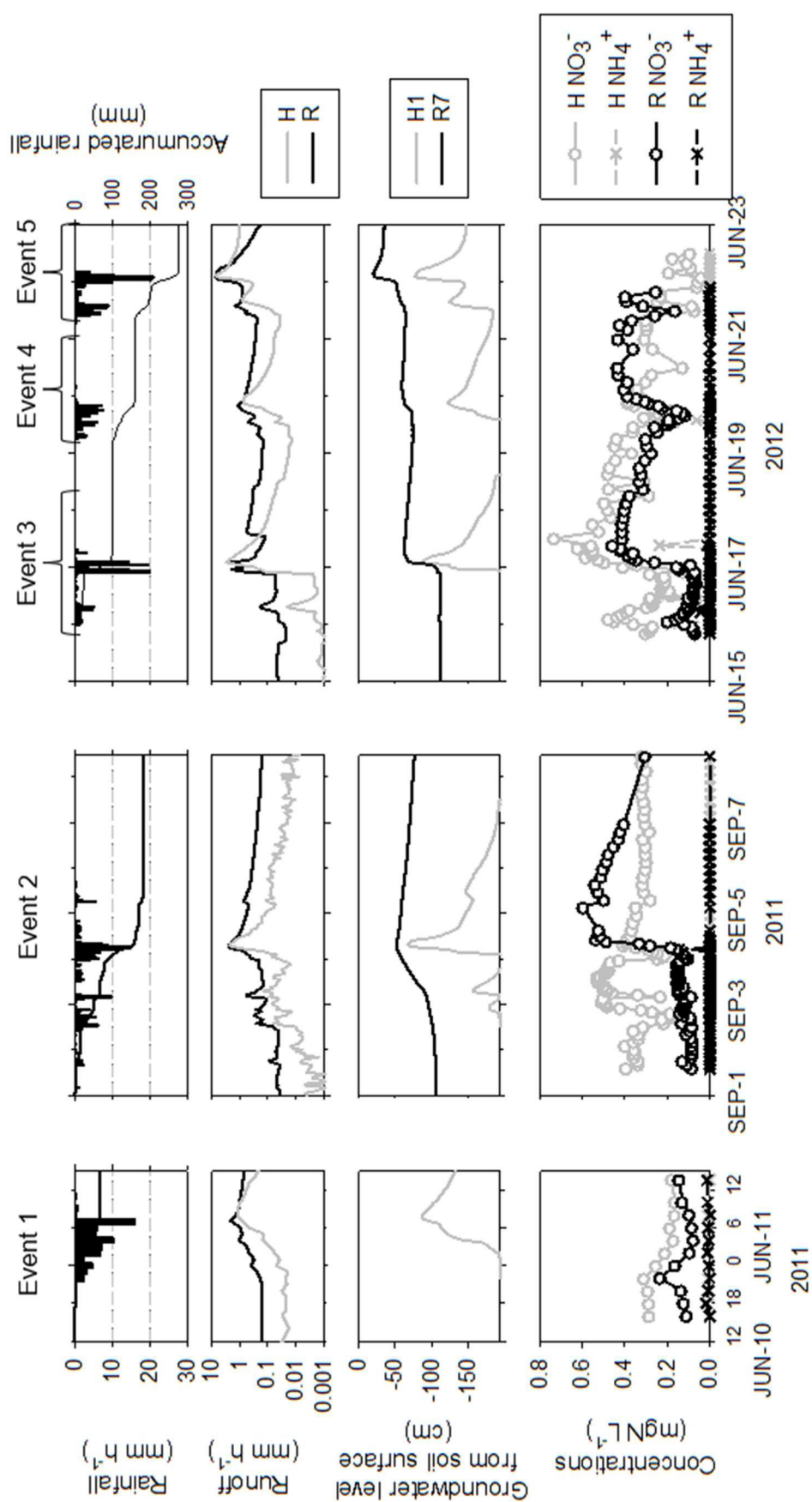


Figure 5.5 Temporal fluctuations of hourly rainfall, runoff, riparian groundwater level, and concentrations of NO_3^- and NH_4^+ in stream water during rainfall events. Groundwater level data at R7 during event 1 were not available.

