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Stable isotopic composition of calcic paleosols of the Early Cretaceous Hasandong Formation, southeastern Korea

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Abstract

Abundant pedogenic carbonate nodules are present in the Cretaceous non-marine Hasandong Formation, southeastern Korea. The oxygen isotope compositions of these pedogenic carbonates range from −14.2 to −18.0‰ (PDB). The meteoric water composition estimated from the oxygen isotope composition of the soil-water in equilibrium with the carbonates is much depleted compared with the previous estimate. This suggests that the oxygen isotope compositions of the Hasandong pedogenic carbonates were modified during diagenesis. The carbon isotopic compositions of the Hasandong carbonates range from −2.4 to −9.3‰ (PDB) with an average of −5.6‰ (PDB) suggesting carbonate formation in soils dominated by C_3 type of vegetation. The estimated average composition of the vegetation is about 6‰ (PDB) enriched compared with the present-day C_3 vegetation. This is probably due to atmospheric influence contributing about 35% of the total CO_2 in the soil. An analysis of this contribution using the model of Cerling (1991) indicates that the partial pressure of CO_2 in the Early Cretaceous Hasandong atmosphere was about 2300 ppmV.

Keywords: Cretaceous; paleosol; carbonate nodule; stable isotopes; vegetation

1. Introduction

Soil carbonates form in soils with a net water deficit, generally in soils where precipitation is less than about 100 cm per year (Jenny, 1980; Birkland, 1984). They are often preserved in continental sediments in paleosols and therefore are a potentially important paleoenvironmental indicator of continental climatic and ecologic conditions (Retallack, 1990). Recent studies have demonstrated that the oxygen and carbon isotope compositions of paleosol carbonates can be used to infer paleoclimate and paleoecology (Anderson and Arthur, 1983). The oxygen and carbon atoms constituting the pedogenic carbonates ($CaCO_3$) are derived from soil-water and soil-CO_2, respectively. Therefore, the isotopic compositions of oxygen and carbon in pedogenic carbonate reflect the compositions of these two sources.

The oxygen isotopic composition of soil-water is related to local meteoric water, whose oxygen isotopic composition is controlled by a number of meterological and orogenic parameters like amount and seasonality of the rainfall, latitude and elevation of the station and its distance from the coast, etc. (Rozanski et al., 1993). On the other hand, the carbon isotopic composition of soil-CO_2 is controlled by the composition of the plant-derived CO_2, the depth

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in the soil profile, difference in the pressures of soil-CO_2 and atmospheric CO_2, etc. (Cerling, 1984). The composition of the soil-CO_2 and its pressure are related to the nature and volume of vegetation and their seasonal activity (Cerling, 1984, 1991; Cerling et al., 1989; Mack et al., 1991).

The Early Cretaceous Hasandong Formation in the Gyeongsang Basin, southeastern Korea (Fig. 1), contains numerous horizons of pedogenic carbonate. The goals of this study are to analyze the stable isotopic compositions of these pedogenic carbonates, to infer the paleovegetational set-up and to estimate the partial pressures of atmospheric CO_2 at the time of the deposition of the Hasandong Formation. The Cretaceous is known as a greenhouse period. Although ancient atmospheric p_{CO_2} levels have been modeled (Berner, 1991), there is a need to gather more field measurements at many stratigraphic levels and localities to verify such modeling results. For the Cretaceous only one study (Ghosh et al., 1995) and two estimates (Cerling, 1991) based on literature have been reported. The result of this study may provide additional information to such a database.

2. Stratigraphy

The Cretaceous sedimentary rocks in the Gyeongsang Basin were deposited in the non-marine environments and are divided into three groups (Chang, 1975). They are the Sindong, Hayang, Yucheon groups in ascending order. The Hasandong Formation is a middle stratigraphic unit of the Sindong Group, which is distributed along the western margin of the Gyeongsang Basin (Fig. 1). It is 550–1400 m thick (Chang, 1975) and consists of sandstones, reddish and gray silty shales, and minor conglomerates which were deposited in the fluvial channels and on the floodplains (Choi, 1985, 1986). The formation is underlain by the Nagdong Formation (alluvial fan to fluvial deposits) and is overlain by the Jinju Formation (lacustrine deposits).

The geologic age of the Hasandong Formation has been generally inferred to be Hauterivian by the charophyte study from the underlying Nagdong Formation (Seo, 1985), although the Aptian to Albian age was suggested by studies on molluscan fauna (Yang, 1982) and paleomagnetism (Doh et al., 1994). Abundant fossils of vascular plants, bivalves, and some gastropods have been reported from mudstones of the Hasandong Formation. Most of them indicate an Early Cretaceous age and non-marine environments. The paleolatitude of the Korean Peninsula during the Cretaceous was similar to the present one (Kim et al., 1993).

