Journal of Hydroscience and Hydraulic Engineering Vol. 11, No. 1, May, 1993, 79-87

FIELD STUDY OF MIGRATION OF SOIL WATER AND GROUNDWATER
BY ISOTOPIC COMPOSITION

BY

Hideshi Ikeda, Takashi Ohsumi and Yoichi Miyanaga
Environmental Science Department
Abiko Research Laboratory
Central Research Institute of Electric Power Industry
1646 Abiko, Abiko-City, Chiba, Japan

SYNOPSTS

To evaluate the buffer capacity of a watershed to acid deposition, it is necessary to understand the runoff of precipitation to stream and the concurrent chemistry changes in the watershed. We observed temporal variations in the chemical and isotopic composition of the groundwater and streamwater in a mountainous watershed during storm flow generation. Based on these results, we separated stream discharge into three components: Precipitation, soil water and groundwater. It was determined that soil water runoff through the aquifer was an important pathway for streamflow generation and for streamwater chemistry changes.

INTRODUCTION

In Europe and the northeastern United States, acidification of some lakewater and streamwater has been reported, and acid deposition is believed to be the cause. While acidification of streams and lakes has not been reported in Japan (7), it is impossible to estimate chemical change in future without understanding the mechanism of acidification. This mechanism is explained by two processes: The first is the runoff of precipitation in the watershed to stream, and the second is the concurrent chemical change of precipitation. It has previously been reported that these phenomena cause a change in temporal variation of groundwater chemistry during storms (6). Therefore, estimation of the ratios of runoff components in stream discharge by hydrograph separation during a storm is an important part of the evaluation models of streamwater acidification, such as MAGIC (2), Birkenes (11), and ETD (10). Prior studies have not attempted to separate the contributions of groundwater and soil water. This paper consists of two field research results. First is chemistry of precipitation, soil water, groundwater and streamwater measured throughout a year. The second is analysis of water migration in the watershed, estimated by chemical and isotopic composition in the streamwater and groundwater during a storm. The purpose of this study is to clarify the mechanism of runoff based on these results, incorporating the three component mass balance tracer model.

OBSERVATION METHOD

Site Description

Topography and observation points in the experimental watershed are shown in Fig. 1. A cross-section of the watershed, including boring wells W1-W3, is shown in Fig. 2. The 3.2 km² watershed is located in a mountainous area in the mid-part of Japan, and is covered by ceder, pine, and beech. The surface soil type is pyroclastic regosol (weathered granodiorite) and the hydraulic conductivity of the aquifer is about $10^{-4}~\rm cm/s$.

Chemical survey throughout a year

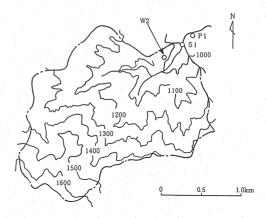
Precipitation, soil water, ground-water and streamwater were sampled and analyzed throughout a hydrological year to understand the chemical properties in the experimental watershed. Observations and sampling schemes are shown in Table 1. Precipitation was sampled by fraction collector. Groundwater and streamwater were sampled at the surface of water. Soil water was sampled by a lysimeter. Water temperature, pH, alkalinity (endpoint of pH 4.8), and chemical composition of major ions (Ca²+, Mg²+, Na+, K+, SO4²-, Cl-, NO3-, and HCO3-) were analyzed.

Hydrograph separation by isotopic composition

Dincer et al.(4), Martinec (8), and Sklash and Farvolden (12) proposed the hydrograph separation method using a conservative tracer, such as ¹⁸0 and ³H. This method was also applied to a Japanese watershed by Matsubaya et al. (9), and we also separated stream discharge into two components using ¹⁸0 (5): Event water (precipitation) and pre-event water (soil water and groundwater). Conservation equations for separation are shown below:

$$Q_0 = Q_1 + Q_2 (1)$$

$$Q_0 R_0 = Q_1 R_1 + Q_2 R_2 \tag{2}$$



Area: 3.2km²
Altitude: 940-1660m
Annual Precipitation:
About 2200mm
P1:Rain Gauge
& Precipitation Sampling Point