5.3.4 Annual inorganic N flux

The parameters a and b in equation (5.2) were determined from both monthly and event-based sampling data in Catchments H and R. The L-Q equation in each catchment was expressed as follows (Figure 5.6):

$$\text{Catchment H: } L_S(t) = 2.75 Q_S(t)^{0.97} \quad (R^2 = 0.95, P < 0.001) \quad (5.4)$$

$$\text{Catchment R: } L_S(t) = 2.82 Q_S(t)^{1.30} \quad (R^2 = 0.88, P < 0.001) \quad (5.5)$$

The annual DIN output in streamflow was estimated using these equations in each catchment.

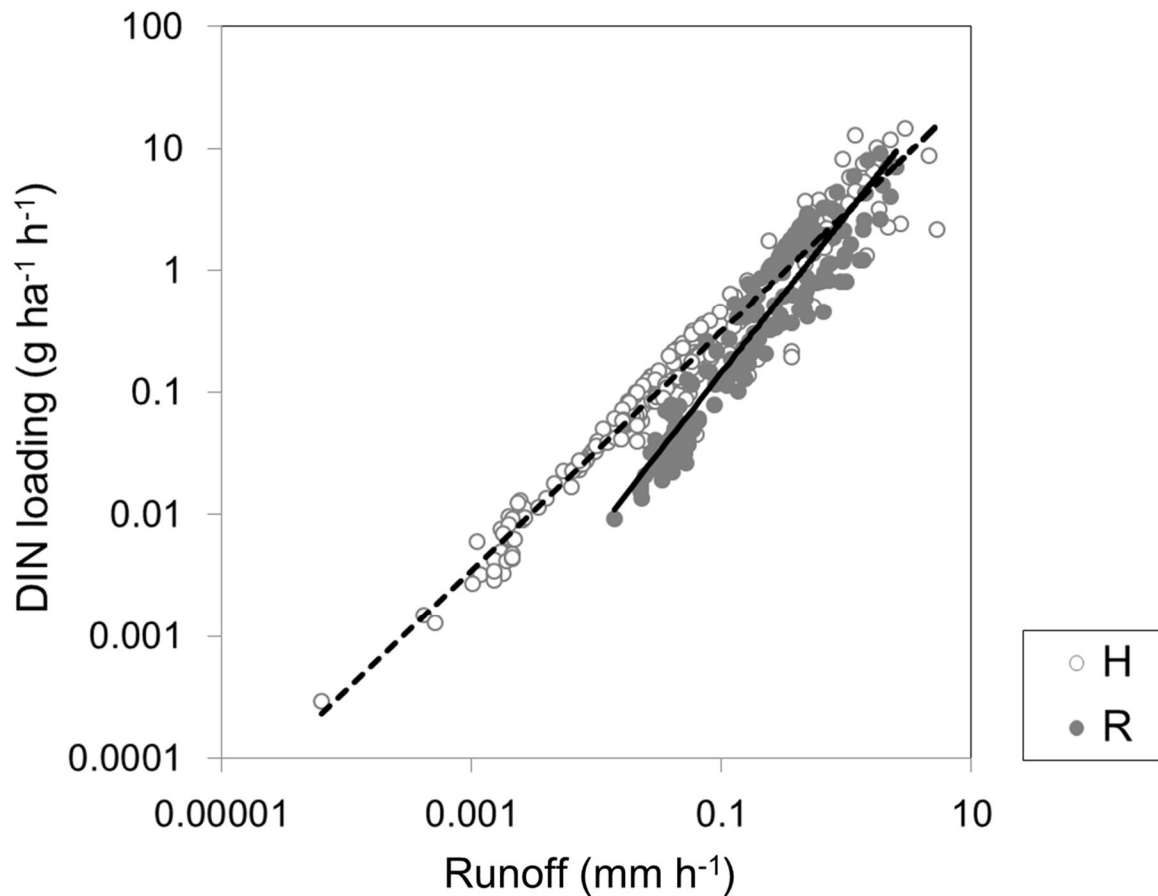


Figure 5.6 Relationship between discharge (Q) and DIN load (L). Broken line, regression line between Q and L in Catchment H: $L = 2.75 \times Q^{0.97}$, $R^2 = 0.95$. Solid line, regression line between Q and L in Catchment R: $L = 2.82 \times Q^{1.30}$, $R^2 = 0.88$.