3. Methods

A total of eighteen samples of pedogenic carbonate nodules developed in the floodplain deposits were collected from various localities (Fig. 1). In all the localities sampling was done for several nodules in single horizons. All the samples consist of caliche nodules (globules) and tubular rhizocretions. The gleyed soils were avoided in order to sample soils of high free-air porosity.

Stable isotopic measurements were carried out on powdered samples collected by a dental drilling machine after the nodules were sliced to expose the fresh surface. Drilling sites were chosen far from any cracks or fractures to avoid contamination from carbonate of non-pedogenic origin. Stable carbon and oxygen isotopes of samples were analyzed by a VG PRISM Series II Model at the Korea Basic Science Center. The analyses were carried out following a technique similar to that described by Swart et al. (1991). The isotopic ratios of carbon and oxygen are presented in standard-notation (‰) in the PDB scale (Table 1). Reproducibility of the analyses was ± 0.1‰ for both the carbon and oxygen isotopes.

4. Description of pedogenic carbonate

A typical paleosol-bearing columnar section of the Hasandong Formation is shown in Fig. 2. In the Hasandong Formation two types of paleosol occur: vertic and calcic paleosols. In calcic paleosols, paleosol horizons consist of common, diffusive and irregular to crudely cylindrical, rounded or discoidal carbonate nodules set in sandy mudstone (Fig. 3A). The enclosing non-calcic sandy mudstones are blocky and show a purple color with greenish-gray mottles. The carbonate nodules vary from a few millimeters to >10 cm across or long and

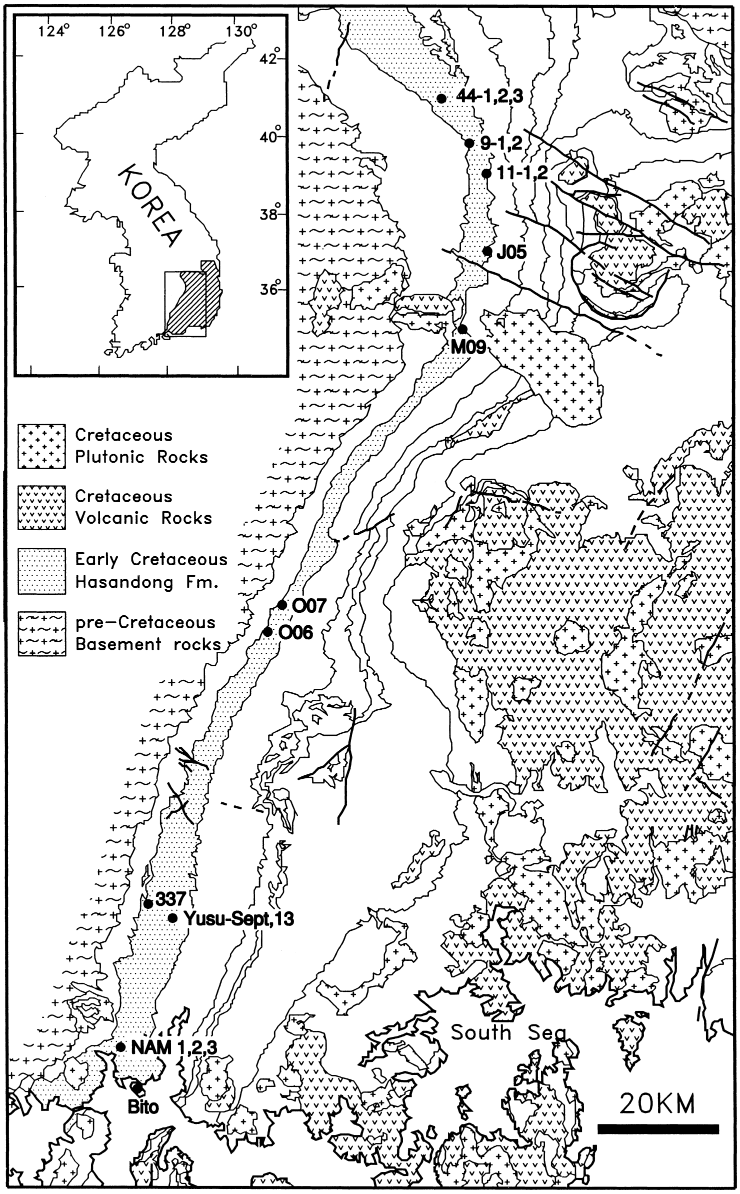

Fig. 1. Simplified geologic map of the Hasandong Formation in the Gyeongsang Basin (modified after Um et al., 1983). Sampling locality is represented by a dot with a sample number.