& recipitation Sampling Folic SI:Gauging Station & Streamwater Sampling Point W2:Soil Water & Groundwater Sampling Point Num:Altitude(m) ---: Watershed Boundary

Fig. 1 Topography and Observation Points in watershed

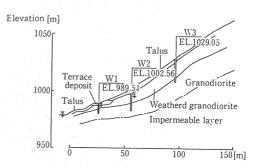


Fig. 2 Cross Section of W1-W3 Line

Table 1 Observation Items and Schemes

		Items and Samples	Observation and Sampling Points	Observation and Sampling Intervals
Hydrology		Precipitation Discharge	P1 S1	Countinuous Countinuous
Chemistry	Annual observation	Precipitation Soil water Groundwater Streamwater	P1 W2* W2 S1	every 2 days month month month
	Observation during a storm	Precipitation Soil water Groundwater Streamwater	P1 W2* W1,W2,W3 S1	every 1 or 2 hours once prior to storm every 3 hours every hour

^{* :} Soil Water were sampled at some depth vertically

where Q = discharge; R = isotopic composition; and subscript 0, 1, and 2 = discharge, event water, and pre-event water, respectively. In our previous study (5), we attempted to determine the concentrations of particular ions in streamwater during a storm using hydrograph separation. However, because pre-event water included soil water and groundwater, chemical concentrations such as ${\rm Ca}^{2+}$ and ${\rm SO_4}^{2-}$ were not vertically uniform. Therefore, chemical concentrations of these ions in pre-event water were not constant during a storm, and calculated concentrations

did not agree well with the measured values.

Therefore, to obtain better agreement between calculated and measured values, we separated discharge into three components: Precipitation, soil water, and groundwater using two conservative and independent isotopic tracers (180 and 3H). Precipitation was considered to be event water in the two component model and soil water and groundwater were considered to be pre-event water. First, the stream discharge is separated into event water and pre-event water using 180 as conservative tracers; second, pre-event water was divided into soil water and groundwater using 3H as a tracer. In this way, stream discharge was divided into three components. The conservation equations used for the separation are shown below:

$$Q_0 = Q_0 + Q_V + Q_g \tag{3}$$

$$Q_0 T_0 = Q_0 T_0 + Q_v T_v + Q_g T_g$$
(4)

where T = concentration of 3H ; subscript e, v, and g = precipitation, soil water and groundwater, respectively. Oxygen-18 is expressed as δ per mil (%) difference relatives to the standard mean of ocean water because it is present in such small amounts (3).

Chemical survey during a storm

Streamwater and groundwater chemistry changes were observed during a storm. Separation results were verified by these results, and migration of soil water and

groundwater is analyzed.

The cations and anions measured in streamwater were Ca^{2+} , Mg^{2+} , SO_4^{2-} , and Cl^- , which are prevalent in streamwater. Concentrations of these ions are calculated by conservation equations (5) and (6), and were compared with the measured values.

Conservation equation by the two component model

$$Q_0C_0 = Q_1C_1 + Q_2C_2 \tag{5}$$

Conservation equation by the three component model

$$Q_0C_0 = Q_0C_0 + Q_vC_v + Q_gC_g$$
(6)

where C_0 = calculated concentrations in streamwater; C_1 and C_2 = measured concentration in precipitation; C_2 = the measured concentration in soil water, which was sampled prior to the storm; and C_2 and C_3 = concentrations in pre-event water and groundwater, respectively, which were assumed to be equal to be those in streamwater before the storm.

RESULTS

Chemical survey throughout a year

Chemical properties, such as pH, alkalinity, and major ion concentrations of precipitation, soil water, groundwater, and streamwater, were measured in the watershed throughout a hydrological year, from October 1989 to September 1990. As shown in Fig. 3, the annual averages of pH, alkalinity, and concentrations of

alkali earth cations, Ca²+ and Mg²+, increase with soil depth. This suggests that acid deposition in precipitation is neutralized during runoff in the watershed to stream. However, the annual average alkalinity of streamwater was

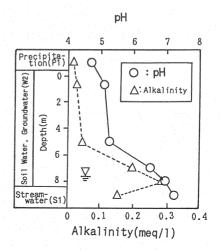
0.14 meq/l, which is smaller than the spatial average value of streamwater in the mid-part of Japan (0.49 meq/l) (1).

