INTERANNUAL VARIATION OF LIMESTONE SOLUTION RATES IN JAPAN

KAZUKO URUSHIBARA-YOSHINO[1] AND RESEARCH GROUP OF SOLUTION RATES IN JAPAN*

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Summary

The solution rates of four tablets made from limestone obtained in Kozina (Slovenia), Guilin (China), Chichibu (Japan), and from the rock at the observation point were measured in seven limestone areas in Japan from 1993 to 1997. Solution rates in the air 1.5m above the ground show a high correlation with (water surplus (WS) - water deficit (WD)) by Thornthwaite's method. The solution rates of limestone tablets in soils show values 1.5 to 5 times higher than those in the air. The solution rates of limestone tablets in the A_3 and B_2 horizons show high correlation with annual precipitation. High solution rates in soils must be related to high CO_2 values under humid and warm periods. In 1993, when cool and humid summer prevailed in almost all parts of Japan, the trend of solution rates increased in accordance with (WS-WD), having the largest range during the five years. In 1994, when an extremely hot and dry summer prevailed, the solution rates increased in accordance with (WS-WD), having with the lowest range during the five years. Solution rates of limestone tablets in the air at each point increased with increasing (WS-WD) during the five years. However, it is interesting to note that the solution rates of limestone increased sharply by the amount of 1,000-1,600mm of (WS-WD). Lithologically, Guilin and Slovenia tablets belong to one group, and Chichibu and Akiyoshi tablets belong to the other group. The group of Guilin and Slovenia show high solution rate under the wet condition and low solution rate under the dry condition but the group of Chichibu and Akiyoshi show low solution rate under wet conditions.

Introduction

Many research workers, who have been interested to karst, have tried to clarify rates of solution and main factors for karstification. Measurements of solution rates using limestone tablets were tried by *Trudgill* (1975) and Jennings (1977). The Commission of International Speleological Union has measured solution rates of limestone tablets from the stand point of global comparison using the Slovenian Cretaceous limestone since 1980 (*Gams, 1985*). Similar limestone tablets made of Guilin Permian limestone were measured in China (*Yuan, 1991*). The solution rates of Slovenian Cretaceous limestone in Japan was measured from 1989 to 1990 by *Urushibara* (1991). Not only the solution rates of Slovenian Cretaceous limestone, but also those of Guilin Permian limestone and Japanese limestone have been measured since 1992. The results of these observation during the three years, 1993, 1994 and 1995, were published *elsewhere (Urushibara-Yoshino et al., 1998)*.

Five years results were also published in Japanese (*Urushibara-Yoshino et al., 1999*). In this study, the effective factors for annual solution rates of limestone in Japan are discussed using measured values.

Study Areas and Methods

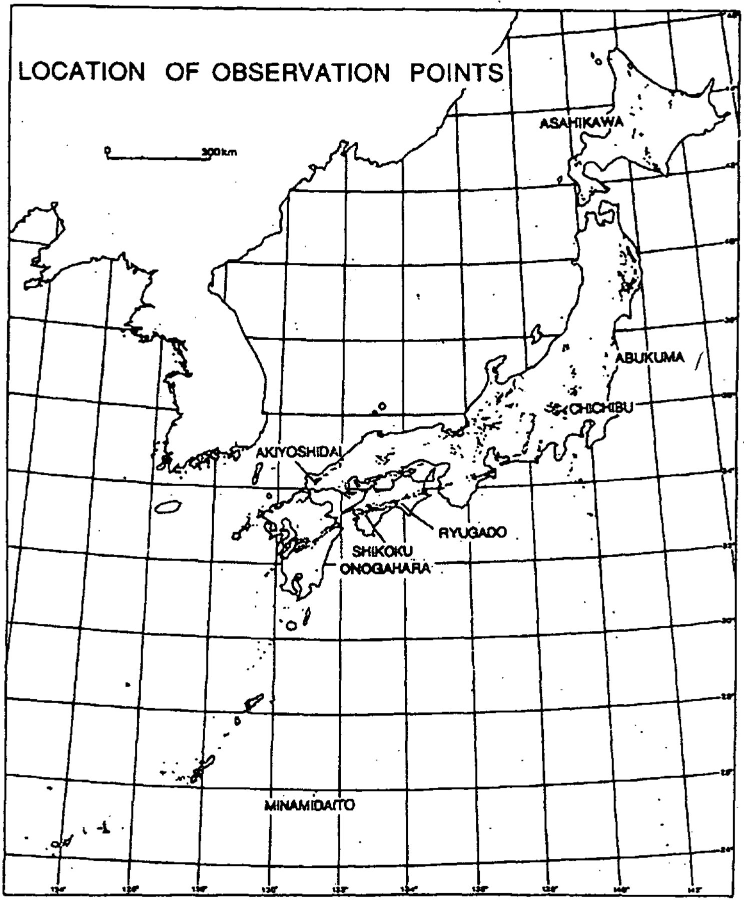

Fig. 1 Limestone areas and location of observation points in Japan.

