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Chemical Characteristics of Precipitation in *Quercus* Forests in Korea and Japan

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Abstract : The major objective of this study was to analyze the difference of the chemical characteristics of acid deposition in *Quercus* forests in Korea and Japan. The pH values of rainfall at the experimental forest of Kangwon National University (KS site) were higher than those at the Foresta Hills in Japan (JP site), and all chemical contents of throughfall and stemflow were much higher than those of rainfall in *Quercus* forest stands at the KS and JP site. The pH values, Ca^{2+}, NO_3^- and SO_4^{2-} concentration of throughfall and stemflow at the KS site showed seasonal variation. While at the JP site, the same pattern was shown in the pH values of throughfall and stemflow, however, did not show any difference among seasons. Also, the annual input of all nutrients in these two contrasting forests varied seasonally. These results can be used to predict the amounts of air pollutant that are washed off and leached by the rainfall and Yellow Sand (Asian dust), including NOx and SOx acid pollutants transported easterly from China in the spring. Therefore, it is necessary to quantify the inputs of dry and wet deposition throughout a full year to gain a more complete understanding of the effects of acid deposition on the nutrient cycles in these forest ecosystems.

Key words **:** *stemflow, throughfall, stream water, Yellow sand, forest ecosystem*

Introduction

In 1997, the International Congress of Acid Snow and Rain (Niigata University, Niigata, Japan) discussed the effect of acid rain and snow on the environment. Evaluating the effects of acid deposition such as acid rain and snow on forest has been a major concern of researchers in Europe, America (Thomas, 1991; Carmicheal, 1997; Uri and Dennis, 2003) and Asia (Sassa *et al.*, 1991; Ishizuka, 1992; Kobayashi *et al.*, 1995; Kim *et al.*, 1996; Han and Lee, 1997; Park and Woo, 1998; Fan *et al.*, 1999; Kim *et al.*, 2001; Sasaki *et al.*, 2001). Quantification of nutrients inputs affecting acidification has been important research subjects.

Recently, Korean and Japanese forest hydrologists have focused on the importance of hydrogeochemistry on forested catchments, especially to understand and predict the effects of acid deposition (Bashkin, 1997; Carmicheal, 1997; Chun *et al.*, 1997; Katoh *et al.*, 1997; Ogawa *et al.*, 1997). Throughfall and stemflow are also important pathways of nutrient to the forest floor and can influence soil physical and chemical properties. Also, the environmental problems caused by Yellow Sand from China have been noticed. Previous studies related to the Yellow Sand in Korea dealt with visibility impairment and dust concentrations during the Yellow Sand event (Chohji *et al.*, 1997) and route of transfer (Chung *et al.*, 2000, 2001), but there has been no study of the effects of Yellow Sand on the nutrient cycle in forests.

Since 1996, in the experimental forests of the College of Forest Sciences, Kangwon National University, we have been analyzed pH, EC, and concentrations of cations and anions in rainfall, throughfall and stemflow collected from the coniferous trees and the broad-leaved trees (Chun *et al.*, 1997, 1998a, 1998b; Kim *et al.*, 1997, 2000, 2001). Moreover, in Foresta Hills located in Toyota city, Aichi-prefecture, Japan, we observed chemical contents to develop a simulation model for study on water cycles of hydrochemical processes in forest catchment from 2001. We analyzed chemical contents in rain-

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fall, throughfall, stemflow and stream water.

The major objective of this study was to analyze the differences of the chemical characteristics of acid deposition in *Quercus* forests in Korea and Japan. We observed a clear seasonal trend in the inputs of various elements from precipitation on forest catchments. For the evaluation on precipitation input in *Quercus* forests, we analyzed results obtained from these Korean contents, and compared with contents collected from Foresta Hills in Toyota city, Aichi Prefecture, Japan.