may locally coalesce with each other. The carbonate nodules are generally concentrated to form calcic horizons that contain from less than 10 to 40 weight percent carbonate. They show a light purple color on weathered surface with some greenish- to dark-gray colors. The calcic horizons are characterized by a gradual upward increase in carbonate nodule size and density. The overall degree of pedogenic carbonate development in the Hasandong Formation ranges from Stage I to III (classification of Machette, 1985). Some nodules contain septarian crystallaria which narrow from the center of the nodule outward (Fig. 3B). Spar-filled circumnodular cracks are also developed around nodules whose diameter is

Table 1
Isotopic data of the Hasandong pedogenic carbonates along with calculated composition of meteoric water and associated vegetation

Sample code	$\delta^{13}C$ (‰)	$\delta^{18}O$ (‰)	$\delta^{18}O$ of soil water (‰) [a]	$\delta^{18}O$ of meteoric water (‰) [a]	$\delta^{13}C$ of soil $C0_2$ (‰) [a]	Estimated $\delta^{13}C$ of plant CO_2 (‰) [a]
NAM 1	−4.5	−16.7	−14.7	−16.7	−14.2	−18.6
NAM 2	−3.9	−15.9	−13.9	−15.9	−13.6	−18.0
NAM 3	−4.0	−15.9	−13.9	−15.9	−13.7	−18.1
9-1	−7.0	−18.0	−16.0	−18.0	−16.7	−21.1
9-2	−4.8	−17.5	−15.5	−17.5	−14.5	−18.9
44-1	−6.2	−17.9	−15.9	−17.9	−15.9	−20.3
44-2	−6.0	−16.3	−14.3	−16.3	−15.7	−20.1
44-3	−3.9	−16.3	−14.3	−16.3	−13.6	−18.0
11-1	−7.6	−17.2	−15.2	−17.2	−17.3	−21.7
11-2	−6.2	−16.8	−14.8	−16.8	−15.9	−20.3
J05	−7.8	−17.9	−15.9	−17.9	−17.5	−21.9
337	−9.3	−5.6 [b]	−3.6	−5.6	−19.0	−23.4
M09	−5.2	−17.6	−15.6	−17.6	−14.9	−19.3
O07	−3.3	−14.2	−12.2	−14.2	−13.2	−17.6
O06	−2.4	−17.5	−15.5	−17.5	−12.1	−16.5
Bito	−6.8	−16.8	−14.8	−16.8	−16.5	−20.9
Yusu-Sept	−6.4	−16.1	−14.1	−16.1	−16.1	−20.5
Yusu-13	−6.0	−15.4	−13.4	−15.4	−15.7	−20.1

[a] These values are calculated assuming soil water in equilibrium with soil carbonate and soil CO_2 at 25ºC. $\delta^{18}O$ values are with respect to SMOW and $\delta^{13}C$ with respect to PDB.
[b] This value is considered as an outlier.

larger than 10 cm. Internally, the carbonate nodules consist of homogeneous microcrystalline calcite and microspar. Micrite constitutes over 90% of the volume of the carbonate nodules, with the remainder being randomly scattered detrital grains, resulting in a carbonate mudstone texture. Sparry calcite, 50 to 100 μm thick, fringing detrital grains of quartz and other rock fragments are frequently observed. Also observed are spar-filled root molds (1–2 cm in diameter) and fine networks of rootlets (<1 mm in diameter) (Fig. 3C). Some carbonate nodules contain fitted peloids, circumgranular and curved cracks filled with crystallaria of sparry calcite, and breccia-tion fabric. *Microcodium* is observed in 10-cm-long carbonate tubules (Paik and Lee, 1995). Some calcic horizons are characterized by selective occurrence of greenish-gray carbonate in mud cracks (Fig. 3D). In plan view, the carbonates are distributed in a polygonal pattern and in cross-section they are cylindrical and tapering downward.

The carbonate nodules in the Hasandong Formation are of pedogenic origin based on the following characteristics: (1) the presence of calcareous root traces and rhizoliths as well as *Microcodium*; (2) the presence of circumgranular and curved cracks, fitted peloids and calcite aureoles around detrital grains; (3) vertical profiles of two or more horizons with the calcic interval present toward the base; and (4) purple horizon colors (e.g., Esteban and Klappa, 1983; Ettensohn et al., 1988; Retallack, 1988, 1990). The poor stratification of the nodule-bearing fines and the presence of intraclast horizons (Fig. 2) resulted from reworking of pedogenic carbonates also support a pedogenic origin of the nodules.