Limestone areas occupy only 0.44% of the all land in Japan (*Urushibara, 1996*). However, the areas are distributed from north to south under a wide range of different climatic conditions. The seven limestone areas were selected from north to south, namely Toma, Abukuma, Chichibu, Akiyoshidai, Shikoku, Ryugado and Minamidaito. The locations of these points are shown in *Fig. 1* and their environmental conditions in *Table 1*. The tablets were made from Slovenian Cretaceous limestone, Guilin Permian limestone, Chichibu Triassic limestones and the local limestone at each location in Japan and were

40mm in diameter with a thickness of 4mm. They were set in the open air 1.5m above the ground and in the soils at the A_3 horizon and B_2 horizon. Their depth was different because of the different depth of horizons. The weights of the tablets ranged from 12.5g - 13.5g. Once a year during the five years, the tablets were cleared and weighted. The results of weight loss had shown in 10^{-4}g to 10^{-5}g then, calculated as the solution rate for the year. By microscopic observation, the upper surfaces of tablets corroded more than the lower surfaces at all observation points. Therefore, because of differential solution between the upper and lower surfaces, the loss of thickness was not calculated but only the loss of weight was measured.

The CO_2 concentrations in soils were paid attention in the present study, because the solution rates are extremely high in soils. In this study, the CO_2 in A_3 and B_2 horizons, where the tablets were set, was measured in at least 4 seasons at each localities. The three methods, Non Dispersive Infrared Gas Analyzer, Gastec and Dräger method (*Miotke, 1974*) were *examined (Urushibara-Yoshino et al., 1998*). To further understand the solution processes of limestone, the water balance in 1993, 1994, 1995, 1996 and 1997 was calculated by the methods of *Thornthwaite* (1948), because it considers the water balance including soil moisture. During the five years measured, the summer of 1993 was very cool and wet, and the summer of 1994 was extremely hot and dry. In June - August, 1994, monthly mean air temperature was the highest in the whole Honshu area during the last 100 years observation period, according to meteorological data.

Table 1 Environmental conditions of the observation points.

Observation points	Location	Elevation	Stratigraphy and Ecological Condition
Asahikawa (Toma)	43°49'30''N 142°37'50''E	220m a. s. l.	Permian limestone mixed forest with needle trees and desidual trees
Abukuma	37°20'30''N 140°40'40''E	640m a. s. l	crystallized limestone by Cretaceous metamorphism deciduous forest and grassland
Chichibu	35°57'30''N 139° 6'10''E	960m a. s. l.	Trias limestone deciduous forest and grassland
Akiyoshidai	31°14'50''N 131°17'40''E	240m a. s. l.	Carboniferous and Permian limestone mixed forest with evergreen and deciduous forest, and grassland
Shikoku (Onogahara)	33°28'24''N 132°52'51''E	1200m a. s. l.	Permian limestone deciduous forest and grassland
Ryugado	33°54'54''N 133°44'53''E	280m a. s. l.	Trias limestone evergreen forest and grassland
Minamidaito	25°50' 7''N 131°14' 1''E	14m a. s. l	Quaternary limestone subtropical evergreen trees and grassland

Solution rates of limestone tablets

In comparing the solution rates of the four limestone tablets (Slovenian limestone, Guilin limestone, Chichibu limestone and local limestone tablets) set in the air, in the A_3 horizon of soils and in the B_2 horizon of soils at each observation points, the following results were obtained. The mean solution rates of four limestone tablets were considered as the best estimate of solution rate at each position at each observation points. The solution rates at 1.5m above the ground were higher in southern Honshu and Shikoku and lower in