Materials and Methods

The experimental forest of Kangwon National University (KS) is located in Gangwon Province (127°48'-52'E, 37°46'-51'N.L) about 100 km away from Seoul, Korea and Foresta Hills (JP) is located in Toyota city, Aichi Prefecture (137° 11E, 35° 02'N.L), Japan (Figure 1). At the KS site, the altitude ranges from 255m to 899m above sea level. The mean temperature is 11.1°C and the mean annual precipitation is approximately 1154.9 mm in 2000 (about 70% of precipitation from April to Sep.). The soils were generally formed from granite parent material. The soil type is a slightly dry brown forest soil (B_B) (Jin *et al.*, 1994). The research area belongs to a temperate secondary forest, where the dominant tree species are *Quercus variabilis, Q. serrata* and *Q. variabilis, Fraxinus rhynchophylla, Cornus controversa, Lindera obtusiloba* and *Lespedeza bicolor* and reforested tree species are *Pinus koraiensis* and *Larix leptolepis*.

The JP site varies from 40m to 105m in altitude, and the annual precipitation is 1272 mm and the mean temperature is 16.8°C in 2001. The research area belongs to a temperate secondary forest, where the dominant tree species are *Q. serrata, Q. variabilis* and *Evodiopanax innovans*. The soil originated from granite bed rock. The soil type is slightly dry brown forest soil (B_B~B_D) (Committee of Forest Soil of Japan, 1983). Table 1 shows the comparison of general description and meteorologic outlook for the research site in Korea (KS) and Japan (JP).

Rainfall was collected using a 20-cm funnel connected to a 20-L polyethylene bottle in an open area of the research forest catchments. Throughfall was collected using the same method as for rainfall but within each plot. Stemflow was collected using a synthetic rubber sheet with an acutely angled edge attached around the stem by silicon to avoid leakage; collected stemflow was stored in a 200-L polyethylene container (KS) and a 20-L polyethylene bottle (JP). From December to February, sampling of the rainfall, throughfall and stemflow at the KS site was not performed because of snow fall.

The pH and EC values were measured within 12 h after sampling by using pH and EC meter (model 1230, Orion Corp., USA and model WM-22WP, TOA, Japan). We analyzed the chemical compositions of rainfall, throughfall, stemflow and stream water sampled from this site after sequential filtrations using 0.2 μm (KS) and 0.45 μm (JP) membrane filters. Cations such as sodium (Na^+), magnesium (Mg^{2+}), calcium (Ca^{2+}) and potassium (K^+) were analyzed using an Atomic Absorption Spectrophotometer (AA-6800, Shimadzu, Kyoto, Japan) and an Inductively Coupled Plasma Spectrophotometer (IRIS ICAP, Nippon Jarrell-Ash, Kyoto, Japan). Anions such as chloride (Cl^-), nitrate (NO_3^-) and sulfate (SO_4^{2-}) in the collected solutions were analyzed using ion chromatography (DX-120, DIONEX Corp., USA and PIA-1000, Shimadzu, Kyoto, Japan).

The study was conducted from April 24 to October 26 in 2000 (KS) and from July 5 in 2002 to October 14 in 2003 (JP). In order to estimate the seasonal contribution of nutrients inputs to the forest floor, all data were divided into three seasons, spring (April-May), summer (June, July, August) and fall (September-October). From the volumes of throughfall and stemflow in each forest stand, the total amount of elemental input to the forest floor was estimated, respectively.

Results and Discussion

1. Variations of pH and EC values, cations and anions concentrations of rainfall, throughfall, stemflow and stream water in *Quercus* forest stands in Korea and Japan.

The mean pH and EC values, and concentrations of cations and anions of rainfall, throughfall and stemflow of *Quercus* forests in Korea and Japan are shown in Table 2. In Korea, the pH values of rainfall ranged from 4.77 to 6.39 (average: 5.62). The pH values of through-

Table 1. Comparison of general description and meteorologic outlook for the research site in Korea and Japan.

Site	Location	Elevation (m)	Mean Temp.	Dominant species	Mean precipitation
Korea (KS)	127° 48' 52"E, 37° 46' 51" N.L.	255~899m	10°C	*Quercus mongolica, Q. variabilis, Q. aliena, Fraxinus rhynchophylla, Cornus controversa, Lindera obtusiloba* and *Lespedeza bicolor*	1154.9 mm
Japan (JP)	137° 11' 27" E, 35° 2' 00"N.L.	40~105m	6.8°C	*Q. serrata, Q. variabilis* and *Evodiopanax innovans*	1272 mm

Table 2. The mean pH and EC values and ion concentrations of rainfall, throughfall, stemflow and stream water in *Quercus* forest stands in Korea and Japan.