5. The stable isotope composition

The $\delta^{13}C$ values of the pedogenic carbonates from the Hasandong Formation have a mean −5.6‰ and standard deviation of 1.7‰. The corresponding mean value of $\delta^{18}O$ is −16.7‰ with a standard deviation of 1.0‰. The $\delta^{18}O$ and $\delta^{13}C$ values are plotted in a correlation diagram in Fig. 4, which shows that except one outlier the values are closely clustered around a $\delta^{18}O$ value of −16.7‰ and a $\delta^{13}C$ value of −5.6‰.

The potential for postpedogenic (i.e., diagenetic) modification of carbonate chemistry has been re-

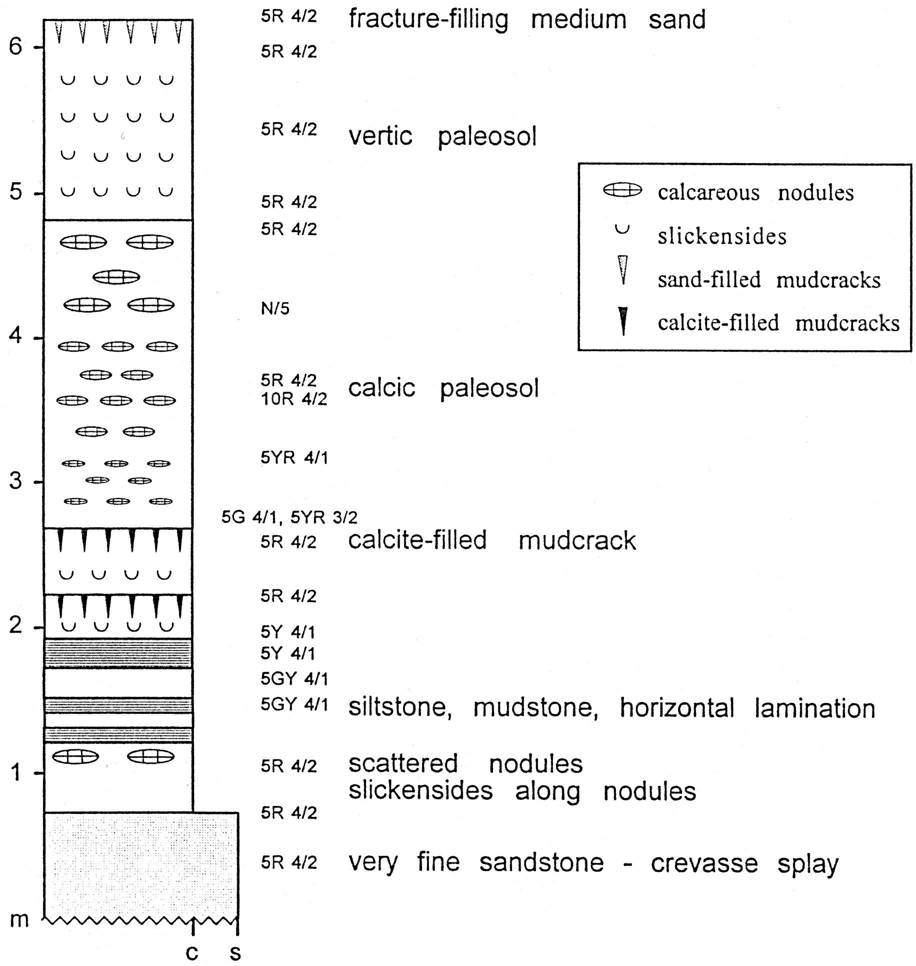

Fig. 2. Typical columnar section of the upper part of the Hasandong Formation at Yusuri, Jinju showing the distribution of pedogenic carbonates: c = clay; s = sand. Color codes are according to Munsell color charts.

garded as a major obstacle to isotopic studies of Mesozoic and Paleozoic paleosols, although diagenesis may significantly shift $\delta^{18}O$ values of calcretes without similarly affecting $\delta^{13}C$ values (cf. Cerling, 1991). Pedogenic carbonate in the Hasandong paleosols does not show the obvious recrystallization fabrics documented in studies of other paleosols (Solomon and Walkden, 1985; Wright and Peeters, 1989; Wright et al., 1993). The dominance of pedogenic micrites and their fabric in the samples indicate that these carbonates have undergone minimal recrystallization and were formed in an active vadose zone.