Chemical and isotopic observations during a storm $% \left\{ 1,2,\ldots ,n\right\}$

The episodic event studied was a storm from September 19 to 21, 1990, and the total amount of precipitation was 140 mm during 16 hours. Temporal variations of precipitation, discharge. and groundwater level are shown in Fig. It began to rain in the afternoon of the 19 September, and discharge reached its peak flow of 4 m3/s at midnight of September 20. Therefore, it decreased from September 20 to 21. Temporal variations of pH in precipitation, streamwater, and groundwater are shown in Fig. 5. Temporal variations of chemical compositions in streamwater are shown in Fig. 6. We can see that streamwater was diluted with precipitation. Furthermore, the groundwater level at Wl. which was shallower than W2 or W3, was increasing during the storm.

Hydrograph separation into two components

Temporal variations of isotopic components (180) in precipitation, streamwater, and groundwater at Wl are shown in Fig. 7, and the vertical profile of the sampled water is shown in Fig. 8. Precipitation was sampled during the storm, and the others were sampled prior to the storm. Clearly, 180 was vertically uniform in soil water and groundwater. Therefore, it is apparent that the isotopic component in pre-event water was constant even if the ratio of soil water and groundwater varied during the storm. Based on this data, it is possible to separate event water and pre-event water in this episodic event using hydrograph separation.



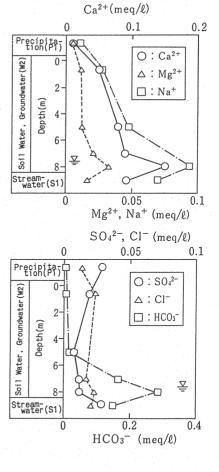


Fig. 3 Vertical Variations in Chemistry in Watershed (pH, Alkalinity, Cations, Anions)

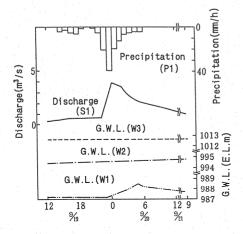


Fig. 4 Temporal Variations in Precipitation, Discharge, and Groundwater Level

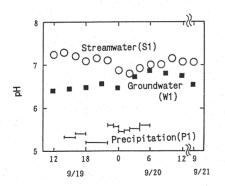


Fig. 5 Temporal Variations in pH of Precipitation, Streamwater, and Groundwater

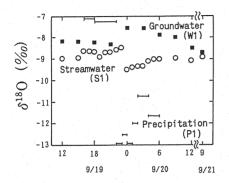
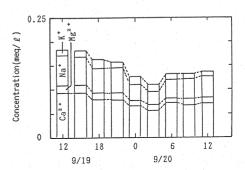


Fig. 7 Temporal Variation in Isotopic Composition (180)



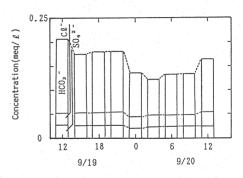


Fig. 6 Temporal Variations in Streamwater Chemistry

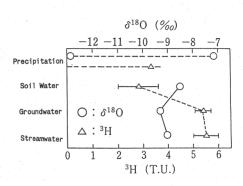
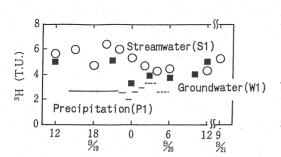


Fig. 8 Vertical Profiles in Isotopic Composition (180, 3H)

Hydrograph separation into three components

Three components are obtained by dividing pre-event water into two components: Soil water and groundwater, using ³H. Temporal variations in concentration of ³H in precipitation, streamwater, and groundwater are shown in Fig. 9 and their vertical profile is shown in Fig. 8. Because the concentration of ³H was not vertically uniform in soil water and groundwater, it is possible to separate the hydrograph into three components: Precipitation, soil water, and groundwater, using the two components described above, and ³H in soil water and groundwater. The results of hydrograph separation are shown in Fig. 10. The maximum discharge was recorded at midnight of September 20. Precipitation accounted for 10% of discharge at that time, and soil water accounted for 50% at 6 0'clock September 20.