Site		pH	EC (mS/m)	Anion (ppm)			Cation (ppm)				period
				Cl^-	NO_3^-	SO_4^{2-}	Ca^{2+}	K^+	Mg^{2+}	Na^+	
KS	Rainfall	5.62	2.47	1.16	6.72	5.57	0.95	1.63	0.73	0.82	2001
	Throughfall	6.13	9.21	1.94	9.36	8.52	3.72	8.76	3.31	1.27	
	Stemflow	5.71	13.0	4.19	23.7	17.6	7.92	11.3	4.15	2.26	
	Stream water	6.63	3.37	1.62	4.14	4.13	0.42	1.08	0.58	2.05	1994-1996*
JS	Rainfall	4.80	2.28	1.02	1.04	1.97	0.37	0.29	0.09	0.49	2002-2003
	Throughfall	5.60	3.26	2.45	1.92	3.03	1.14	3.57	0.46	0.81	
	Stemflow	5.10	2.33	2.00	1.26	2.78	0.95	2.40	0.27	0.67	
	Stream water	6.59	3.85	3.23	0.39	3.16	0.86	0.91	0.33	5.88	

*Kim (1997) averaged 1994-1996.

fall and stemflow ranged from 4.72 to 6.30 (average: 6.13) and 4.33 to 6.01 (average: 5.71), respectively. In Japan, the pH values of rainfall ranged from 4.77 to 6.39 (average: 4.80). The mean pH value of throughfall and stemflow ranged from 4.72 to 6.30 (average: 5.60) and 4.33 to 6.01 (average: 5.10). The mean pH values of rainfall were significantly lower than those of throughfall and stemflow in Korea and Japan. The mean EC values of rainfall, throughfall and stemflow at the KS and JP site were 2.47 mS/m, 9.21mS/m and 12.97 mS/m, and 2.28 mS/m, 3.26 mS/m and 2.33 mS/m, respectively. In particular, the EC values of throughfall and stemflow were much higher than that of rainfall at the KS site, and these facts indicated canopy and stem were washed off and leached by rainfall.

In particular, NO_3^- and SO_4^{2-} concentrations of rainfall, throughfall and stemflow in Korea were about 5 times to 15 times higher than those in Japan. The high NO_3^- and SO_4^{2-} concentrations in the throughfall and stemflow of *Quercus* forests were influenced in absorption with leaf, branch and stem by atmosphere pollution brought out from industrializations and urbanization. Takenaka and Win (1998) and Takenaka *et al.* (1998) emphasized that the NO_3^- and SO_4^{2-} concentrations in the throughfall and stemflow of coniferous trees were influenced in absorption with leaf and branch. The KS site is located in 100 km from Seoul and the Sihwa industrial complex, and is thus likely to be influenced by high levels of atmosphere pollution caused by industrialization and urbanization. Also, the tendencies of increase in pH values and chemical components were found at the JP site. Such trends appear more at the KS site than at the JP site, and these findings can be used for demonstrating that fertility of soil by nutrients input from forest.

Comparing the results from the two sites, the chemical properties of rainfall at the KS and JP site were lower than those of throughfall and stemflow in *Quercus* forest. The fluxes of nutrients in rainfall, throughfall and stemflow showed the same trends at the KS and JP site: SO_4^{2-} and NO_3^->K^+>Ca^{2+}>Mg^{2+}. The ion concentrations of rainfall, throughfall and stemflow at the KS site had about 2~8 times higher than at the JS site. Our results support these earlier reports that the contents of stemflow vary with tree species in Korea (Chun *et al.*, 1998a, 1998b; Park and Woo, 1998; Joo *et al.*, 1999; Kim *et al.*, 2000, 2001) and Japan (Gao *et. al.*, 2001; Sassa *et al.*, 1991; Takahashi, 1996). There were remarkable differences in chemical property between the two regions. These facts suggested that the rates of dry depositions in Korea were higher than those in Japan. Carlisle *et al.* (1966) analyzed the nutrient return in throughfall in an Oak stand over a complete year and in comparison with other pathways of nutrient returns, they emphasized the importance of throughfall in nutrient cycling.

