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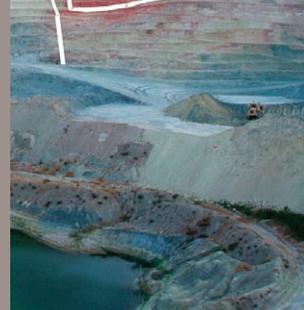
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Geological Aspects and Genesis of Bentonites

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1811-5209/08/0005-0093\$2.50 DOI: 10.2113/gselements.5.2.93



Bentonites are clay rocks consisting predominantly of smectite. They form mainly from alteration of pyroclastic and/or volcanoclastic rocks. Extensive deposits, linked to large eruptions, have formed repeatedly in the past. Bentonite layers are useful for stratigraphic correlation and for interpreting the geodynamic evolution of our planet. Bentonites generally form by diagenetic or hydrothermal alteration, favoured by fluids that leach alkali elements and by high Mg content. Smectite composition is partly controlled by parent rock chemistry. Recent studies have shown that bentonite deposits may display cryptic variations in layer charge – i.e. the variations are not visible at the macroscopic scale – and these correlate with physical properties.

KEYWORDS: bentonite, smectite, glass alteration, stratigraphic correlation, cryptic variation, clay

INTRODUCTION

Bentonites are clay rocks that form from volcanic glass and that consist predominantly of clay minerals of the smectite group. Bentonites are used industrially because they have important physical and chemical properties, including crystal structure, chemical composition, small crystal size (hence large specific surface area), high capacity for ion exchange, variety in the type of exchangeable cations, hydration and swelling behaviour, colloidal properties, dehydration, and reactions with organic and inorganic reagents (Odom 1984). Because of these properties, bentonites are used in a variety of applications, such as drilling, foundry processes, iron ore pelletization, civil engineering, adsorbents, filtering, decolourizing and clarifying. The most recently developed application is the production of nanocomposites.

The purpose of this article is to describe the formation of bentonite. Some of the questions we aim to answer are: What controls smectite formation during alteration of volcanic glass? Are there variations in bentonite that are invisible at the macroscopic scale (i.e. cryptic variations)? How can smectite-rich bentonites form from rocks of unfavourable composition? Does parent rock composition control smectite composition? Did bentonites form in previous geological epochs and, if so, how can they be used for stratigraphic correlation?

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PARAMETERS CONTROLLING BENTONITE FORMATION

Mechanisms

The alteration of volcanic pyroclastic rocks to smectite is a common process, often producing bentonite deposits of commercial value. Evidence for the volcanic nature of bentonite precursors is the presence of primary igneous minerals (β -quartz, biotite, sanidine, zircon, apatite, ilmenite, magnetite); the presence of glass shards that may be fresh, partially altered or pseudomorphically replaced by

smectite; and the distribution pattern of characteristic trace components such as the rare earth elements (REE). Alteration of glass may occur through vapour-phase crystallization often associated with welding of ignimbrites, burial diagenesis, contact metamorphism, hydrothermal activity, and by hydrolysis either in alkaline lakes and in marine sediments or by percolating groundwater. Vapour-phase crystallization is not an important process in bentonite formation. Bentonites form as a result of the alteration of glass by a fluid phase, so they form in aqueous environments, usually in shallow seas or lakes. However, alteration of volcanic glass to smectites is frequently incomplete and fresh glass shards may be preserved even in old volcano-sedimentary strata (Weaver 1989). In fact, most commercial bentonite deposits contain fresh glass shards.

Economic deposits of bentonite may be formed by any of three mechanisms: (1) diagenetic alteration of volcanic glass, (2) hydrothermal alteration of volcanic glass, and (3) formation of smectite-rich sediments in salt lakes and sabkha environments usually from dissolution of detrital smectites, which is often associated with sepiolite and/or palygorskite. This last process does not necessarily require pyroclastic or volcanoclastic precursors and is observed in arid climates. Sedimentary deposits contain trioctahedral smectites (saponite and stevensite) in small amounts, so their quality is inferior.