5.1. Oxygen isotope composition

The $\delta^{18}O$ values of soil-water can be calculated assuming that carbonates were precipitated in isotopic equilibrium with soil water at given temperatures. The expected oxygen isotopic composition of the soil-water in equilibrium with the Hasandong carbonates formed at 25°C ranges from −12.2‰ to −16.0‰ SMOW (excluding one outlier: −3.6‰).

The composition of soil-water associated with the Hasandong pedogenic carbonates can be used to derive the local meteoric water composition if the effect of evaporation is taken into account. In general the composition of pedogenic carbonates is about 2–

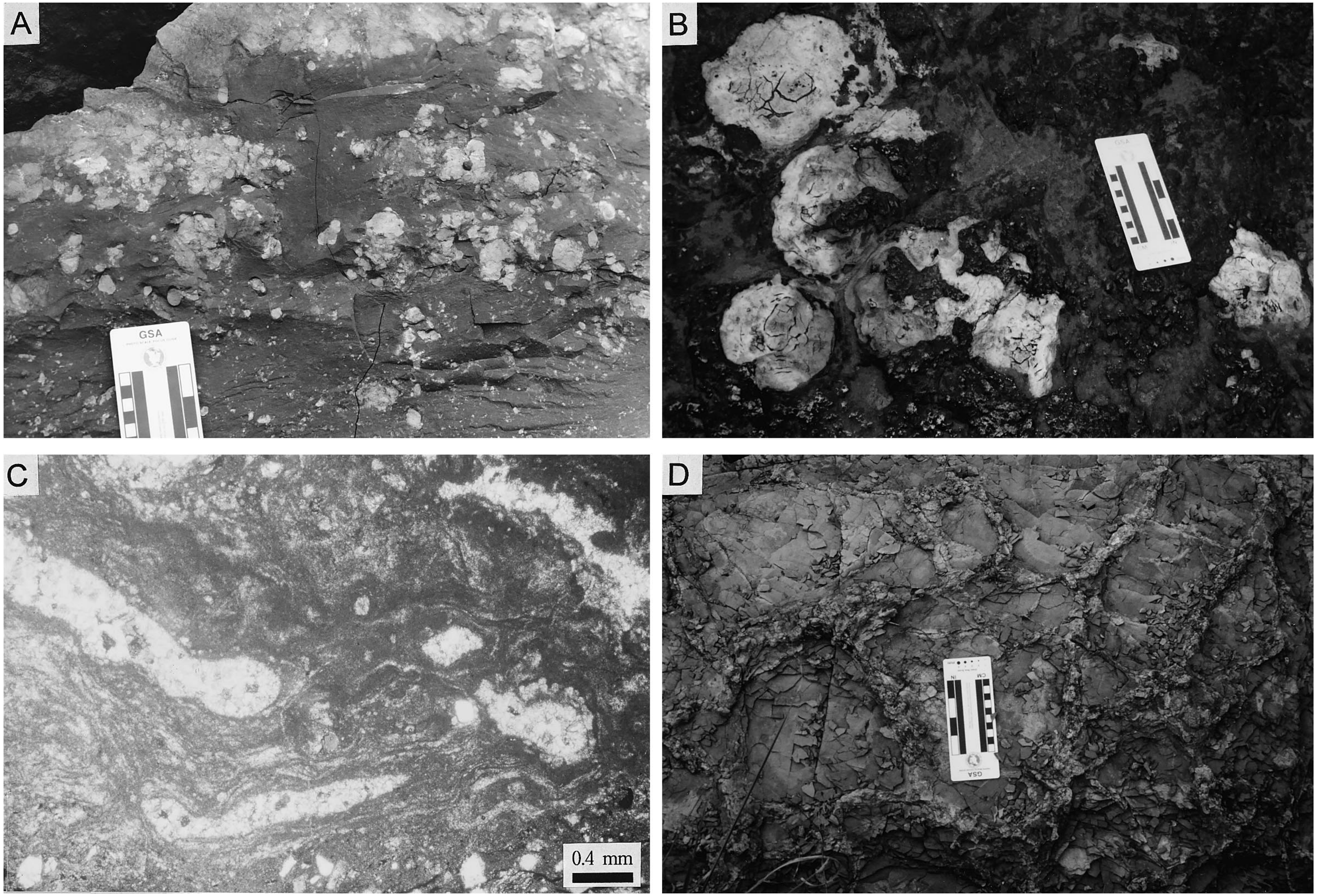

Fig. 3. The Hasandong pedogenic carbonates. Field photographs showing (A) calcic paleosols and (B) septarian crystallaria. (C) Photomicrograph of spar-filled root mold and rootlets. (D) Mud-cracks filled with carbonates in vertic paleosols.