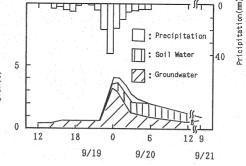


Fig. 9 Temporal Variation in Isotopic Composition (³H)

Fig.10 Result of Hydrograph Separation into three Components

Determination of chemical composition

Streamwater chemistry changes were determined based on the hydrograph separation results. Concentrations of the most prevalent cations and anions in streamwater (Ca^{2+} , Mg^{2+} , SO_4^{2-} , Cl^-) were calculated by conservation equations (5) and (6), and the values obtained were compared with the measured values.

In Fig. 11, measured concentrations in streamwater and the corresponding calculated values are compared. As shown in Fig. 11, calculated concentrations of Cl⁻, using two and three components, agree with measured values. Cl⁻ is supplied from the atmosphere to the watershed as wet and dry deposition and is not supplied from the soil. Therefore, Cl⁻ is a conservative anion and its concentration is spatially uniform in soil water and groundwater in a watershed. On the other hand, calculated values of $SO_4^{\ 2^-}$ and Na^+ in streamwater using three components agree with measured values better than those obtained using two components. In this way, the temporal variation in streamwater chemistry during the storm is determined more accurately by three components than two components, and the hydrograph separation result of the three component model is verified. However, agreement between the calculated and measured concentrations of Ca²⁺ using three components is worse than with two components.

In this analysis, soil water was sampled at W2 only prior to the storm, as shown in Table 1. Therefore, we were unable to ascertain the ability of the models to determine spatial variation in soil water chemistry. As shown in Fig. 11, the calculated concentrations of Na $^+$, SO $_4$ 2 -, and Cl $^-$ in streamwater agree with the measured values, and concentrations of these ions in soil water are in theory spatially uniform in watersheds. On the other hand, Ca 2 + is an alkali earth cation and is very reactive with soil, and its calculated concentrations did not agree with the measured values well. Therefore, the concentration of sampled soil water at W2 is expected to be higher than the spatial average in a given watershed. Furthermore, where the soil water and groundwater runoff into stream, it is thought that chemical reaction occurs by the release of CO $_2$ into atmosphere. These phenomena need to be studied in the future.

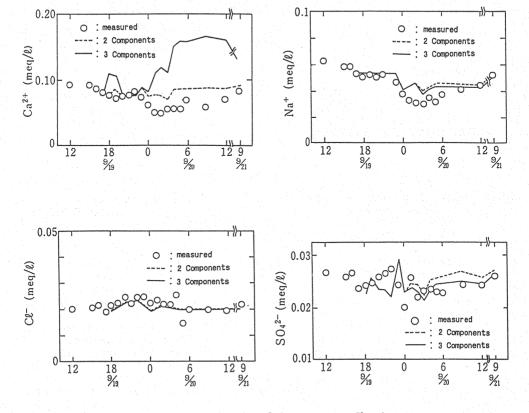


Fig.11 Determination of Streamwater Chemistry (Upper Left: Ca²⁺, Upper Right: Na⁺ Lower Left: Cl⁻, Lower Left: SO₄²⁺)

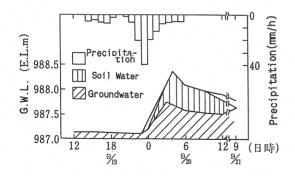
Separation of groundwater

In this section, the pathways of soil water and groundwater into stream are analyzed by separation of groundwater. The groundwater at W1 was chosen for analysis because its temporal variation of groundwater level during the storm was more drastic than W2 or W3. The groundwater is separated using the conservation tracers, ¹⁸O and ³H (Figs. 7, 8, and 9), in the same way as hydrograph separation.