Regardless of the formation process, leaching of alkali elements and high $(\text{Mg}^{2+})/(\text{H}^+)$ are required to form smectites instead of zeolites during the alteration of volcanic glass (Senkayi et al. 1984; Christidis 1998). Magnesium is often supplied by the fluid phase, especially when the parent rock is acidic (Si-rich). Mass balance calculations show that the large water to rock ratios of an open system are necessary to form smectites, regardless of the parent rock (Christidis 1998). High water to rock ratios are facilitated by high permeability. Low permeability, thus low water to rock ratio, favours zeolites. Smectites have been

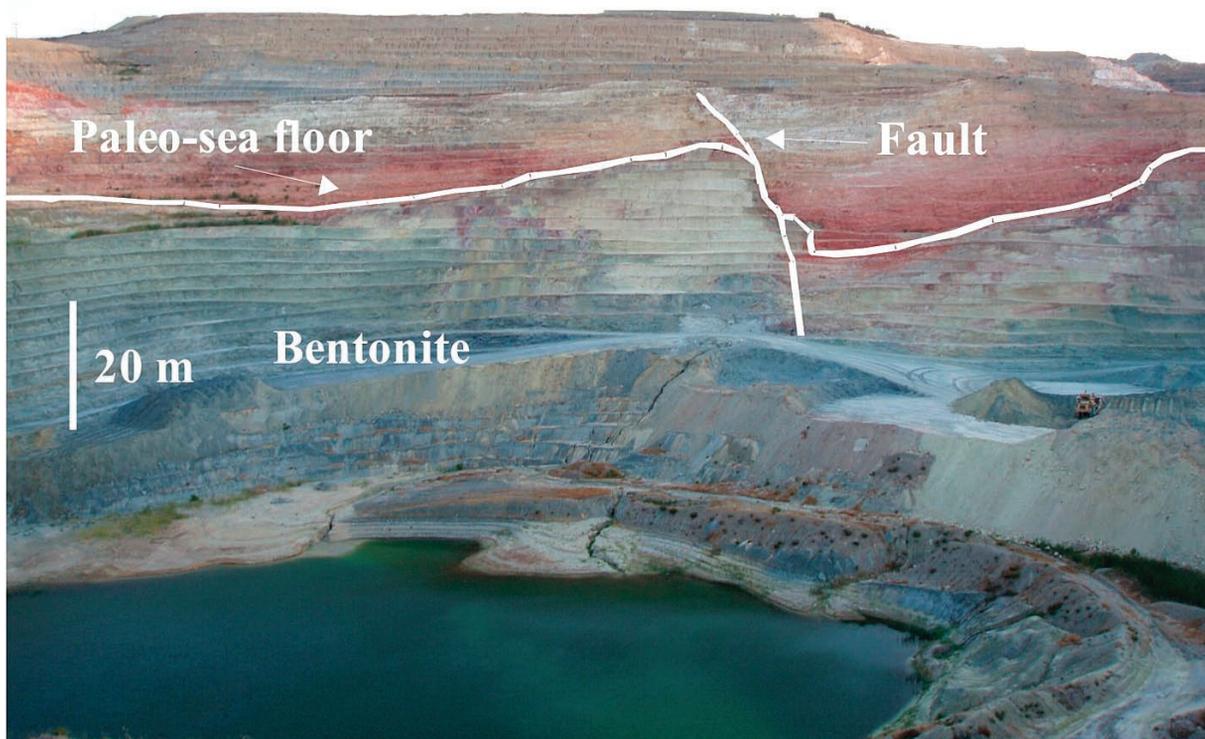


FIGURE 1 Bentonite deposit formed from pyroclastic flows, Milos Island, Greece. Deposition and alteration of the parent rocks took place in a marine environment. Individual flows can be identified by variation in colour. Red marl overlies the bentonite. The deposit is ~60 m thick but its lateral extent is limited by the dimensions of the pyroclastic flows.

reported to form from poorly crystalline precursors, usually gels, in a variety of environments (Zhou and Fyfe 1989; Christidis 2001), but such a precursor phase is not always essential.

Diagenetic alteration of volcanic glass shards deposited in aqueous environments yields thin beds a few centimetres to a few metres thick and extending over large areas (Elzea and Murray 1994). For this reason, bentonite layers that formed in previous epochs are useful for stratigraphic correlation (Huff et al. 1999). Parent rocks are usually volcanic ash deposits derived from highly explosive eruptions (i.e. subplinian or plinian) or transported as volcanoclastic sediments. Bed thickness, the particle size of the glass shards and the relict igneous minerals depend on the type of volcanic eruption. Diagenetic alteration of volcanic glass to bentonite is facilitated by convective flow, which is sustained by hydraulic gradients and controlled by permeability. Temperature gradients are not considered important for supporting convective flow because thermal equilibrium is established shortly after deposition of the volcanic fragments. This is because the volcanic ejecta travel long distances and cool before deposition. Typical examples of well-known diagenetic bentonites are the deposits in Wyoming, Montana and Texas (USA), the fuller's earth in England, and the bentonites in Bavaria, Germany, etc.