Shown in Table 2, the mean pH values of stream water at the KS and JP site were 6.63 and 6.59, respectively. The table showed that the stream water in both regions is neutral water, and consequently was not different by regions. These pH values of stream water at the KS and JP site were similar to that of other stream in forest watersheds in Japan (Ohrui and Mitchell, 1998; Toda *et al.*, 2000), and were higher than those in America sites recorded below 6.2 (Ohte *et al.*, 2001). This indicated that any symptoms of acidification of stream waters were not found at the KS and JP site.

This pattern was showed in EC values, SO_4^{2-}, K^+ and Mg^{2+} concentrations, but Na^+, Ca^{2+}, Cl^- and NO_3^- concentrations were considerably different in level between the sites; NO_3^- concentration was high in KS while Na^+, Ca^{2+} and Cl^- concentrations were high in JP. In particular, the high NO_3^- concentration of stream water in KS suggested that Korean forest catchments in the high throughfall and stemflow were influenced by acid depositions. In contrast, Na^+, Ca^{2+} and Cl^- concentrations in JP site suggested that Japanese forest watersheds were influenced by coastal ele-

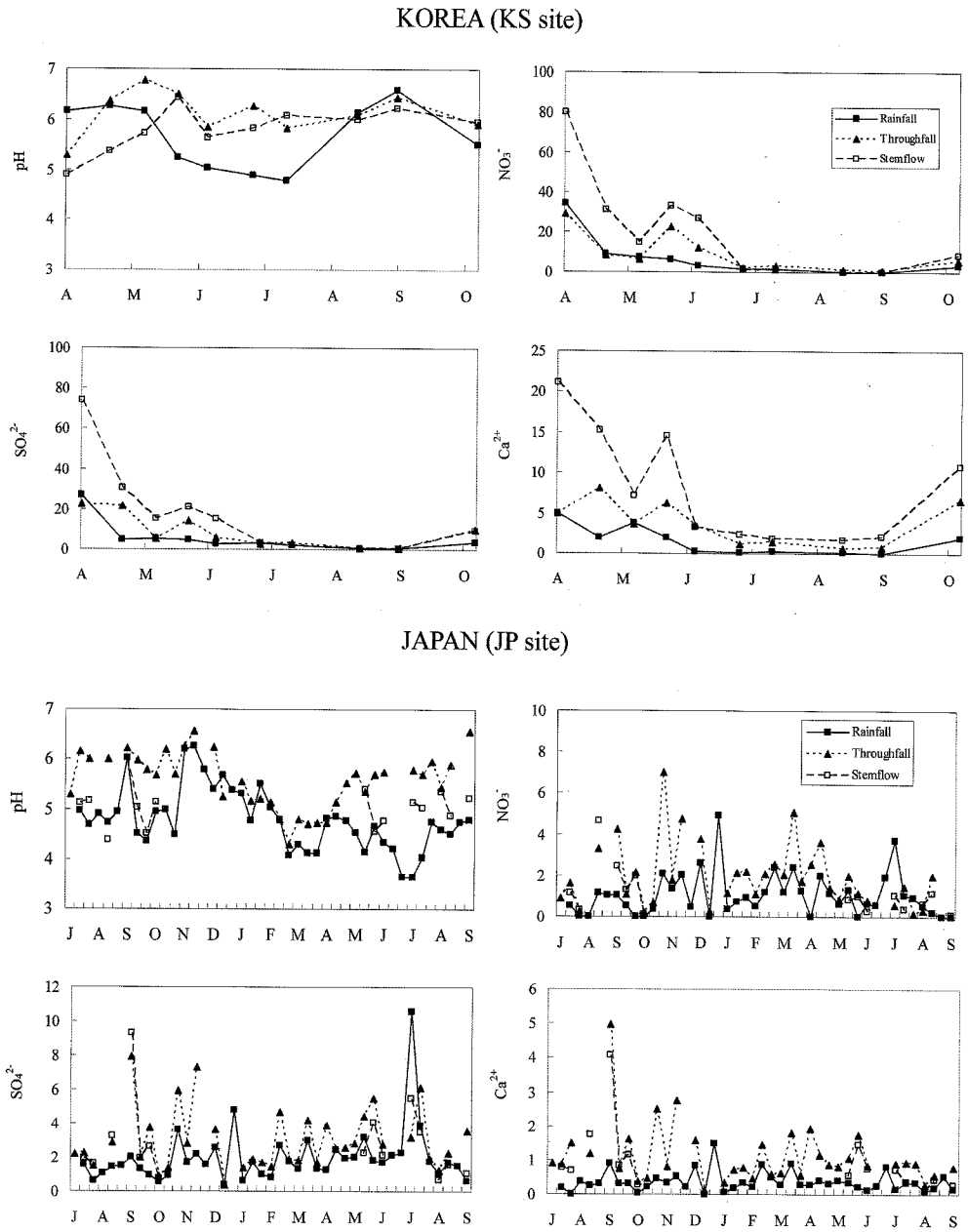

Figure 1. The variation of pH values, NO_3^-, SO_4^{2-} and Ca^{2+} concentrations in Korea and Japan.

ments. Ohrui and Mitchell (1996) reported that the high pH with high stream NO_3^- concentration has been reported for several sites in Japan.

2. Seasonal variations in rainfall, throughfall and stemflow in *Quercus* forest in KS and JP sites

Changes in the pH values, SO_4^{2-}, NO_3^- and Ca^{2+}concentrations of rainfall, throughfall and stemflow of *Quercus* forest in KS and JP sites are illustrated in Figure 1. In Figure 1, the lowest pH value for rainfall at the KS and JP sites occurred in July and August, and those under 4 at the JP site appeared particularly in 2003. The report by Takenaka *et al.* (1998) and Kim *et al.* (2001) found that the pH values, SO_4^{2-}, NO_3^- and Ca^{2+}concentrations of rainfall shown differences by seasons. In particular, the pH values of throughfall and stemflow at the KS showed seasonal patterns; It was low in spring while high in summer and fall. These patterns agreed with the previous report by Kim *et al.