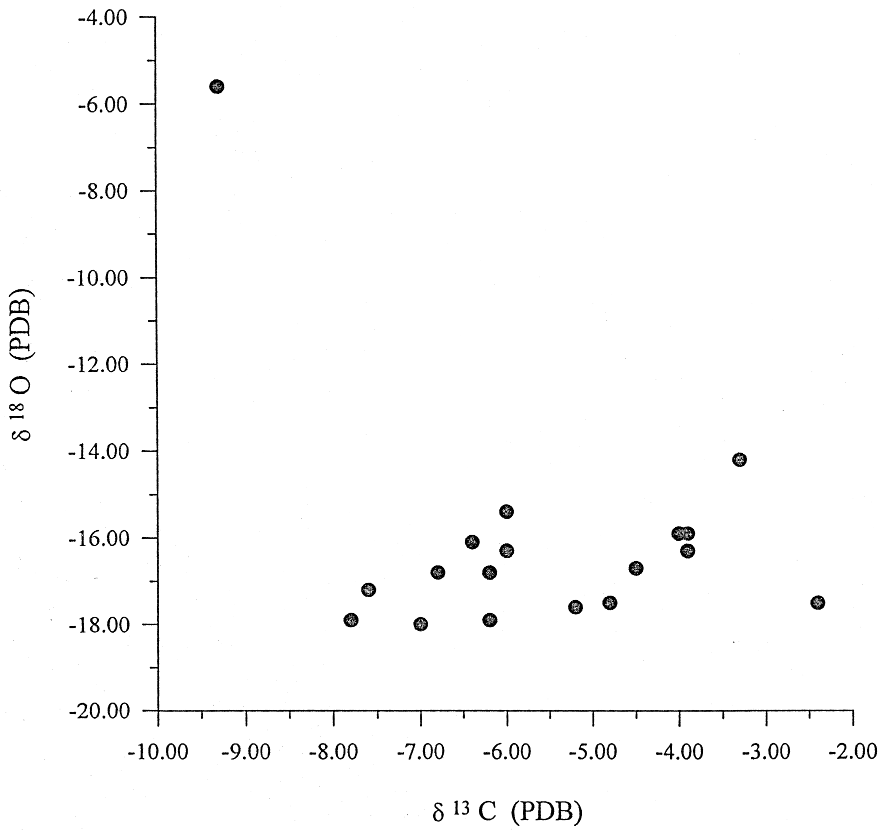

Fig. 4. $\delta^{18}O$ vs. $\delta^{13}C$ stable isotope cross plot for the Hasandong pedogenic carbonates analyzed in this study.

3‰ enriched compared to the expected value based on the present-day meteoric water composition (Salomons et al., 1978). If 2‰ enrichment is assumed, to derive the meteoric water composition one has to subtract 2‰ from the δ-value of the soil-water (Table 1). The Hasandong soil-water composition of −12.2 to −16.0‰ would thus correspond to meteoric water compositions of −14.2 to −18.0‰ with a mean of −16.7‰.

However, these estimates for the Hasandong meteoric water composition are lighter by about 10‰ compared with the estimated δ-values of the Early Cretaceous meteoric waters for the Gyeongsang Basin by Lee (1995). He estimated that the oxygen isotope composition of the Early Cretaceous meteoric water was about −6‰ SMOW. Cerling (1991) reported depletion of similar magnitude in $\delta^{18}O$ values of pedogenic carbonate due to the effects of recrystallization during diagenesis. The enrichment of ^{16}O in the calculated Hasandong meteoric waters may indicate that the Hasandong pedogenic carbonates have undergone recrystallization under elevated temperature conditions even though apparent textural fabrics of recrystallization are not observed. It has been inferred that the Gyeongsang Supergroup experienced paleotemperatures higher than 100°C (Lee, 1995; Son et al., 1994).

5.2. Soil CO_2–atmospheric CO_2 in the Hasandong pedogenic carbonate

The carbon-isotope compositions of pedogenic carbonate can be influenced by many factors, including the type of vegetation, the temperature and depth of carbonate formation, the soil respiration rate, and the mean CO_2 production rate (cf. Cerling, 1991). Fortunately, some of these important factors are constrained in this study; they are paleoclimate (semiarid to arid, seasonally wet–dry precipitation pattern; Paik and Lee, 1995) and parent material (red terrigenous sandy clay derived from adjacent eroding highland regions to the northwest). Thus, it can be assumed that carbon isotopes in soil carbonate reflect variable contributions from atmospheric- and soil-derived CO_2 (plant-CO_2).