Hydrothermal alteration is another important process for bentonite formation. It also includes deuteric alteration caused by gases and vapours after emplacement of pyroclastic rocks (Grim and Güven 1978). Two types of hydrothermal alteration are considered: (1) alteration where the energy source for heating the water is located at depth and associated with a magmatic body and (2) alteration where the energy source is the cooling pyroclastic rock itself. In the first case, alteration occurs via fluid flow through fracture zones or faults, forming elongated or elliptical bentonite bodies that are often zoned parallel to the axis of the structural feature (Ddani et al. 2005; Yildiz and Kuşcu 2007). In the second case, cooling of the parent rock sustains a hydrothermal system, driving alteration. The hydrothermal system is active as long as the temperature

difference between the cooling rock and the aqueous phase (seawater or fresh water) can maintain fluid flow. In other words, this type of alteration is favoured by high temperatures during emplacement, such as is common in pyroclastic flows (300 to 800°C according to Cas and Wright 1988). Most of the bentonite deposits of Milos Island, Greece, are derived from the alteration of pyroclastic flows. They show some important differences compared with their diagenetic and hydrothermal (magmatic heat) counterparts. They are stratiform, as are the diagenetic bentonites, but they are much thicker and are spread over distances limited by the dimensions of the pyroclastic flow (Fig. 1). Such a deposit may exceed 60 m in thickness in a series of successive flows.

Formation of Bentonites from Si-Rich Rocks

It is curious that high-quality, smectite-rich bentonites form from rocks of unfavourable composition. In fact, although dioctahedral smectites form by alteration of nearly all types of volcanic rocks, trachyandesites are the most suitable precursors (Grim and Güven 1978). Acidic rocks are not suitable, because their high SiO_2 to Al_2O_3 ratios favour SiO_2 polymorphs such as opal-CT (i.e. composed of cristobalite and/or tridymite) and alkaline zeolites (heulandite-type zeolites and mordenite). However, high-quality, opal-CT-free bentonites formed from acidic precursors have also been observed (Christidis 1998; Berry 1999; Ddani et al. 2005). On the other hand, trachyandesitic or even basaltic-andesitic rocks may also yield bentonites with abundant opal-CT that is at least partly biogenic (Christidis 2006).

Answers to questions about how bentonites form from rocks of unfavourable composition can be drawn from experiments designed to simulate volcanic glass alteration in closed or quasi-open systems. Ca, Mg and Si are mobile elements (Shiraki et al. 1987; Daux et al. 1997; Fiore et al. 1999). In experiments simulating open systems, the amount of available Si depends on the fluid flow rate, decreasing with increasing flow rate, and on the accumulation of Si in the fluid phase; the precipitation rate of free SiO₂ decreases with increasing flow rate (Daux et al. 1997). In contrast, Al leaching is not significant. This raises a question about the parameters controlling the rate of fluid flow during bentonite formation. The answer can be traced to the processes that form bentonites. Alteration of permeable, hot, pyroclastic flows sustains water circulation and convective flow. As long as the temperature gradients within the volcanic rock and between the volcanic rock and the bulk fluid can maintain convective flow, the system remains active. The temperature gradients control (1) the order of formation of the neoformed minerals (smectite, opal-CT, zeolites, authigenic feldspar) and (2) the distribution of layer charge and charge heterogeneity of smectites within the bentonite, as is detailed in the next section.