* (2000, 2001) and Chun *et al.* (1997, 1998b) from 1996 to 2000 conducted at the KS site on the same study area. In contrast, at the JP site, the same pattern was shown in the pH values of throughfall but those of stemflow had not differences among the seasons. Furthermore, the contents of rainfall, throughfall and stemflow show the same pattern to pH values, and Ca^{2+}, NO_3^- and SO_4^{2-} concentrations of rainfall, throughfall and stemflow were recorded remarkably at the KS site. This tendency appeared more in stemflow rather than throughfall and rainfall. As reported by Lian and Zhang (1998), annual pattern of nutrient inputs of

stemflow showed similar trends to bulk precipitation, with a peak in April - May. However, those at the JP site had not shown any difference among the seasons.

As described above, the elements measured at the KS site were extremely high compared with the data obtained from JP site. These results indicated that the contribution of this Yellow Sand appeared to determine the chemical composition of rainfall, throughfall and stemflow in spring (Kim *et al.*, 2001, 2005). Consequently, in spring time, deposition of Yellow Sand currently causes serious problem in Korea and Japan (Chohji *et al.*, 1997). It has been reported that Yellow Sand contains not only silicates such as $CaCO_3$ or CaO, but also acidic pollutants containing nitrate and sulfate (Chung *et al.*, 2000, 2001).

3. Seasonal nutrients inputs in *Quercus* forest floor between Korea and Japan

In order to clarify the seasonal contribution of nutrients inputs to the forest floor, all data were divided into four seasons spring (March - May), summer (June - August), fall (September - November) and winter (December - February). In Figure 2, the seasonal inputs of all nutrients in these two contrasting forests remained relatively stable throughout the observation period, although the components of total nutrient deposition varied seasonally; More than nearly 40% of the total inputs of nitrate (NO_3^-), sulfate (SO_4^{2-}), calcium (Ca^{2+}) and sodium (Na^+)