The annual DIN flux in Catchments H and R is shown in Table 5.2. The DIN input in throughfall on 2011 and 2012 was estimated to be 6.70 and 9.50 kg-N ha⁻¹ yr⁻¹, respectively, in Catchment H and 10.97 and 11.30 kg-N ha⁻¹ yr⁻¹, respectively, in Catchment R. The DIN output in streamflow in Catchment R was more than twice that of Catchment H. In Catchment H, the estimated deep

percolation of DIN from 2010 to 2012 was 0.60–3.07 kg-N ha⁻¹ yr⁻¹. The deep percolation of DIN from 2011 to 2012 was estimated to be -0.38–0.21 kg-N ha⁻¹ yr⁻¹ in Catchment R, where the deep percolation of water was smaller than in Catchment H and its minimum value was negative. The estimated deep percolation of DIN from 2010 to 2012 corresponded to 34–76% and -18–8% of the total DIN output in Catchments H and R, respectively.

Table 5.2. Annual DIN flux in Catchments H and R.

Year	Annual DIN flux (kgNha ⁻¹ yr ⁻¹)											
	Throughfall		Stream water		Deep percolation				Total DIN output			
	H R		H R		H		R		H		R	
					Min	Max	Min	Max	Min	Max	Min	Max
2010	—	—	1.12	—	0.64	2.92	—	—	1.76	4.04	—	—
2011	6.70	10.97	0.96	2.54	0.67	3.07	-0.38	-0.04	1.64	4.04	2.16	2.51
2012	9.50	11.30	1.16	2.36	0.60	2.75	-0.02	0.21	1.76	3.91	2.34	2.57

5.4 Discussion

5.4.1 Effects of bedrock return flow on deep percolation

Annual deep percolation of water was considerably larger in Catchment H than R (Table 5.1). According to the conceptual diagram of runoff process in KEW (Figure 5.1), there are two possible processes that causes the different deep percolation of water between Catchments H and R: (1) Larger amount of bedrock infiltration of water at the hillslope in H than in R. (2) Smaller amount of bedrock return flow at the riparian zone in H than in R. However, the difference in the amount of bedrock infiltration of water is not likely to occur because the two catchments are neighboring each other and have the same geology. On the other hand, the difference in the amount of bedrock return flow is more likely to occur because the contribution of bedrock groundwater to stream water varied greatly between small granite catchments (Uchida and Asano 2010). Katsuyama et al. (2005) showed that bedrock return flow during baseflow period occurred between the zone where the soil layer was saturated transiently during the wet season and the zone where the soil layer was always saturated in a sub-catchment in KEW. In Catchment R, almost all groundwater samples in the SGW (R1 and R6) had Na⁺ concentrations higher than 4 mg L⁻¹. In contrast, almost all groundwater samples in TGW (R7) had Na⁺ concentrations lower than 4 mg L⁻¹ (Figure 5.2), although the distance between R7 and R6 was only 5 m (Figure 2.5). This result indicates that bedrock groundwater with Na⁺ concentrations

higher than 4 mg L^{-1} (Figure 5.2) re-emerged into the soil layer as bedrock return flow between R7 and R6 in Catchment R. In contrast, almost all samples of TGW and stream water in Catchment H had Na^+ concentrations lower than 4 mg L^{-1} (Figure 5.2). This result indicates that bedrock return flow hardly occurred in Catchment H. Thus, the difference in annual deep percolation between Catchments H and R were mainly caused by different amount of bedrock return flow.

5.4.2 Effects of bedrock return flow on DIN concentrations in stream water

DIN concentrations in stream water was considerably different between Catchments H and R; DIN concentrations in stream water in R were lower during baseflow period than H (Figure 5.3), although they became close to or higher than H after a significant amount of rainfall (Figure 5.5). Katsuyama et al. (2004) reported that baseflow stream water had a lower NO_3^- concentration than hillslope groundwater due to the mixing of bedrock groundwater with a low NO_3^- concentration in a sub-catchment in KEW. My observations showed that bedrock groundwater had mean DIN concentrations lower than UGW (Figure 5.4). In addition, Osaka et al. (2010) found that denitrification processes depleted NO_3^- in groundwater in the saturated zone formed by bedrock return flow (Katsuyama et al. 2005) in KEW. These results suggest that the DIN concentration of baseflow stream water in Catchment R was lower than in Catchment H due to the mixing of bedrock groundwater and/or denitrification in the anoxic environment formed by bedrock return flow. On the other hand, I showed the increasing relative contribution of UGW in Catchment R after a significant amount of rainfall by EMMA in Chapter 3. This result suggests that increasing contribution of hillslope groundwater with high DIN concentrations raised DIN concentrations of stream water after a significant amount of rainfall in Catchment R. Thus, bedrock return flow was suggested to reduce DIN concentrations of stream water in Catchment R during baseflow, although its effect became smaller after a significant amount of rainfall due to increasing contribution of hillslope groundwater. Therefore, different DIN concentrations of baseflow stream water do not mean the differences in N status between the two catchments.