At the onset of alteration, temperature gradients between the volcanic rock and the fluid phase are at their highest, maximizing fluid flow and leaching Si relative to Al. Opal-CT and zeolites are not favoured because Si and alkali elements leached from the volcanic glass are removed. Hence formation of high-quality bentonites from acidic rocks is likely when high temperature gradients are maintained for an adequate time, keeping (Mg²⁺)/(H⁺) high and H₄SiO₄ activity low in the pore fluids. In hydrothermal systems characterized by high rates of seawater flow through fracture zones and faults, these conditions are rather common. Pyroclastic rocks deposited at low temperatures cannot sustain high flow rates for a long time, so fluids reach saturation, amorphous SiO₂ precipitates, and converts to opal-CT. Similar processes are expected in hydrothermal systems with moderate flow rates. In all systems, the rate of smectite crystallization versus the rate of amorphous SiO₂ precipitation determines the final mineral assemblage. Finally, during diagenesis, high-grade bentonites are produced if fluid flow is maintained.

CRYPTIC VARIATIONS IN BENTONITE DEPOSITS

Cryptic variations are not visible at the macroscopic scale. Smectites in bentonites derived from rocks whose composition varies from rhyolite to basaltic andesite vary considerably in composition at the micrometre scale (Christidis 2006 and references therein); it is also well known that the physical properties of bentonites can vary considerably within a single deposit (Odom 1984). Compositional heterogeneity has been attributed to heterogeneity of the microenvironment during their formation (Christidis 2006 and references therein), but until recently, evidence of systematic compositional variability was lacking. Two recent reports demonstrate cryptic variation of smectites in bentonites. Variation that is not visible macroscopically can be seen in microscopic trends in composition and, more specifically, layer charge. Data from a 1 m thick bentonite bed in Charente, France (Meunier et al. 2004) show that (Al₂O₃+Fe₂O₃)/MgO in smectite decreases from the centre towards the margin of the deposit, whereas the cation exchange capacity (CEC) increases (Fig. 2). Because layer charge is derived mainly from substitution in the octahedral sheet (Meunier et al. 2004), the observed compositional trend also suggests that layer charge increases toward the margins.

A similar, clearer trend was observed by Christidis and Makri (2007) in a bentonite from Milos, Aegean, Greece (Fig. 3). The layer charge increases towards the top of the deposit, although the parent rock was different from the previous case. In Charente, the bentonite formed from a thin ash fall, whereas in Milos, a thick pyroclastic flow was the parent rock. Also there is a pronounced upward increase in tetrahedral charge, i.e. an increase in the beidellite component, in the Milos deposit. Neither deposit shows a systematic change in macroscopic characteristics (colour, rock texture) that would reflect a systematic change in composition. Future studies may clarify layer charge distribution and other such cryptic variations. This information is expected to be useful for interpreting the variability of physical properties and the controls on the genesis of the deposits. Clearer understanding of local variability is also expected to contribute to the optimization of routine industrial processes such as the exchange of Na for Ca and/or Mg in Ca- and/or Mg-bentonites, a process known as Na activation.

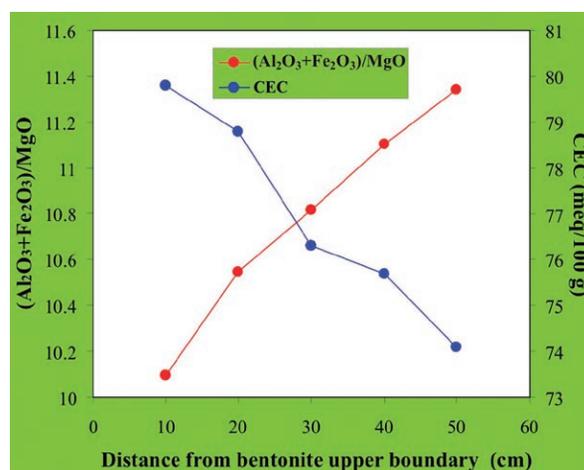


FIGURE 2 Variation of cation exchange capacity (CEC) (milliequivalents/100g) and the (Al₂O₃+Fe₂O₃)/MgO ratio with depth in smectites from a bentonite deposit in Charente, France. The CEC increases as the (Al₂O₃+Fe₂O₃)/MgO ratio decreases toward the upper boundary, suggesting increasing smectite layer charge from the centre of the bentonite bed to the top. DATA FROM MEUNIER ET AL. (2004)