The isotopic composition of plant-CO_2 is dependent on the proportion of different types of vegetation since the $\delta^{13}C$ values of the plants are determined by the photosynthetic pathways (Cerling, 1991; Mack et al., 1991). Plants display three distinct carbon isotopic groupings. Virtually all trees, most shrubs and herbs, and cool-season and montane grasses are C_3 plants which have a mean $\delta^{13}C$ value of about −27‰, but display a range from −35‰ to −20‰ (Ehleringer, 1989). C_4 plants include grasses favored by warm growing seasons and a few shrubs in the families Euphorbiaceae and Chenopodiaceae. C_4 grasses range between −6‰ and −19‰, having an average of −13‰ (Smith and Epstein, 1971). CAM (Crassulacean Acid Metabolism) plants include succulents like cactus and some yuccas; they show a natural range of $\delta^{13}C$ from −10‰ to −20‰ (Lerman, 1972). The present carbon isotopic composition of the atmosphere is about −8‰, being depleted relative to the pre-industrial atmosphere which was about −6.5‰ (Friedli et al., 1986). In pre-industrial conditions the average isotopic composition of C_3 and C_4 plants should have been about −26‰ and −12‰, respectively. This is confirmed by the fact that Mesozoic and Cenozoic terrestrial plants have mean $\delta^{13}C$ values of about −26‰ (Popp et al., 1986).

The $\delta^{13}C$ values of Hasandong pedogenic carbonates range from −2.4 to −9.3‰ with an average of $-5.6 \pm 1.7‰\ (1\sigma)$. $\delta^{13}C$ values of Hasandong soil- and plant-CO_2 were estimated by assuming that soil CO_2 is 4.4‰ enriched in ^{13}C relative to soil-respired CO_2 (Cerling, 1984). Estimated $\delta^{13}C$ values of plant-CO_2 for the Hasandong vegetation have a range from −23.9‰ to −16.5‰ (Table 1) and only half of them fall in the range of C_3 type plants. They have a higher average value of −20‰ compared to the expected average of −26‰. This difference can be due to: (1) the presence of C_4 and CAM type flora intermixed with the C_3 type; (2) contributions from atmospheric CO_2.

Although there is a debate on the presence of non-C_3 plants in the Phanerozoic (Wright and Vanstone, 1991; Cerling et al., 1992), it is generally believed that the plants utilizing C_4 or CAM photosynthetic pathways did not evolve until the Miocene (Cerling, 1991; Mack et al., 1991; Morgan et al., 1994). Recently, however, the possible presence of non-C_3 plants in the latest Cretaceous was suggested by Bocherens et al. (1994). Assuming that this case can be extended back to the Early Cretaceous and considering that the total biomass of CAM plants is usually insignificant in most ecosystems except in some of the deserts, the $\delta^{13}C$ values of the Hasandong pedogenic carbonates suggest some possible contribution from C_4 type plants. However, more data on the evolution of C_4 plants are needed to support this interpretation.

Alternatively, the heavy carbon isotope composition of the Hasandong pedogenic carbonates might have been significantly influenced by atmospheric CO_2. The contribution of atmospheric CO_2 can be estimated by assuming that organic carbon is derived from a C_3 flora whose average $\delta^{13}C$ value is −26‰. The results indicate a 25–50% (av. 35%) contribution of atmospheric CO_2 in the Hasandong pedogenic carbonates. This estimation of atmospheric CO_2 contribution is consistent with that of using fig. 3 of Cerling (1984).

Cerling (1984) reported that pedogenic carbonates having a significant atmospheric component above 10% are observed from regions where the soils annually freeze to the depth of carbonate formation. This implies that the Hasandong pedogenic carbonates might have formed during periods of low soil respiration rates. However, during the Early and Middle Cretaceous the intensified greenhouse conditions in the atmosphere were formed due to increased atmospheric CO_2 levels caused by extensive mid-plate basalt volcanism (Schlager et al., 1981; Rampino and Stothers, 1988; Larson, 1991). The resulting general warming and widening of tropical climate belts toward high latitudes might have provided different situations than today, which implies that the high atmospheric component in pedogenic carbonates does not necessarily represent freezing conditions during carbonate precipitation.

In summary, the stable isotopic evidence for Hasandong indicates an ecosystem dominated by C_3 plants, probably a dry woodland.