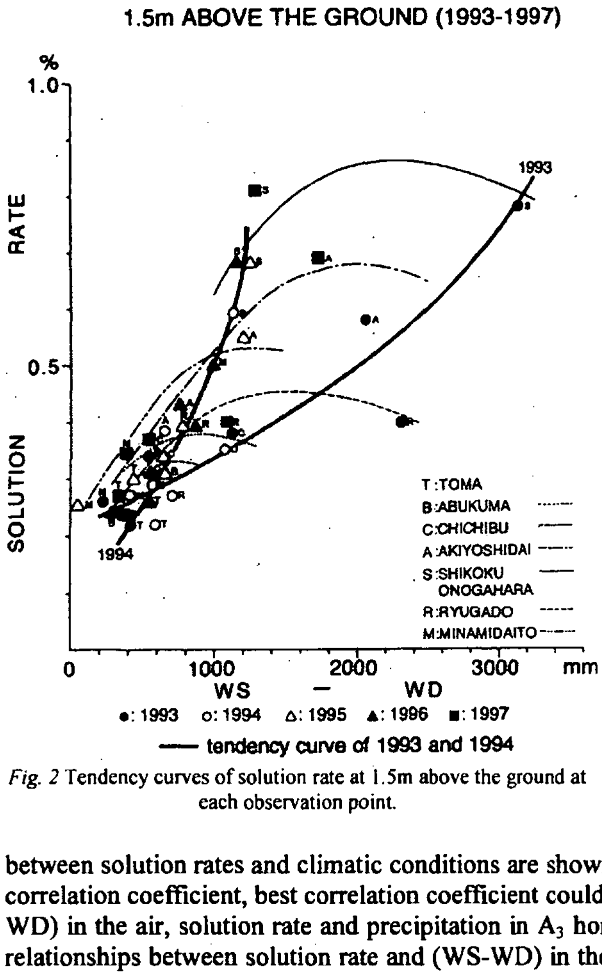

Fig. 2 Tendency curves of solution rate at 1.5m above the ground at each observation point.

northern Honshu and Hokkaido. The solution rates in southern Japan are generally higher than those in northern Japan. However, they are not the highest in Minamidaito in air, because of dry periods that appear during summer. The inter-annual fluctuations of the solution rates in Japan are very clear from 1993 to 1997. Extremely high solution rates appeared in 1993, a very wet and cool summer, and the smallest in 1994, an exceptionally dry and hot summer. As an example of these two years, mean annual solution rate at the three points (Toma, Akiyoshi, Minamidaito) in 1993 and 1994 are shown in *Table 2*. Original weight of tablets was in the range of 12.5g-13.5g as mentioned above. Therefore, the mean annual solution rate of four tablets at various situation of the location was used for the discussion of differences location to location and year to year fluctuations. The relationships between solution rates and climatic conditions are shown in *Table 3*. Showing the matrix of correlation coefficient, best correlation coefficient could be obtained solution rate and (WS-WD) in the air, solution rate and precipitation in A_3 horizon and B_2 horizon. The trends of relationships between solution rate and (WS-WD) in the air 1.5m above the ground at each observation point during the five years are shown in *Fig. 2*. The solution rate increases in accordance with (WS-WD) at values lower than 1000-1600mm for each point. However, they decrease slightly at the higher values over the maximum points. The maximum solution rates are different at each location, but appear within the range of 1000-1600mm of (WS-WD). At the observation points of Shikoku, Akiyoshidai and Ryugado the maximum values are higher than those of the other four points.

Comparing the values year by year, it is clearly shown that the solution rates at the seven observation points increase in accordance with (WS-WD). In 1993, with cool and humid summer, the trend shows the largest (WS-WD) during the five years. In contrast, it shows the lowest (WS-WD) in 1994 with extremely hot and dry summer during the five years. Under the influence of monsoon climate in Japan, solution rate of limestone shows values higher than 0.5%, when (WS-WD) over 1000mm. In the range of (WS-WD) from 0 to 1000mm, the solution rate increases with positive relation taking the values of 0.2 to 0.5% with (WS-WD).

Table 2 Annual solution rate (%) in Toma, Akiyoshi and Minamidaito.