5.4.3 Effects of deep percolation on DIN budget

DIN output from Catchments H and R was estimated from DIN output in streamflow, annual water budget, and DIN concentrations of bedrock groundwater, each of which has certain degree of uncertainty. My approach minimized or incorporated this uncertainty because: (1) the DIN output in streamflow was calculated from monthly sampling data as well as event-based sampling data, (2) I used annual evapotranspiration observed independently by one of the most reliable methods (eddy covariance method) when the range of annual precipitation (1150–1971 mm; Matsumoto et al., 2011)

included the annual precipitation from 2010 to 2012 (Table 5.1) as the range of annual evapotranspiration, (3) I used the range of mean DIN concentrations of bedrock groundwater collected from the wells with different depths.

Deep percolation of DIN corresponded to 34–76% and -18–8% of the total DIN output in Catchments H and R, respectively (Table 5.2). Thus, in Catchment H where deep percolation of water was at least 37% (Table 5.1), deep percolation of DIN was not negligible, even for the minimum value observed. Verry (2003) analyzed the data of stream discharge response to tree harvesting treatment in 32 headwater catchments and estimated that annual deep percolation was 45% of streamflow in average. Considerable deep percolation of water was also reported in many other forested headwater catchments by the observation of water balance and chloride balance (Table 1.4; Figure 1.1). These results indicate that deep percolation of water and DIN like the case of Catchment H is not rare in forested headwater catchments.

DIN output in streamflow was widely used to evaluate the response of forest N cycle to disturbance (Likens et al. 1970; Swank et al. 1981; Likens and Bormann 1995; Campbell et al. 2004; Tokuchi et al. 2013). In Catchment R, with a smaller deep percolation of water than Catchment H (Table 5.1), the annual DIN output without consideration for deep percolation, i.e., streamflow flux, was more than twice that of Catchment H (Table 5.2). However, I could not find the difference between total DIN output (streamflow + deep percolation flux) in Catchments H ($1.64 - 4.04 \text{ kgNha}^{-1}\text{yr}^{-1}$) and R ($2.16 - 2.57 \text{ kgNha}^{-1}\text{yr}^{-1}$) (Table 5.2). Therefore, DIN output without consideration for deep percolation does not reflect N status in forested headwater catchments if deep percolation of water was large.

In KEW, an increasing contribution of bedrock groundwater to stream water with expanding catchment scale to second-order (i.e. the entire KEW) was shown by performing EMMA (Chapter 3). In addition, Matsumoto et al. (2011) showed that deep percolation of water corresponded to 0-10% (average 5%) of precipitation in the entire KEW by annual water budget analysis. These facts indicate that DIN deep percolating in Catchment H discharges mainly into a lower elevation catchment in KEW and some of it may discharge into further downstream, i.e., outside KEW. Thus, we need to take account of interbasin bedrock groundwater discharge when we predict the effects of disturbance in Catchment H on DIN export from the entire KEW or a further downstream catchment.

5.5 Conclusions

It was estimated that the deep percolation of water was 37–45% and -6–3% of annual precipitation and the deep percolation of DIN corresponded to 34–76% and -18–8% of the total DIN output in the zero-order catchment and first-order catchment, respectively. The negative value of the minimum deep percolation of water and DIN indicates that interbasin bedrock groundwater discharge from the other

catchments might occur in the first-order catchment. Although the highly weathered bedrock groundwater and the moderately weathered bedrock groundwater had smaller DIN concentrations of stream water compared to hillslope groundwater, the deep percolation of DIN was not negligible for the estimation of the total output of DIN in the zero-order catchment. Moreover, as demonstrated in Chapter 3, the contribution of bedrock groundwater to stream water increased with increasing catchment scale, which suggests deeply percolated water and DIN discharge in higher-order catchments. Therefore, in the headwater catchments where the water budget is not balanced, it is necessary to estimate the deep percolation of DIN in order to evaluate its effects on downstream stream water quality, and the assessment of deep percolation based on individual measurement of water budget components such as rainfall, streamflow, and evapotranspiration can be helpful for understanding the N cycle.

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CHAPTER 6

Summary and Conclusions

6.1 Summary

This study examined the effects of bedrock groundwater dynamics on the hydro-biogeochemical processes in granitic headwater catchments from the perspective of geographic source, flow path, and nutrient budget.