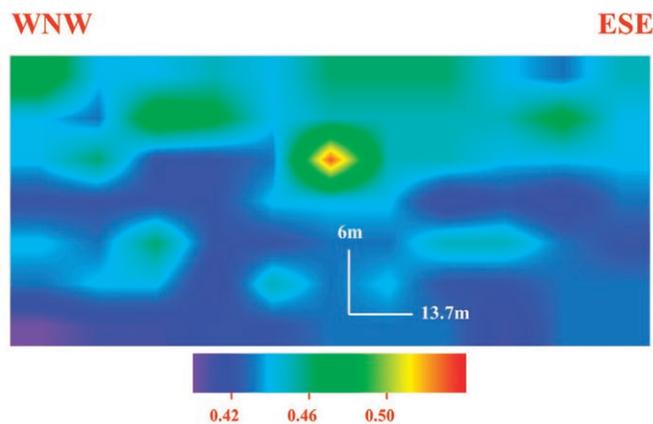


FIGURE 3 Cross-section of a bentonite profile (21 m × 120 m) from a deposit in Eastern Milos, Greece, showing evolution of smectite layer charge (the colour scale indicates charge equivalents per half unit cell). Smectites have layer charge between 0.3 and 0.6 charge equivalents, increasing towards the top of the profile. Smectite layer charge is defined as low for less than 0.425 equivalents per half unit cell (blue), intermediate between 0.425 and 0.47 (blue to green) and high for more than 0.47 equivalents (green to red).

DOES THE PARENT ROCK CONTROL SMECTITE COMPOSITION?

Composition in the microenvironment is an important parameter controlling the composition of the smectite, but the composition of the parent rock is also important. However, this relationship has been addressed in only a few studies of natural systems (Christidis 1998, 2006). Published data on the composition of parent rocks, which are characterized as acidic, intermediate or basic on the basis of their geochemical affinities, show that smectites can be distinguished by their average iron and, to a lesser extent, by their average Si contents. Smectites derived from the alteration of basic rocks have, on average, 0.63 Fe³⁺ atoms per half unit cell (phuc), whereas those derived from intermediate and acidic rocks have, on average, 0.21 and 0.12 Fe³⁺ atoms phuc, respectively. Smectites derived from altered acidic rocks have, on average, 3.92 Si atoms phuc, whereas those derived from intermediate and basic rocks have 3.82 and 3.73 Si atoms phuc. Hence smectites derived from altered basic volcanic rocks are mainly Fe-montmorillonites and Fe-beidellites.

FIGURE 4 shows the results of a multivariate statistical analysis of smectites derived from a variety of parent rocks using the canonical discriminant analysis method (Christidis 2008). The chemical composition of 91% of the smectites studied correlated with parent rock composition. This means that smectites derived from precursors with acidic to basic geochemical affinities may have different crystal compositions. The model can also be used to estimate parent rock composition from the composition of the smectite. Hence, using the canonical discriminant analysis, a score (value) can be obtained for any smectite derived from volcanic rocks of unknown composition according to its crystal composition. From this score, the nature of the parent rock may be estimated. The main reason for incorrect allocation of a limited number of smectites in FIGURE 4 is the uncertainty in the geochemical affinities of the precursor volcanic rocks. This is because, with a few exceptions, only a broad characterization of the geochemical affinities of the parent rocks was available in the data set used. Moreover, in several commercial bentonites, contribution from different volcanic sources has been proven (Christidis et al. 1995; Bertog et al. 2007). A larger data set for smectites derived from known precursors will allow for verification and refinement of the model and substantiate the validity of canonical discriminant analysis for estimating the parent rock composition of bentonites.

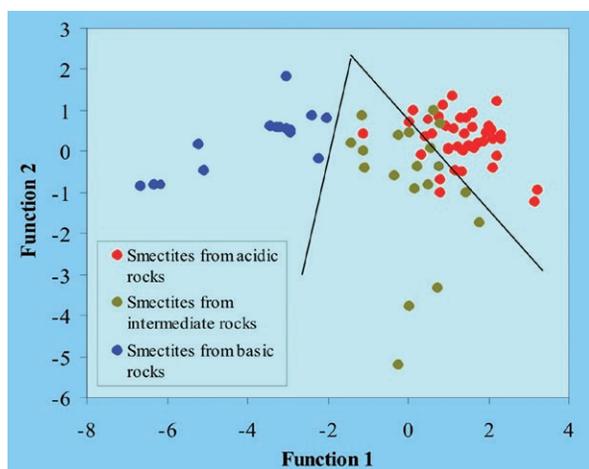


FIGURE 4 Canonical discriminant analysis (straight lines) of the data set consisting of 75 smectites in bentonites derived from basic, intermediate and acidic precursors. Using two canonical discriminant functions, smectites can be separated into three distinct groups (coloured dots) according to the composition of the precursor rocks. Sixty-eight out of the 75 smectites (91%) were allocated correctly, according to the analysis. REPRODUCED WITH PERMISSION FROM THE MINERALOGICAL SOCIETY