TOMA (ASAHIKAWA)

	1993	1994
AIR	**0.2173***	**0.2228**
A3 HORIZON	**0.4056**	**0.4651**
B2 HORIZON	**0.5476**	**0.6341**

AKIYOSHIDAI

	1993	1994
AIR	**0.5834**	**0.3782**
A3 HORIZON	**2.8828**	**1.2498**
B2 HORIZON	**2.6578**	**1.3951**

MINAMIDAITO

	1993	1994
AIR	**0.2623**	**0.2660**
A3 HORIZON	**1.0407**	**1.2819**
B2 HORIZON	**1.3714**	**1.2984**

* 0.2173% : mean solution rate of 4 tablets

Table 3 Correlation matrix for 5 years (1993-1997) between solution rates of limestone tablets and climatic factors

	PREC.	WS	WS - WD	P - E
1.5m ABOVE THE GROUND	r = 0.695	r = 0.744	r = 0.749	r = 0.745
A_3 HORIZON	r = 0.485	r = 0.426	r = 0.409	r = 0.406
B_2 HORIZON	r = 0.562	r = 0.436	r = 0.418	r = 0.416

CO_2 in soils

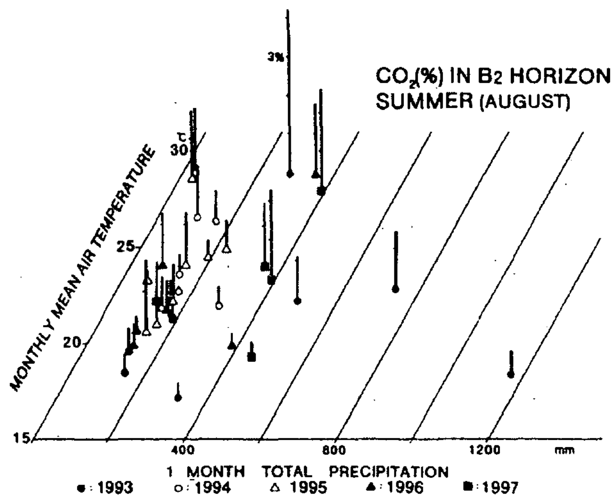

Fig. 3 Soil CO_2 in the B_2 horizon, monthly mean air temperature and one month precipitation prior to measurement, August 1993-1997.

In this part of the study, carbon dioxide concentrations in the A_3 and B_2 horizons measured at the seven points are discussed. The CO_2 in the B_2 horizon shows higher values than the A_3 horizon in almost all cases, even though A_3 horizon is the most productive horizon. The detailed results from 1993 to 1995 have been reported in *Urushibara-Yoshino* (1998) and *Urushibara-Yoshino et al.* (1999). It is considered that CO_2 gas accumulated and was retarded longer in the B_2 horizon than in the A_3 horizon. Therefore the solution rates in

B_2 horizon were higher than that of in A_3 horizon in almost all cases. Carbon dioxide concentrations of the B_2 horizon in summer (August) are shown in *Fig. 3*. In summer, CO_2 concentrations show higher values, when monthly mean air temperature is over 22 and monthly precipitation before the measurements are lower than 400mm. A monthly precipitation higher than 400mm is less effective for increasing CO_2%. In winter (February), observation point at Minamidaito shows a high CO_2 value from 1 to 2%, because of higher monthly mean air temperatures over 17. However, the other points show lower CO_2 less than 2,000ppm with reduced temperature, because of less assimilation and less CO_2 production. It can be summarised that these high CO_2 contents in the B_2 horizon during the warm period can support higher solution rates of limestone tablets in soils. However, the solutional activity in winter is extremely small, because of low CO_2 in B_2 horizon.

Solution of tablets at Location Akiyoshi

The tablets at Location Akiyoshi show typical results of solution rate from year to year and lithological differentiation as mentioned above. Therefore, as examples of the results measured at Locations Akiyoshi, the four tablets are taken for discussion. The climatic condition from 1993 to 1997 at Location Akiyoshi are shown in *Table 4*.

Table 4 Climatic conditions of the Akiyoshidai station from 1993 to 1997

	1993	1994	1995	1996	1997
Precipitation	2773	1411	1992	1488	2458
Annual temperature	13,1	14,3	13,1	13,2	13,7
Evapotranspiration	720,7	788,6	737,7	741,0	752,8
Water surplus	2052,3	695,5	1206,1	747,0	1705,2
Water deficit	0	40,9	0	0	0

Lithological differences

The four tablets, namely those from Regions Akiyoshi, Chichibu, Slovenia and Guilin were measured at Location Akiyoshi. The particle sizes of calcite of each tablets and geological ages are as follows:

- Tablets from Region Akiyoshi: Carboniferous, Permian; average size of calcite diameter 7.5μm; and maximum size of diameter 78.2μm.
- Tablets from Region Chichibu: Trias; average size of calcite diameter 7.5μm; and maximum size of diameter 60.9μm.
- Tablets from Region Slovenia: Cretaceous; average size of calcite diameter 4.0μm and 10.6μm; and maximum size of diameter 74.2μm.
- Tablets from Region Guilin: Devonian; average size of calcite diameter 3.6μm and 11.6μm; and maximum size of diameter 100.5μm.