In Chapter 1, the background and objective of this study were presented. Previous studies have shown that bedrock groundwater dynamics affect the geographic source and the flow path of stream water mainly during baseflow periods. However, the effects of bedrock groundwater dynamics on geographic source and the flow path of stream water during storm flow periods, the differences in them between neighboring catchments with different spatial scale, and the effects of bedrock groundwater dynamics on nutrient budgets have not yet been systematically studied. This study aimed to clarify the effects of bedrock groundwater on hydro-biogeochemical processes in granitic headwater catchments. The effects of bedrock groundwater dynamics on stream geographic source (Chapter 3), stream flow path (Chapter 4), and nutrient budget (Chapter 5) during not only baseflow periods but also storm flow periods were discussed. The differences in them among neighboring catchments with different spatial scales were also examined.

In Chapter 2, the study site and the methods used in this study were described. The study site was the Kiryu Experimental Watershed (KEW), Shiga Prefecture, Japan. The catchment is underlain by weathered granite. Five hydrological observation nests were established within the KEW. Observations were conducted in a second-order catchment that corresponded to the entire KEW and four sub-catchments: a first-order catchment, a zero-order catchment without a perennial groundwater body within the riparian soil layer (perennial riparian groundwater body), a zero-order catchment with a small volume of perennial riparian groundwater, and a zero-order catchment with a large volume of perennial riparian groundwater. Continuous measurements of precipitation, runoff, and groundwater level were conducted together with monthly and storm-based water chemistry observations of throughfall, groundwater, and stream water in each catchment.

In Chapter 3, stream geographic source during baseflow and storm flow periods was estimated using end-member mixing analysis using dissolved inorganic materials as tracers in the neighboring or nested five granitic headwater catchments. Throughfall, hillslope groundwater, highly weathered bedrock groundwater, and moderately weathered bedrock groundwater were identified as

end-members. The perennial riparian groundwater body was recharged by highly weathered bedrock groundwater and/or moderately weathered bedrock groundwater. During baseflow periods and recession periods after rainfall, the contribution of highly weathered bedrock groundwater to stream water was the smallest in the zero-order catchment without perennial riparian groundwater, followed in order by the zero-order catchments with perennial riparian groundwater, and the first-order catchment. In the second-order catchment, bedrock groundwater, deeper than that found in the zero- and first-order catchments contributed to stream water during baseflow periods and recession periods after rainfall. The change of stream geographic source with increasing catchment-scale was attributed to the storage of highly weathered bedrock groundwater and/or moderately weathered bedrock groundwater within the perennial riparian groundwater body during the baseflow period. From these findings, it was shown that bedrock groundwater dynamics affect the change of stream geographic source with increasing catchment scale during not only baseflow but also storm flow periods by stored within perennial riparian groundwater body.

In Chapter 4, the dominant flow paths of peak runoff were examined in the neighboring or nested five granitic headwater catchments. In the two zero-order catchments where a perennial riparian groundwater body was absent or small, the dominant flow path of peak runoff changed from saturation overland flow and rapid groundwater discharge in the riparian zones (riparian runoff) or channel precipitation to hillslope runoff, as antecedent moisture conditions became wet. In contrast, the dominant flow path of peak runoff remained as riparian runoff or channel precipitation even during large rainfall events in the zero-order catchment with a large perennial riparian groundwater body and in the first- and second-order catchments. The size of the perennial riparian groundwater was related to the differences in the dominant flow path of peak runoff between the catchments. In Chapter 3, it was shown that the perennial riparian groundwater body was recharged by highly weathered bedrock groundwater and/or moderately weathered bedrock groundwater. The lag time from peak rainfall to peak runoff was longer in the hillslope runoff-dominated catchments compared to channel precipitation- or riparian runoff-dominated catchments. These results indicate that bedrock groundwater dynamics affect the dominant flow path of peak runoff by stored within the perennial riparian groundwater body and the effects are reflected by the lag time from peak rainfall to peak runoff.