BENTONITES OF OLDER GEOLOGICAL EPOCHS

Ordovician K-Bentonites: Examples from the Geologic Record

As with every Phanerozoic system, Ordovician successions contain a number of so-called K-bentonites, which represent episodes of explosive volcanism. K-bentonites begin as smectite rich, but they gradually convert to interstratified illite-smectite clay-rich beds over time. This reduces their value for industrial applications, and Paleozoic bentonites are generally not commercially mined. However, they do have enormous value as stratigraphic marker horizons and as remnants of large caldera-forming tectonic events. FIGURE 5 shows the general global stratigraphic and geographic distribution of documented beds for the Ordovician. Numerous beds have been reported from North and South America, Asia and Europe. These beds are not uniformly distributed through time, a reflection of both the episodic nature of explosive volcanism and the periodic nature of suitable ash-preservation conditions. The Ordovician successions of North America are known to contain nearly 100 K-bentonite beds, several of which are distributed over areas as large as 1.5×10^6 km² (Kolata et al. 1996). Approximately 150 beds are recorded in the Baltic region of northern Europe (Bergström et al. 1995), and several hundred beds are known in South America and China. Two prominent K-bentonites occur throughout the eastern and central United States: the Millbrig K-bentonite or T-4 bed of Wilson (1949) and the Deicke K-bentonite or T-3 bed (Wilson 1949). Using geophysical logs and chemical fingerprinting techniques with field mapping, Huff and Kolata (1990) and Kolata et al. (1996) correlated the Deicke and Millbrig throughout the eastern midcontinent region of the United States. FIGURE 6 shows examples of what these beds look like in outcrop and drill core sections.

In northern Europe, one especially prominent ash bed, the Kinnekulle K-bentonite, has been identified across Scandinavia and the Baltic states and locally in Great Britain (Bergström et al. 1995). Known occurrences of the Kinnekulle bed extend from the Oslo area in Norway, across approximately 900 km in southern Sweden and northern Denmark, to eastern Estonia and most likely into western Russia. The Millbrig and Kinnekulle beds have similar mineral and bulk chemical compositions and appear to have formed at the same time, which led Huff et al. (1992) to propose they were derived from the same gigantic eruption.

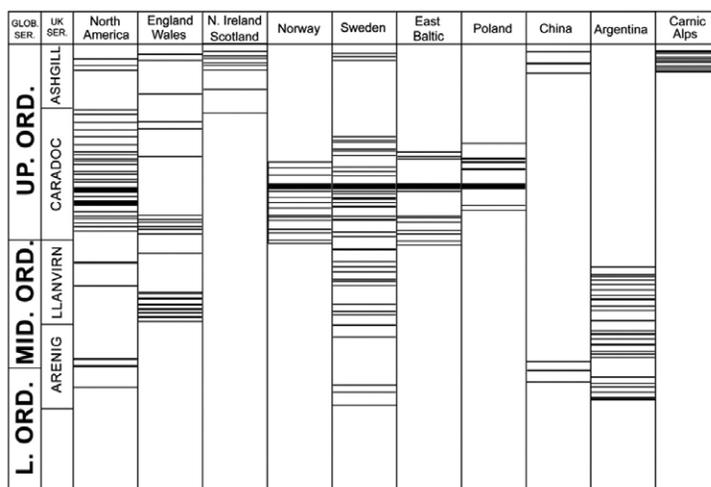


FIGURE 5 Global distribution of Ordovician K-bentonites. The thicker bands represent the Deicke and Millbrig K-bentonites in North America and the Kinnekulle K-bentonite in northern Europe.

The Products of Supervolcanoes?