Inter-annual variation of solution rate of four different lithological tablets

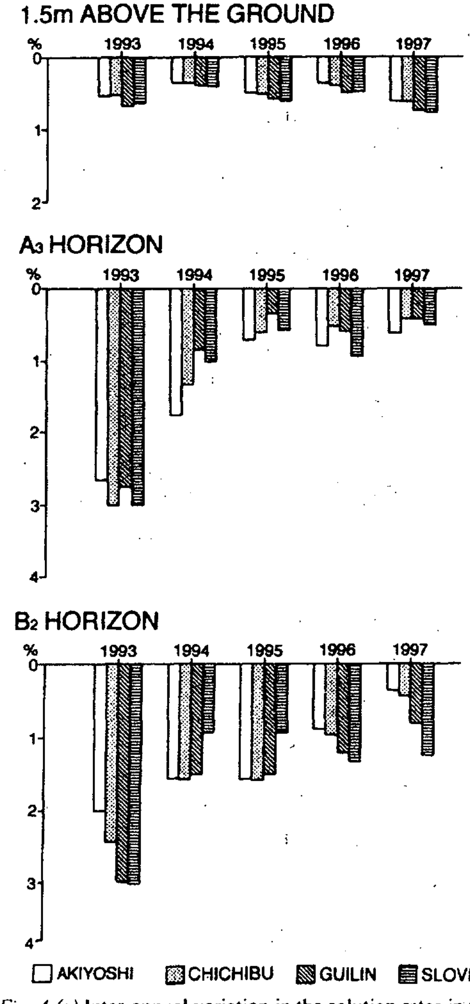

Fig. 4 (a) Inter-annual variation in the solution rates in the four tablets in the air 1.5m above the ground, and A_3 and B_2 horizons in the soils.

Four different lithological tablets, namely those from Regions Akiyoshi, Chichibu, Guilin and Slovenia, were compared by taking inter-annual variation of solution rate from 1993 to 1997.

1. In the air

Solution rates of four tablets show clear tendency in the air. The values of solution rates are in the range of 0.2% in a year. The tablets from Akiyoshi and Chichibu show similar tendency of inter-annual solution rates. The tablets from Guilin and Slovenia show higher solution rates than those from Akiyoshi and Chichibu in every year as shown in *Fig. 4 (a)*. Especially, the tablets from Guilin and Slovenia show greater solution rates in 1993, wet year. In the air, inter-annual solution rates are divided clearly into two groups. The cumulative solution rates show also that the 5 years results of the tablets from Guilin and Slovenia are bigger than those of value of the tablets from Akiyoshi and Chichibu as shown in *Fig. 4 (b)*.

2. In the A_3 horizon

The maximum value of inter-annual solution rates appeared in 1993, wet year. In 1993, the solution rate of the tablet from Akiyoshi shows minima among the four tablets. The tablets from Slovenia showed maximum solution rates in 1993. On the other hand, the solution rates of the four tablets were not minima in 1994, the extremely dry year. The minimum values appeared in 1995. This tendency can be seen also in B_2 horizon. It suggests that the condition of soils or activity of organic decomposition in soils appeared slightly later than those under the conditions in the air.

In 1994 and 1995, the solution rates of the tablets from Chichibu and Akiyoshi were higher than those from Guilin and Slovenia. The solution rates were different