In Chapter 5, deep percolation of dissolved inorganic nitrogen (DIN) was quantified in the zero-order catchment without perennial riparian groundwater, and the neighboring first-order catchment. It was estimated that the deep percolation of water was 37–45% and –6–3% of annual precipitation and the deep percolation of DIN corresponded to 34–76% and –18–8% of the total DIN output in the zero-order catchment and first-order catchment, respectively. The negative value of the

minimum deep percolation of water and DIN indicates that interbasin bedrock groundwater discharge from the other catchments might occur in the first-order catchment. Although the highly weathered bedrock groundwater and the moderately weathered bedrock groundwater had smaller DIN concentrations compared to hillslope groundwater, the deep percolation of DIN was not negligible for the estimation of the total output of DIN in the zero-order catchment. Moreover, as demonstrated in Chapter 3, the contribution of bedrock groundwater to stream water increased with increasing catchment scale, which suggests deeply percolated water and DIN discharge in higher-order catchments. Therefore, in the headwater catchments where the water budget is not balanced, it is necessary to estimate the deep percolation of DIN in order to evaluate its effects on downstream stream water quality, and the assessment of deep percolation based on individual measurement of water budget components such as rainfall, streamflow, and evapotranspiration can be helpful for understanding the N cycle.

Conceptual diagram of runoff generation processes and bedrock groundwater contributions to stream water in granitic headwater catchments.

Figure 6.1 shows a conceptual diagram of runoff generation processes and bedrock groundwater contributions to stream water in the granitic headwater catchments of the KEW revealed by the current study. In this diagram, the changes of bedrock groundwater contributions to stream water with increasing catchment scale from zero- to second-order are described. Differences between baseflow, peak flow, and recession periods are also taken into consideration.

During baseflow periods, moderately weathered bedrock groundwater (Source 4 in Figure 6.1a) had almost no contribution to stream water in zero-order catchments (Chapter 3). Contributions of highly weathered bedrock groundwater to stream water (flow from Source 3 to stream in Figure 6.1a) were also smaller in the zero-order catchment without a perennial riparian groundwater body, compared to the other catchments (Chapter 3). In the zero-order catchment, highly weathered bedrock groundwater (Source 3 in Figure 6.1a) and moderately weathered bedrock groundwater (Source 4 in Figure 6.1a) left the catchment as deep percolation (Chapter 5). This caused considerable deep percolation of DIN in the zero-order catchment (Chapter 5). On the other hand, contributions of highly weathered bedrock groundwater to stream water (flow from Source 3 to stream in Figure 6.1b) increased as the spatial scale increased to first- and second-order catchments during baseflow periods (Chapter 3). Contributions of moderately weathered bedrock groundwater to stream water (flow from Source 4 to stream in Figure 6.1b) also increased with increasing catchment scale from first- to second-order catchment (Chapter 3). Larger contribution of highly weathered bedrock groundwater to stream water (flow from Source 3 to stream in Figure 6.1b) caused almost negligible deep percolation of DIN in the

first-order catchment (Chapter 5). Hillslope groundwater, highly weathered bedrock groundwater, and moderately weathered bedrock groundwater was stored within the perennial riparian groundwater body (Sources 2, 3, and 4 within the blue zone in Figure 6.1b) (Chapter 3). Which geographic source was stored the most within the perennial riparian groundwater body differed between catchments depending on the amount of bedrock groundwater seeping into the riparian soil layer (Sources 2, 3, and 4 within the blue zone in Figure 6.1b) (Chapter 3).

During peak flow periods, the throughfall component (Source 1 in Figure 6.1c) became the dominant geographic source of stream water in the zero-order catchments with either no or a small riparian perennial groundwater body (Chapter 3). The throughfall component (Source 1 in Figure 6.1c) mainly reached the stream water via the hillslope soil layer (Chapter 4). The throughfall component (Source 1 in Figure 6.1d) also became the dominant geographic source of stream water in the zero-order catchment with a large riparian perennial groundwater body, the first-order catchment, and a second-order catchment (Chapter 3). However, the throughfall component (Source 1 in Figure 6.1d) mainly reached the stream water via only the riparian soil layer and/or the stream channel (Chapter 4). The difference in the dominant flow path of peak runoff between catchments was affected by the different size of the perennial riparian groundwater body (blue zone in Figure 6.1d) recharged by bedrock groundwater (Sources 3 and 4 in Figure 6.1d) (Chapter 4). Different dominant flow paths of peak runoff caused the difference in lag time from peak rainfall to peak runoff between neighboring catchments (Chapter 4).