The Millbrig covers an estimated area of at least $2.2 \times 10^6 \text{ km}^2$ in North America, and the Kinnekulle at least $6.9 \times 10^5 \text{ km}^2$ in northwestern Europe. The volume of silicic magma that produced them is estimated to have been 1509 km^3 for the Millbrig and 972 km^3 for the Kinnekulle (Huff et al. 1996). These values do not include the vast quantity of volcanic ash presumably deposited in the Iapetus Ocean between Baltica and Laurentia and subsequently subducted. The truly enormous size of these ash falls is made clear when one considers that the volume of the 1980 Mount St. Helens eruption is estimated at 0.2 km^3 (Sarna-Wojcicki et al. 1981). These huge Ordovician ash falls are among the largest recorded in the Earth's Phanerozoic history, easily classifying them as supervolcano events. A supervolcano is defined as a volcano that ejects more than a trillion tons of material. Supervolcanoes can cover an entire continent in ash. The explosion of a supervolcano is about 30 times stronger than the largest volcanic eruption in recorded human history, which occurred at Krakatau, Indonesia, in 1883. The last time a supervolcano exploded was at Toba in Sumatra around 74,000 years ago. Ten thousand times bigger than Mount St. Helens, the Toba event created a global catastrophe dramatically affecting life on Earth. Ash blotted out the Sun's rays, and humanity came close to extinction. Anthropologists estimate that only about 5000 humans survived to reproduce in the aftermath of this event. Another famous supervolcano is located directly underneath Yellowstone National Park, in the United States, and is so large that it is visible from space. FIGURE 7 compares the measured volumes of some of the largest historic and prehistoric volcanic eruptions known. The *Elements* "Supervolcanoes" issue and particularly Miller and Wark (2008) provide more details about supervolcanoes.

Large-magnitude events tend to occur with lower frequency than smaller-magnitude eruptions, but they are more likely to result in ash beds that are preserved in the stratigraphic record because of their aerial extent and the volume of material. The Millbrig-Kinnekulle supereruption appears to have been what is also called an ultraplinian event, in reference to Pliny the Younger who wrote about the famous eruption of Mt. Vesuvius in 79 AD (Walker 1980). The largest previously documented events had eruption columns $>50 \text{ km}$ high, discharge rates of $\sim 106 \text{ m}^3/\text{s}$ (Walker 1980) and estimated eruption velocities at the vent on the order of 500 m/s . Low- to medium-magnitude eruptions have dispersal patterns dominated by the prevailing tropospheric wind currents, but some ultraplinian eruptions have mushroom-shaped dispersal patterns that may override prevailing wind directions (Carey and Sparks 1986). We interpret the Millbrig-Kinnekulle eruption to have been of this type.

Do Such Events Affect the Climate?

Can bentonite-forming events significantly impact global climate? Of the various gases that make up any volcanic eruption, sulfur dioxide (SO_2) causes the strongest effect on the environment. It reacts with oxygen and water to produce tiny droplets of sulfuric acid (H_2SO_4). By blocking sunlight, these droplets are the main cause of the dramatic climatic cooling that would grip the planet in the wake of a supereruption. In 1996, investigators studying ice cores from Greenland and Antarctica found evidence for the sulfuric acid peak that followed the supereruption at Toba (Zielinski et al. 1996), which ejected nearly 3000 km^3 of lava and ash and reduced average global temperatures by an estimated 5 to 15°C . Based on their magnitude, it is expected that the supereruptions that produced the Ordovician bentonites had an even greater impact on the global climate.