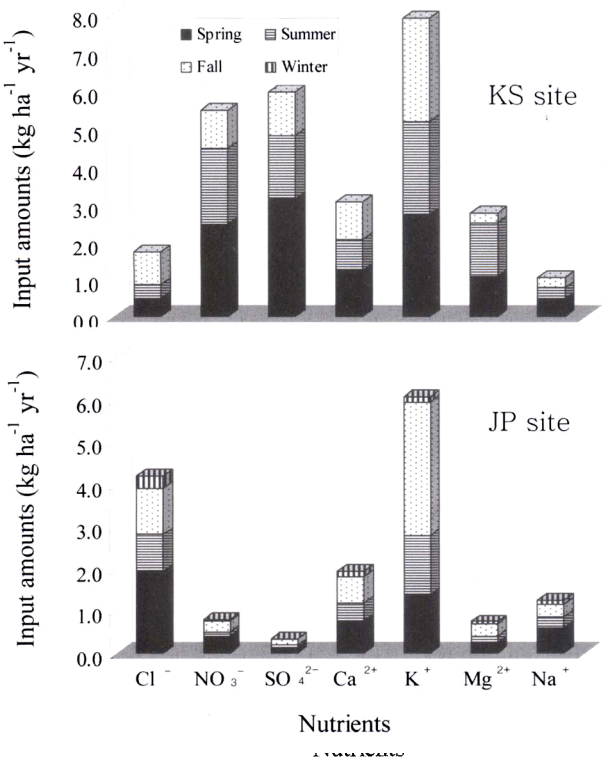

Figure 2. Seasonal input of various elements into *Quercus* forests stand in Korea and Japan.

were estimated to be supplied in spring. The total inputs at the KS site were higher than those at the JP site. In particular, the chemical analysis of precipitation in *Quercus* forests at the KS and JP site showed the regional importance of intensive inputs of nitrate, sulfate and calcium in spring.

In contrast, the inputs of potassium at the KS and JP site were not different among seasons and it is considered due to the characteristic of deciduous trees that potassium leaching from leaves increases as leaves age (Parker, 1983). Comparing data of all nutrients inputs between KS and JP site, the net fluxes of nutrients showed the same trends in *P. koraiensis* and *L. leptolepis* plantations in Korea (Kim *et al.*, 2005), and the input of nutrients in this study were lower than those in *P. koraiensis* and *L. leptolepis* plantations, however the inputs in KS site still were higher than those in JP site. In addition, the inputs of throughfall were the greatest compared with stemflow in the two contrasting forests. However, throughfall and stemflow determined seasonal peak of nutrient inputs. This result was consistent with the results reported by Callaway and Nadkarni (1991). Stemflow returns a small amount of nutrients to the forest floor compared with throughfall. Its proportional contribution to fluxes of net rainfall nutrients is between 1 and 20% (Parker, 1983) compared with 5.2 ~ 20.0% (KS) and 0.8 ~ 7.5% (JP) recorded in this study. Also, the high inputs of soluble nutrients from throughfall and stemflow would supply increasing nutrient availability.

Conclusion

We evaluated the effects of the difference in the chemical characteristics of acid deposition and nutrients cycles in *Quercus* forests in Korea and Japan. The pH values of rainfall at the KS site were higher than those at the JP site, and all chemical contents of throughfall and stemflow at the KS and JP site were much higher than those of rainfall in *Quercus* forest stands. The pH values, Ca^{2+}, NO_3^- and SO_4^{2-} concentration of throughfall and stemflow at the KS site showed seasonal variation. While at the JP site, the same pattern was shown in the pH values of throughfall, those of stemflow, however, did not show any difference among seasons. Also, the annual input of all nutrients in these two contrasting forests varied seasonally, but the inputs of potassium at the KS and JP site were not different among seasons. These results can be used to predict the amounts of air pollutant that are washed off and leached by the rainfall and Yellow Sand (Asian dust), including NOx and SOx acid pollutants transported easterly from China in the spring.

Furthermore, the neutral pH values with high NO_3^-

and SO_4^{2-} of stream water were shown at the KS and JP site and this suggests that forest catchments at the KS and JP do not show any symptoms of acidification of stream water. These results also indicated that the high Acid Neutralization Capacity (ANC) of forest ecosystem at the KS and JP site appeared in stream water via soil layers. Therefore, it is necessary to quantify the inputs of dry and wet deposition throughout a full year to gain a more complete understanding of the effects of acid deposition on the nutrient cycles in these forest ecosystems.

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