During the recession periods after rainfall, the throughfall component (Source 1 in Figure 6.1e) remained the dominant geographic source of stream water in the zero-order catchment without a perennial riparian groundwater body (Chapter 3). This was because hillslope runoff with a high proportion of throughfall under continuous rainfall conditions (flow from Source 1 from the hillslope in Figure 6.1e) directly discharged into stream water without mixing with groundwater stored within the perennial riparian groundwater body in the zero-order catchment (Chapter 3). On the other hand, hillslope runoff with a high proportion of throughfall (flow from Source 1 from the hillslope in Figure 6.1f) was buffered by the groundwater stored within the riparian soil layer (Sources 2, 3, and 4 within the blue zone in Figure 6.1f) in the zero-order catchments with a perennial riparian groundwater body, first-order catchment, and second-order catchment (Chapter 3). The dominant geographic source of stream water during recession periods was determined by which geographic sources had stored the most within the perennial riparian groundwater body before the rainfall (Sources 2, 3, and 4 within the blue zone in Figure 6.1f). The geographic sources that stored the most were the hillslope groundwater (Source 2 within the blue zone in Figure 6.1f) in the zero-order catchment with a perennial riparian groundwater body, and the highly weathered bedrock groundwater (Source 3 within the blue zone in

Figure 6.1f) in the first- and second-order catchments (Chapter 3). In the second-order catchment, storage of moderately weathered bedrock groundwater within the riparian soil layer (Source 4 within the blue zone in Figure 6.1f) was larger than in the zero- and first-order catchments (Chapter 3).