The separated results are shown in Fig. 12, and were verified by Na⁺ concentration in groundwater (Fig. 13). Through this analysis, we concluded that precipitation reached the aquifer only when rain intensity was large. While groundwater level is decreasing, the aquifer was cultivated by the soil water. However, because the rise of groundwater level at Wl was too large to be attributed to the soil water near Wl alone, it seems likely that the aquifer around Wl was cultivated by the soil water upper of Wl.

Moreover, as shown in Figs. 10 and 12, the ratio of soil water in streamwater increased after the ratio of soil water in groundwater increased. This suggests that soil water in the watershed first percolated into the aquifer near the stream,

then flowed out to the stream.



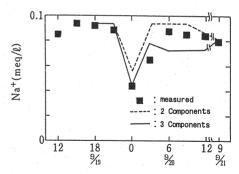


Fig.12 Result of Groundwater Separation into three Components

Fig.13 Determination of Groundwater Chemistry

CONCLUSION

(1) Stream discharge in a mountainous watershed with area of 3.2 km² during a storm was separated into three components: Precipitation, soil water, and groundwater by conservative isotopic tracers 180 and 3H. Based on the results, discharge was comprised of 10% precipitation at time of peak flow, and 50% soil water at 6 hours later.

(2) Calculated concentrations of SO₄²⁻ and Cl⁻ in the streamwater, using the separation result by the method proposed here, agree with measured values. On the other hand, the calculated concentration of Ca^{2+} exceeded the measured values. This is attributed to the spatial variability of Ca2+ concentrations in the soil Soil water should be sampled at several points to analyze the spatial distribution and reaction of Ca^{2+} between soil water and soil in the watershed in detail.

(3) The results show that runoff of soil water through aquifer is an important pathway in stream discharge during the episodic event under study.

REFERENCES

1. Chikasui Handbook (Groundwater Handbook) p.132, 1979.

2. Cosby, B.J., R.F. Wright, G.M.Hornberger, and J.N. Galloway: Modeling the effects of acid deposition: Estimation of long-term water quality response in a small forested catchment, Water Resources Research, Vol.21, No.11, pp.1591-1601, 1985.

Craig, H.: Standard for reporting concentrations of deuterium and oxygen-18 in

natural waters, Science, No.133, pp.1833-1834, 1961.
Dincer, T., B.R. Payne, T. Florkowski, J. Martinec, and E. Tongiorgi: Snowmelt runoff from Measurements of tritium and oxygen-18, Water Resources Research, Vol.6, No.1, pp.110-124, 1970.

Ikeda, H., T. OHSUMI, and Y. MIYANAGA: Relationship between runoff components and streamwater chemistry during storm (synopsis in English), Proceedings of

Hydraulic Engineering, J.S.C.E, Vol.35, pp.567-572, 1991.

6. Itoyama, T.: Compositional change of groundwater caused by precipitation (synopsis in English), Chikyuu Kagaku (Geochemistry), Vol.18, pp.15-19, 1984.

7. Kiyono, M. and Y. ISHIKAWA: CRIEPI Report, No.E484002, pp.8-14, 1985.

8. Martinec, J.: Subsurface flow snowmelt traced by tritium, Water Resources Research, Vol.11, No.3, pp.496-498, 1975.

Matsubaya, O., M. Yoshida and K. Tanaka-Miyamoto: Runoff analysis by means of multiple isotope tracers in Iwami river drainage, Akita, Japan, Mass Spectroscopy, Vol.38, No.6, pp.331-339, 1990.

10. Nikolaidis, N.P., H. Rajaram, J. Schnoor and K. Georgakakos: A generalized softwater acidification model, Water Resources Research, Vol.24, No.12, pp.1983-1996, 1988.

11. Seip, M., N. Christophersen and S. Rusted: Predicted changes in streamwater following reduced/increased sulfur deposition using the "Birkenes model",

Water, Air and Soil Pollution, Vol.31, pp.239-246, 1986.

12. Sklash, M.G. and R.N. Farvolden: The role of groundwater in storm runoff, J. Hydrology, Vol.43, pp.45-65, 1979.

(Received November 26, 1992; revised May 13, 1993)