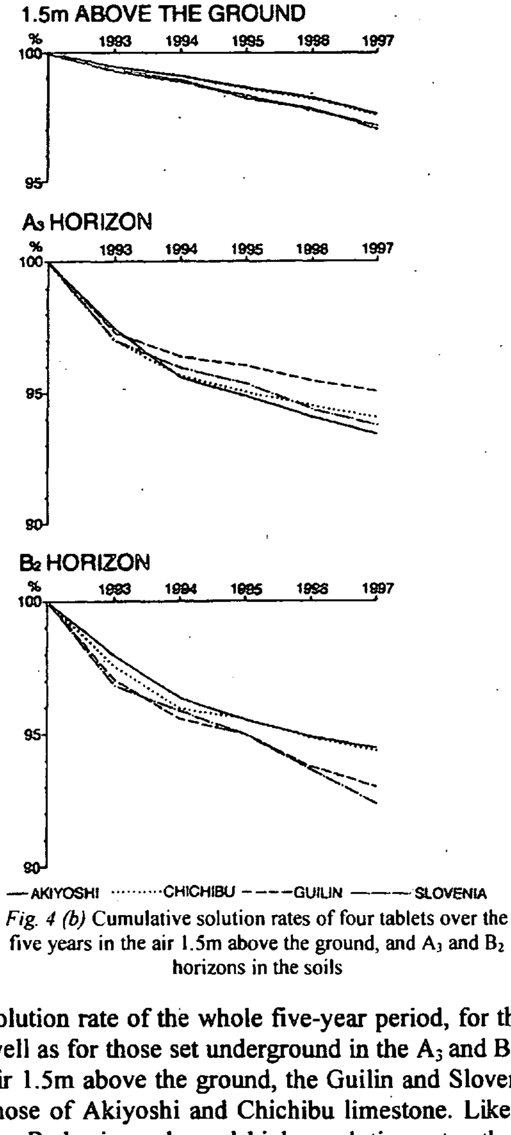

Fig. 4 (b) Cumulative solution rates of four tablets over the five years in the air 1.5m above the ground, and A_3 and B_2 horizons in the soils

lithologically; namely they show opposite tendencies in the soils at dry year, as compared with those in the air.

3. In the B_2 horizon

The maximum solution rates of the four tablets appeared in 1993. The values in B_2 horizon were 3 to 5 times higher than those in the air. The range of values of B_2 horizon was the biggest in 1993. In 1993, the groups of the tablets from Akiyoshi and Chichibu show lower rates than those from Guilin and Slovenia. The groups of Guilin and Slovenia show maximum solution rates regardless the conditions of the five years. The longer period of wet condition in B_2 horizon will be accelerated effectively for the Slovenia and Guilin tablets. In the dry year, 1994, the solution rates of the four tablets showed lower values, but did not show minimum. As same to A_3 horizon, the minimum values appeared in 1995.

Solution rates of extreme year tendency at Location Akiyoshi

The year 1993 was an exceptionally wet year, having the highest annual precipitation among the five-year period as shown in *Table 4.* The year 1993 exhibited the maximum solution rate of the whole five-year period, for the tablets set at 1.5m above the ground as well as for those set underground in the A_3 and B_2 horizons. Of the tablets suspended in the air 1.5m above the ground, the Guilin and Slovenian tablets had higher solution rates than those of Akiyoshi and Chichibu limestone. Likewise the Guilin and Slovenian planted in the B_2 horizon showed higher solution rates than the B_2 horizon Akiyoshi and Chichibu tablets. However, this difference between Guilin, Slovenian, and Akiyoshi, Chichibu tablets were not clear for the tablets planted in the A_3 horizon.

The year 1994 was a dry year; it had the least precipitation in the five-year period as shown in *Table 4*. For the tablets suspended 1.5m above the ground, the solution rates of all four types of tablets were smaller in the five-year period. For the tablets suspended in the air, the solution rates of the Guilin and Slovenian tablets were less than those of the Akiyoshi and Chichibu tablets. However, the year 1994 was not the smallest solution rate for the tablets planted in the B_2 horizons; the year 1995 exhibited the minimal solution rate in the five-year period. The measurement of the CO_2 in the soil revealed that 1995 had shorter periods of high CO_2 content. This suggests that the solution rates of 1995 reflect the CO_2 content of the soil. The 1995 solution rate of the Guilin tablets in the A_3 horizon was the smallest of the five years, while under dry conditions probably because the grain size of their calcite is smaller than that in the other types of tablet.

CO_2 effect at Location of Akiyoshi

Over the five-year period, the solution rates of the tablets planted in the A_3 and B_2 horizons were three to five times greater than those of the tablets suspended in the air. As shown in *Fig. 5*, CO_2 in A_3 and B_2 horizons had a clear relationship to daily precipitation. It can be understood that the higher solution rate was occurred due to the persistence of water contents in the soil and the development of high CO_2 contents in the wet soil. High CO_2 contents develops in the soil in the warm season, after it rained for five days consecutively with precipitation of 50mm to 100mm per day.