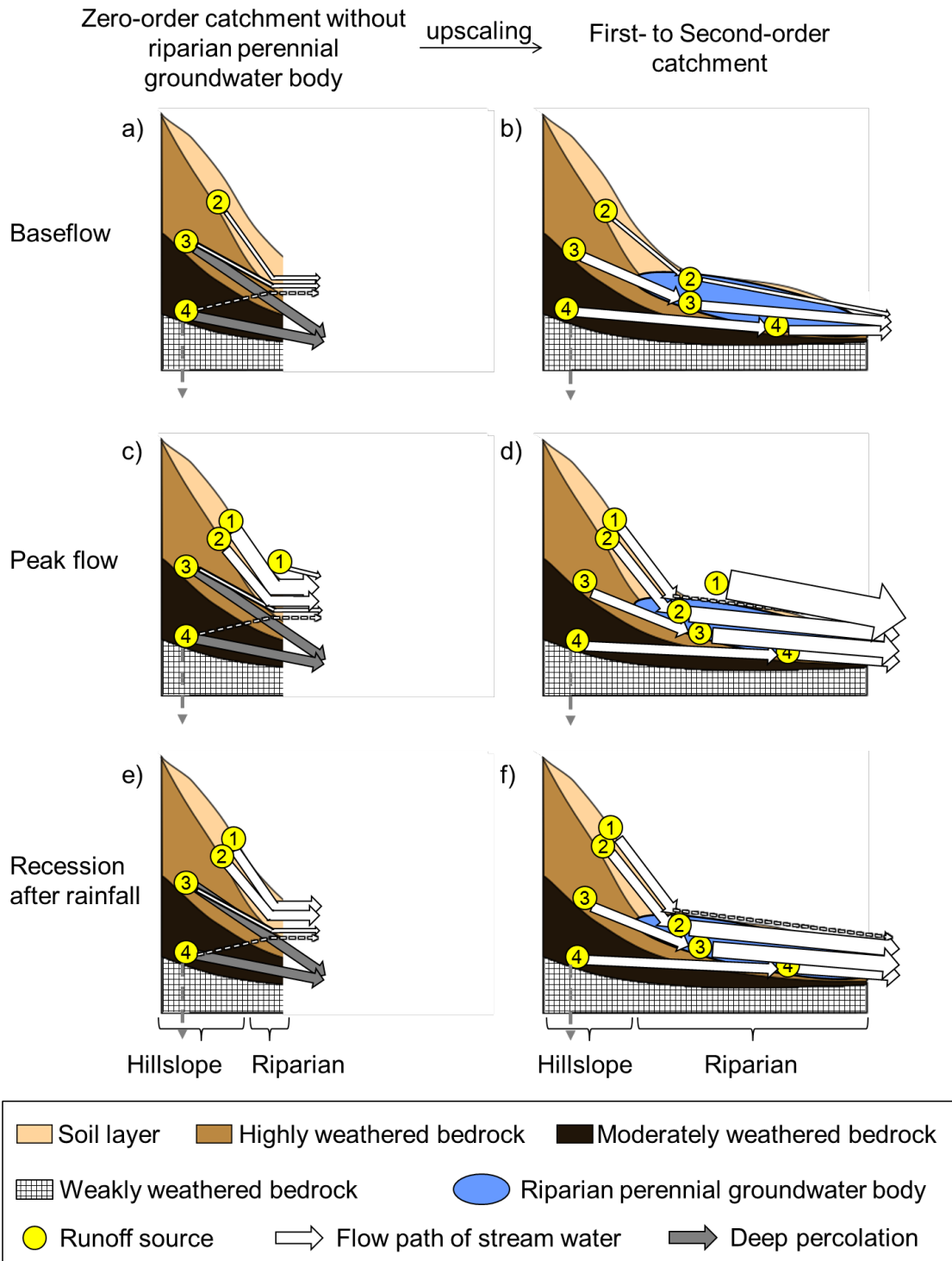


Figure 6.1 Conceptual diagram of runoff generation processes and bedrock groundwater contributions to stream water in granitic headwater catchments, Kiryu Experimental Watershed (KEW), Japan. Numbers in circles indicate the different geographic sources: 1 = throughfall; 2 = hillslope groundwater; 3 = highly weathered bedrock groundwater; and 4 = moderately weathered bedrock groundwater. The width of each arrow indicates the relative volume of each flow path.

6.2 General conclusions

This study investigated the effects of bedrock groundwater dynamics on hydro-biogeochemical processes in granitic headwater catchments by considering the differences between baseflow and storm flow periods as well as differences resulting from spatial scale, from zero- to second-order catchments. Contributions of bedrock groundwater to stream water increased with increasing catchment scale during baseflow and recession periods. Bedrock groundwater that was stored within a perennial riparian groundwater body during baseflow periods contributed to stream water during the recession periods. The storage of bedrock groundwater within a perennial riparian groundwater body also affected the differences in the dominant flow path of peak runoff, resulting in different lag times from peak rainfall to peak runoff between neighboring catchments. These findings indicate that the storage of bedrock groundwater within a riparian perennial groundwater body plays an essential role in bridging the gap of spatial scale between hillslope-scale and catchment-scale. Even if the bedrock groundwater does not rapidly respond to rainfall at hillslope-scale, the bedrock groundwater can affect the geographic source and flow path of stream water during storm flow periods by the rapid response of the perennial riparian groundwater body. Thus, the storage of bedrock groundwater within a riparian perennial groundwater body should be the key to connect hillslope-scale bedrock groundwater dynamics and catchment-scale stream runoff and chemistry. The current study was conducted in granitic headwater catchments. The effects of bedrock groundwater dynamics on hydro-biogeochemical processes in headwater catchments may differ depending on the geology of the catchment. In future, simultaneous observations of bedrock groundwater dynamics at hillslope-scale and stream geographic sources and flow paths at catchment-scale in study sites with various geology are necessary in order to understand the effects of bedrock groundwater on stream runoff and chemistry more deeply. Future study, focusing on residence time in addition to runoff source and the flow path of stream water is also necessary. Bedrock groundwater with various residence times via various flow paths may be stored within a perennial riparian groundwater body and may simultaneously contribute to stream water during storm flow periods. Thus, storage of bedrock groundwater within riparian perennial groundwater body may affect the residence time of stream water.

This study also revealed the existence of a zero-order catchment where deep percolation of DIN was not negligible. This finding indicates that the assumption of negligible deep percolation of nutrients should be reconsidered in headwater catchments where the nutrient budget has been estimated in previous studies. The most reliable way to verify this assumption is the investigation of water budgets by observing runoff, precipitation, and evapotranspiration in each catchment. This study suggested that deeply percolated water and DIN discharge in higher-order catchments. Deeply percolated water

should take more time than stream water to contribute to the stream water in higher-order catchments. In addition, nutrient uptake by plants cannot occur and a reductive environment might be formed within the bedrock layer. Compared to stream water, different biogeochemical processes may be involved in determining the chemistry of deeply percolated water. To assess the influence of forest disturbance in headwater catchments on stream water chemistry in higher-order catchments, the time required for deep percolating water to discharge in higher-order catchments, and the biogeochemical processes within the bedrock layer should be taken into consideration in future.

In conclusion, this study revealed that the changes of stream geographic source, stream flow path, and DIN budget with increasing spatial scale from zero- to first- or second-order catchments were controlled by bedrock groundwater dynamics. It was found that the storage of bedrock groundwater within a perennial riparian groundwater body and the deep percolation of water and nutrients should be considered when predicting stream water runoff and chemistry at the scale of higher-order catchments, based on observations at the scale of 10^{-2} – 10^{-1} ha.

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