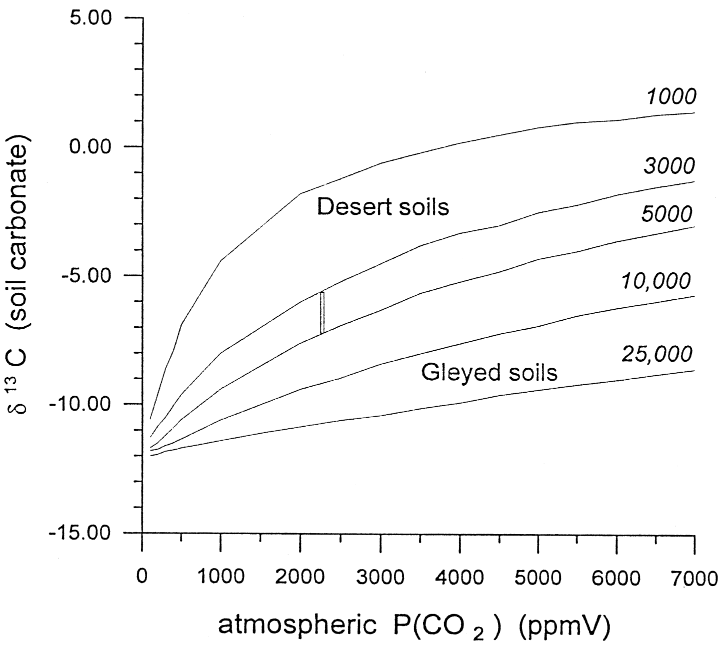

Fig. 5. Atmospheric p_{CO_2} inferred from $\delta^{13}C$ of Hasandong paleosol by using the model of Cerling (1991). Isopleths are values of soil CO_2 minus atmospheric CO_2 in ppmV. Considering the semi-arid climate set-up during the Hasandong period values of 3000–5000 ppmV were used in this study.

5.3. p_{CO_2} partial pressure of the Hasandong atmosphere

Cerling (1991) developed a diffusion-reaction model of estimation of atmospheric p_{CO_2} using the carbon-isotope composition of pedogenic carbonate. It was indicated that the Hasandong soils were developed in a warm and semi-arid climate with seasonal rainfall, and a considerable amount of vegetation was supported by these soils (Paik and Lee, 1995). The p_{CO_2} values of soil-CO_2 and the atmospheric CO_2 usually have a difference of about 3000 ppmV or less in desert soils. Considering the semi-arid climatic set-up during the Hasandong period this difference is expected to lie between 3000 and 5000 ppmV. Taking these values as limits and the average $\delta^{13}C$ value of −5.6‰ for the Hasandong carbonates, fig. 7 of Cerling (1991) can be used to derive about 2300 ppmV for the p_{CO_2} of the Hasandong atmosphere (Fig. 5). The seven-times higher partial pressure of CO_2 in the Hasandong atmosphere compared with the present-day value corroborates the estimates of Berner (1991) of atmospheric p_{CO_2} during the Early Cretaceous. Also, this estimate fits in nicely with the other estimates for the Cretaceous: i.e., 2500–3300 ppmV for the Early Cretaceous Trinity Group, Texas (estimation after Cerling, 1991), 1600–2600 ppmV for the Early Cretaceous Cameros Basin, Spain (estimation after Cerling, 1991) and 800–1200 ppmV for the Late Cretaceous Lameta Formation, India (Ghosh et al., 1995).

6. Conclusions

The Early Cretaceous Hasandong Formation, Gyeongsang Basin, Korea contains well developed paleosols and pedogenic carbonates. The pedogenic carbonates from these paleosols were analyzed for oxygen and carbon isotopic composition to infer the paleoclimatological conditions. The oxygen isotope compositions of the carbonates range from −14.2 to −18.0‰. Assuming the fractionation model and a formation temperature of 25°C in soil, these data indicate that the oxygen isotope compositions of the Hasandong pedogenic carbonates were modified due to the effects of recrystallization during diagenesis.

The carbon isotopic composition of the Hasandong carbonates range from −2.4 to −9.3‰ with an average of −5.6‰. These isotopic compositions suggest a series of soils dominated by a C_3 type of vegetation. The estimated average composition of the vegetation is enriched by about 6‰ compared with the present-day C_3 vegetation. This is probably due to an admixture of atmospheric CO_2 ($\delta^{13}C$ −6.5‰) contributing about 35% of the total CO_2 in the soil. The carbon isotopic composition suggests that the Hasandong paleoecosystem was probably a C_3-dominated, dry woodland. The contribution of atmospheric CO_2 allows one to estimate the partial pressure of CO_2 in the Hasandong atmosphere using the model of Cerling (1991) under certain assumptions. The paleo-p_{CO_2} level of the Hasandong is estimated to be around 2300 ppmV.

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