Especially, it is clearly shown that high CO_2 content in A_3 horizon was appeared earlier after rainfall. In contrast, high CO_2 content in B_2 horizon was appeared slowly after rainfall. The CO_2 content in B_2 horizon was higher than in A_3 horizon. It seems that the tight layer of B_2 horizon can keep CO_2 gas longer than that of A_3 horizon.

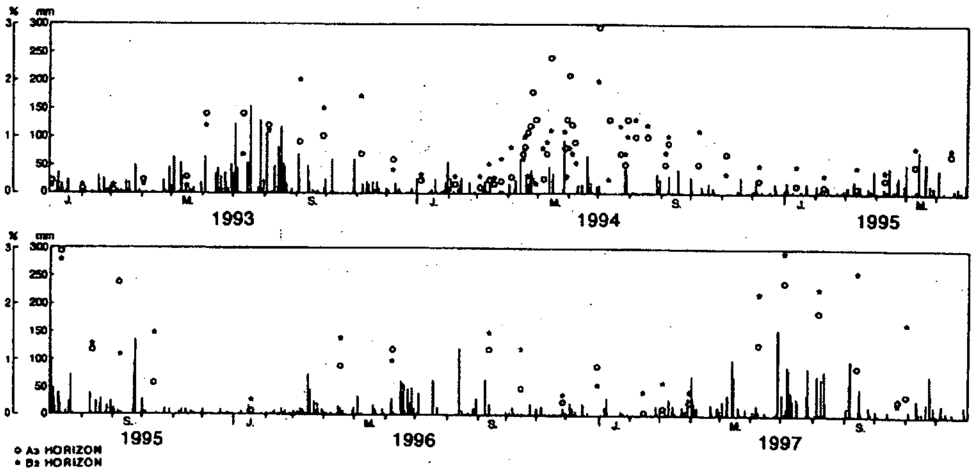

Fig. 5 CO_2 % in A_3 and B_2 horizons and rainfall at Akiyoshidai station from 1993 to 1997

Conclusions

Solution rates of four tablets made from limestone obtained in Slovenia, Guilin, Chichibu and from the limestone at each of seven observation points in Japan were measured from 1993 to 1997. The solution rates were measured for estimating karstification rate of bare karst and soil covered karst in monsoon climate in the temperate and subtropical zones. The results are summarised as follows:

- Solution rates of limestone tablets in soils are 1.5 to 5 times higher than those in the air 1.5 m above the ground. This means that karstification will progress with a high rate in the soil than on the bare karst (without soil) in Japan.
- The correlation coefficient between solution rates in the air and (WS-WD) is the highest for the five years. On the other hand, in the A_3 and B_2 horizons, correlation coefficients between solution rate and annual precipitation were highest in the five years observed.
- In the five years, solution rate of limestone at the four observation points shows inter-annual fluctuations. Especially, the solution rates in the air 1.5m above the ground show clear trend with (WS-WD). At each observation point, the solution rate increases in accordance with (WS-WD), for the range 1,000-1,600mm. Then solution rates decrease in the range above 1,600mm in (WS-WD).
- The following considerations summing up the facts obtained from the experimental measurements can be possible. The dominant solution factor is climate. The solution rates reveal inter-annual variation according to atmospheric climatic conditions. The inter-annual variation of the solution rates of tablets planted underground is a function of the rate of precipitation and the CO_2 content of the soil. The subsidiary factor for solution rate is the lithological properties of the limestone. For the limestone suspended in the air, the five year cumulative solution rates of the Guilin and Slovenian tablets were 20% higher than the Akiyoshi and Chichibu tablets. The Guilin and Slovenian tablets planted in the B_2 horizon had slightly higher cumulative solution rates than the Akiyoshi and Chichibu tablets suspended 1.5m in the air.
- From these results, Shikoku and Akiyoshidai have the best water balance condition for the solution of limestones in air in the monsoon temperate zone. Because of these situations with good water balance condition, karstified limestone plateaus with dense dolines develop relatively better in these areas of Japan. Of course, for the development of dolines, bedding planes, thickness of layers, fissure pattern in limestone also assist karstification. However, in Minamidaito island, solution rates of limestone in the air are not the highest, because of dry periods in summer. In the soils of Minamidaito, the solution rate of limestone tablets show the highest values except in 1995. This means that walls rugged karst development is possible under present climatic conditions in Minamidaito.

Acknowledgements

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