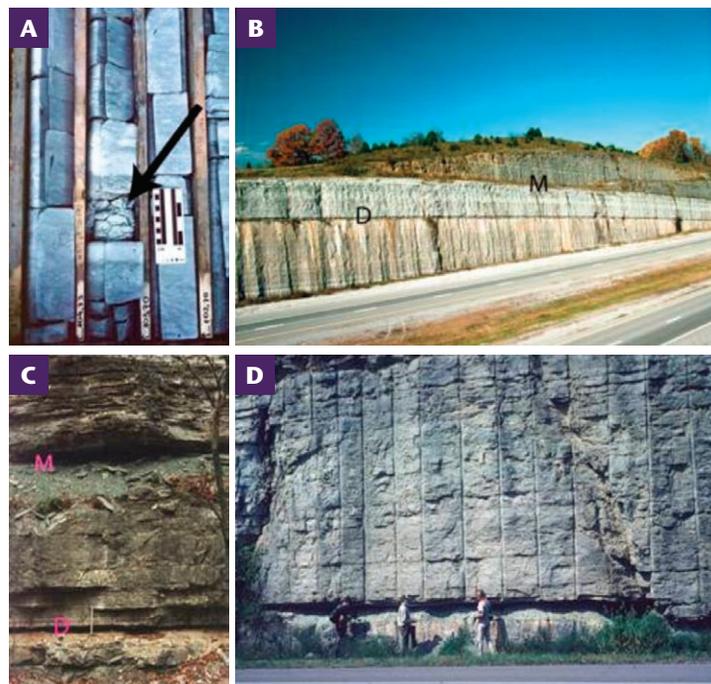


FIGURE 6 What do K-bentonites look like? Here are some examples. (A) Drill core showing a K-bentonite bed (arrow) in a carbonate section; (B) a road cut near Gladeville, Tennessee, showing the centimetre-thick Deicke (D) and Millbrig (M) K-bentonites separated by about 4 m of limestone; (C) the Deicke (D) and Millbrig (M) K-bentonites exposed near Minke Hollow, Missouri; (D) the Deicke K-bentonite in a road cut near Carthage, Tennessee

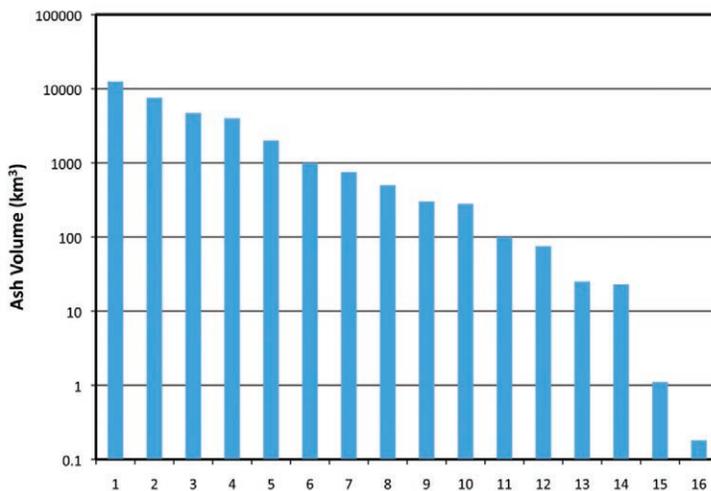


FIGURE 7 A comparison of ash volumes of some of the largest-known historic and prehistoric volcanic eruptions: (1) Millbrig-Kinnekulle (454 Ma), (2) Millbrig K-bentonite (454 Ma), (3) Deicke K-bentonite (456 Ma), (4) Toba (74,000 years), (5) Yellowstone – Huckleberry Ridge (2 Ma), (6) Yellowstone – Lava Creek (650,000 years), (7) Los Chocoyos (85,000 years), (8) Bishop (730,000 years), (9) Upper Bandelier (1 Ma), (10) Mesa Falls (1.3 Ma), (11) Tambora (1815), (12) Campanian (33,000 years), (13) Shikotsu (1909), (14) Taupo (25,000 years), (15) Mt. St. Helens (1980), (16) Hekla (1104)

CONCLUSIONS

Bentonites have formed throughout Earth's history, because explosive volcanic activity has played an important role in the evolution of our planet. However, only those formed after the Jurassic, and especially those in the Cenozoic, have economic importance. Nevertheless, older bentonites, in which smectite has converted to mixed-layer illite-smectite, are important stratigraphic markers used for

correlation purposes. The volcanic events that yielded major bentonite deposits are linked with supereruptions, which affected the global climate.

ACKNOWLEDGMENTS

G. Christidis thanks the Greek State Scholarship Foundation (SSF), the Fulbright Institution and the Greek Secretariat for Research and Development (GSRT) (contract no. 03–209)

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ADVANCED MINERAL TECHNOLOGIES

We have a large deposit (40+ million tons drilled and blocked out) of volcanic clay in southern Oregon classified as rectorite. This unique, ordered illite-smectite deposit is the largest reserve known. We are pursuing federal funds to develop the reserves and seek to identify possible future partners and customers.

Rectorite has potential uses in radioactive waste containment, as heat-resistant drilling mud, and in water treatment. Rectorite from this deposit has been shown to kill bacteria (in vitro) harmful to humans, such as *MRSA (S. aureus)*, *E.coli* and *Salmonella*.

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